



Low-cost adsorbents for heavy metals uptake from contaminated water: a review

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Abstract

In this article, the technical feasibility of various low-cost adsorbents for heavy metal removal from contaminated water has been reviewed. Instead of using commercial activated carbon, researchers have worked on inexpensive materials, such as chitosan, zeolites, and other adsorbents, which have high adsorption capacity and are locally available. The results of their removal performance are compared to that of activated carbon and are presented in this study. It is evident from our literature survey of about 100 papers that low-cost adsorbents have demonstrated outstanding removal capabilities for certain metal ions as compared to activated carbon. Adsorbents that stand out for high adsorption capacities are chitosan (815, 273, 250 mg/g of Hg^{2+} , Cr^{6+} , and Cd^{2+} , respectively), zeolites (175 and 137 mg/g of Pb^{2+} and Cd^{2+} , respectively), waste slurry (1030, 560, 540 mg/g of Pb^{2+} , Hg^{2+} , and Cr^{6+} , respectively), and lignin (1865 mg/g of Pb^{2+}). These adsorbents are suitable for inorganic effluent treatment containing the metal ions mentioned previously. It is important to note that the adsorption capacities of the adsorbents presented in this paper vary, depending on the characteristics of the individual adsorbent, the extent of chemical modifications, and the concentration of adsorbate.

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1. Introduction

Since its first introduction for heavy metal removal, activated carbon has undoubtedly been the most popular and widely used adsorbent in wastewater treatment applications throughout the world. In spite of its prolific use, activated carbon remains an expensive

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material since higher the quality of activated carbon, the greater its cost. Activated carbon also requires complexing agents to improve its removal performance for inorganic matters. Therefore, this situation makes it no longer attractive to be widely used in small-scale industries because of cost inefficiency.

Due to the problems mentioned previously, research interest into the production of alternative adsorbents to replace the costly activated carbon has intensified in recent years. Attention has been focused on the various adsorbents, which have metal-binding capacities and are able to remove unwanted heavy metals from contaminated water at low cost. Because of their low cost and local availability, natural materials such as chitosan, zeolites, clay, or certain waste products from industrial operations such as fly ash, coal, and oxides are classified as low-cost adsorbents.

Chitosan has received considerable interest for heavy metals removal due to its excellent metal-binding capacities and low cost as compared to activated carbon. In Asian countries such as Thailand, Japan, and China, fishery wastes such as shrimp, lobster, and crab shells have been developed into one of the promising options to produce chitosan. These wastes could be obtained for free from local fishery industries. Since such wastes are abundantly available, chitosan may be produced at low cost. Consequently, chitosan offers a lot of promising benefits for wastewater treatment applications today.

Natural zeolites also gained a significant interest among scientist, mainly due to their valuable properties such as ion exchange capability. Large deposits of natural zeolites in many countries such as Greece, UK, Italy, Mexico, Iran, and Jordan, provide local industries some promising benefits such as cost efficiency since they are able to treat wastewater contaminated with heavy metal at low cost.

Clay is one of potential alternatives to activated carbon as well. Similar to zeolites, clay minerals are also important inorganic components in soil. Their sorption capabilities come from their high surface area and exchange capacities. The negative charge on the structure of clay minerals gives clay the capability to attract metal ions. The USA and the former Republics of Soviet Union such as Lithuania, Georgia, and Kazakhstan are well known for their large deposits of natural clay minerals.

Industrial waste is also one of the potentially low-cost adsorbent for heavy metal removal. It requires little processing to increase its sorptive capacity. Generally industrial wastes are generated as by-products. Since these materials are locally available in large quantities, they are inexpensive. In India, various types of industrial wastes such as waste slurry, lignin, iron(III) hydroxide, and red mud, have been explored for their technical feasibility to remove heavy metals from contaminated water.

Low rank coal, such as lignite, is capable of having ion exchange with heavy metals due to its carboxylic acid and phenolic hydroxyl functional groups. These materials exist as large deposits in many countries, notably Australia and India.

Other low-cost adsorbents, such as agricultural wastes, have been studied less extensively due to their local availability. Although many research works have been done recently to find the potential of using various alternative adsorbents, so far no efforts have been made to obtain a comparative overview of all adsorbents mentioned previously in terms of their removal performance, adsorption capacity, and cost effectiveness.

An overview of some low-cost adsorbents based on recent publications is presented in this paper and their removal performance is compared. Adsorbents that stand out for

high removal efficiencies and adsorption capacities are compared with the activated carbon.

2. Low-cost adsorbents and removal of heavy metals

2.1. Chitosan

Among various biosorbents, chitin is the second most abundant natural biopolymers after cellulose. However, more important than chitin is chitosan, which has a molecular structure similar to cellulose. Presently, chitosan is attracting an increasing amount of research interest, as it is an effective scavenger for heavy metals.

Chitosan is produced by alkaline *N*-deacetylation of chitin, which is widely found in the exoskeleton of shellfish and crustaceans. It was estimated that chitosan could be produced from fish and crustaceans at a market price of US\$ 15.43/kg [1]. The growing need for new sources of low-cost adsorbent, the increased problems of waste disposal, the increasing cost of synthetic resins undoubtedly make chitosan one of the most attractive materials for wastewater treatment.

Various researches on chitosan have been done in recent years. In 1988, the utilization of chitosan for cadmium removal was intensively investigated [2]. It was demonstrated that an adsorption capacity of 5.93 mg of Cd²⁺/g of chitosan was achieved at a pH range of 4.0–8.3 and that the presence of ethylene diamine tetra acetic acid (EDTA) significantly decreased the cadmium removal by chitosan since EDTA, a stronger chelating agent than chitosan, suppressed the metal uptake by chitosan. It was also reported that in the presence of EDTA, the affinity of Cd²⁺ for the amino groups was drastically reduced since the EDTA masked the presence of Cd²⁺ in aqueous solution, causing their removal from the solution to become difficult.

Another similar research evaluated the sorption of some metal ions onto chitosan [3]. It was found that the maximum adsorption capacities of chitosan for Hg²⁺, Cu²⁺, Ni²⁺, and Zn²⁺ were 815, 222, 164, and 75 mg/g, respectively. However, the result of mercury removal was different from that obtained in the latter study [4], which indicated that an adsorption capacity of 430 mg of Hg²⁺/g was achieved by chitosan. This difference occurs due to the fact that the latter study used chitosan, with particle size ranging from 1.25 to 2.5 mm (against 0.21–1 mm in the former study [3]). Such reduction in particle size of chitosan, of course, increases its surface area to be adsorbed by the metal ions and it results in higher removal efficiency of heavy metal. Therefore, the extent of surface occupancy by adsorbate has a decisive influence upon the removal efficiency of chitosan.

The interaction between chitosan and hexavalent chromium was intensively investigated [5]. It was observed that an adsorption capacity of 273 mg of Cr⁶⁺/g chitosan was achieved at pH of 4.0. In 1996, a comparative study on the adsorption capacity of chitosan for various metal ions such Cu²⁺, Cd²⁺, Ni²⁺, Pb²⁺, and Hg²⁺ was conducted [6]. It was found that chitosan exhibits the highest binding capacity for Hg²⁺ (Table 1).

The adsorption of copper on chitosan was also studied and it was found that 1 g chitosan could adsorb 4.7 mg of Cu²⁺ at pH of 6.2 [7]. This result is significantly different from that obtained in a previous study [8]. It was reported that pH of 5.5 was found to be

Table 1
Adsorption capacities (mg/g) of chitosan for various heavy metals

Material	Sources	Cr ⁶⁺	Ni ²⁺	Pb ²⁺	Hg ²⁺	Zn ²⁺	Cu ²⁺	Cd ²⁺	Pt ⁶⁺
Chitosan	[2]							5.93	
	[3]				815	75	222		
	[4]				430				
	[5]	273							
	[6]		2.40	16.36	51.55		16.80	8.54	
	[7]						4.70		
	[8]						13.00		
Chitosan beads	[11]							250.00	
Non-crosslinked chitosan	[12]	80					85		
Crosslinked chitosan	[12]	50					86		
Crosslinked chitosan with GD	[10]								280
	[13]						60		
EC	[13]						62		
EGDE	[13]						46		

optimum for copper removal and about 13 mg of Cu²⁺ could be adsorbed by 1 g chitosan at equilibrium condition. The difference in chitosan adsorption capacity between the two studies could be due to the fact that a bigger particle size of chitosan (200 mesh) was used in the former study [7] (against 50 mesh in the latter study [8]). Such increase in particle size of chitosan, of course, decreases surface area available for adsorption, resulting in lower adsorption.

