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Review

Utilization of agro-industrial and municipal waste materials as potential adsorbents for water treatment—A review

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ABSTRACT

Adsorption process has been proven one of the best water treatment technologies around the world and activated carbon is undoubtedly considered as universal adsorbent for the removal of diverse types of pollutants from water. However, widespread use of commercial activated carbon is sometimes restricted due to its higher costs. Attempts have been made to develop inexpensive adsorbents utilizing numerous agro-industrial and municipal waste materials. Use of waste materials as low-cost adsorbents is attractive due to their contribution in the reduction of costs for waste disposal, therefore contributing to environmental protection. In this review, an extensive list of low-cost adsorbents (prepared by utilizing different types of waste materials) from vast literature has been compiled and their adsorption capacities for various aquatic pollutants as available in the literature are presented. It is evident from the literature survey that various low-cost adsorbents have shown good potential for the removal of various aquatic pollutants. However, there are few issues and drawbacks on the use of low-cost adsorbents in water treatment that have been discussed in this paper. Additionally, more research is needed to find the practical utility of low-cost adsorbents on commercial scale.

1. Introduction

Water is a source of life and energy, although millions of people worldwide are suffering with the shortage of fresh and clean drinking water. Rapid pace of industrialization, population expansion, and unplanned urbanization have largely contributed to the severe water pollution and surrounding soils. The main sources of freshwater pollution can be attributed to discharge of untreated sanitary and toxic industrial wastes, dumping of industrial effluent, and runoff from agricultural fields. It is well known that 70–80% of all illnesses in developing countries are related to water contamination, particularly susceptible for women and children [1]. Pollutants discharged in wastewaters can be toxic to aquatic life and cause natural waters to be unfit as potable water sources.

As a result of development of advanced analytical techniques and better health monitoring technologies, the acceptable minimum concentration of these chemicals is progressively decreasing. As such, stringent regulations have been introduced by many countries with respect to the presence of these chemicals in water which binds industries to treat industrial effluents properly before discharging it into the natural water-bodies containing clean water. In view of the importance of water pollution control, a number of technologies have been developed.

2. Technologies for water treatment

With the ever-increasing use of water for municipal and industrial purposes, it has become more important to appraise water quality on a continuous basis. Water treatment process selection is a complex task involving the consideration of many factors which include, available space for the construction of treatment facilities, reliability of process equipment, waste disposal constraints, desired finished water quality and capital and operating costs. The treatment of wastewaters to make them suitable for subsequent use requires physical, chemical and biological processes. A number of technologies are available with varying degree of success to control water pollution. Some of them are coagulation [2], foam flotation [3], filtration [4], ion exchange [5], aerobic and anaerobic treatment [6,7], advanced oxidation processes [8], solvent extraction [9], adsorption [10], electrolysis [11], microbial reduction [12], and activated sludge [13]. However, most of them require substantial financial input and their use is restricted because of cost factors overriding the importance of pollution control.

Among various available water treatment technologies, adsorption process is considered better because of convenience, ease of

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operation and simplicity of design [10]. Further, this process can remove/minimize different type of pollutants and thus it has a wider applicability in water pollution control.

2.1. Adsorption

The term adsorption refers to the accumulation of a substance at the interface between two phases such as solid and liquid or solid and gas. The substance that accumulates at the interface is called ‘adsorbate’ and the solid on which adsorption occurs is ‘adsorbent’. Although certain phenomenon associated with adsorption were known in ancient times, the first quantitative studies were reported by C.W. Scheele in 1773 [14] on the uptake of gases by charcoal and clays. This was followed by Lowitz’ observations who used charcoal for decolorization of tartaric acid solutions. Larvitz in 1792 and Kehl in 1793 observed similar phenomenon with vegetable and animal charcoals, respectively. However, the term ‘adsorption’ was proposed by Bois-Reymond but introduced into the literature by Kayser [15]. Ever since then, the adsorption process has been widely used for the removal of solutes from solutions and gases from air atmosphere.

