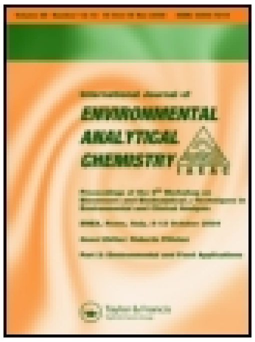

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Adsorption of heavy metal ions by various low-cost adsorbents: a review

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ABSTRACT

Environmental pollution, particularly from heavy metal ions in the wastewater, is one of the most serious concerns of the world. In the pursuit of remedial action, various conventional methods such as ion exchange, chemical precipitation, coagulation, membrane separation, reverse osmosis, and adsorption methods have so far been used for the removal of heavy metal ions. A good variety of adsorbents have been developed to remove different heavy metal ions from wastewater in particular those which have been detrimental to living organisms. Adsorption processes have been very demanding for high removal efficiency of heavy metal ions even at trace levels and they are low cost as compared to conventional methods. It has therefore been crucial to develop low cost and readily available adsorbents for the adsorption of heavy metal ions from wastewater. The adsorbents may be collected from agricultural and animal waste and industrial by-products. All adsorbents, by their intrinsic nature, have functional groups to play the key role in metal ion adsorption. Generally, chemically modified adsorbents enhance the surface area of the adsorbent and exhibit higher adsorption capacity than unmodified adsorbents. In this review, a series of natural waste materials and their modified forms have been evaluated for the removal of various metals from potable and wastewater. The major focus has been an accumulation of comprehensive knowledge on of the use of the low-cost adsorbents for removal of heavy metal ions.

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Waste product; adsorption; removal of heavy metal; low-cost adsorbents

1. Introduction

A number of pollutants – both organic and inorganic – causes harm to the environment. The present world faces the risk of heavy metal ions most since they are very toxic and carcinogenic in nature. The pollution from heavy metals is a result of human activities and the consequence on the food chain of the ecosystem. Heavy metals are released into environment from various sources such as chemical industries, textiles, tanneries, plastics, mining, battery manufacturing, paints and pigments, paper and pulp industries, etc., [1–4]. Release of toxic metals into water stream is a serious concern, which may affect the quality of water supply [5]. Some of the hazardous heavy metal ions which pose potential danger

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threat to human health are arsenic, mercury, chromium, cadmium, and lead. Heavy metal ions noted are not recyclable and accumulate in living organisms [6]. Several past disasters for the contamination of heavy metals in waterbodies include: 'Itai–Itai' for pollution of cadmium in Jintsu river of Japan and 'Minamata tragedy' due to methyl mercury contamination in Japan [7]. Heavy metals are released into water stream at higher concentrations beyond the limit of the prescribed value by the industrial activity. Heavy metals exhibit their toxicity at low concentrations of about 1.0–10.0 mg/L [8].

These have been an ever increase in the use of heavy metal ions due to rise in industrial activities to cause increased pollution of water streams day by day. The heavy metal ions are strong toxicants since they are non-biodegradable. Moreover, heavy metal ions are carcinogenic in natures. Alarmingly, the heavy metal ions in water systems are at higher concentrations than the permissible limits to cause numerous diseases [9]. Toxicity and the permissible limits of certain metal ions are listed in Table 1.

A numbers of technologies are available to treat of heavy metals polluted wastewater. Traditional techniques are chemical precipitation, ion exchange, ultrafiltration, reverse osmosis, nanofiltration, coagulation, flocculation, flotation. However, these have some limitations such as low removal efficiency, high operational cost and sludge production. Comparatively, the adsorption method is more advantageous for low energy consumption, ease of operation and high removal efficiency [10]. In the category of low-cost adsorbents, both natural adsorbents and bio-adsorbents [11] are used. In natural materials, zeolites, clay, chitosan and red mud are utilised as adsorbents. The main sources of bio-adsorbents are agricultural and animal waste materials.

This review provides detailed information about the use of low-cost materials as adsorbents for the removal of heavy metal ions. A crucial analysis of low-cost adsorbent materials has been made and their features and advantages have been described.

Table 1. Heavy metal ions, their adverse effect and permissible limits according to World Health Organisations.

S. No.	Heavy Metals	Health hazards	Permissible Limit (mg/L) (WHO)	References
1.	Cadmium (Cd)	Kidney damage, renal disorder, human carcinogen, emphysema.	0.003	[41]
2.	Mercury (Hg)	Neurological damage, paralysis, blindness, rheumatoid arthritis and anorexia.	0.001	[42]
3.	Arsenic (As)	Skin, lung, bladder and kidney cancer, neurological disorder, muscular weakness and nausea.	0.01	[43]
4.	Lead (Pb)	Brain damage, anaemia, anorexia, vomiting, disease of circulatory and nervous systems.	0.05	[44]
5.	Chromium (Cr)	Headache, diarrhoea, nausea, vomiting, carcinogenic, lung tumour.	0.05	[45]
6.	Cobalt (Co)	Asthma like allergy, damage to the heart, damage to the thyroid and liver, carcinogenic.	0.1	[46]
7.	Copper (Cu)	Liver damage, Wilson disease, Insomnia.	2.5	[47]
8.	Zinc (Zn)	Depression, lethargy, neurological signs, dehydration anaemia and increased thirst.	5.0	[48]
9.	Manganese (Mn)	Syndrome of motor dysfunction, memory loss resembling, Parkinson disease.	0.5	[49]
10.	Iron (Fe)	Headache, Brittle nails, Depression, Constipation, Tinnitus, Gastrointestinal complains.	0.3	[50]
11.	Nickel (Ni)	Dermatitis, nausea, chronic asthma, coughing and cancer of the lung.	2.0	[51]

2. Sources and toxicity effects of heavy metal ions

2.1. Arsenic

Arsenic is a metalloid element and it is highly toxic and hazardous. Generally arsenic is found in four oxidation state- +5 (arsenate), +3 (arsenite), 0 (arsenic) and −3 (arsine). As(V) (arsenate) is highly toxic, whereas in anaerobic conditions, As(III) (arsenite) form dominates [12]. Various sources, such as insecticides, herbicides, phosphate fertilisers, mining, semi-conductor industries, coal combustion, etc., cause arsenic contamination [13,14]. Ingestion of arsenic-contaminated water for a prolonged period causes various diseases such as arsenicosis, skin, lung, bladder and kidney cancer, neurological disorder, muscular weakness, and nausea [15]. In India, various states such as West Bengal, Assam, Manipur, Bihar, Jharkhand, Uttar Pradesh and Chhattisgarh are reported to be affected by arsenic-polluted water. It may be worth mentioning that according to World Health Organisation (WHO) the permissible limit of drinking water is 0.01 mg/L (10 ppb) [16,17].

2.2. Mercury

Mercury is one of the most hazardous heavy metal ions. The highly toxic form of mercury is +2 oxidation state. It is released into environment by both natural and anthropogenic sources. Mercury contamination originates from natural sources, such as volcanic action and geological activities, and anthropogenic sources such as pharmaceutical and cosmetic preparation, paper and pulp industries, thermometers, batteries and medical drugs, etc., [18,19]. It causes neurological damage, paralysis, blindness, rheumatoid arthritis and anorexia. Due to its high toxicity European Union (EU) set the permissible limit of drinking water and wastewater as 0.001 and 0.005 mg/L, respectively, [20].

2.3. Cadmium

Cadmium is non-essential and highly toxic element and a lesser amount is used in our daily life. Generally it can exist in +2 oxidation state. Cadmium is used in a broad variety of industries such as cadmium-nickel batteries, metal plating, mining, phosphate fertilisers, pigments, stabilisers, and alloy industries [21]. The detrimental effects of cadmium are itai–itai disease, kidney damage, renal disorder, human carcinogen, and emphysema [22,23]. The permissible level of cadmium fixed by WHO is 0.003 mg/L and according to United States Environmental Protection agency (EPA) is 0.005 mg/L [24].

2.4. Lead

Lead is a common hazardous heavy metal and generally found in industrial wastewater. It can exist in divalent form. Lead contamination exists in water stream of many industries such as battery manufacturing, ammunition, bronze products and pipe, ceramic and glass industries. Effects of lead poisoning are brain damage, anaemia, anorexia, vomiting, and disease of circulatory and nervous systems [25]. According to WHO the permissible level of lead is 0.05 mg/L [26].

2.5. Chromium

Chromium may exist in Cr(III) (trivalent) and Cr(VI) (hexavalent) oxidation states in water sample. Toxicity of Cr(VI) is very high and it is carcinogenic [27]. Chromium is released in large quantities from various industries and pollutes the environment. Sources of chromium contamination in water stream is steel fabrication, paints and pigments, chemical industries, textile industries, ceramics and wood treatments units [28]. It causes cancer in the digestive tract, headache, diarrhoea, nausea, vomiting, lung tumour, and haemorrhage [29]. EPA has set the permissible limit of Cr(VI) for drinking water is 0.1 mg/L [30]. According to WHO and FAO the permissible limit of Cr(III) for drinking water is less than 5 mg/L [31].

2.6. Copper

Copper is basically used in various industries such as paints and pigments, tanneries, fertilisers and cleaning and plating baths, etc., [32,33]. Ingestion of higher concentration of copper continuously may cause Wilson disease, liver damage, insomnia, renal damage, vomiting, diarrhoea, and lung cancer [34]. The maximum permissible limit of copper set by the U.S. EPA is 1.3 mg/L [35].

2.7. Nickel

Nickel is a commonly used heavy metal ion and it is also a toxic element. Its toxicity is dispersed from chemical industries, electroplating, mining, refining, and paints, and ink formulation unit [36]. It causes adverse effects of health such as dermatitis, nausea, chronic asthma, coughing and cancer. The U.S. EPA has set the permissible level of nickel for drinking water as 0.015 mg/L [37].

2.8. Zinc

Zinc is generally found in +2 oxidation state. It is an essential element but excess amount of zinc is harmful for life. Zinc is used for numerous industrial activities such as mining, galvanisation, stabilisers, steel production, burning of coal, and purification of zinc [38]. The detrimental effects of zinc are depression, lethargy, neurological signs, and increased thirst and loss of appetite [39]. WHO has fixed the maximum permissible limit of zinc as 5.0 mg/L [40].

Table 1 summarises different heavy metal ions, their adverse effect and permissible limits according to WHO.

3. Conventional methods for removal of heavy metal ions

Various methods are available for removing heavy metal ions which include ion exchange [52,53], chemical precipitation [54,55], coagulation [56], membrane separation [57], electro-coagulation [58], and reverse osmosis [59,60] (Figure 1).