Chemical modifications of chitosan were also conducted to improve its removal performance and adsorption capacity for metal ions. In 1994, it was evaluated whether chemical modification of chitosan promotes selectivity in vanadium sorption or not [9]. It was reported that chitosan and its oxo-2-glutaric acid substitute form are effective to adsorb 450 mg of vanadium/g and that the sorption preferentially follow the Freundlich isotherm. It was also found that pH of 3.0 was found to be optimum for vanadium uptake.

The removal of platinum using chitosan-derived sorbents was also carried out [10]. It was found that glutaraldehyde-crosslinked chitosan was very effective for removing Pt⁶⁺ and that its adsorption capacity was about 280 mg of Pt⁶⁺/g. It was also reported that the optimum pH for Pt⁶⁺ sorption is around 2.0.

In 1998, the crosslinking effect of glutaric aldehyde on the removal of Cd²⁺ using chitosan beads was demonstrated [11]. It was reported that the adsorption capacity of the crosslinked gel beads exponentially decreased 60% from 250 mg of Cd²⁺/g.

The crosslinking effects of chitosan were also investigated [12]. It was found that non-crosslinked chitosan has potential to adsorb 30 mg more of Cr⁶⁺/g of chitosan. This is consistent with the fact that cross-linking reduces the adsorption capacities of chitosan, but this loss of capacity may be necessary to ensure the stability of chitosan.

The latest similar experimental work demonstrated the adsorption of Cu²⁺ onto chitosan crosslinked with glutaraldehyde (GD), epichlorohydrin (EC), and ethylene glycol diglycidyl ether (EGDE) [13]. It was pointed out that pH of 6.0 was found to be optimum for Cu²⁺

removal and that the uptake of Cu^{2+} on chitosan crosslinked with GD, EC, and EGDE beads were 59.67, 62.47, and 45.94 mg/g, respectively.

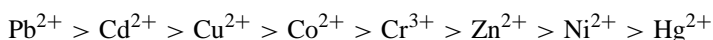
Overall, the results mentioned previously indicate that chitosan is a good adsorbent for all heavy metals. It is widely known that the excellent adsorption behaviors of chitosan for heavy metal removal is attributed to: (1) high hydrophilicity of chitosan due to large number of hydroxyl groups, (2) large number of primary amino groups with high activity, and (3) flexible structure of polymer chain of chitosan making suitable configuration for adsorption of metal ions.

2.2. Zeolites

Basically zeolites are a naturally occurring crystalline aluminosilicates consisting of a framework of tetrahedral molecules, linked with each other by shared oxygen atoms. During 1970s, natural zeolites gained a significant interest among scientists due to their ion-exchange capability to preferentially remove unwanted heavy metals such as strontium and cesium [14]. This unique property makes zeolites favorable for wastewater treatment. The price of zeolites itself is considered very cheap about US\$ 0.03–0.12/kg, depending on the quality of the zeolites itself [15].

Zeolites consist of a wide variety of species such as clinoptilolite and chabazite. Clinoptilolite is most abundant in nature and is readily available from more than 40 natural zeolites species [16]. Among the most frequently studied natural zeolites, clinoptilolite was shown to have high selectivity for certain heavy metal ions such as Pb^{2+} , Cd^{2+} , Zn^{2+} , and Cu^{2+} .

In 1990, the removal of heavy metals from wastewater using clinoptilolite was studied [17]. The results indicated that the ion exchange loading values could range from 1.6 mg/g for Pb^{2+} to 0 mg/g for Cr^{3+} . The selectivity of the series of the heavy metals studied was determined to be as follows:



Research has been conducted to show the effectiveness of clinoptilolite to remove lead and cadmium as well [18]. It was indicated that clinoptilolite is more selective for Pb^{2+} , but Cd^{2+} is also exchanged at satisfactory level. Approximately 1.4 mg/g of Pb^{2+} and 1.2 mg/g of Cd^{2+} were removed. Concerning the effect of temperature on the adsorption process, it was further mentioned that the metals uptake is favored at higher temperature [19] since a higher temperature activates the metal ions for enhancing adsorption at the coordination site of zeolites (Table 2).

In 1992, a further investigation on the use of carbonized zeolites for removing lead from wastewater was also conducted [20]. The strategy behind creating carbonized zeolites is to combine lyophilic and lyophobic surfaces that can bind with organic and inorganic substances, which are found in waste streams. It was reported that the carbonized zeolites removed about 99% of 260 ppm lead solution.

The influence of pretreatment upon the cation exchange capacity (CEC) and selectivity of zeolites for metal ions was investigated by a number of researchers. It was demonstrated that the CEC of clinoptilolite depends on the pretreatment method and that conditioning improves its ion exchange ability and removal efficiency [21]. Their findings were in

Table 2
Metal uptake by clinoptilolite at different temperatures and particle size [19]

Particle size of clinoptilolite (μm)	$\text{Pb}^{2+ \text{a}}$		$\text{Cd}^{2+ \text{a}}$	
	25 °C	50 °C	25 °C	50 °C
<600	1.31	1.41	1.06	1.20
160–600	1.29	–	1.02	–
600–1000	1.28	1.31	1.01	1.12
1000–2000	1.27	1.29	0.98	1.08

^a Pb^{2+} and Cd^{2+} taken at equilibrium (meq/g).

agreement with [22–24]. It was also reported that conditioning of zeolites with NaOH solution improved removal efficiency. Chabazite and clinoptilolite treated with sodium hydroxide performed best with Pb^{2+} and Cd^{2+} exchange capacity exceeding 100 mg/g and the performance of chabazite's CEC was indicated to be superior to that of clinoptilolite for both the ions.

In a further study, the removal performance of clinoptilolite and chabazite was compared [25]. The two zeolites were evaluated with respect to their performance for treating effluents contaminated with Pb^{2+} , Cd^{2+} , Cu^{2+} , Zn^{2+} , Ni^{2+} , and Co^{2+} . It was reported that both the zeolites exhibited 100% removal efficiency at the metal concentration of 10 mg/l. It was also found that clinoptilolite and chabazite exhibited different selectivity for all metals studied except Pb^{2+} , for which both performed exceptionally well. Finally, it was concluded that the superiority of chabazite's CEC was mainly due to the fact that chabazite has a higher Al substitution of Si than clinoptilolite. This provides chabazite a negative framework favorable for higher exchange capability.

The interactions of Pb^{2+} , Cd^{2+} , and Cr^{6+} competing for ion exchange sites in clinoptilolite was also investigated [26]. It was reported that dissolved Pb and Cd were effectively removed in acidic pH range. It was also found that the presence of Cr^{6+} diminishes the removal efficiency of Pb^{2+} and Cd^{2+} . It was suggested that decreased removal performance is due to the presence of ligands that form complexes with reduced accessibility and/or affinity for ion exchange.