At the surface of the solids, there are unbalanced forces of attraction which are responsible for adsorption. In cases where the adsorption is due to weak van der Waals forces, it is called physical adsorption. On the other hand, there may be a chemical bonding between adsorbent and adsorbate molecule and such type of adsorption is referred as chemisorption. Readers interested in a detailed discussion about the theory and applications of adsorption should refer to an excellent comprehensive review by Dąbrowski [15].

3. Commercial adsorbents

A number of materials have been extensively investigated as adsorbents in water pollution control. Some of the important ones include silica gel, activated alumina, zeolites and activated carbon, etc.

3.1. Silica gel

Silica gels are classified into three types: regular, intermediate and low density gels. Regular density silica gel is prepared in an acid medium and shows high surface area (e.g. 750 m^2/g). Intermediate and low density silica gels have low surface areas (300–350 and 100–200 m^2/g, respectively). The gel is considered a good adsorbent and is used in many industries [16,17]. Modified forms of silica have also been widely explored for the removal of different pollutants [18–20].

3.2. Activated alumina

The activated alumina comprises a series of nonequilibrium forms of partially hydroxylated alumina oxide, Al_2O_3. In general, as a hydrous alumina precursor is heated, hydroxyl groups are driven off leaving a porous solid structure of activated alumina. It is also used to remove water from organic liquids including gasoline, kerosene, oils, aromatic hydrocarbons and many chlorinated hydrocarbons, having the surface area ranging from 200 to 300 m^2/g. Activated alumina is receiving renewed attention as an adsorbent and a wealth of information has been published [21–25] on its adsorption characteristics. Readers interested in a detailed discussion of the application of alumina in water treatment should refer to an excellent comprehensive review by Kasprzyk-Hordern [26].

3.3. Zeolites

Zeolites are aluminosilicates with Si/Al ratios between 1 and infinity. There are 40 natural and over 100 synthetic zeolites. They are also considered as selective adsorbents. Zeolites-based materials are extremely versatile and their main use include detergent manufacture, ion-exchange resins (i.e. water softeners), catalytic applications in the petroleum industry, separation process (i.e. molecular sieves) and as an adsorbent for water, carbon dioxide and hydrogen sulfide. Various zeolites have been employed for the removal of pollutants [27–30]. Recently, Wang and Peng [31] discussed the role of natural zeolites as effective adsorbents in water and wastewater treatment.

3.4. Activated carbon

Activated carbon has undoubtedly been the most popular and widely used adsorbent in wastewater treatment throughout the world. Charcoal, the forerunner of modern activated carbon has been recognized as the oldest adsorbent known in wastewater treatment. Activated carbon is produced by a process consisting of raw material dehydration and carbonization followed by activation. The product obtained is known as activated carbon and generally has a very porous structure with a large surface area ranging from 600 to 2000 m^2/g. Activated carbon has been found to be a versatile adsorbent, which can remove diverse types of pollutants such as metal ions [32–35], anions [36–38] dyes [39–43], phenols [44–49], detergents [50,51], pesticides [52,53], humic substances [54,55], chlorinated hydrocarbons [56,57] and many other chemicals and organisms [58–64]. In spite of abundant uses of activated carbon, its applications are sometime restricted due to its higher cost. Therefore, researchers are looking for low-cost adsorbents for water pollution control, where cost factors play a major role. As such, for quite sometime, efforts have been directed towards developing low-cost alternative adsorbents. Low-cost alternative adsorbents can be prepared from a wide variety of raw materials, which are abundant and cheap, having high organic (carbon) content and low inorganic content and these can be easily activated [65].

The preparation of low-cost adsorbents from waste materials has several advantages, mainly of economic and environmental nature. A wide variety of low-cost adsorbents have been prepared from different waste materials utilizing agricultural as well industrial and municipal wastes. Although many review articles have been published [66–77] so far discussing the importance of low-cost adsorbents in water pollution control, many of them are generally either adsorbate-specific (metals, dyes, phenols, etc.) or adsorbent-specific. However, one of the aims of the present review is to compile and present the adsorption potential (adsorption capacities) of various low-cost adsorbents developed from agricultural and industrial wastes for different aquatic pollutants . This review provides the recent literature of past 10–15 years, to demonstrate the usefulness of low-cost adsorbents in water treatment. A summary of relevant published data (in terms of adsorption capacities of different low-cost adsorbents for the removal of various pollutants) with some of the latest important findings, and a source of up-to-date literature is presented and the results have been discussed.