These methods, however, are not generally used due to their high cost and low feasibility. However, adsorption techniques [61] are one of the preferential methods for removal of heavy metals because of their high efficiency and low cost. Adsorption techniques are the most widely used and low-cost alternative technology [62], but the

Figure 1. Conventional methods for the removal of heavy metal ions.

Table 2. Comparison of current treatment methods for the removal of various heavy metal ions [24].

S. No.	Methods	Advantages	Disadvantages	Reference
1.	Ion exchange	High-quality of metal removal, metal selective	Expensive, cannot be used on large scale	[87]
2.	Chemical precipitation	Easy to use, most of metals can be removed	Excess amount of chemical required, huge amount of sludge production, disposal problem	[88]
3.	Coagulation	Suitable to large scale waste water treatment	Expensive, large amount of sludge production and difficulties of disposal.	[89]
4.	Membrane separation	Less amount of chemical used, less sludge production	High maintenance and operational cost	[90]
5.	Reverse osmosis	Effective removal	Required chemicals cost is high, consumption of high power	[91]
6.	Adsorption	High efficiency, highly effective for removal of heavy metal ions	High cost in case of activated carbon	[92]

search for effective adsorbents is still on. Each conventional method has its own advantages and disadvantages (Table 2) [63].

3.1. Ion exchange

Last few decades experienced the use of ion exchange method for the removal of impurity from water [64]. It is a physical/chemical process; in the process of ion exchange method, cations are exchanged with metal ions in wastewater [65–67] (Figure 2).

Synthetic resin and natural zeolites are used in ion exchange process. Zeolites are generally used due to their inexpensive nature and capacity of selective metal removal [68]. The As(V) can be easily removed through by the use of strong base anion exchange resins [69,70]. In ion exchange process removal of As(V) depends on pH of the solution,

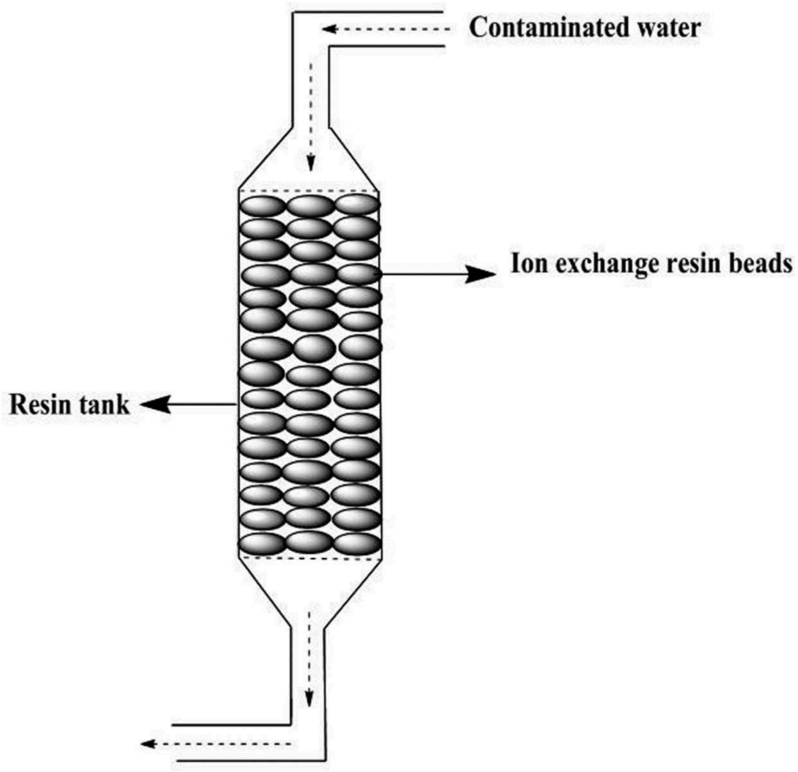


Figure 2. Removal of heavy metal ions by ion exchange process.

concentration of ions, alkalinity and type of resin. The disadvantages of this process are that it is affected by the pH of the solution and it cannot be used on a large scale. In addition, synthetic resins are expensive in nature.

3.2. Chemical precipitation

Chemical precipitation is a commonly used conventional process for removal of heavy metal ions from contaminated water because of its easy use and inexpensive nature [71] (Figure 3). In this process, chemical reagents react with metal ions to form insoluble

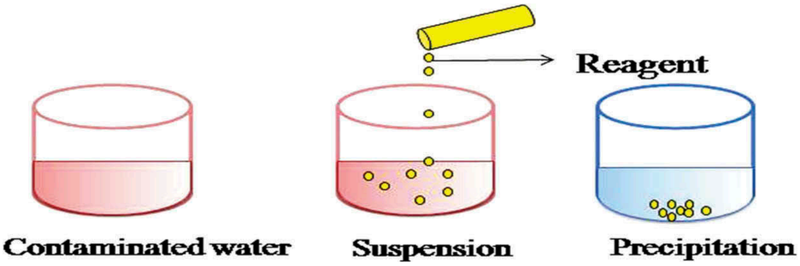


Figure 3. Schematic diagram showing chemical precipitation for removal of heavy metal ions.

precipitates, which can be separated by filtration. Precipitation of metals is obtained by the addition of chemical reagents like alum, lime and limestone. The major disadvantages of chemical precipitation are the requirement of an excess amount of chemical reagents for the treatment of heavy metals, production of sludge to a large quantity, and high cost for disposal of sludge [72].

3.3. Coagulation

Coagulation is another important process for the treatment of contaminated water [73] (Figure 4). In the process of coagulation, some coagulates are used to prepare colloids such as aluminium and ferric chloride. In this process, removal of metal ions depends on initial metal ion concentration, coagulant dosage, and pH of the solution. This process can be used on a large scale for wastewater treatment. The disadvantages of this method are the requirement of in high cost reagent and production of excessive sludge and problem of disposal of this toxic sludge in the environment [74].

3.4. Membrane separation

In membrane separation process, different types of membranes are used for the removal of heavy metal ions in wastewater which are based on size of the particle and the methods are ultra-filtration [75], nanofiltration [76] and electrodialysis [77]. The advantages of these methods are less amount of chemicals used and less production of sludge, but the process suffers from the drawback of high maintenance and operational cost [78].

3.5. Reverse osmosis

Reverse osmosis is used for the separation of heavy metal ions from aqueous medium with the help of a semi-permeable membrane (Figure 5). Upon application of pressure from concentrated side of membrane, purified water flows into the dilute side and the impurities from the high concentration side can be washed out with water.

This method can be used for the removal of different industrial effluents such as electroplating, textile, tanneries, and pulp and paper industries [79]. It has some disadvantages: cost of chemicals is high and it consumes high power for pressure in pumping.

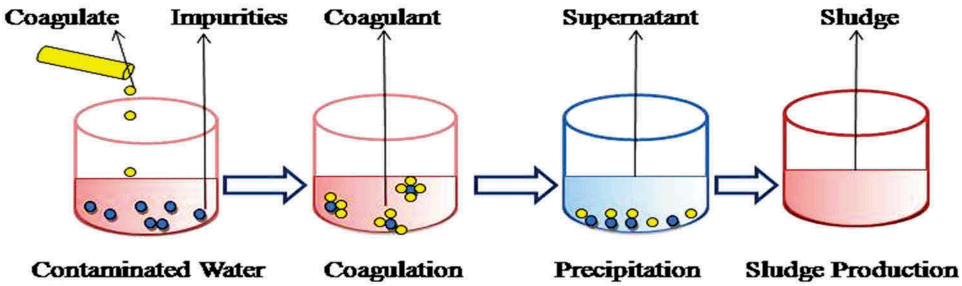


Figure 4. Schematic diagram showing coagulation process used for removal of heavy metal ions.

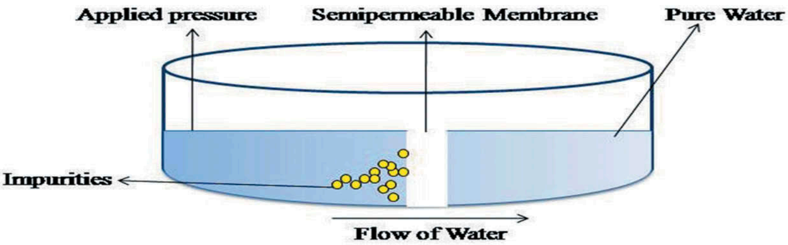


Figure 5. Schematic diagram showing removal of heavy metal ions by reverse osmosis process.

3.6. Adsorption

Adsorption is the accumulation of atoms or ions from a gas, liquid or solid to a surface (Figure 6). This process creates a layer of the adsorbate (metal ions) on the surface of the adsorbent. This process is extensively used due to its high efficiency of effective removal of heavy metal ions and low cost.

Various conventional methods are used for the removal of heavy metal ions, but these methods have many drawbacks such as high cost, huge amount of toxic sludge production and disposal problem, high energy consumption and requirement of an excess amount of chemicals. Therefore, there is a need to develop a suitable treatment method, which is simple, effective and eco-friendly with low cost [80]. In comparison with other methods, adsorption is most preferential method due to their high efficiency and low cost but it has some disadvantages: requirement of chemical regeneration, involvement of

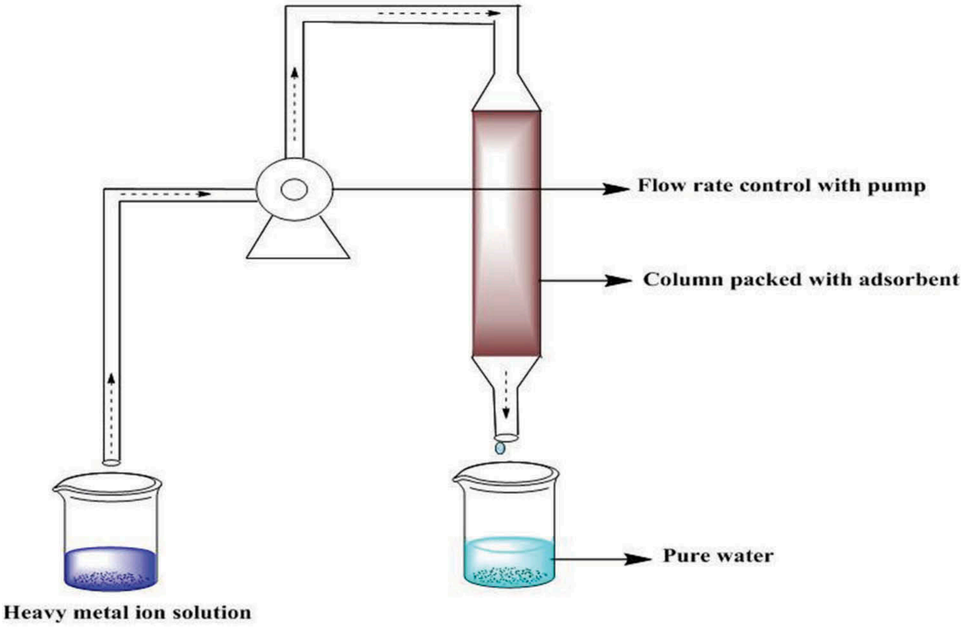


Figure 6. Schematic diagram showing adsorption process by continuous column for heavy metal ions removal.

high cost for preparation of adsorbent such as in case of activated carbon, loss of adsorption capacity by the adsorbent at each regeneration cycle [78].