In 1991, the removal of Cr^{3+} from industrial wastewater using Italian natural zeolite tuffs containing phillipsite and chabazite was evaluated in column operation [27]. Table 3 describes the operating conditions set for column experiments. It was reported that a lower

Table 3
Operational parameters for column experiments [27]

Operational parameters (units)	Value
Column diameter (cm)	1.43
Bed depth (cm)	38–48
Dose of adsorbent (g)	47.10–50.90
Feed composition of Cr^{3+} (mg/l)	52–56
Flow rate (ml/min)	1.83–7.00
Contact time (min)	8.7–33.3

Table 4
Summary of column runs for Cr³⁺ uptake by Italian zeolites [27]

Type of tuff sample	CEC (meq/g)	Flow rate (ml/min)	Working CEC (meq/l)	Efficiency (%)	Adsorption capacity (mg of Cr ³⁺ /g)
GS	2.11	7.00	56	9.0	3.3
NA–GS	2.11	7.00	52	15.2	5.5
NP	2.66	7.00	52	10.2	4.7
NP	2.66	1.83	52	15.4	7.1

Table 5
Column operation conditions [28]

Operational parameters (units)	Value
Column internal diameter (cm)	4.5
Bed depth (cm)	25
Bed weight (g)	400
Average bed density (g/cm ³)	1
Bed volume (cm ³)	400
Flow rate (ml/min)	15

flow rate (1.83 ml/min) is more favored by the Na ions of zeolites for a higher cation exchange capacity (CEC) with Cr³⁺ (Table 4). It can be explained due to the fact that more physicochemical interactions occurred between zeolites and metal species during column operation. The results indicated that flow rate is the most crucial characteristic in evaluating the effectiveness of an adsorbent for chromium adsorption.

The effect of flow rate on the adsorption capacity was also demonstrated in the latter study [28]. The specific conditions of column operation are listed in Table 5. From the Table 6, it can be seen that the Cr³⁺ uptake by zeolites in column operation is significantly higher in the previous study [27] than that in the latter study [28] due to the effect of flow rate. During column operation, a flow rate of 15 ml/min was used in the latter study [28], but a lower flow rate of 1.83 ml/min was applied in the former study [27].

It was also reported that at the same flow rate, zeolites had a higher cation exchange capacity with Ni²⁺ and Cu²⁺ than that with Zn²⁺, Cr³⁺, and Fe²⁺ (Table 7) [28]. It can

Table 6
Adsorption capacities (mg/g) of zeolites for some heavy metals

Material	Source	Cd ²⁺	Cr ³⁺	Cr ⁶⁺	Co ²⁺	Ni ²⁺	Zn ²⁺	Cu ²⁺	Pb ²⁺
Clinoptilolite	[17]	2.40	0		1.42	0.48	0.50	1.64	1.60
	[18]	1.20							1.40
	[21]	70.00							62.00
	[25]	3.70		2.40	1.50	0.90	2.70	3.80	6.00
Chabazite	[21]	137							175
	[25]	6.70		3.60	5.8	4.50	5.50	5.10	6.00
Chabazite–phillipsite	[27]		7.10						
	[28]		0.25			0.56	0.04	0.37	

Table 7
Summary of column runs for Ni²⁺, Zn²⁺, Cu²⁺, Cr³⁺, and Fe²⁺ uptake by Jordanian zeolites [28]

Parameters	Ni ²⁺	Zn ²⁺	Cu ²⁺	Cr ³⁺	Fe ²⁺	Remarks
Vb (BV)	23.70	27.50	25.00	27.50	27.50	Vb: volume at breakthrough point in bed volume (BV)
Vt (BV)	52.50	60.00	47.50	62.50	65.00	Vt: total volume at exhaustion point expressed in BV
MTZ (cm)	21.80	18.50	15.50	19.40	20.20	MTZ: mass transfer zone
CEC (meq/g)	1.12	0.08	0.76	0.74	0.13	CEC: CEC for individual cation at the exhaustion point
WEC (meq/g)	0.53	0.04	0.41	0.35	0.06	WEC: CEC for individual cation at breakthrough point
Efficiency (%)	29.60	2.20	22.70	19.70	3.10	Efficiency (%) = (WEC/total CEC) × 100
Selectivity (%)	63.60	4.60	41.10	42.40	7.10	Selectivity (%) = (CEC/total CEC) × 100

be explained due to the fact that the Ni²⁺ and Cu²⁺ were more preferable for zeolites due to their higher concentrations. Therefore, both ions had greater opportunity to have ion exchange with the Na ions of zeolites than others.

Overall, the results presented above show that instead of using costly activated carbon, zeolites hold great potential to remove heavy metal species from industrial wastewater effluents. However, low permeability of zeolites requires an artificial support when used in column operations.

2.3. Clay

It is widely known that there are three basic species of clay: smectites (such as montmorillonite), kaolinite, and micas; out of which montmorillonite has the highest cation exchange capacity and that its current market price (about US\$ 0.04–0.12/kg) is considered to be 20 times cheaper than that of activated carbon [29]. Therefore, a number of studies have been conducted using clays, mainly montmorillonite, to show their effectiveness for removing metal ions such as Zn²⁺, Pb²⁺, and Al³⁺ from aqueous solutions [30–32].

In 1989, the removal performance of montmorillonite and kaolinite for lead and cadmium was compared [33]. It was found that the adsorption capacity of Pb²⁺ and Cd²⁺ is greater on montmorillonite (Pb: 0.68, Cd: 0.72 mg/g) than on kaolinite (Pb: 0.12, Cd: 0.32 mg/g). It was also indicated that the presence of cationic surfactant reduces the uptake of both ions, while the anionic surfactants enhance their removal.

The adsorption of montmorillonite on Cd²⁺ and Zn²⁺ was also evaluated [34]. It was found that the Zn²⁺ is adsorbed in larger amounts than Cd²⁺ due to the fact that zinc has higher ionic potential than cadmium. Therefore, Zn²⁺ adsorption was favored over Cd²⁺ by cationic interchange.

The removal of zinc from wastewater using China clay, which mainly consists of aluminosilicates, was studied [35]. The essential characteristic of kaolinite group is that they do not swell with the addition of water. It was reported that an adsorption capacity of 1.25 mg

Table 8
Adsorption capacity of China clay and wollastonite for Pb^{2+} at different temperatures [37]

Type of adsorbent	Temperature (K)	Adsorption capacity (mg/g)	ΔG° (kJ/mol)	Remarks
China clay	293	0.411	-8.08	$\Delta H^\circ = -77.95$ kJ/mol
	303	0.395	-4.53	$\Delta S^\circ = -238.46$ J/K mol
	313	0.346	-3.07	
Wollastonite	293	1.680	-2.36	$\Delta H^\circ = -16.40$ kJ/mol
	303	1.290	-1.58	$\Delta S^\circ = -47.92$ J/K mol
	313	1.100	-1.05	

of Zn^{2+} /g was achieved by kaolinite and that the maximum removal efficiency was found to be at pH of 8.0.

Using a homogenous mixture of adsorbents, which consists of China clay–fly ash, wollastonite–fly ash, and China clay–wollastonite, the removal of copper from aqueous solution was also evaluated [36]. It was demonstrated that the highest adsorption capacity of about 1.18 mg of Cu^{2+} /g was achieved by fly ash–wollastonite.

The effect of temperature on the removal of lead using China clay and wollastonite was investigated [37]. It was found that the amount of Pb^{2+} removed is highly dependent on the temperature and that the sorption follows Langmuir isotherm. It was also reported that the maximum adsorption capacity of China clay and wollastonite was found to be 0.411 and 1.680 mg of Pb^{2+} /g, respectively (Table 8). It was indicated that a higher metal removal is observed at lower temperature. The negative value of enthalpy change (ΔH°) for both adsorbents indicated that the adsorption process was exothermic.

In 2001, the adsorption of lead on Thai kaolin and clay consisting mainly kaolinite and illite was studied [38]. It was reported that the maximum adsorption capacity of both materials was found to be 1.41 and 4.29 mg of Pb^{2+} /g, respectively, and that their adsorption followed both Langmuir and Freundlich models of isotherm. It was also observed that lead adsorption increased with an increase in pH. However, the presence of co-ions such as Cd^{2+} , Cr^{6+} , Cu^{2+} , Ni^{2+} , and Zn^{2+} , reduces the lead uptake from aqueous solution due to the fact that the co-ions bind strongly with organic matter present in clay to form a complex.