4. Low-cost alternative adsorbents

4.1. Agricultural wastes as low-cost adsorbents

Agricultural materials particularly those containing cellulose shows potential sorption capacity for various pollutants. The basic components of the agricultural waste materials include hemicellulose, lignin, lipids, proteins, simple sugars, water, hydrocarbons, and starch, containing variety of functional groups. Agricultural waste materials being economic and eco-friendly due to their unique chemical composition, availability in abundance, renewable nature and low cost are viable option for water and wastewater remediation. Agricultural waste is a rich source for activated carbon production due to its low ash content and reasonable hardness [78], therefore, conversion of agricultural wastes into low-cost adsorbents is a promising alternative to solve environmental problems and also to reduce the preparation costs.

In the last several decades, various agricultural wastes have been explored as low-cost adsorbent. Some of them include the shells and/or stones of fruits like nuts [79–81], peanuts [82], olive wastes [83], almonds [84], apricots stones [85] and cherries [86]; and wastes resulting from the production of cereals such as rice [87], maize [88] and corn [89] as well as sugar cane bagasse [90] and coir pith [91]. These agricultural waste materials have been used in their natural form or after some physical or chemical modification.

4.1.1. *Adsorbents from rice and wheat waste*

Rice is the seed of a monocot plant *Oryza sativa*. Rice is one of the major crops grown throughout the world. As a cereal grain, it is the most important staple food for a large part of the world's human population. World production of rice has risen steadily from about 200 million tonnes of paddy rice in 1960 to 600 million tonnes in 2004. Consequently, rice industry produces several by-products, e.g. rice husk, rice hull, rice husk/hull ash, rice bran.

Rice husks were tested for arsenic removal from water by Nasir et al. [92]. Maximum adsorption was reported on treatment with 0.01 mol/L of HNO_3, HCl, H_2SO_4 or $HClO_4$ using 1.0 g of adsorbent for 5.97×10^{-3} mol/L of arsenic in 5 min. The Freundlich isotherm was followed over a concentration range of 8.69×10^{-5} to 1.73×10^{-3} mol/L arsenic ($l/n = 0.83$ and $K = 4.43$ mmol/g). The uptake of arsenic was found to increase with increasing temperature.

The adsorption behavior of rice husks for antimony ions from aqueous solutions has been investigated by Khalid et al. [93]. The adsorption was studied as a function of appropriate electrolyte, equilibration time, hydrogen ions, amount of adsorbent, concentration of adsorbate, effect of diverse ions and temperature. Quantitative adsorption of antimony from aqueous solutions on rice husks was achieved within a short contact time of 10 min. The reaction was found to be endothermic and occurred in presence of a variety of anions, with maximum adsorption in the presence of 0.01 M HNO_3. The same researchers also studied the removal of Pb(II) and Hg(II) using rice husk as an adsorbent [94,95].

The sorption of Cd(II) from aqueous solution by rice husk was investigated by Kumar and Bandyopadhyay [96]. They used some simple and low-cost chemical modifications which resulted in increasing the sorption capacity of raw rice husk (RRH) from 8.58 to 11.12, 20.24, 16.18 mg/g and the equilibrium time was reduced from 10 h of RRH to 2, 4 and 1 h for epichlorohydrin treated rice husk (ERH), NaOH treated rice husk (NRH), sodium bicarbonate treated rice husk (NCRH), respectively. The researchers suggested that NCRH could be an excellent alternative for the removal of heavy metal by sorption process in view of low cost and the rapid uptake of Cd(II).

Rice hull ash has also been explored as an adsorbent for the removal of lead(II) ion from aqueous solution by Wang and Lin [97]. Experimental results indicated that the kinetics and the equilibrium adsorption could be increased by increasing the initial lead concentration, pH, stroke speed, or adsorption temperature. The data of adsorption kinetics indicated that the process was physisorption controlled and the pseudo-second-order rate equation suitably interpreted the overall process. The acid-treated (H_3PO_4) rice bran was used as a low-cost sorbent for the removal of nickel by Zafar et al. [98]. The maximum adsorption capacity of 102 mg/g was reported at pH 6 for nickel removal.