Natural materials or certain waste materials from industrial and agricultural activities have capacity as inexpensive adsorbent alternatives for removal of heavy metal ions. Cost is an important factor for comparing other adsorbent materials. In general, adsorbents could be assumed to be 'low-cost adsorbent' if they require little processing and are abundantly available, or are waste material from another industry or agricultural by-product [81]. Many examples of natural adsorbents for waste water treatment have been reported in the literature to remove toxic pollutants as colourants and heavy metals and they include sugar beet pulp [82], coal fly ash [83], coir pith [84], rice husk cellulose [85], and castor tree leaf powder [86], etc.

4. Types of adsorbents

4.1. Commercially available adsorbents

Commercial adsorbents are efficient adsorbents for the removal of heavy metals from wastewater due to their high surface area and the various functional groups present on their surface. Commercial adsorbents, such as graphene [93], activated carbon [94] and carbon nanotube [95] are used as complexing adsorbents in order to remove heavy metal ions from wastewater, but these adsorbents are expensive.

4.2. Natural materials

Different types of adsorbent are classified into natural materials such as clays [96], zeolites [97] and siliceous materials [98]. These natural materials are low cost, abundantly available and have significant capability for modification of their adsorption capacity.

4.3. Bio-adsorbents

Adsorption of toxic pollutants from aqueous solutions by the use of biological materials is known as bio-adsorption and these biological materials are termed as bio-adsorbents. These bio-adsorbents and their derivatives consist of various functional groups which can bind the heavy metal ions. Chitins and chitosan [99], peat [100], yeasts [101], fungal biomass [102] or bacterial biomass [103], etc., are used as bio-adsorbents or chelating adsorbents [104].

4.4. Waste materials or by product from agriculture, animal and industry

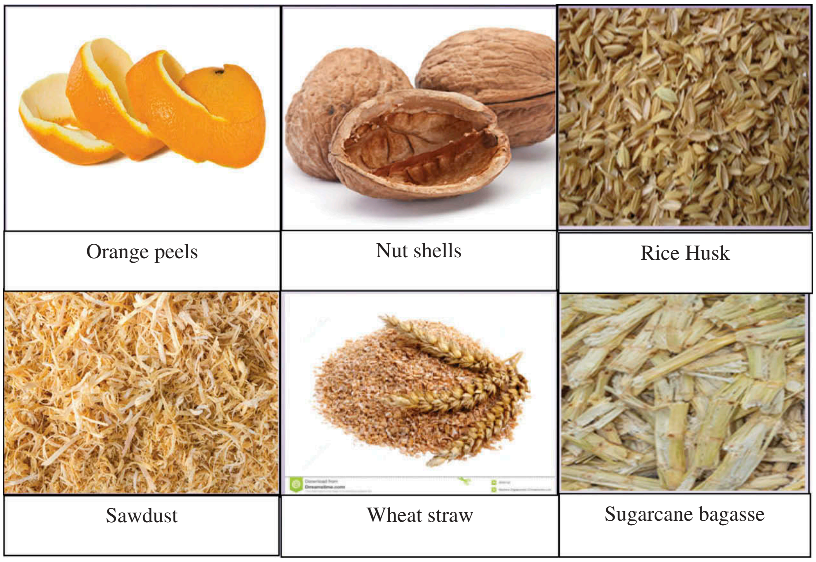
The waste materials from the agriculture, animals and industries could be assumed to be the low-cost adsorbents since they are abundantly available, low cost, and require little processing. There are a number of types of adsorbents that are used for the efficient removal of heavy metals removal from wastewater.

5. Natural waste materials as low-cost adsorbents

The use of low-cost adsorbents to purify heavy metal contaminated water has become more and more popular through the past decade, since they are inexpensive, environmental friendly, highly efficient, and abundant [105]. These low-cost adsorbent materials can be classified into five categories as natural mineral (coal, peat, clays, etc.), agricultural waste (peels, nutshells, rice husk, wheat straw, maize bran, etc.), forest waste (bark, leaves and seeds, etc.), animal waste (chicken feathers, eggshells and crab shells, etc.), and industrial waste (fly ash, red mud and furnace slag, etc.). The main objective of this study is to search for less expensive adsorbents and their contribution for the removal of heavy metal ions present in water.

5.1. Agricultural waste

The occupationally available activated carbon is generally used for the removal of heavy metal ions. Activated carbon is impressive for adsorption of heavy metal ions but its cost is expensive and hence its use is limited. It is therefore necessary to develop low cost and readily available adsorbents for the adsorption of heavy metal ions from wastewater. Agricultural waste is abundantly available in the world and it has improved metal sorption capacity (Figure 7). Its adsorption capacity is less than activated carbon but cost is very low. It is environmental friendly, economically feasible, and highly efficient for removal of heavy metal ions from wastewater. Agricultural waste consists of cellulose, hemicelluloses, lignin, sugar, and starch. Many of agricultural wastes contain numerous functional groups like amine, aldehyde and keto group, etc. These qualities improve the efficiency of agricultural waste for removal of toxic pollutants [106]. Several agricultural solid wastes have so far been suggested as adsorbents to remove heavy metal ions due to the impressive adsorption capacities. Wan Ngah et al. reported the use of chemically modified



Orange peels
Nut shells
Rice Husk
Sawdust
Wheat straw
Sugarcane bagasse

Figure 7. Various agricultural wastes used as low-cost adsorbents.

plant waste as adsorbents to remove heavy metal ions from wastewater. In general, chemically modified adsorbents showed higher adsorption capacity than unmodified adsorbents [107].

5.1.1. Fruit/Vegetable peels

Fruit peels are abundant agricultural wastes which are obtained from juice industry. Fruit peels are very cheap and readily available. Generally, fruit peels have been used for the sorption of heavy metal ions. Feng et al. [108] reported that orange peel is composed of cellulose, pectin, hemicelluloses and lignin. They prepared chemically modified orange peel from hydrolysis of the grafted copolymer and synthesised by interaction of methyl acrylate with cross-linking orange peel. Comparison between modified orange peels and unmodified orange peel in removing Cu(II) ion showed that modified biomass the highest adsorption capacity in wastewater. The maximum adsorption capacity of modified orange peels for Cu(II) ion was reported to be 289.0 mg/g. In another experiment Feng et al. [109] observed that chemically modified reactions including cross-linking and functionalization of orange peel increased the adsorption capacity than the unmodified orange peel. They obtained the maximum adsorption capacity of modified orange peel for Pb^{2+}, Cd^{2+} and Ni^{2+} ions as 476.1, 293.3, and 162.6 mg/g, respectively, at pH 5.5 for a contact time of 150 min. They observed that the adsorption equilibrium data were best described by Langmuir isotherm than the Freundlich isotherm. Liang et al. [110] reported that pristine orange peel reacted with carbon disulphide in alkaline medium and formed orange peel xanthate for the adsorption of Pb^{2+} ion from wastewater. The authors reported that orange peel xanthate showed higher adsorption capacity than the pristine orange peel. They found maximum adsorption capacity of orange peel xanthate as 204.50 mg/g, under optimal conditions (pH = 5.0, contact time = 20 min, temperature = 30°C) and the pseudo-second-order kinetics was applicable to kinetic equilibrium data. In another work, Lugo-Lugo et al. [111] demonstrated the removal of Cr(III) and Fe(III) in single and binary systems using orange peel as an adsorbent with the help of batch experiments. The results indicated that Langmuir adsorption isotherm could be applied for equilibrium experimental data and the maximum adsorption capacity of orange peel for Cr(III) and Fe(III) was 9.43 and 18.19 mg/g, respectively. Moreover, Castro et al. [112] suggested the use of banana peel for the extraction of lead and copper ions from wastewater. By applying Langmuir adsorption isotherm they determined the maximum adsorption capacities of Cu(II) and Pb(II) to be around 20.97 mg/g and 41.44 mg/g, respectively, at pH 3 within 10 min. According to Bhatnagar et al. [113], lemon peel can be utilised as a low-cost adsorbent for the removal of Co(II) ions from wastewater. They obtained maximum adsorption capacity of lemon peel as an adsorbent for Co(II) was 22 mg/g at 25°C and the kinetic experimental data could be described more suitably by the pseudo-second-order kinetic model. Huang et al. [114] reported that muskmelon peel chemically modified with alkaline solution of calcium hydroxide by saponification process could be utilised for the removal of Pb(II) ion. Calcium hydroxide activated the functional group. Muskmelon peel consists of pectic acid and it was responsible for the adsorption of lead ions from aqueous solution. The finding show that the maximum adsorption capacity was 0.81 mol/kg for Pb(II) ions at the pH of 4.5 and reach the equilibrium within 10 min. Sudha et al. [115] prepared activated carbon of citrus limettioides peel and seed for the removal of Pb(II) ions by the use of batch experiment. The reported value of the highest removal

Table 3. Adsorption capacities of fruit peels for the removal of heavy metal ions from waste water.

S. No.	Fruit Peels	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	References
1.	Banana peel	Cu(II)	49.5	92.5	[117]
		Pb(II)	45.6	89.35	
		Cd(II)	30.7	91.89	
		Cr(VI)	25.2	82.88	
2.	Banana peel	Cd(II)	5.71	89.2	[118]
		Pb(II)	2.18	85.3	
3.	Alkali treated citrus limetta peels	Pb(II)	630	-	[119]
4.	Cassava peel	Pb(II)	5.80	73	[120]
		Cu(II)	8.00	79	
5.	Alkali treated lemon peel	Co(II)	35.71	-	[121]
6.	Oak acorn peel	Cr(VI)	47.39	100	[122]
7.	Potato peel	Co(II)	405	92	[123]
8.	Modified litchi peel	Cr(VI)	9.55	100	[124]
9.	Yam peel	Pb(II)	98.36	81.24	[125]

efficiency of citrus limettioides peel and seed was 92% and 31%, respectively. Citrus limettioides peel consists of hydroxyl, carboxylic, and sulphonic acid groups, which showed the ion exchange property. Based on the analysed results, the adsorption data could be applied with the Langmuir isotherm model to confirm monolayer adsorption and the maximum adsorption capacity citrus limettioides peel and seed was found to be 166.67 and 142.86 mg/g, respectively. They also observed that pseudo-second-order kinetics well fitted with the experimental data, and showed chemisorptions process. In another research work, Mallampati et al. [116] reported that tomato peels consists of pectin, carotene, and phenolic compounds. Many of these contain several amino, hydroxyl and carboxylic functional groups, which act as an adsorption site for heavy metal ions. The authors reported that the maximum adsorption capacity of Ni(II) ions was 8.7 mg/g and Langmuir and Freundlich isotherm models were used to describe the adsorption process. Table 3 summarises some of the important results of heavy metal ions adsorption using fruit peels.