Another material from clay minerals to adsorb metal is bentonite, which mainly consists of clay, silt, and sand. This material is highly valued for its tendency to absorb water in the interlayer sites. The adsorption of Cd^{2+} and Zn^{2+} from aqueous solution was investigated using natural bentonite [39]. It was reported that the Cd^{2+} are adsorbed two times more than Zn^{2+} (Table 9) due to the fact that Cd^{2+} have less polarizing effects to the surface charge of bentonite than zinc ions.

In 1995, the adsorption of Cr^{6+} on bentonite at different temperatures ranging from 20 to 40 °C was investigated [40]. It was reported that the sorption is favored at higher temperature since the sorption of Cr(VI) is an endothermic process as indicated by positive enthalpy change (ΔH°) value listed in Table 10. It was also observed that the maximum adsorption capacity of 0.572 mg of Cr^{6+} /g bentonite was achieved at pH of 2.0.

The sorption of strontium was also studied using bentonite [41]. It was found that an adsorption capacity of 32.94 mg of Sr^{2+} /g bentonite was achieved at pH of 8.5 and that the sorption process followed Langmuir isotherm. It was also reported that the sorption

Table 9
Adsorption capacities (mg/g) of clay for different heavy metals

Material	Sources	Cu ²⁺	Pb ²⁺	Cd ²⁺	Zn ²⁺	Cr ⁶⁺	Sr ²⁺
Montmorillonite	[33]		0.68	0.72			
	[34]			4.78	4.98		
Kaolinite	[35]				1.25		
	[33]		0.12	0.32			
	[38]		1.41				
Illite	[38]		4.29				
Bentonite	[36]			11.41	4.54		
	[40]					0.57	
	[41]						32.94
	[42]				52.91		
	[43]			20			
Fly ash–wollastonite	[36]	1.18					

process of strontium on bentonite is endothermic as higher metal removal is more favorable at higher temperature (Table 11).

In 1997, the use of bentonite for zinc removal was evaluated [42]. It was found to be 52.91 mg of Zn²⁺/g bentonite as given by Langmuir model. In the latter study, outstanding removal capability of bentonite clay to uptake Pb²⁺ was demonstrated [43]. It was reported that adsorption capacities of 20 mg of Pb²⁺/g were achieved by bentonite at pH of 3.4. The usage of bentonite was also carried out for removal of radioactive waste [44] and cesium [45].

When clay minerals are used for industrial application, the swelling factor should be taken into account since it may cause remarkable pressure drop due to their different structural characteristics and ion-exchange mechanism. This is different from zeolites, which do not

Table 10
Adsorption capacity of bentonite for hexavalent chromium at different temperatures [40]

Temperature (K)	Adsorption capacity (mg/g)	ΔG° (kJ/mol)	Remarks
293	0.33	-0.24	$\Delta H^\circ = +5.62$ kJ/mol
303	0.45	-0.44	$\Delta S^\circ = +0.02$ kJ/K mol
313	0.57	-0.64	

Table 11
Adsorption capacity of bentonite for Sr²⁺ at different temperatures [41]

Temperature (K)	Adsorption capacity (mg of Sr ²⁺ /g)	ΔG° (kJ/mol)	Remarks
298	28.65	-10.69	$\Delta H^\circ = +30.62$ kJ/mol
303	30.49	-11.37	$\Delta S^\circ = +0.14$ kJ/K mol
308	32.94	-12.08	

Table 12
Adsorption capacities (mg/g) of peat moss for some metals

Material	Sources	Cu ²⁺	Cr ⁶⁺	Cd ²⁺	Zn ²⁺	Ni ²⁺
Eutrophic peat	[47]	12.07		20.23	11.12	11.15
	[48]	19.56				
Oligotrophic peat	[47]	12.07		22.48	13.08	11.74
	[48]	6.41				
Sphagnum peat moss	[49]		132			
	[50]		43.9			

show any swelling when embedded in liquid medium. Although the removal efficiency of clays for heavy metals may not be as good as that of zeolites, their easy availability and low cost may compensate for the associated drawbacks.

2.4. Peat moss

Peat moss, a complex soil material containing lignin and cellulose as major constituents, is a natural substance widely available and abundant, not only in Europe (British and Ireland), but also in the US. Peat moss has a large surface area (>200 m²/g) and is highly porous so that it can be used to bind heavy metals. Peat moss is a relatively inexpensive material and commercially sold at US\$ 0.023/kg in the US [46].

In 1986, the use of peat to remove heavy metals was investigated [47]. It was observed that peat moss plays an important role in treatment of metal-bearing industrial effluents such as Cu²⁺, Cd²⁺, Zn²⁺, and Ni²⁺ using eutrophic and oligotrophic peat. Eutrophic peat is poor in cellulose, but rich in humic substances. Oligotrophic peat is a sphagnum one, which is more acidic than eutrophic peat and contains more organic matter. Both peats contain about 85% of humic acid and 15% of fulvic acid. Their adsorption capacity for different metals is listed in Table 12.

Using both peats mentioned previously, the exchange properties of peat for copper removal were investigated [48]. It was reported that an adsorption capacity of 19.56 mg of Cu²⁺/g eutrophic peat was exhibited and that eutrophic peat showed higher adsorption capacity than oligotrophic peat (6.41 mg of Cu²⁺/g), although the latter is richer in cellulose.

The removal of Cr⁶⁺ using sphagnum peat moss was explored [49]. The adsorption capacity of sphagnum peat moss was found to be 132 mg of Cr⁶⁺/g at a pH range of 1.5–3.0. The most attractive advantage of this adsorbent in treatment is the simplicity of the system, low cost, and the ability to accept a wide variation of effluent composition.

In an extended study, the adsorption of Cr⁶⁺ was also investigated using sphagnum peat moss [50]. It was found that the adsorption capacity is higher at lower pH. At pH of 2.0, peat is able to adsorb 20% Cr⁶⁺ higher (about 43.9 mg of Cr⁶⁺/g) than that at pH of 2.5.

Overall, the results mentioned previously indicate that peat moss is a good adsorbent for all metals. It is widely known that peat moss exhibited a high CEC and complexities towards metals due to the presence of carboxylic, phenolic, and hydroxylic functional groups.

Table 13
Adsorption capacities (mg/g) of fly ash for some metals

Material	Sources	Cu ²⁺	Cr ⁶⁺	Hg ²⁺
Fly ash	[51]	1.39		
	[53]			2.82
Fly ash–wollastonite	[52]	1.18	2.92	
Fly ash–China clay	[52]		0.31	

2.5. Fly ash

Fly ash, an industrial solid waste of thermal power plants located in India, is one of the cheapest adsorbents having excellent removal capabilities for heavy metals such as copper ions [51]. It was reported that an adsorption capacity of 1.39 mg of Cu²⁺/g was achieved by fly ash at pH of 8.0 (Table 13). It was also found that the adsorption capacity increases with an increase in temperature.

Other studies have been conducted to show the effectiveness of fly ash on the removal of Cr⁶⁺ from aqueous solution using a homogenous mixture of fly ash and wollastonite (1:1) [52]. It was reported that an adsorption capacity of 2.92 mg of Cr⁶⁺/g could be achieved at pH of 2.0 and that the adsorption process followed Langmuir model of isotherm. This mixed adsorbent performed better than fly ash–China clay, where the maximum adsorption capacity was found to be 0.31 mg of Cr⁶⁺/g at pH of 2.0. This difference could be due to the fact that the adsorptive force between adsorbate and mixture (fly ash–wollastonite) is stronger than that of the other one (fly ash–China clay and adsorbate) so that Cr⁶⁺ is adsorbed more effectively.

In 1987, research on the adsorption of mercury using fly ash was carried out [53]. It was reported that the maximum adsorption capacity of 2.82 mg of Hg²⁺/g took place at a pH range of 3.5–4.5 and that adsorption followed the Freundlich model.

It is also known from various studies that fly ash could be easily solidified after the heavy metals are adsorbed. However, since it also contains heavy metals, the possibility of leaching should be considered and evaluated.