Rice husk has also been explored for dyes and phenols removal from water. McKay et al. [99] examined the potential of rice husk for the removal of two basic dyes, safranine and methylene blue, and adsorption capacity of 838 and 312 mg/g, respectively was reported. Sumanjit and Prasad [100] studied the removal of acidic dyes, viz. acid violet 54, acid violet 17, acid blue 15, acid violet 49 and acid red 119 from aqueous solutions using rice husk ash. The adsorption capacity was found to vary from 99.4 to 155 mg/g, suggesting rice husk ash as a good adsorbent for dyes removal.

The possible utilization of rice husk ash as an adsorbent for methylene blue dye from aqueous solutions has been investigated by Chandrasekhar and Pramada [101]. The highest adsorption capacity was found to be approximately 690 mg/g. Rice husk ash as an adsorbent was also tested for Indigo Carmine dye removal from aqueous solution by Lakshmi et al. [102]. An adsorption capacity in the range of 29.3–65.9 mg/g was reported at different temperatures (293–323 K).

Mahvi et al. [103] investigated the potential of rice husk and rice husk ash for phenol removal from aqueous solution. It was observed by them that rice husk ash was more effective than rice husk for phenol removal and can be used as an efficient adsorbent for the removal of phenolic compounds from wastewater. Chemically and thermally treated rice husk (RHT) was used as an adsorbent for the removal of 2,4-dichlorophenol from aqueous solution by Akhtar et al. [104]. The maximum sorption (98 ± 1.2%) was achieved for RHT from 6.1×10^{-5} mol/dm^3 of sorbate solution using 0.1 g of rice husk for 10 min agitation time at pH 6 and 303 K, which was comparable to commercial activated carbon (96.6 ± 1.2%), but significantly higher than chemically treated rice husk (65 ± 1.6%) and untreated rice husk (41 ± 2.3%). Several other researchers also studied the application of rice husk as adsorbent for the removal of different pollutants from water [105–109].

Rice bran was also tested for the removal of organochlorine compounds and benzene from industrial wastewater by Adachi et al. [110]. Dichloromethane was found to be successfully removed from water samples with an average removal efficiency of 70% after 60 min when rice bran was added to water samples containing 0.006–100 mg/L dichloromethane. It was proposed by the workers that the removal of organochlorine compounds and benzene by rice bran was due to the uptake by intracellular particles called spherosomes. The removal of 22 different pesticides using rice bran was examined by Adachi et al. [111]. The pesticides with high lipophilicity were found to be easily removed by rice bran. Further, the removal efficiencies varied from 22.2% to 98.8% in their study.

Wheat (*Triticum* spp.) is an another important staple food generally used to make flour. In 2007 world production of wheat was 607 million tonnes, making it the third most-produced cereal after maize and rice. Similar to the rice industry, wheat industry also produces some by-products such as wheat bran, wheat husk, etc. which have been examined as adsorbents. Wheat bran has been utilized for the removal of cadmium(II) from wastewater by Singh et al. [112]. The maximum removal of cadmium(II) was reported to be 87.15% with initial Cd(II) concentration of 12.5 mg/L at 20 °C and pH 8.6.

The adsorption of Pb(II) ions from aqueous solutions on wheat bran has also been reported by Bulut and Baysal [113]. The adsorption of Pb(II) was studied between the pH range of 1.85–7.01 with an initial Pb(II) concentration range of 0–1000 mg/L. The lowest Pb(II) uptake was observed at pH 2.0, while a significant increase in the adsorption of Pb(II) on wheat bran was found in a pH range of 4–7. The maximum sorption capacities of 69.0, 80.7 and 87.0 mg/g of Pb(II) on wheat bran at 20, 40 and 60 °C, respectively were reported.