5.1.2. Sawdust

Sawdust is generally obtained by sawmills in large quantities as a solid waste. It is easily available and the price is very low. It is composed of lignin, cellulose and hemicelluloses. Sawdust contains polyphenolic groups, which are binders of heavy metal ions. Acar et al. [126] reported sawdust treated with H_2SO_4 at 150°C for 24 hours as adsorbent. The authors demonstrated that the activated poplar sawdust enhanced adsorption capacity than the unmodified sawdust for the removal of Cu(II) ions from aqueous solutions. The maximum adsorption capacities of sawdust and modified sawdust were 5.432 mg/g and 13.495 mg/g, respectively. They also observed that, the maximum removal efficiency of sawdust and activated sawdust were 47.05% and 92.38% at pH 4.0 and 5.0, respectively. According to Ahmad et al. chemically treated sawdust enhanced the adsorption result with increasing pH, temperature, and amount of adsorbent. Langmuir adsorption isotherm could be applied for adsorption data [127]. In another experiment Shukla et al. [128] investigated dye loaded ground nut shell and saw dust showed higher potential for adsorption of Cu(II), Ni(II) and Zn(II) ions than unloaded ground nut shell and saw dust.

Table 4. Adsorption capacities of sawdust for the removal of heavy metal ions from waste water.

S. No.	Sawdust	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	Reference
1.	Modified sawdust	Cr(VI)	8.84	100	[80]
2.	Acid treated sawdust	Cu(II)	286	-	[132]
		Ni(II)	74.1	-	
3.	sodium hydroxide treated sawdust	Cd(II)	73.62	-	[133]
4.	Formaldehyde treated sawdust	Cr(VI)	8.2	-	[134]
5.	Modified wood sawdust	Zn(II)	105	89	[135]
6.	Modified sawdust	Cr(VI)	238.6	-	[136]
		As(V)	71.23	-	
7.	Beech sawdust	Cu(II)	4.5	-	[137]
		Ni(II)	4	-	
		Zn(II)	2	-	
8.	Picea smithiana sawdust	Pb(II)	6.35	95.2	[138]
		Cr(VI)	3.37	87.7	
		Cd(II)	2.87	83.3	

Adsorption capacities of dye loaded sawdust of Cu(II), Ni(II) and Zn(II) were 8.07 mg/g, 9.87 mg/g and 17.09 mg/g respectively, which were higher than unloaded sawdust. In addition, Setyono et al. [129]reported that chemically modified sawdust can remove arsenic from aqueous solution. La or Zr oxide was included on the surface of sawdust and used for the removal of arsenite and arsenate anions. The maximum adsorption capacities of lanthanum oxide (La_2O_3) treated sawdust for arsenite and arsenate were 22 and 28 mg/g respectively, while zirconium oxide (ZrO_2) treated sawdust were 29 and 12 mg/g for arsenite and arsenate, respectively, at pH 7. These experiments showed that surface modified sawdust has higher adsorption capacity. According to Argun et al. [130] acid (HCl) treated oak sawdust was appropriate for the removal of Cu(II), Ni(II), and Cr(VI) ions from wastewater. The reported value of maximum removal efficiencies of oak sawdust were 93% for Cu(II) at pH 4, 82% for Ni(II) at pH 8, and 84% for Cr(VI) at pH 3. They also analysed that the kinetic experimental data followed by the pseudo-second-order kinetics. Besides, Rabago et al. [131] reported white pine sawdust chemically treated with citric, malonic, and tartaric acids and used for the removal of Pb(II) ions. The functional carboxylic group improved the adsorption capacity. They observed that the citric acid modified sawdust had higher adsorption capacity than the other modifications. Maximum adsorption capacity of citric acid modified sawdust was 304 mg/g at pH 5 and 25°C. Adsorption process was dependent on the pH of the solutions. Table 4 presents some data on the adsorptive capacities of the sawdust for different heavy metal ions reported in literatures.

5.1.3. *Nut shells/Hulls*

Nut shells are abundant in the world and are one of the popular low-cost adsorbents for the adsorption of heavy metal ions. Pehlivan et al. [139] observed that the shells of walnut (WNS), hazelnut (HNS) and almond (AS) have a capacity to remove Cr(VI) ion by the batch experiments. Adsorption capacity of the shells was dependent on the pH of the chromium solutions. The highest adsorption capacities of WNS, HNS and AS were obtained at pH values of 3.5 (8.01 mg/g), 3.5 (8.28 mg/g) and 3.2 (3.40 mg/g), respectively, and hazelnut shell showed highest adsorption capacity. They also showed that, the maximum removal

Table 5. Adsorption capacities of nut shells for the removal of different heavy metal ions from waste water.

S. No.	Nut shells/Hulls	Heavy metals	Adsorption capacity (mg/g)	Maximum removal efficiency (%)	Reference
1.	Alkali treated almond shell	Pb(II)	9	-	[145]
		Cd(II)	7	-	
2.	Cashew nut shell	Cd(II)	22.11	80.13	[146]
3.	Palm shell	Hg(II)	83.33	-	[147]
4.	Wheat shell	Cu(II)	0.83	99	[148]
5.	Hazelnut shell	Cu(II)	58.27	-	[149]
6.	Walnut shell	Cu(II)	6.74	80.3	[150]
7.	Alkali treated walnut shell	Cd(II)	14.29	-	[151]
8.	Hazelnut Shell activated carbon	Cr(VI)	170	-	[152]

efficiency of WNS, HNS and AS were 85.32, 88.46 and 55.0%, respectively, at a concentration of 0.5 mM in 100 min. In another research work, Ahmadpour et al. [140] used chemically modified almond green hull for the removal of Co(II) ions from wastewater by batch technique. They observed that the experimental data followed Langmuir and Freundlich isotherms and pseudo-second-order model were applied to the kinetic data. They achieved maximum Co(II) ions adsorption capacity of 45.5 mg/g in 1 min. Senthilkumar et al. [141] investigated adsorption of Cu(II) ions by cashew nut shell by batch technique. They found maximum adsorption capacity of 20.00 mg/g for Cu(II) ions, under optimal conditions (pH = 5, contact time = 30 min, temperature = 30ºC, initial concentration of metal ion = 20 mg/L, adsorbent dose = 3 g/L). They also analysed that the adsorption process followed Langmuir and Freundlich isotherm and pseudo-second-order kinetics. In the similar study, Senthilkumar et al. [142] investigated the shell of cashew nut utilised for the removal of Ni(II) ions. The adsorption of Ni(II) were studied at different parameters such as pH of solution, adsorbent dose, contact time, initial concentration of Ni(II) ions, and temperature. The maximum adsorption capacity was 18.86 mg/g and highest removal efficiency was 73.89% at pH 5, the contact time 30 min, the temperature of 30ºC, initial Ni(II) concentration of 20 mg/L and adsorbent dose of 3 g/L. The adsorption data was applied to the Langmuir adsorption isotherm that confirmed monolayer adsorption. Furthermore, Yin et al. [143] prepared chemically modified adsorbent from agricultural residues buckwheat hulls chemically treated with 1- hydroxylethylidenediphosphonic acid for the removal of Au(III) ions from wastewater. The maximum adsorption capacity for Au(III) was 450.45 mg/g at 35°C and adsorption data was consistent with Langmuir isotherm. Ali et al. [144] utilised peanut hull as an adsorbent for the removal of Cu(II) ions from aqueous solutions by batch experiments. The reported value of the highest adsorption capacity of Ni(II) ions was 14.13 mg/g and Langmuir and Freundlich adsorption models were used for describing the adsorption process. Table 5 shows that various heavy metal ions could be adsorbed by nut shells.

5.1.4. Husks

Rice husk is an agricultural waste, a by-product obtained from rice mill. This is composed of cellulose (32.24%), hemicelluloses (21.34%), lignin (21.44%), and mineral ash (15.05%) [153]. It has been used for the removal of heavy metal ions of modified and unmodified form. Song et al. [154] reported that rice husk functionalised with sulphur (RH-CS) and

Table 6. Adsorption capacities of husks for the removal of heavy metal ions from wastewater.

S. No.	Husks	Heavy Metals	Adsorption Capacities (mg/g)	Maximum removal efficiency (%)	References
1.	Peanut husk biochar	Cd(II)	28.99	99.9	[158]
2.	Sulphuric acid treated peanut husk	Pb(II)	29.14	-	[159]
		Cr(III)	7.67	-	
		Cu(II)	10.15	-	
3.	Sulphuric acid treated Groundnut husk with silver impregnation	Cr(VI)	11.4	97	[160]
4.	Hazelnut husk	Cd(II)	20.9	-	[161]
5.	Acid treated groundnut husk	Pb(II)	31.62	-	[162]
		Cd(II)	29.78	-	

organosilane grafted rice husk (RH-GM) exhibit maximum adsorption capacity of Hg(II) 89 and 118 mg/g, respectively. The value of point of zero charge (pH_{pzc}) for RH-Raw, RH-NaOH, RH-CS and RH-GM was 5.2, 6.8, 7.4 and 7.1, respectively. After modification pH_{pzc} increased, which indicate that the adsorbent become more positive. In addition, Sobhanardakani et al. [63] used unmodified rice husk, as an organic solid waste for the adsorption of Cr(III) and Cu(II) from wastewater. The highest adsorption capacities of 30.0 and 22.5 mg/g of Cr(III) and Cu(II), respectively, were obtained. The maximum removal efficiency was 100% for both the metal ions at pH 5–6 for a contact time of 60 min and adsorbent dose 5 and 4 g, respectively. In another experiment Rehman et al. [155] demonstrated that chemically modified rice husk and sawdust with polyaniline have the potential for adsorption of Cd(II) from wastewater with the help of batch technique. They used polyaniline in three forms, simple polyaniline, rice husk modified with polyaniline and sawdust treated with polyaniline. Among these, modified rice husk showed the highest adsorption capacity. Coconut husk consists of coconut pith and coconut fibre. Johari et al. [156] used this as a low-cost adsorbent for the removal of Hg^0 from wastewater. They reported that chemically treated coconut husk enhanced the adsorption capacity. They modified coconut pith and fibres in different forms, in which sodium hydroxide treated coconut pith exhibited maximum adsorption capacity of 956.28 ng/g. Pseudo-second-order kinetics was applicable to the experimental data. According to Ricordel et al. [157] carbon of peanut husk is a valuable adsorbent due to its economic feasibility and the capacity for the removal of Pb(II), Cd(II), Zn(II) and Ni(II) ions. Adsorption process was based on particle size distribution and amount of adsorbents. The surface area of peanut husk was found to be 485 m^2/g and pore diameters were less than 3.5 nm. The experimental study was performed in a batch technique. Table 6 lists the adsorption capacities of heavy metal ions onto husks.