2.6. Coal

In 1984, the removal of Cd²⁺ using Giridih coal (GC) was intensively investigated [54]. It was reported that an adsorption capacity of 0.91 mg of Cd²⁺/g GC was achieved. It was also found that sorption followed Freundlich isotherm and that it decreased beyond pH 10 due to the formation of soluble hydroxy complexes.

A similar study for Hg²⁺ sorption using Giridih bituminous coal (GBC) was also conducted [55]. It was noted that chemical pretreatment for bituminous coal with nitric acid significantly enhanced mercury sorption to a level higher than that exhibited by activated carbon in terms of initial rate of sorption and adsorption capacity. It was also reported that an adsorption capacity of 10 mg of Hg²⁺/g was observed at a pH range of 7.0–8.5. It was found that the sorption reaction involves a rapid interaction between Hg²⁺ and active sites on the external surface of coal.

Table 14
Comparative study of chrome dye removal using mixed fly ash and coal (1:1) and activated carbon [56]

Adsorbents	Temperature (°C)	Removal efficiency (%)	Adsorption capacity (mg/g)	Optimum contact time (min)	Unit price (US\$/kg)
Fly ash + coal (1:1)	30	92.70	0.76	100	0.03
	40	82.48	0.69		
	50	70.80	0.61		
Activated carbon (commercial grade)	30	98.41	2.09	60	0.82
	40	84.13	1.72		
	50	71.43	0.98		

Gupta et al. studied the removal of chrome dye from aqueous solutions using a homogeneous mixed adsorbent consisting of fly ash and coal [56]. It was reported that adsorption capacity of 0.76 mg of chrome dye per gram was observed at pH of 2.0 and that it was nearly three times less than that by activated carbon (Table 14). It was also noted that the sorption process follows Langmuir isotherm and that lower temperature, ranging from 30 to 50 °C, favored higher removal efficiency of chrome dye as the sorption process is exothermic.

2.7. Natural oxide

In 1985, a study on the use of aluminium oxide to remove Cr^{6+} from aqueous waste was conducted [57]. It was reported that the ultimate adsorption capacity of 11.7 mg of Cr^{6+} /g alumina was observed at pH of 4.0. It is important to note that the adsorptive capacity of alumina significantly reduced in the presence of CN^- anions. It can be explained due to the fact that cyanide has a strong anionic influence upon the sorption characteristics of alumina. Therefore, CN^- anions are competitively adsorbed covering the surface sites of alumina, which in turn prevent the Cr^{6+} to be adsorbed on the internal surface of adsorbent.

The removal of Pb^{2+} and Cd^{2+} from aqueous solutions using aluminium oxide and goethite, an iron oxide was also explored [58]. It was found that goethite exhibits a better sorption capacity for both ions than aluminium oxide and that the uptake of Pb^{2+} is higher than that of Cd^{2+} (Table 15).

A further study was conducted using iron oxide coated with sand to remove Cr^{6+} [59,60]. It was reported that this adsorbent was found to be effective for removing metal from solution and that about 99% of 0.038 mM Cr^{6+} removal was achieved consistently [59]. In column operation, it was noted that this adsorbent showed good performance in treating

Table 15
Adsorption capacities (mg/g) of natural oxides for some metals

Material	Sources	Cd^{2+}	Pb^{2+}	Cr^{6+}
Aluminium oxide	[57]			11.7
	[58]	31	33	
Ferric oxide	[58]	72	230	

cadmium-plating wastes, but poor in treating chromium-plating waste [60]. It was also reported that cadmium-plating waste is adsorbed 28% higher than that of chromium (about 2.70 mg/l).

In 1996, the removal of arsenic ions from ground water in home treatment units using iron oxide coated with sand was investigated [61]. It was reported that this adsorbent was a promising medium to remove As^{3+} and As^{5+} from ground water since it could remove about 80–85% of 1.0 ppm arsenic solution. The process itself costs US\$ 8 to produce 700–800 l water free from any arsenic compound.

The use of manganese oxide, (cost about US\$ 0.05/kg), for removing arsenic compounds from ground water was demonstrated [62]. Removal efficiency of almost 100% was reported for both As^{3+} and As^{5+} at a pH range of 2.0–8.0. It was also found that the presence of bivalent cations such as Ni^{2+} , Co^{2+} , and Mg^{2+} in ground water enhances the adsorption capacity of manganese oxide due to the fact that these co-ions provide a framework to which the metal ions can be affixed by the surface of adsorbent.

2.8. Industrial waste

Iron(III) hydroxide waste is one of waste material from fertilizer industries. It has been extensively investigated for removing Cr^{6+} from wastewater [63]. It was reported that the maximum adsorption capacity of iron(III) hydroxide was found to be 0.47 mg of Cr^{6+} /g at pH of 5.6. This result is not in agreement with the previous study [64], which observed that HCrO_4^- is effectively adsorbed at pH of 8.5. This difference can be explained due to the fact that adsorption of Cr^{6+} in the latter study is suppressed by the presence of both SO_4^{2-} and SCN^- anions, which compete for adsorption sites.

Waste slurry is also one of the industrial by-products generated in fertilizer plant showing good sorptive capacities. In 1989, the use of waste slurry to remove Cu^{2+} , Cr^{6+} , Hg^{2+} , and Pb^{2+} from aqueous solution was investigated [65]. It was reported that this product exhibits outstanding adsorption capacity for Cr^{6+} , Hg^{2+} , and Pb^{2+} (Table 16). A similar study evaluated also the removal of Cu^{2+} and Cd^{2+} using waste slurry generated in seafood processing [66]. The adsorption capacity of waste slurry was found to be 20.97 and 15.73 mg/g for Cu^{2+} and Cd^{2+} , respectively.

Table 16
Adsorption capacities of industrial waste (mg/g)

Material	Sources	Ni^{2+}	Cu^{2+}	Pb^{2+}	Hg^{2+}	Cr^{6+}	Zn^{2+}	Cd^{2+}
Waste slurry	[65]			1030	560	640		
	[66]		20.97					15.73
Iron(III) hydroxide	[63]					0.47		
Lignin	[67]			1865			95	
Blast-furnace slag	[69]			40		7.5		
Sawdust	[70]		13.80					
Activated red mud	[73]	160						
	[72]					1.6		
Bagasse fly ash	[71]					260		

Table 17
Metals uptake by lignin at different temperatures [67]

Type of metal	Temperature (K)		ΔG° (kJ/mol)		Remarks
	303	313	30	40	
Pb ²⁺	1586	1865	-26.17	-28.47	$\Delta H^\circ = +43.6$ kJ/mol, $\Delta S^\circ = 0.23$ kJ/K mol
Zn ²⁺	73.24	94.83	-32.49	-36.79	$\Delta H^\circ = +97.8$ kJ/mol, $\Delta S^\circ = 0.43$ kJ/K mol

In 1994, research on the adsorption of Pb²⁺ and Zn²⁺ onto lignin extracted from black liquor was carried out [67]. Black liquor, a waste product originated from paper industry, could be purchased at US\$ 1.00/ton and the lignin could be processed for US\$ 60/t, comparable to activated carbon sold at US\$ 100/t. It was reported that the adsorption capacity of lignin for both Pb²⁺ and Zn²⁺ was found to be 1865 and 95 mg/g, respectively, at 40 °C. Results indicated that the high adsorption capacity of lignin is due to the presence of polyhydric phenol groups on the surface of lignin. It was also indicated that the adsorption is an endothermic process since a higher removal was favored at higher temperature (Table 17).

Another low-cost adsorbent showing capability to adsorb heavy metals is blast-furnace slag, an industrial by-product generated in steel plants. In 1996, the sorption of Cu²⁺, Ni²⁺, and Zn²⁺ using blast-furnace slag was studied [68]. It was found that metal ions sorption takes place in the form of hydro-oxo complexes and that the high sorption capacity is related to the formation of soluble compounds on the internal surface of adsorbent.