Activated wheat husk has also been found efficient as an adsorbent for the adsorption of dye, Reactofix Navy Blue 2 GFN, from aqueous solution by Gupta et al. [114]. The contact time of 40 min was found sufficient to achieve equilibrium conditions. Moreover, low pH and low temperature were found suitable for maximum adsorption. The same adsorbent was also used for the removal of Reactofix golden yellow 3 RFN dye from aqueous solution [115] and was found efficient for dye removal. The adsorption of dyes, viz. reactive blue 19 (RB19), reactive red 195 (RR 195) and reactive yellow 145 (RY 145) was examined by wheat bran by Fatma et al. [116]. The monolayer coverage capacities of wheat bran for RB19, RR 195 and RY 145 dyes were found as 117.6, 119.1 and 196.1 mg/g at 60 °C, respectively.

4.1.2. Adsorbents from tea and coffee waste

Tea is the product of the leaves, leaf buds, and internodes of the *Camellia sinensis* plant, prepared and cured by various methods. Tea is the most popular drink in the world in terms of consumption and world tea production was 3.15 million tonnes annually in 2003. Waste/spent tea leaves have been investigated as adsorbent by various researchers for the removal of different types of pollutants from water.

Waste tea leaves have been used for the removal of lead, iron, zinc and nickel from water by Ahluwalia and Goyal [117]. The order of adsorption of metal was: Pb > Fe > Zn > Ni from 5 to 100 mg/L of metal solution. Adsorption of copper and lead ions onto tea waste from aqueous solutions was studied by Amarasinghe and Williams [118]. The highest metal uptake of 48 and 65 mg/g was observed for Cu and Pb, respectively. Mozumder et al. [119] also investigated the potential of tea-leaves waste for Cr(VI) removal and reported that adsorption of Cr(VI) was highly pH dependent, and the removal efficiency dropped sharply from 95% to 10% when pH of the system increased from 2 to 5. The adsorption ability of Turkish tea waste (fibrous) was investigated for the removal of Cu(II) and Cd(II) from single (non-competitive) and binary (competitive) aqueous systems [120]. Adsorption of the investigated heavy metal ions by tea waste was found to be strongly dependent on pH, contact time, initial concentration of the heavy metal ions and adsorbent dosage. The maximum adsorption capacities of Cu(II) and Cd(II) were calculated as 8.64 and 11.29 mg/g for single and 6.65 and 2.59 mg/g for binary systems, respectively. Batch adsorption studies were carried out for evaluating the suitability of tea factory waste as a low-cost adsorbent for the removal of zinc [121]. The effects of various parameters like pH, adsorbent dose, initial concentration of zinc and temperature, on adsorption performance were studied using tea factory waste (TFW). The maximum adsorption capacity of TFW was calculated as 8.9 mg/g for Zn(II).

Besides metal ions, spent tea leaves were used for cationic dye (methylene blue) removal by Hameed [122]. The high adsorption capacity (300.05 mg/g) of this adsorbent was reported for methylene blue dye removal at 30 °C. The potentiality of tea waste for the adsorptive removal of methylene blue from aqueous solution was also studied by Uddin et al. [123]. The nature of the adsorbent and methylene blue interactions was examined by the Fourier transform infrared (FTIR) technique. Adsorption equilibrium of tea waste was reached within 5 h for methylene blue concentrations of 20–50 mg/L. The sorption was found to follow a pseudo-second-order kinetic model. The extent of the dye removal increased with increasing initial dye concentration. The adsorption capacity of methylene blue onto tea waste was found to be 85.16 mg/g. Besides these, several other researchers also investigated the potential of tea waste as adsorbent for the removal of different pollutants from water [124,125].

Coffee is another most popular beverages globally. Worldwide, 6.7 million metric tons of coffee were produced annually in 1998–2000, and the forecast is a rise to 7 million metric tons annually by 2010. Brazil is the world leader in the production of green coffee, followed by Vietnam and Colombia. Wastes from coffee industry have been explored as adsorbents for water treatment. The adsorption behavior of heavy metals on arabica and robusta roasted coffee beans was investigated by Minamisawa et al. [126]. The results showed that the adsorption percentage of the heavy metal ions was above 90% for all coffee beans examined. Further, the adsorption capacities of Cu(II) and Cd(II) ions onto blend coffee were about 2.0 mg/g.