5.1.5. Straws

Rice straws are cheap and produced worldwide. As an adsorbent, rice straw exhibits high removal capacity due to its high composition of cellulose, hemicellulose and lignin [163]. Wu et al. [164] used the chemically modified rice straw for the removal of Cr(VI) and Ni(II) ions from single and binary systems. Modified rice straw had a much higher adsorption capacities for Cr(VI) and Ni(II) ions as compared to unmodified rice straw. Under optimum conditions in terms of initial metal ion concentration (200 and 80 mg/L for Cr(VI) and Ni(II), respectively), temperature (318 K), adsorbent dosage (10 g/L) and pH (2 and 7 for Cr(VI) and

Ni(II), respectively), maximum adsorption capacities were 15.82 mg/g for Cr(VI) and 3.95 mg/g for Ni(II) ions from modified rice straws. Functionalization with amine group enhanced the adsorption capacities of rice straw for Cr and Ni ions. According to Song et al. [165] synthesised mercaptan-grafted rice straw (RS-GM) to improve the adsorption capacity of Hg(II) ions. The specific surface area of RS-Raw and RS-GM was obtained as 8.46 m^2/g and 11.46 m^2/g, respectively. The maximum adsorption capacity was found to be 103.10 mg/g and 161.30 mg/g for RS-Raw and RS-GM, respectively. The experimental study was performed in a batch technique and the pH of the solution, amount of adsorbent, contact time and temperature influenced the adsorption behaviour. They found the maximum removal efficiency of 96% for Hg(II) ions by using the RS-GM. In another experiment, Pehlivan et al. [166] reported that barley straw has capability to remove Cu(II) and Pb(II) ions from aqueous solution by batch technique in adsorption process. Moreover, experiments were performed on the influence of different parameters such as pH (2–9), contact time (10–240 min), initial metal ion concentration (0.1–1 mM), and the adsorbent dosage (0.1–1.0 g). Finally, under optimum condition (pH of 6, contact time 2 h, and adsorbent amount of 1 g/L), they obtained maximum removal efficiency of 69 and 88% for the Cu(II) and Pb(II) ions, respectively. The removal efficiency of metal ions increased with an increase in amount of chemically treated barley straw and pH of metal solutions. The reported value of maximum adsorption capacity was 4.64 mg/g and 23.20 mg/g for Cu(II) and Pb(II) ions, respectively. In a similar experiment, Pehlivan et al. [167] reported that barley straw treated with citric acid enhanced the adsorption capacity for Cu(II) ions. The adsorption capacities were reported as 4.64 mg/g and 31.71 mg/g for raw barley straw and citric acid barley straw respectively. The maximum removal efficiency of citric acid barley straw was found to be 88.1% at pH 7 in 120 min for Cu(II) ions from aqueous solution. Adsorption capacity was dependent on the functional carboxylic group of modified barley straw. Results show that the adsorption process followed Langmuir, Freundlich, Scatchard and D-R (Dubinin-Radushkevich) isotherm models. Wheat straw is obtained in large amount and at low cost. It is composed of 34–40% cellulose, 20–35% hemicelluloses, 8–15% lignin and sugars [168]. In another work, Gorgievski et al. [169] observed that wheat straw can be utilised as an adsorbent to remove heavy metal ions such as Cu(II), Ni(II) and Zn(II) ions from aqueous solution. FTIR analysis of wheat straw indicated that in the hydroxyl group helps binding the Cu(II) metal ion and is responsible for its removal from aqueous solution. Adsorption by wheat straw followed Langmuir isotherm model and the maximum adsorption capacity was found to be 5 mg/g, 2.5 mg/g and 3.25 mg/g for Cu(II), Ni(II), and Zn(II) ions respectively. Table 7 summarises the results of the important heavy metal ions investigated using wheat straws.

Table 7. Adsorption capacities of wheat straws for the removal of heavy metal ions from wastewater.

S. No.	Straws	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	Reference
1.	Wheat straw	Cu(II)	7.05	-	[170]
2.	Wheat straw treated with urea and microwave radiation	Pb(II)	31.85	-	[171]
3.	Wheat straw	Cd(II)	14.56	-	[172]
		Cu(II)	11.43	-	
4.	Wheat straw	Pb(II)	599.1	99.8	[173]
5.	Wheat straw	Cr(III)	21.0	-	[174]

5.1.6. Brans

Wheat bran is the shell of wheat seed. This bran is a by-product obtained from wheat milling industries. Wheat bran has not only nutritions, it has been reported to have some anticancer characteristics [175]. Bulut et al. [6] reported that wheat bran has potential to remove Pb ions from wastewater by batch technique. The adsorption capacities of wheat bran for removal of Pb(II) ions were 69.0 mg/g, 80.7 mg/g and 87.0 mg/g at 20°C, 40°C and 60°C, respectively, within 60 min. Adsorption process was endothermic in nature. In addition, Ozer et al. [176] showed that sulphuric acid treated wheat bran improve the adsorption capacity due to enhancement of the surface area by the acid. The maximum adsorption capacity of Cd(II) ions was 101.0 mg/g at pH of 5.4, contact time of 4 h, temperature of 25°C, and initial Cd(II) ion concentration of 100 mg/L. They also achieved the maximum removal efficiency of 86.2% for Cd(II) ion. Langmuir isotherm was applicable to the experimental data and the thermodynamic studies confirmed the adsorption process as an exothermic one. Singh et al. [177] used wheat bran for the removal of Cd(II) ions. The maximum removal efficiency was 87.15% for Cd(II) ions, under optimum conditions (pH = 8.6, temperature = 20°C, and initial Cd(II) concentration = 12.5 mg/L) and indicating the adsorption process to be monolayer adsorption. The adsorption was spontaneous and exothermic. In another work, Singh et al. [178] utilised maize bran as a low-cost adsorbent for the adsorption of Pb(II) ion from water sample and reported value of maximum removal efficiency as 98.4% at pH 6.5 for an initial concentration of 100 mg/L and 20°C. The adsorption behaviour was explained well by Langmuir isotherm model, Thermodynamic studies proved the process to be spontaneous and exothermic. Rice bran is a by-product of rice milling industries. It is widely used as a fertiliser. Rice bran consists of hemicellulose A, hemicellulose B and hemicellulose C, which have metal binding capacity. Hu et al. [179] reported that hemicellulose B showed the highest adsorption capacity than the hemicelluloses A and hemicellulose C. The authors determined the capacity of rice bran hemicellulose A, B, C and rice bran dietary fibres binding to heavy metal ions such as Pb, Cu, Cd, and Zn. Zafar et al. [180] used acid (H_3PO_4) treated rice bran as adsorbent. The maximum adsorption capacity of Ni(II) ion was reported as 102 mg/g, under optimal conditions (pH = 6, initial concentration of Ni(II) ion = 100 mg/L, and adsorbent dosage = 0.25 g) and they observed Metal adsorption capacity decreased at lower pH values. Adsorption process was described well by Langmuir and Freundlich isotherms. Results on heavy metal ions adsorption onto brans from different researchers are shown in Table 8.

Table 8. Adsorption capacities of brans for the removal of heavy metal ions from wastewater.

S. No.	Brans	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	References
1.	Wheat bran	Cd(II)	15.71	-	[181]
2.	Acid treated wheat bran	Cr(VI)	5.28	90	[182]
3.	Rice bran	Pb(II)	416.61	74.54	[183]
4.	Maize bran	Cr(VI)	312.52	-	[184]
5.	Sodium hydroxide treated rice bran	Ni(II)	153.6	-	[185]

5.1.7. Tea/coffee waste

Tea is the most popular beverage in India. Tea wastes are obtained by tea processing factories and cafeterias. Tea leaves are composed of cellulose, hemicelluloses, lignin, and condensed tannins with some functional groups such as carboxylate, aromatic carboxylate, phenolic, hydroxyl and oxyl groups [186]. These functional groups are responsible for removal of heavy metal ions. In recent years, black tea wastes have been utilised as low-cost adsorbents for removal of some heavy metal ions due to its high adsorption capacity. Weng et al. [187] treated black tea waste with base (NaOH) solution and used as adsorbent for the removal of Cu(II) ions. The base treated tea waste has higher adsorption capacity than the activated carbon. The maximum adsorption capacity of Cu(II) ion was 43.18 mg/g at pH 4.4. Functional groups – OH, – CH and – C = C were responsible for Cu(II) metal ion binding. The maximum removal efficiency was 90% for Cu(II) ion within 10 min. Thermodynamic studies showed adsorption process to be endothermic in nature. In another work, Amarasinghe et al. [188] prepared adsorbent using tea waste for the adsorption of Cu(II) and Pb(II) ions from wastewater. Adsorption efficiency increased with rise in solution pH with a value around 5–6. The adsorption process was described by Langmuir and Freundlich isotherm model and the maximum adsorption capacities were reported as 48 mg/g and 65 mg/g for Cu(II) and Pb(II) ions, respectively. Cu(II) and Pb(II) ions removal efficiency was fast ca. 90% removal within 15–20 min of contact time. Prabhakaran et al. [189]reported that spent tea and coffee dusts have capacity for the adsorption of Cr(VI) from water sample. The Cr(VI) ions are reduced to Cr(III) ions and removal of Cr(III) ions depended on the pH of the solutions. The maximum chromium removal exhibited by spent tea and coffee dusts were 44.9 mg/g and 39.0 mg/g, respectively, at pH 4. Langmuir isotherm and Toth isotherm models were used to describe adsorption method, among them Toth isotherm better explained the adsorption process. Mohammed et al. [190] used black tea waste in removal of heavy metal ions such as Co, Cd and Zn from wastewater. Maximum adsorption capacity was reported around 15.39 mg/g, 13.77 mg/g and 12.24 mg/g for Co, Cd and Zn ions, respectively, at pH 6 in 180 min. Langmuir and Freundlich isotherm models were used to explain experimental adsorption data and adsorption kinetics could be better explained by the pseudo-first-order reaction. Besides, Cordova et al. [191] modified the spent coffee grains with citric acid solutions; carboxylic functional group present was reported to enhance the adsorption capacity of metal ions. The batch technique was used for the removal of lead and copper from aqueous solution at 30°C. Langmuir adsorption model was used for elucidation of the adsorption process and maximum adsorption capacities reported were 0.77 mmol/g at pH 4 and 1.53 mmol/g at pH 5 for Pb(II) and Cu(II) ions, respectively. Table 9 reports a summary of adsorption capacities for heavy metal ions onto tea/coffee waste.