A further study was also conducted to investigate the removal of Pb²⁺ and Cr⁶⁺ using blast-furnace slag sold at US\$ 38/t [69]. It was found that the maximum metals uptake observed under identical conditions is 40 and 7.5 mg/g of Pb²⁺ and Cr⁶⁺, respectively. It can be concluded that its removal performance is comparable to the cheapest commercial activated carbon (cost about US\$ 1000/t), as 1 g of activated carbon can adsorb 32.4 mg of Pb²⁺.

The role of sawdust, collected from a timber working shop, for copper removal was evaluated [70]. It was reported that an adsorption capacity of 13.80 mg of Cu²⁺/g sawdust was achieved. It was suggested that sawdust is a good adsorbent for copper removal.

The use of bagasse fly ash, an industrial waste generated in the sugar industry, to remove Cr⁶⁺ from electroplating wastewater was investigated [71]. From the Table 18, it was found that the sorption capacity decreases with an increase in temperature as the adsorption process is exothermic. It was also reported that the adsorption of Cr⁶⁺ on bagasse fly ash followed both Freundlich and Langmuir isotherm and that the sorption capacity of bagasse fly ash was found to be 260 mg of Cr⁶⁺/g as given by the Langmuir model.

Table 18
Hexavalent chromium uptake by bagasse fly ash at different temperatures [71]

Temperature (K)	Adsorption capacity (mg/g)	ΔG° (kJ/mol)	Remarks
303	260.00	-16.26	$\Delta H^\circ = +50.43$ kJ/mol
313	123.76	-15.14	$\Delta S^\circ = -112.76$ kJ/K mol

In 1999, research on the usage of activated red mud to adsorb hexavalent chromium from aqueous solution was carried out [72]. Red mud, a by-product of the aluminum industry, is composed mainly of the particles of silica, aluminium, iron, and titanium oxide. It was found that an adsorption capacity of 1.6 mg of Cr^{6+} /g red mud was achieved at pH of 5.2. However, the adsorption capacity of red mud was significantly different from that obtained in a previous study [73], which investigated the use of red mud for Ni^{2+} removal. It was reported that an adsorption capacity of 160 mg of Ni^{2+} /g red mud was accomplished. It can be explained due to the fact that there was pretreatment for red mud. Such pretreatment, of course, could increase its effectiveness to adsorb the targeted metal and consequently, red mud has a higher cation exchange capacity with Ni^{2+} .

2.9. Miscellaneous adsorbents

Other low-cost adsorbents have been studied less extensively such as xanthate, rice husk carbon (RHC), and coconut shell. Xanthate is one of the effective low-cost adsorbents. It is a group of sulfur-bearing compounds, which have a high affinity for heavy metals and formed when an organic hydroxyl-containing substrate reacts with carbon disulfide. A study conducted by Tare et al. compared the removal performance of soluble and insoluble starch xanthate for heavy metal such as Cd^{2+} and Cr^{3+} [74]. It was found that the performance of insoluble starch xanthate is better in terms of adsorption capacity and ease of operation. It was also reported that soluble starch xanthates cost only US\$ 1.0/kg.

The use of RHC for removing Cr^{6+} was also demonstrated [75]. The activated carbon was derived from rice husk using carbonization with sulfuric acid. The maximum adsorption capacity of RHC was found to be 45.6 mg of Cr^{6+} /g at pH 2.5.

Alaerts et al. [76] reported that coconut shell-based activated carbon could be used for Cr^{6+} removal. It was shown that optimum chromium removal was achieved at pH < 7.0. It was also found that removal efficiency increases with a decrease in chromium concentration and that the adsorption follows the Freundlich isotherm.

In 2000, the use of activated alumina to remove arsenite and arsenate from ground water was studied [77]. It was reported that the uptake of arsenite is much less than that of arsenate for alumina grain at optimum pH. The adsorption capacity of alumina grain for arsenite and arsenate were found to be 3.48 and 12.34 mg/g at pH 6.9 and 2.6, respectively. The difference may be due to the fact that under most pH conditions for natural water, arsenate is present in negatively ionic form and arsenite is in non-ionic form.

Ajmal et al. [78] carried out an adsorption study on *Citrus reticulata*, an agricultural waste originated from the fruit peel of orange, for the removal of Ni^{2+} from electroplating wastewater. It was reported that maximum removal of Ni^{2+} occurred at pH of 6.0 and that the adsorption followed the Langmuir isotherm. It was also found that an adsorption capacity of 158 mg of Ni^{2+} /g was achieved by *Citrus reticulata* at 50 °C and that the sorption of Ni^{2+} was endothermic, as shown by the negative value of free energy (ΔG°) (Table 19). It can be concluded that the adsorption capacity of the waste increases with an increase in temperature.

In 2002, the potential of Parthenium, an Indian agricultural waste, for removing Ni^{2+} from aqueous solution was also investigated [79]. It was reported that the adsorption capacity of Parthenium was found to be significantly lower (about 54.35 mg of Ni^{2+} /g) than the previous study [78], although both are agricultural wastes.

Table 19
Adsorption capacity of *Citrus reticulata* for Ni²⁺ at different temperatures [78]

Temperature (K)	Adsorption capacity (mg/g)	ΔG° (kJ/mol)	Remarks
303	80	−8.24	$\Delta S^\circ = -0.06$ kJ/K mol
313	119	−8.95	$\Delta H^\circ = 10.37$ kJ/mol
323	158	−9.49	

A comparative study on the removal of Cr⁶⁺ from aqueous solution was also carried out using low-cost adsorbents derived from used tyres (TAC), sawdust (SPC), and granular activated carbon (GAC) type Filtrasorb 400 [80]. It was found that the adsorption capacities of TAC are comparable to that of GAC at optimum pH of 2.0 (Table 20). However, the adsorption capacity of SPC is significantly lower than that of both TAC and GAC. It can be explained due to the fact that both TAC and GAC have smaller particle size (0.2 mm) than SPC (0.65 mm). Such reduction in particle size of adsorbents increases its surface area for metal adsorption and it results in higher removal efficiency on Cr⁶⁺. It was also indicated that the adsorption of Cr⁶⁺ was more favorable at higher temperature.

The use of diatomite to remove Cr⁶⁺ from aqueous solution was demonstrated as well [81]. Microemulsions treated diatomite are quite efficient in removal process of metallic ions. From Table 21, it was reported that an adsorption capacity of 1.68 mg of Cr⁶⁺/g diatomite was achieved at pH of 2.95.

An investigation on the use of spheroidal cellulose to remove chromium was also conducted in China [82]. Cellulose is the most abundant among renewable and natural polymers and it has three reactive hydroxyl groups. The adsorption capacity of spheroidal cellulose was found to be 73.46 mg of Cr⁶⁺/g at pH of 6.0. This investigation is useful to develop advanced technology for wastewater treatment facility since the process is economically feasible and easy to carry out. Presently its market price is about US\$ 1.07/kg.

2.10. Activated carbon

Based on its size and shape, activated carbon is classified into four types: powder (PAC), granular (GAC), fibrous (ACF), and clothe (ACC). Due to the different sources of raw

Table 20
Comparison of the adsorption capacities between low-cost adsorbents (TAC and SPC) and GAC at different temperatures [80]

Temperature (°C)	Type of adsorbent	Adsorption capacity (mg/g)	ΔG° (kJ/mol)
22	TAC	48.08	−12.33
	SPC	1.93	−1.17
	GAC	44.44	−2.37
30	TAC	55.25	−12.92
	SPC	2.16	−3.30
	GAC	48.54	−4.35
38	TAC	58.48	−13.86
	SPC	2.29	−5.06
	GAC	53.19	−6.94

Table 21
Adsorption capacities (mg/g) of miscellaneous adsorbents

Material	Source	Cd ²⁺	Cr ³⁺	Cr ⁶⁺	As ³⁺	As ⁵⁺	Ni ²⁺
Cellulose xanthate	[74]	19.88					
Soluble starch xanthates	[74]	33.27	17.57				
Rice husk carbon	[75]			45.6			
Activated alumina	[77]				3.48	12.34	
Diatomite	[81]			1.68			
Spheroidal cellulose	[82]			73.46			
Agricultural waste (<i>Citrus reticulata</i>)	[78]						158
Parthenium	[79]						54.35
Waste tyre	[80]			58.48			
Sawdust	[80]			2.29			

materials, the extent of chemical activation, and the physicochemical characteristics; each type of activated carbon has its specific application as well as inherent advantages and disadvantages in wastewater treatment.