Coffee residue has been used as raw material in the preparation of powder activated carbon by chemical activation with zinc chloride for the sorption of Pb(II) from dilute aqueous solutions [127]. Batch experiments were conducted to study the effects of the main parameters such as contact time, initial concentration of Pb(II), solution pH, ionic strength and temperature. The maximum uptake of Pb(II) on prepared adsorbent at 25 °C was about 63 mg/g at pH 5.8, initial Pb(II) concentration of 10 mg/L, agitation speed of 200 rpm and ionic strength of 0.005 M. The kinetic data followed pseudo-second-order model. Besides these, several other researchers also investigated the potential of coffee waste as adsorbent for the removal of different pollutants [128–134].

4.1.3. Adsorbents from coconut waste

The coconut (*Cocos nucifera*) is highly nutritious and rich in fiber, vitamins, and minerals. Coconut palms are grown in more than 80 countries of the world, with a total production of 49 billion nuts. Coconut wastes such as coir pith, coconut bunch waste, coconut husk, copra meal, male flowers of coconut tree, etc. have been widely explored as adsorbents for the removal of various pollutants from water. Coir pith waste was used for the adsorptive removal of Pb(II) from aqueous solution by Kadirvelu and Namasivayam [135]. The adsorption capacity was reported to be 263 mg/g for Pb(II) removal which was found to increase with increasing pH from 2 to 4 and remained constant up to pH 10.

Coir pith was also investigated as adsorbent for Co(II), Cr(III) and Ni(II) adsorption in both single and multi-component systems by Parab et al. [136]. The maximum adsorption capacity of coir pith was found to be 12.82, 11.56 and 15.95 mg/g for cobalt, chromium and nickel, respectively. Optimum pH values for maximum metal-ion adsorption were found as 4.3 for cobalt, 3.3 for chromium and 5.3 for nickel. Namasivayam et al. [137] investigated coir pith as an adsorbent for the adsorption of rhodamine B and acid violet dyes. The low adsorption capacities, viz. 2.56 and 8.06 mg/g of the adsorbent for rhodamine B and acid violet, respectively were reported. Coir pith was also examined for congo red dye removal and an adsorption capacity of 6.72 mg/g was reported [138].

The coconut bunch waste (CBW) was used to remove methylene blue from aqueous solution [139]. The monolayer adsorption capacity was found to be 70.92 mg/g at 30 °C. The kinetic data obtained at different concentrations fitted very well with the pseudo-second-order kinetic model.

The adsorption of 2,4,6-trichlorophenol (TCP) on coconut husk-based activated carbon prepared under optimized conditions was evaluated by Hameed et al. [140]. Adsorption capacity was found to increase with increase in initial concentration and agitation time, while acidic pH was more favorable for the adsorption of TCP. The maximum monolayer adsorption capacity of 716.10 mg/g was observed at 30 °C. The adsorption kinetics was found to follow the pseudo-second-order kinetic model. The mechanism of the adsorption process was determined from the intraparticle diffusion model. Boyd plot revealed that the adsorption of TCP on the activated carbon was mainly governed by particle diffusion.

Adsorption isotherm and kinetics of methylene blue on activated carbon prepared from coconut husk were determined from batch tests [141]. The effects of contact time (1–30 h), initial dye concentration (50–500 mg/L) and solution temperature (30–50 °C) were investigated. The equilibrium data were best represented by Langmuir isotherm model, showing maximum monolayer adsorption capacity of 434.78 mg/g. The kinetics was found to follow pseudo-second-order kinetic model.

Fibers taken from coconut husks were functionalized with the thiophosphoryl (P═S) group by means of the direct reaction with $Cl_3P═S$, $(CH_3O)_2ClP═S$ or $(CH_3CH_2O)_2ClP═S$ in order to test their potential for Cd^{2+} removal [142]. Adsorption isotherms for Cd^{2+} fitted well with the Langmuir model, with binding capacities of 0.2–5 mmol/g at 25 °C.

Extracted babassu coconut (*Orbignya speciosa*) mesocarp (BCM) was examined as sorbent for the removal of various dyes, Blue Remazol R160 (BR 160), Rubi S2G (R S2G), Red Remazol 5R (RR 5), Violet Remazol 5R (VR 5) and Indanthrene Olive Green (IOG) dye solutions [143]. The maximum adsorption was found at pH 1.0 for all dyes. The ability of babassu coconut mesocarp to adsorb dyes was in the order: R S2G > VR 5 > BR 160 > IOG > RR 5.