Table 9. Adsorption capacities of tea/coffee waste for the removal of heavy metal ions from waste water.

S. No.	Tea/Coffee waste	Heavy metal	Adsorption capacity (mg/g)	Maximum removal efficiency (%)	Reference
1.	Waste tea leaves	Pb(II)	73	96	[192]
2.	Exhausted ground coffee waste	Cr(VI)	10.2	-	[193]
3.	Spent tea leaves	Pb(II)	83–130	98–99	[194]
4.	Coffee residue	Pb(II)	9.7	96	[195]
		Zn(II)	4.4	44	
5.	Coffee grounds	Cd(II)	15.65	-	[196]

5.1.8. Sugarcane bagasse

Sugarcane bagasse is a by-product and fibrous material. It is a low-cost material remaining after juice extracting. The lignocellulosic agricultural waste consists of 40% cellulose, 24% hemicellulose and 25% lignin [197]. Various functional groups such as carboxylic, carbonyl, amine, and hydroxyl groups are present in it, and bind to the metal ions. Ramos et al. [198] used chemically treated sugarcane bagasse with phthalic anhydride for the removal of Co(II), Cu(II) and Ni(II) ions from aqueous solution. The authors used batch technique for adsorption of these heavy metal ions. The monocomponent isotherm model was depicted by the Langmuir isotherm and the reported adsorption capacities were 0.561 mg/g for Co(II), 0.935 mg/g for Cu(II) and 0.932 mg/g for Ni(II) ions. In a similar experiment Ramos et al. [199] prepared modified sugarcane bagasse with trimellitic anhydride for the removal of Co(II), Cu(II) and Ni(II) ions. The maximum adsorption capacities were 1.140 mmol/g, 1.197 mmol/g and 1.563 mmol/g for Co(II), Cu(II) and Ni(II) ions, respectively. According to Yu et al. [200] modified sugarcane bagasse have better adsorption capacity than raw sugarcane bagasse. Sugarcane bagasse chemically treated with pyromellitic dianhydride were used for the removal of Pb(II), Cd(II), Cu(II) and Zn(II) ions from aqueous solution. The maximum adsorption capacities of the modified sugarcane bagasse were 1.06, 0.93, 1.21, and 1.00 mmol/g for Pb(II), Cd(II), Cu(II) and Zn(II) ions, respectively, whereas, raw sugarcane bagasse exhibited the adsorption capacities of 0.04, 0.13, 0.10 and 0.07 mmol/g in that order. Homagai et al. [201] prepared charred sugarcane bagasse with carbon-disulphide (CS_2) in alkaline medium for the removal of cadmium, lead, nickel, zinc, and copper from aqueous solution. The physical and chemical characterisation of adsorbent was confirmed by SEM, FTIR, and TG/DTA analysis. The maximum adsorption capacity of charred xanthated sugarcane bagasse were evaluated at 225, 318, 144, 164, and 178 mg/g for Cd(II), Pb(II), Ni(II), Zn(II) and Cu(II), respectively. Therefore, the results showed that the reaction of xanthate group improves the adsorption capacity of untreated sugarcane bagasse. Table 10 shows a summary of adsorption capacities of sugarcane bagasse for different heavy metal ions.

5.2. Animal waste materials

Animal waste materials are distributed in urban areas and countryside (Figure 8). These materials are obtained from animal processing industries, houses, restaurants and comprise some fibrous protein, calcium carbonate, chitin and organic protein. Metal ions can

Table 10. Adsorption capacities of sugarcane bagasse for the removal of heavy metal ions from wastewater.

S. No.	Sugarcane Bagasse	Heavy Metals	Adsorption Capacities (mg/g)	Maximum removal efficiency (%)	Reference
1.	Sugarcane bagasse	Hg(I)	35.71	97.58	[202]
2.	Sugarcane bagasse treated with thiophosphoryl chloride	Cd(II)	74	-	[203]
3.	Sugarcane bagasse modified with triethylenetetramine	Cu(II)	133	-	[204]
		Pb(II)	313	-	
		Cd(II)	313	-	
4.	Acid (H_2SO_4) treated sugarcane bagasse	Pb(II)	7.297	-	[205]
5.	Immobilised sugarcane bagasse	Cr(VI)	-	80.6	[206]

Figure 8. Various animal wastes using as low-cost adsorbents.

be removed by using natural animal waste materials (poultry waste like hen feathers, eggshell and membrane, crab shell, mollusc shells, *Catla catla* fish scale and bone composite, etc.) as an adsorbent [207–212]. The materials also have potential to adsorb heavy metal ions in a large amount.

5.2.1. Chicken feathers

Several million pounds of waste feathers are obtained by the poultry processing plants. They are very light weight and consist of keratin protein nearly 90–91%. These keratin proteins contain carboxyl, hydroxyl, and amine groups. They act as binders of the metal ion and remove them from wastewater. Villarreal et al. [213] reported that raw chicken feathers could be utilised for the removal of zinc ions from aqueous solutions by the batch and continuous systems. The maximum adsorption capacity was found to be 4.31 mg/g of Zn(II) at 30°C with pH 5. Al-Asheh et al. [214] found that natural and chemically treated chicken feathers have the potential to remove heavy metal ions such as copper and zinc ions from wastewater sample. The authors reported that chemically treated chicken feathers such as alkaline solution of 0.2 M NaOH gave the enhanced adsorption and anionic surfactant dodecyl sulphate also promoted adsorption as compared to untreated chicken feathers. In another experiment, Khosa et al. [215] reported ground chicken feather chemically modified with four dopants such as poly (ethylene glycol) diglycidyl ether (PEG), poly (N- isopropylacrylamide) (PNIPAM), allyl alcohol (AA), and trisilanolphenyl polyhedral oligomeric silsesquioxanes (POSS). After modification authors compared the modified biopolymers and unmodified chicken feathers by different characterisation techniques. The modified biopolymers were used for the removal of As(III) ion in wastewater. The authors observed that AA and POSS treated biosorbents have higher adsorption capacity for As(III) ion of nearly 11.5×10^{-2} and 11.0×10^{-2} mg/g in arsenic-contaminated water, respectively. A study by Rosa et al. [216] showed that chicken feathers can be used as a natural sorbents for adsorption of Pb from wastewater. Maximum adsorption capacity of Pb^{2+} ion was 0.8–8.3 mg/g at 30°C and pH 5. The authors found that metal sorption capacity depends on pH of metal solutions. Freundlich isotherm model and pseudo-second-order kinetics were applied in the adsorption experimental data. Sun et al. [217] demonstrated that chemically treated chicken feathers have higher adsorption capacity than raw chicken feathers. They showed that NaOH treated chicken feathers (CFs) have low sorption capacities for the removal of Cr(VI) ions but the capacity was higher than the raw chicken feathers and this process was physical in nature. The authors showed that CFs cross linked with epichlorohydrin (Epi) have higher adsorption capacity for the adsorption of Cr(VI) ion and this process was chemisorption. Finally, the authors treated CFs with ethylenediamine (EA) in aqueous NaOH solution and cross-linked with epichlorohydrin. Ethylenediamine epichlorohydrin chicken feathers EAEpiCF showed 90% capacity for the removal of Cr(VI) ions. Reynel-Avila et al. [218] showed that adsorption of Cd^{2+} and Ni^{2+} ions by chicken feathers was mainly dependent on pH and temperature. Thermodynamic studies showed that removal of Cd^{2+} and Ni^{2+} ions was controlled by chemisorptions process. The extent of adsorption was 0.039 mmol/g 0.065 mmol/g for Cd^{2+} and Ni^{2+} ions, respectively, at pH 5 and 30°C.

5.2.2. Eggshells

Hen eggshell wastes are obtained from poultries, homes, restaurants and bakery industries. It has some adsorption characteristics such as developed porosity, $CaCO_3$ and protein acid mucopolysaccharides. This protein acid mucopolysaccharides contain various functional groups such as hydroxyl (–OH), carboxyl (–COOH), amino ($–NH_2$), thiol (–SH) and amide ($–CONH_2$) etc., which can be used for binding of heavy metal ions [219]. Wang et al. [220] synthesised eggshell membrane through ammonium thioglycolate and functionalised with thiol group. The thiol functional group is an outstanding ligand for binding various heavy metal ions due to its strong attraction power of Lewis acid–base interactions. Modified eggshell membrane was used for the removal of heavy metal ions such as Cr(VI), Hg(II), Cu(II), Pb(II), Cd(II) and Ag(I) from water sample. The authors demonstrated that the adsorption capability of the thiol functionalised eggshell membrane of Cr(VI), Hg(II), Cu(II), Pb(II), Cd(II), and Ag(I) enhanced 1.6, 5.5, 7.7, 12.4, 12.7, and 21.1 fold, respectively, than the untreated eggshell membrane. Cano et al. [221] investigated the adsorption capacity of eggshell for adsorption of Cd(II) ions from water sample, and various characterisation techniques were used for analysis of eggshell in detail. The authors examined $CaCO_3$ as the main component of eggshell. The eggshell membrane and calcareous layers morphologies were examined through the SEM analysis. Amines and amides functional groups were present in the eggshell membrane and carbonate was found in the calcareous layer. The adsorption capacity was improved with increasing temperature and pH of the solutions. In another work, Liao et al. [222] synthesised carbonate hydroxyapatite by eggshell waste, and utilised for the removal of Pb(II) ions from aqueous solutions. The adsorption of Pb(II) ions were studied at different parameters such as pH, contact time, and initial concentration, etc., through the batch technique. The maximum adsorption of Pb(II) ions was at pH 6.0 and 60 min to reach the equilibrium. The maximum removal of Pb(II) ions was found to be 101 mg/g. Aldaco et al. [223] expelled calcium solution from eggshell waste and then chemically modified with acid (H_3PO_4) for use as low-cost adsorbent. Eggshell wastes after

Table 11. Adsorption capacities of eggshell waste for the removal of heavy metal ions from wastewater.