Although a significant number of low-cost adsorbents from various materials have been found, commercial activated carbon (CAC) has still been used intensively today. A large number of researchers are still studying the use of activated carbon for removing heavy metals such as mercury [83], copper [84], lead [85], chromium [86–91], cadmium [92,93], Ni [94,95], zinc [96–98], and lithium [99]. Recently the market price of activated carbon for industrial grade is considered to be very expensive (about US\$ 20–22.00/kg), depending on the quality of activated carbon itself [100].

Various types of CAC for heavy metals removal have been reported. In 1984, the removal efficiency of mercury using different brands of PAC such as Nuchar SA and SN was evaluated [83]. It was reported that about 99–100% of total 0.2 mM Hg²⁺ removal was attained by both the types at pH of 4.0–5.0.

Another similar research was carried out to compare the phenomenon of copper and cobalt adsorption by GAC from aqueous solution [84]. It was reported that the removal capability of CAC for both metals was found to be significantly different. It was also demonstrated that at pH 4.0, GAC could remove 99% of 10 ppm of cobalt solution, but only 93% of copper solution at the same concentration.

The use of GAC for lead removal was also evaluated [85]. It was demonstrated that the adsorption capacity of activated carbon was found to be 30 mg of Pb²⁺/g and that the amount of Pb²⁺ not removed corresponded to the amount that was complexed by EDTA.

The use of activated carbon for Cr⁶⁺ removal was also intensively investigated in recent years. In 1995, the removal efficiency of different types of PAC prepared from different raw materials such as leather, olive stone, and almond shell was evaluated [86]. It was indicated that the extent of the adsorption process depends on the pretreatment of activated carbon and that the highest removal performance was obtained with those prepared by physical activation. It was also reported that at pH of 1.0, the retention of Cr⁶⁺ was affected by its reduction to Cr³⁺.

Table 22
Adsorption capacities (mg/g) for activated carbon

Type of activated carbon	Source	Cr ⁶⁺	Cr ³⁺	Pb ²⁺	Zn ²⁺	Cd ²⁺	Ni ²⁺	Cu ²⁺	Li ⁺	Hg ²⁺
PAC-Nuchar SA	[83]									40.12
	[99]								0.45	
GAC-HD 400	[85]			30						
GAC-C	[97]				18					
GAC	[98]	6.84			9.90			38		
GAC-Filtrisorb 400	[87]	145								
	[80]	53.19								
	[89]	0.18								
	[91]		30							
	[92]					8				
GAC-LB 830	[89]	0.13								
As-received ACF	[93]					146				
	[95]	40								
	[94]						2	9		
Oxidized ACF						10	30			
ACC	[96]				2	3.75				65

In 1996, the removal of Cr⁶⁺ from aqueous solution using GAC type Filtrasorb 400 was also studied [87]. It was reported that an adsorption capacity of 145 mg of Cr⁶⁺/g was achieved at a pH range of 2.5–3.0. This result is not in agreement with that obtained in the latter study [89] conducting a similar comparative study using activated carbon LB 830 and Filtrasorb 400. It was reported that the maximum adsorption capacity of Filtrasorb 400 in the latter study is only 0.18 mg of Cr⁶⁺/g (Table 22).

Chemical modification on the surface of GAC with oxidizing agent such as nitric acid was also conducted to improve its adsorption capacity [90]. It was found that the amount of chemisorbed oxygen on the carbon surface increased after oxidative treatment. It is interesting to note that the adsorption capacity of Cr³⁺ on the oxidized carbon is enhanced about 300% of 30 mg of Cr³⁺/g. This may be attributed to the fact that the surface of oxidized GAC has a larger negative charge than that of non-oxidized one. Due to the electrostatic attractive interactions between them, the change in the negative charge on the carbon surface, and that of the positive charge of Cr³⁺ in the solution favor more adsorption of metal ions.

Another similar research was also conducted to evaluate the removal of Cr⁶⁺ by ACFs plated with copper metal [91]. It was reported that the introduction of Cu²⁺ on ACFs significantly lead to an increase in the surface basicity, resulting in the adsorption capacity of Cr⁶⁺ from an aqueous solution regardless of a decrease in surface area. It was pointed out that the adsorption of chromium ions was essentially dependent on surface properties, rather than by surface area and porosity of ACFs.

The adsorption of Cd²⁺ from the aqueous solution using GAC was also studied [92]. It was reported that maximum adsorption capacity of GAC was found to be 8 mg of Cd²⁺/g at pH 8.0. It was also observed that the amount of Cd²⁺ adsorbed was reduced about three

times by increasing the temperature from 10 to 40 °C. This indicated that the adsorption process was exothermic in nature.

The oxidative effects of nitric acid for ACFs were also investigated for Cd²⁺ removal [93]. Due to more electrostatic attractions between the positive charge of Cd²⁺ and the negative charge of ACFs, it was found that a significant increase in ion-exchange capacity was achieved after oxidative treatment. The maximum adsorption capacity of ACFs was found to be 146 mg of Cd²⁺/g at pH 5.0–6.0.

The use of oxidized ACFs for Ni²⁺ and Cu²⁺ adsorption was also evaluated and compared to that of as-received ACFs [94,95]. It was demonstrated that the adsorption capacity of oxidized ACFs on both the metals was indicated to be superior to that of as-received ACFs and that the adsorption capacity of ACFs was higher for Cu²⁺ than that of Ni²⁺ (Table 22). This indicated that oxidative treatment increases the acidity of the surface functional group of ACFs so that more electrostatic attractions occur between the negative charge of ACFs and the positive charge of cations.

In the latest research [96] investigating the adsorption of various heavy metals such as Zn²⁺, Cd²⁺, and Hg²⁺ onto ACC, it was reported that the adsorption capacity of ACC was found to be significantly higher for Hg²⁺ than that for Zn²⁺ and/or Cd²⁺ (Table 22). It was also indicated that the amount of adsorbed metals increases with an increase in pH.

Other similar work for Zn²⁺ removal was also conducted using different types of GAC such as C, F-300, F-400, and Centaur HSL [97]. It was found that C has higher adsorption capacity for Zn²⁺ than other types of GAC. It was also reported that the adsorption capacity of GAC type C is about 18 mg of Zn²⁺/g at pH 7.0.

Chemical modifications of CAC using tetrabutyl ammonium iodide (TBAI) and sodium diethyl dithiocarbamate (SDDC) were also carried out [98]. It was reported that the TBA-carbon adsorbent was found to have an effective adsorption capacity of approximately five times than that of as-received carbon and that using SDDC-carbon column, heavy metals such as Cu²⁺, Zn²⁺, and Cr⁶⁺ could be eliminated with a removal capacity of 38, 9.9, and 6.84 mg/g, respectively. It was also suggested that the technique of TBA and SDDC modifications optimize the existing properties of activated carbon, giving greater removal capacity to the as-received activated carbon.

In 1996, the use of PAC for Li⁺ removal was evaluated using an electric current [99]. It was reported that the adsorption capacity of PAC is very low (about 0.45 mg of Li⁺/g). This can be explained due to the fact that electrochemical polarization of the carbon material modifies the surface functional groups and exerts a significant influence on the sorption of Li⁺. Thus, indicating that the surface chemical effects appear to dominate, although the specific surface area may be important.