Coconut copra meal, a waste product of coconut oil production, was investigated as a sorbent for cadmium removal from aqueous solution [144]. The adsorption capacity of copra meal for cadmium ions was calculated to be 4.99 mg/g at 299 K. Adsorption of crystal violet, a basic dye, onto phosphoric and sulfuric acid activated carbons (PAAC and SAAC), prepared from male flowers of coconut tree has been investigated [145]. The adsorption capacities of both the carbons were found to be 60.42 and 85.84 mg/g for PAAC and SAAC, respectively. Besides these, several other researchers investigated the potential of coconut waste as adsorbent for the removal of different pollutants [146–149].

4.1.4. Adsorbents from peanut or groundnut waste

The peanut, or groundnut (*Arachis hypogaea*), is a species in the legume “bean” family. The world production of 34.43 million metric tons has been reported in 2008–2009. India and China are the world’s largest producers of peanuts. Peanut/groundnut’s husk/hull have been widely used as adsorbents for the removal of various pollutants from water.

The removal of cadmium and lead ions from aqueous solutions by groundnut husks modified by EDTA ($\%N = 12.05$) was examined by equilibrium sorption studies at 29 °C [150]. The maximum metal-ion binding capacity of the EDTA-modified husk was found to be 0.36 and 0.19 mmol/g for Cd(II) and Pb(II) ions, respectively at pH 6.8. It was found that the rate of sorption was particle-diffusion controlled.

Peanut hull was converted into an adsorbent by Namasivayam and Periasamy [151] by treating it with concentrated sulfuric acid, followed by its carbonization in air and further treating with 1% sodium bicarbonate overnight. The treated material was used as an adsorbent for the removal of Hg(II) ions from aqueous solutions and the adsorption was found to conform to both Freundlich and Langmuir isotherms. Chamarthy et al. [152] also prepared an adsorbent from peanut shell by thermal treatment in presence of phosphoric acid or citric acid and used it for the adsorption of Cd(II), Cu(II), Ni(II), Pb(II), and Zn(II). Their study showed that phosphoric acid-modified shells adsorbed metal ions in larger amounts compared to citric acid-modified shells.

Untreated and silver impregnated groundnut husk carbon were tested for the removal of chromium(VI) [153]. Approximately, 97% of hexavalent chromium was removed at pH 3 within 5 h. It was found that adsorbents chemically modified with an oxidizing agent demonstrated better chromium(VI) removal as compared to untreated adsorbents in terms of their adsorption rate. It was concluded that groundnut husk carbon oxidized with silver treatment has higher chromium(VI) adsorption capacity. Activated carbon prepared from peanut shell (PAC) was used for the removal of Pb^{2+} from aqueous solution [154]. The impacts of the Pb^{2+} adsorption capacities on the acid-modified carbons oxidized with HNO_3 were also investigated. A comparative study with commercial granular activated carbon (GAC) showed that PAC was 10.3 times more efficient compared to GAC based on Langmuir maximum adsorption capacity. Further analysis results by the Langmuir equation showed that HNO_3 [20% (by mass)] modified PAC has larger adsorption capacity for Pb^{2+} from aqueous solution (ca. 35.5 mg/g). The adsorption capacity enhancement ascribed to pore widening, increased cation-exchange capacity by oxygen groups, and the promoted hydrophilicity of the carbon surface.

The use of peanut hulls, for copper and lead removal was studied by Oliveira et al. [155]. The influence of initial pH (3–5) was evaluated and set between 4.0 and 4.5. The maximum sorption capacities obtained from the Langmuir model were 0.21 and 0.18 mmol/g for copper and lead, respectively. In bi-component systems, competitive sorption of copper and lead was verified, the total amount adsorbed was around 0.21 mmol/g in both mono- and bi-component systems. In the kinetic studies equilibrium was reached after 200 min contact time using a 400 rpm stirring rate, achieving 78% and 58% removal, in mono-component system, for copper and lead, respectively. The adsorption followed pseudo-second-order kinetics. Peanut hull was also used for the removal of Cu(II) from aqueous solutions [156]. The extent of adsorption was investigated as a function of pH, contact time, adsorbate concentration and reaction temperature. The Cu(II) removal was found to be pH dependent, reaching a maximum at pH 5.5. The sorption process followed pseudo-second-order kinetics and equilibrium was attained in 2 h. The equilibrium sorption capacity of Cu(II) was 21.25 mg/g at 30 °C.