S. No.	EggShell Waste	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	Reference
1.	Eggshell	Cr(III)	160	-	[224]
2.	Eggshell	Cd(II)	329	-	[225]
3.	Thiol-functionalised eggshell membrane	Hg(II)	71.9	96	[226]
4.	Eggshell waste	Cd(II)	111.1	94	[227]
		Cu(II)	142.86	93.17	
5.	Eggshell membrane	Au(I)	147	-	[228]
		Au(III)	618	-	
6.	Eggshell nano-particle	Hg(II)	116.27	-	[229]

modifications showed improved adsorption properties. The authors used Taguchi method to optimise conditions of adsorbent. The modified eggshell wastes exhibited maximum adsorption capacities of Cd(II), Ni(II) and Zn(II) ions at pH 5 with 30°C and the results may increase up to 15 times than unmodified adsorbent. Table 11 shows the maximum adsorption capacities of eggshell waste used as an adsorbent for the removal of heavy metal ions.

5.2.3. Crab shells/chitosan

Crab shell particles are generally obtained from crabmeat processing plant. The main components of crab shells are calcium carbonate 58%, chitin 17%, protein 10%, moisture 13%, and other substances 2% [230]. Among these, calcium carbonate plays a major role for the treatment of wastewater since it has strong efficiency to forms complexes with chelating agents and metal-carbonate precipitates [231]. Chitin is a natural polysaccharide, consisting of poly-β-(1,4)-N-acetyl-D-glucosamin and chitosan, a polyglucosamin, which can be synthesised by the N-deacetylation of chitin [232]. Chitosan is hydrophilic and a natural cationic polymer. It consists of a large number of amino functional groups, which enhances the adsorption ability of chitosan as compared to the chitin. Pradhan et al. [233] investigated crab shell waste partially converted into chitosan and used for the removal of Ni(II) ions from wastewater. Chitosan is a chelating agent and can bind transition metal ions. The authors reported that adsorption process cannot be well described by the Langmuir and Freundlich isotherms and mechanism appeared to be complicated. Vijayaraghavan et al. [234] used crab shell particles (CSP) for the removal of copper and cobalt from wastewater. Different parameters such as amount of biosorbents, particle size, and initial metal concentration affected the metal removal capacity of CSP. Adsorption process was dependent on pH of the solutions. Based on the results of the CSP, the maximum adsorption capacities were 243.9 mg/g and 322.6 mg/g of Cu(II) and Co(II) respectively, at pH 6, biosorbent dosage 5 g/L, time 2 h, and particle size 0.767 mm. In another experiment Vijayaraghavan et al. [235] used crab shell particles (CSP) for the adsorption of Mn(II) and Zn(II) ions from aqueous medium. Adsorption of both the metal ions on crab shell particles were confirmed by EDX analysis. Adsorption capacity was dependent on pH of the solutions. CSP could effectively bind metal ions with the maximum adsorption capacities of 69.9 mg/g and 123.7 mg/g for Mn(II) and Zn(II) ions respectively, at pH 6 as determined by the Langmuir isotherm model. the adsorption process was fast (90% removal in 120 min for Mn(II) and 90 min for Zn(II) at an initial metal concentration of 500 mg/L). Table 12 presentS the maximum adsorption capacities of crab shell/chitosan for removal of heavy metal ions.

Table 12. Adsorption capacities of crab shell/chitosan for the removal of heavy metal ions from wastewater.

S. No.	Crab Shells/chitosan	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	Reference
1.	Crab shell particles	Co(II)	510	-	[230]
2.	Crab shell particles	Zn(II)	71.46	-	[236]
3.	Chitosan beads	Cr(III)	30.03	-	[237]
		Cr(VI)	76.92	-	

5.2.4. Fish scales

Fish scales are obtained in large volume everyday from fish markets. Most fish scales contain organic protein (collagen) and show almost similar removal capacity of heavy metal ions. Collagen consists of many functional groups such as phosphate, carboxyl, amine, and amide to offer the potential for biosorption of heavy metal ions. Mustafiz et al. [238] reported that fish scales of Atlantic cod were a better adsorbent to the removal of lead, arsenic and chromium from waste water. Zayadi et al. [239] studied fish scale of Tilapia Mossambica as adsorbent for the removal of zinc from waste water. Batch technique was used for adsorption process and they studied the influence of different parameters such as pH of solution, amount of adsorbents, contact time and initial metal ion concentrations on adsorption. The maximum removal efficiency of zinc ion was reported as 93.52% at pH 6, within 3 h and 0.02 g of fish scale in fixed metal concentration of 10 ppb. Srividya et al. [240] reported that Catla catla fish scales have capacity to remove Cr(VI) ions from aqueous solutions. Freundlich isotherm model and pseudo-second-order kinetics better described biosorption of Cr(VI) ions. The authors reported FTIR analysis of biosorbents to indicate that O–H, N–H and C–O groups bind to the Cr(VI) ions.

5.3. Industrial wastes

In many countries, the lack of adequate treatment of industrial waste, has been severe problem and requires urgent attention. This solid waste can be beneficially reused; it can be converted to 'low-cost adsorbents'. Industrial wastes are also feasible to remove the pollutants from wastewaters [241]. Generally industrial wastes are produced as by-products. These materials are abundant and inexpensive (Figure 9). Various types of industrial waste materials are available such as fly ash, red mud, lignin, blast furnace sludge, waste slurry, etc. and many more are currently under investigation. Chemically modified industrial wastes demonstrate higher adsorption capacity.

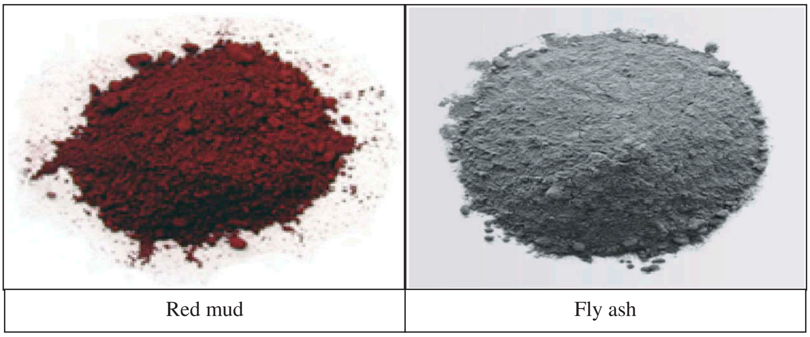

Red mud
Fly ash

Figure 9. Industrial wastes using as low-cost adsorbents.

5.3.1. Fly ash

Fly ash obtained in a large amount from combustion of coal is generally grey in colour, alkaline and refractory in nature. The chemical composition of fly ash has high percentage of silica (60–65%), alumina (25–30%), magnetite, Fe_2O_3 (6–15%) [242]. Cement industries use fly ash as raw materials for the production of concrete. Nowadays, fly ash has been extensively used as low-cost adsorbents. Untreated fly ash has low adsorption capacity than modified fly ash; therefore it is necessary to modify them by physical and chemical treatments [243]. Soco et al. [244] used coal fly ash for the removal of Cu(II) and Ni(II) ions, and used Langmuir and Freundlich isotherm models to describe adsorption behaviours. Adsorption of Cu(II) and Ni(II) occurred through particle diffusion mechanism. The authors reported physisorption and monolayer adsorption for their case. Mishra et al. [245] reported the adsorption capacity of fly ash to adsorb Pb(II), Cu(II) and Zn(II) ions from wastewater with different parameters: pH of solution, contact time, and temperature. Adsorption of metal ions by fly ash was found to be enhanced with increasing pH of the solution and at lower temperatures. The reported removal efficiency of Pb(II), Cu(II) and Zn(II) was 79%, 53% and 80% by fly ash from wastewater. Pehlivan et al. [246] reported fly ash particles and activated carbon adsorbed Cu(II) and Ni(II) ions at equilibrium from aqueous solutions. Adsorption process followed Langmuir and Freundlich isotherm models. The authors studied the influence of different parameters such as pH, contact time, adsorbent dose, and initial metal concentrations and compared the result of activated carbon in same stipulation. The maximum adsorption capacities reported were Cu(II) 7.0 mg/g, Ni(II) 5.9 mg/g for fly ash, while 6.9 mg/g for Cu(II) and 5.4 mg/g for Ni(II) ions, for activated carbon. They examined that metal removal efficiency of fly ash is higher than the activated carbon. Eventually, under optimal conditions (pH = 5 and adsorbent dosage = 8 g/L at a fixed equilibrium time in 60 min), they obtained maximum Cu(II) and Ni(II) removal efficiencies of 97.2% and 78.2%, respectively, using fly ash, while 95.0% and 63% of Cu(II) and Ni(II) removal efficiencies, respectively, using activated carbon in a optimum conditions (pH 4 for Cu(II) and pH 6 for Ni(II) and adsorbent dosage 6 g/L for both the metal ions within 60 min). The adsorption capacities of various heavy metal ions onto fly ash are reported in Table 13.

5.3.2. Red mud

Red mud is obtained through the extraction of alumina from bauxite by the Bayer process and the ground ore is treated with a hot concentrated base (NaOH) [250]. Annually, 90 million tonnes of red mud is generated. Red mud is alkaline in nature and the pH is 10–13 since sodium hydroxide solution is used in the refining process. It is mainly composed of aluminium, iron, silicon, titanium oxide, and hydroxides. The red colour is caused by the presence of oxidised iron [251]. Red mud is a toxic solid waste due to its caustic nature, so it is necessary to neutralise them before utilisation as an adsorbent [252]. Khan et al. [253] investigated iron oxide activated red mud as an adsorbent for the removal of Cd(II) ions from aqueous medium. Adsorption process was better described

Table 13. Adsorption capacities of fly ash for the removal of heavy metal ions from waste water.

S. No.	Fly Ash	Heavy Metals	Adsorption Capacity (mg/g)	Maximum removal efficiency (%)	References
1.	Coal fly ash	Cu(II)	20.92	100	[247]
		Cd(II)	18.98	100	
2.	Coal fly ash	Zn(II)	6.5–13.3	-	[248]
3.	Coal fly ash	Cu(II)	48.8	-	[249]

Table 14. Adsorption capacities of red mud for removal of heavy metal ions from wastewater.

S. No.	Red Mud	Heavy metals	Adsorption capacity (mg/g)	Maximum removal efficiency (%)	References
1.	Red mud	Pb(II)	64.79	-	[255]
		Cr(VI)	35.66	-	
2.	Red mud	Cu(II)	5.35	-	[256]

through Freundlich isotherm than Langmuir isotherm model. The adsorption capacity from Langmuir isotherm was reported as 117.64, 116.28 and 107.53 µg/g, whereas Freundlich isotherm adsorption capacity was 3.83, 3.68 and 3.07 µg/g at 293, 298 and 308 K, respectively. The maximum monolayer adsorption capacity was found to be 117.64 µg/g under optimal conditions (pH = 6.0, adsorbent dosage = 6.0 g/L, contact time = 90 min, initial concentration = 400 µg/L, and temperature = 300 K). The adsorption could be explained through second-order-kinetics. The adsorption process was spontaneous and exothermic in nature. Genc et al. [254] showed that sea water-neutralised red muds (Bauxsol) have potential to remove As(V) (arsenate) from aqueous medium by using batch experiments. Adsorption process followed Langmuir isotherm model. The amount of adsorption increased with higher amount of adsorbent dosage, decreasing value of pH, and initial metal ion concentrations. In the presence of HCO_3^- ions adsorption of As(V) decreased, whereas Ca^{2+} ions enhanced the adsorption. The adsorption capacities of various heavy metal ions onto red mud are shown in Table 14.