3. Comparison of commercial activated carbon with low-cost adsorbents

Fig. 1 illustrates the adsorption capacity of some outstanding adsorbents mentioned in the above study. It is evident from our literature survey and Fig. 1 that some low-cost adsorbents such as chitosan, zeolites, waste slurry, and lignin have demonstrated outstanding removal capabilities for heavy metals, which is far better than commercial activated carbon. Most toxic heavy metals such as copper, zinc, cadmium, and mercury ions, for instance, have

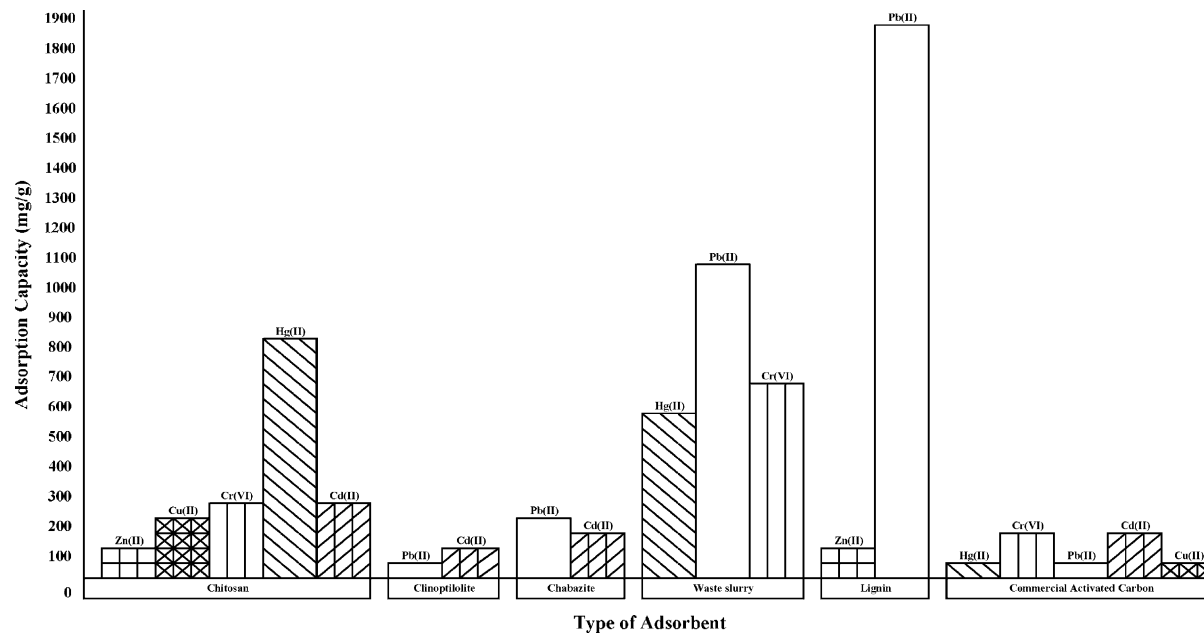


Fig. 1. Summary of some adsorbents with high adsorption capacities (mg/g).

been effectively removed from highly diluted solutions using chitosan. Even for Cr^{6+} , Hg^{2+} , Cu^{2+} , and Cd^{2+} removal, chitosan performs significantly better than any types of commercial activated carbon (CAC) in terms of metal-loading capacity. However, chitosan is considered as the most expensive alternative adsorbent since its market price is nearly comparable to that of CAC. Recently market price of chitosan is US\$ 16/kg and that of CAC (depending on its grade and quality) is about US\$ 20.0–22.0/kg.

Clinoptilolite, one of abundant natural zeolites species, is a good low-cost adsorbent for cadmium and lead removal. Nevertheless, chabazites has better cation exchange capacity than clinoptilolite. Both Pb^{2+} and Cd^{2+} are adsorbed by chabazites nearly two times more than those by clinoptilolite. Although the zeolites' current commercial price (less than US\$ 1.0/kg) is now considered nearly 20 times cheaper, the adsorption capacity of chabazites for Cd^{2+} is comparable to that of CAC. In fact, its adsorption capacity for Pb^{2+} is four times higher than that of CAC type HD 400.

Waste slurry is another alternative adsorbent that is able to remove Cr^{6+} , Pb^{2+} , and Hg^{2+} effectively from aqueous solutions at low cost. As for Cr^{6+} removal, the adsorption capacity of waste slurry is the highest among other low-cost adsorbents and is nearly four times higher than that of CAC type Filtrasorb 400. In fact, its adsorption capacity for Pb^{2+} and Hg^{2+} is more than 20 and 10 times higher than that of any type of CAC, respectively. However, there is no available information for the commercial price of waste slurry as it can be obtained for free or at inexpensive cost from fertilizer plants.

Last but not the least, lignin is considered as the best low-cost adsorbent for lead and zinc removal. Lignin is able to remove both the ions effectively under temperatures ranging from 30 to 40 °C. It adsorbs Pb^{2+} nearly two times higher than waste slurry and its removal capability for Zn^{2+} is comparable to that of chitosan. Even its adsorption capacity for Pb^{2+} is significantly more than 30 times higher than that of CAC. In spite of its inexpensive commercial price (about US\$ 0.06/kg in 1994), lignin undoubtedly has excellent metal-adsorbent binding capacities, which are comparable to that of chitosan. Moreover, the adsorption capacity of lignin for Pb^{2+} is the most outstanding compared to CAC and other low-cost adsorbents such as chitosan, zeolites, or waste slurry. Due to the reasons mentioned previously, a cost–benefit analysis of using lignin for lead and zinc removal in wastewater treatment applications needs to be conducted to judge the economic feasibility of its practical use and the potential for its commercial applications in the future.

4. Conclusions and recommendations

A wide range of low-cost adsorbents has been studied worldwide for heavy metal removal. It is evident from our literature survey that inexpensive and locally available materials could be used instead of commercial activated carbon.

A few adsorbents that stand out for high adsorption capacities are chitosan (815, 273, 250, 222, 75 mg/g of Hg^{2+} , Cr^{6+} , Cd^{2+} , Cu^{2+} , and Zn^{2+} , respectively), zeolites (175 and 137 mg/g of Pb^{2+} and Cd^{2+} , respectively), waste slurry (1030, 560, 640 mg/g of Pb^{2+} , Hg^{2+} , and Cr^{6+} , respectively), and lignin (1865 and 95 mg/g of Pb^{2+} and Zn^{2+} , respectively). These adsorbents are efficient and can be effectively used for inorganic effluent treatment containing metal ions.

Among their current commercial prices, zeolites are undoubtedly the most inexpensive alternative adsorbents compared to others. It is considered 15 times cheaper than chitosan (US\$ 15.43/kg). It is expected that the price of chitosan will slowly go down since more industries worldwide may consider using it in wastewater treatment due to its higher removal efficiencies.

To improve removal efficiencies and adsorption capacities, chemical modifications of low-cost adsorbents such as coconut shell charcoal needs to be conducted using coating process. Coconut shell charcoal has low removal efficiencies and adsorption capacities for metals removal. It is expected that coating coconut shell charcoal with chitosan may significantly improve its removal performance.

So far, cost information of other adsorbents such as fly ash, coal, ferric oxide, and waste slurry is seldom reported in any of the publications since the expense of individual adsorbents varies, depending on the processing required and local availability. This situation makes a comprehensive comparison among alternative adsorbents difficult to materialize due to inconsistencies in data presentation.

In spite of the scarcity of consistent cost information, the widespread uses of low-cost adsorbents in industries for wastewater treatment applications today are strongly recommended due to their local availability, technical feasibility, engineering applicability, and cost effectiveness. If low-cost adsorbents such as chitosan, zeolites, waste slurry, and lignin perform well in removing heavy metals at low cost, they can be adopted and widely used in industries not only to minimize cost inefficiency, but also improve profitability. Due to the reasons mentioned previously, a cost–benefit analysis of using low-cost adsorbents for heavy metal removal needs to be conducted to judge the economic feasibility of its practical use in wastewater treatment applications today.

Last but not the least, if the alternative adsorbents mentioned previously are found highly efficient for heavy metal removal, not only the industries, but the living organisms and the surrounding environment will be also benefited from the potential toxicity due to heavy metal. Thus, the use of low-cost adsorbents may contribute to the sustainability of the surrounding environment. Undoubtedly low-cost adsorbents offer a lot of promising benefits for commercial purpose in the future.

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