6. Adsorption isotherm

The adsorption isotherm models are used to describe how the adsorbent will interact with the adsorbate and to understand the mechanism of adsorption. The isotherm models provide information on adsorption capacity and surface properties. The Langmuir and Freundlich isotherms models were generally used to explain observed adsorption phenomena of heavy metal ions onto low-cost adsorbents.

6.1. Langmuir isotherm model

The Langmuir model assumes that the adsorption of metal ions take a place by monolayer adsorption on a homogenous surface, without any interaction between the adsorbate on adjacent sites [257].

The Langmuir equation can be expressed as:

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \tag{1}$$

The linear form of the equation can be written as:

$$\frac{1}{q_e} = \frac{1}{q_m} + \left(\frac{1}{q_m K_L}\right)\left(\frac{1}{C_e}\right) \tag{2}$$

where q_e is the amount of metal ions adsorbed per specific amount of adsorbent (mg/g), C_e is the equilibrium concentration of metal ions in the solution (mg/L), q_m is the maximum adsorption capacity and K_L is the Langmuir constant (L/mg).

Table 15. Isotherm studies of heavy metal adsorption by various adsorbents.

S. No.	Adsorbents	Heavy metals	Fitted isotherm model	References
1.	Modified orange peel	Cu(II)	Langmuir isotherm	[108]
2.	Modified sawdust	Cr(VI)	Freundlich isotherm	[80]
3.	Hazelnut shell activated carbon	Cr(VI)	Langmuir isotherm	[152]
4.	Modified rice straw	Hg(II)	Langmuir isotherm	[165]
5.	Base treated black tea waste	Cu(II)	Langmuir isotherm	[187]
6.	Immobilised sugarcane bagasse	Cr(VI)	Langmuir isotherm	[206]
7.	Chicken feathers	Cu(II), Zn(II)	Freundlich isotherm	[214]
8.	Egg shell nano-particle	Hg(II)	Freundlich isotherm	[229]
9.	Crab shell particles	Cu(II), Co(II)	Langmuir isotherm	[234]
10.	Neutralised red mud	As(V)	Langmuir isotherm	[254]

6.2. Freundlich isotherm model

The Freundlich isotherm model relies on the assumption that the adsorption of metal ions on a heterogeneous surface, with interaction between the adsorbed molecules [258]. The Freundlich model is represented using the following equation:

$$\mathrm{q_e} = K_{\mathrm{F}} C_e^{1/n} \tag{3}$$

The linear form of the equation can be written as:

$$\log\ \mathrm{q_e} = \log\ K_{\mathrm{F}} + \frac{1}{\mathrm{n}} \log\ \mathrm{C_e} \tag{4}$$

where $\mathrm{q_e}$ is the amount of metal ions adsorbed per specific amount of adsorbent (mg/g), $\mathrm{C_e}$ is the equilibrium concentration of metal ions of the solution (mg/L), K_{f} (mg/g)(mg/L)$^{\mathrm{n}}$ and n are the Freundlich adsorption isotherm constants. The value of the linear regression correlation coefficient (R^2) one of the essential parameters, if the value of the R^2 closer to 1, gives an indication as to which model can be chosen to give the best-fit. Table 15 shows the isotherm results of metal adsorption by various adsorbents.

7. Adsorption kinetics

In the adsorption process, contact time from experimental results can be used to study the rate-limiting steps. These steps are most important aspects and the rate can be calculated from kinetic study. Lagergen's first order equation and Ho's second-order equation are the most popular kinetic models and these models are commonly used to describe adsorption kinetic study. Thus, the kinetics of metal ions onto different adsorbent materials was analysed using kinetic models which are shown below.

7.1. Pseudo-first-order model

The linear form of Lagergren's (1898) pseudo-first-order equation is given as [259]:

$$\log(\mathrm{q_e} - \mathrm{q}_t) = \log\ \mathrm{q}_e - \frac{k_1}{2.303} t \tag{5}$$

where '$\mathrm{q_e}$' and '$\mathrm{q_t}$' (mg/g) are the amount of metal ions adsorbed at equilibrium, at time t, respectively, and k_1 is the pseudo-first-order rate constant.

Table 16. Kinetic studies of heavy metals adsorption by different adsorbents.

S. No.	Adsorbents	Heavy metals	Fitted kinetics model	References
1.	Modified orange peel	Cu(II)	Pseudo-second order	[108]
2.	Formaldehyde treated sawdust	Cr(VI)	Pseudo-second order	[134]
3.	Cashew nut shell	Cd(II)	Pseudo-second order	[146]
4.	Modified rice straw	Hg(II)	Pseudo-second order	[165]
5.	Protonated rice bran	Ni(II)	Pseudo-second order	[180]
6.	Black tea waste	Cd(II), Co(II), Zn(II)	Pseudo-first order	[190]
7.	Sugarcane bagasse	Hg(I)	Pseudo-second order	[202]
8.	Chicken feathers	Zn(II)	Pseudo-second order	[213]
9.	Egg-shell nano-particle	Hg(II)	Pseudo-second order	[229]
10.	Crab shell particles	Cu(II), Co(II)	Pseudo-second order	[234]
11.	Red mud	Pb(II), Cr(VI)	First-order	[255]

7.2. Pseudo-second order model

Pseudo-second-order kinetic models are used for chemisorptions process involving chemical bonding between metal ions and functional groups of the adsorbent [260]. Ho and Mckay (1999) presented the pseudo-second-order kinetic reaction and it is expressed as:

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \tag{6}$$

Integrating Equation (6) and applying the boundary conditions, we get:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{7}$$

where k_2 is the pseudo-second-order rate constant (g/mg min^{-1}) and 'q_e' and 'q_t' (mg/g) are the amount of metal ions adsorbed at equilibrium, at time t, respectively. Generally the experimental data is better represented by the pseudo-second-order kinetic model for most adsorption systems. Table 16 shows a summary of the studies on the application of kinetic models for the removal of heavy metals using different adsorbents.

8. Adsorption thermodynamics

Temperature plays an important role for the adsorption of metal ions associated with the thermodynamics of the adsorption process. Thermodynamic parameters of heavy metal adsorption on different adsorbents reported in the literatures have been summarised in Table 17. Generally, the thermodynamic nature of heavy metal adsorption on low-cost adsorbent can be explained as either exothermic or endothermic adsorption processes [261]. If the adsorption decreases with increasing temperature, the adsorption is an exothermic adsorption process whereas if the adsorption increases with the increasing temperature it is an endothermic process. Three thermodynamic parameters, Gibbs free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) can be often used to determine the adsorption thermodynamics behaviour [262]. The following equations are used:

$$K_d = \frac{C_A}{C_B} \tag{8}$$

$$\Delta G^0 = -RT\ln K_d \tag{9}$$

Table 17. Thermodynamic studies of heavy metals adsorption by different adsorbents.

S. No.	Adsorbent	Heavy metals	ΔH^0 (KJ/mol)	ΔS^0 (J/mol/K)	ΔG^0 (KJ/mol) 303 K	313 K	323 K	Adsorption type	Reference
1.	Modified litchi peel	Cr(VI)	19.03	−81.92	43.85	46.92	47.82	Endothermic	[124]
2.	Formaldehyde treated sawdust	Cr(VI)	9.34	33.3	−0.76	−1.10	−1.43	Endothermic	[134]
3.	Cashew nut shell	Cu(II)	−10.05	−20.33	−3.91	−3.65	−3.45	Exothermic	[141]
4.	Hazelnut shell activated carbon	Cu(II)	18.77	40.4	6.66	6.03	5.71	Endothermic	[149]
5.	Modified rice straw	Hg(II)	72.56	249.95	−3.05	−5.93	−8.03	Endothermic	[165]

$$\ln K_d = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \tag{10}$$

where K_d is the thermodynamic equilibrium constant. C_A and C_B are the concentration on the adsorbate on the adsorbent and residual concentration at equilibrium, respectively. T is the absolute temperature in Kelvin (K) and R is the universal gas constant (8.314×10^{-3} kJ/mol K). The positive value of enthalpy (ΔH°) indicates the endothermic nature of the adsorption process, while the negative value of enthalpy (ΔH°) indicates the exothermic nature of the adsorption process. On the other hand, the negative value of ΔG° indicates the adsorption process is spontaneous and thermodynamically feasible at all temperatures. The positive value of ΔS° confirmed an increased randomness at the solid–liquid interface during the adsorption process, while a negative value of ΔS° indicates a decreased randomness during the adsorption process.

9. Future perspective

Waste materials have promising applications in environmental applications and can serve as alternatives to replace activated carbon for removal of heavy metal ions from wastewater or for water pollution treatment. The waste materials are easily available and low cost. Therefore, removal of heavy metal ions using low-cost adsorbents is expected to gain an upsurge of interest in the near future.

10. Conclusion

Rapid industrialisation has resulted in increased utilisation of heavy metal ions in last few decades to make the serious worldwide environmental issue. Due to its hazardous and bioaccumulative nature, there have been numerous attempts to find a suitable remedial action. Various conventional methods such as ion exchange, chemical precipitation, coagulation, membrane separation, reverse osmosis and adsorption methods have so far been used for the removal of heavy metal ions, but these methods could not be effective due to several issues and drawbacks. Among them, adsorption process is very useful and effective for removal of heavy metal ions, even at low concentrations. Activated carbon, for instance, is highly impressive for adsorption of heavy metal ions but its use is somewhat limited due to consideration of economy. It has therefore been crucial to develop low cost and readily available adsorbents for the adsorption of heavy metal ions from wastewater. The utilisation of low-cost adsorbents has some advantages such as high adsorption capacity, easy separation, abundance, low cost and renewability.

This review presented a variety of low-cost adsorbents such as agricultural waste materials, animal waste materials, and industrial by-products in the removal of heavy metal ions from aqueous solutions. All adsorbents have functional groups that play a major role in metal ion adsorption. Waste materials are associated with functional groups, in part, carboxyl, hydroxyl, and phenol groups, which are oxygen-containing sites and play an important role in the metal ion removal. Generally, chemically modified adsorbents enhance the surface area of adsorbent and exhibited higher adsorption capacity than unmodified adsorbents.

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