Groundnut shell was used as an adsorbent by chemical activation with $ZnCl_2$ under optimized conditions and its comparative characterization was conducted with commercially available powdered activated carbon (CPAC) for its physical, chemical and adsorption properties [157]. The groundnut shell based powdered activated carbon (GSPAC) had higher surface area, iodine and methylene blue numbers compared to CPAC. Both the carbons were used for the removal of malachite green dye from aqueous solution and the effect of various operating variables, viz. adsorbent dose (0.1–1 g/L), contact time (5–120 min) and adsorbate concentrations (100–200 mg/L) were examined for the removal of dye. The experimental results indicate that at a dose of 0.5 g/L and initial concentration of 100 mg/L, GSPAC showed 94.5% removal of the dye in 30 min equilibrium time, while CPAC removed 96% of the dye in 15 min.

Adsorption of Neutral Red (NR) onto peanut husk in aqueous solutions was investigated at 295 K [158]. Experiments were carried out as a function of pH, adsorbent dosage, contact time, and initial dye concentration. The adsorption capacity of peanut husk for the removal of NR was found to be 37.5 mg/g at 295 K. The adsorption kinetic data followed pseudo-first-order and pseudo-second-order kinetic models. The intraparticle diffusion model was also used to explain the adsorption process at the two-step stage.

Besides these, several other researchers also investigated the potential of peanut or groundnut waste for the removal of different pollutants from water [159–163].

4.1.5. Adsorbents from peels of different agricultural waste

Peel, also known as skin, is the outer protective layer of a fruit or vegetable, currently gaining wide attention as adsorbent in water treatment. Peels of different fruits such as, orange, banana, watermelon, cassava, mango, etc. have been used as adsorbents for the removal of different pollutants from water. Orange peel was examined for Ni(II) removal from electroplating wastewater [164]. The extent of Ni(II) removal was found to be dependent on sorbent dose, initial metal concentration, pH and temperature. The process was endothermic showing monolayer adsorption of Ni(II), with a maximum adsorption of 96% at 50 °C for an initial concentration of 50 mg/L at pH 6. The preparation of chemically modified orange peel adsorbents and their sorption behavior for Co(II), Ni(II), Zn(II) and Cd(II) have also been studied [165]. Effects of different chemical modifications on the adsorbent properties including different alkalis saponification (NaOH, NH_4OH, and $Ca(OH)_2$) and acids ($C_6H_6O_7{\cdot}H_2O$, $H_2C_2O_4$, and H_3PO_4) modification after saponification with NaOH were investigated. The maximum adsorption capacities of chemically modified orange peel by different treatments were in the range of 1.13–1.28 mol/kg for the studied metal ions.

The effectiveness of orange peel in adsorbing acid violet 17 dye from aqueous solutions was studied by Sivaraj et al. [166] as a function of agitation time, adsorbent dosage, initial dye concentration and pH. The adsorption capacity was 19.88 mg/g at initial pH 6.3. Adsorption was found to increase with increasing pH. Furthermore, maximum desorption (60%) of dye was achieved in water medium at pH 10.0.

Orange peel waste was also examined by Namasivayam et al. [167] for the removal of congo red, procion orange and rhodamine B dyes. Acidic pH was found to be favorable for the adsorption of three dyes. Arami et al. [168] also studied the use of orange peel as low-cost adsorbent for the removal of textile dyes from aqueous solutions. Direct Red 23 and Direct Red 80 were used as model compounds. The adsorption capacity was found to be 10.72 and 21.05 mg/g for the two studied dyes at initial pH 2.

Banana peel, a commonly produced fruit waste, was examined as adsorbent for the removal of Cd(II) from environmental and industrial wastewater by Memon et al. [169]. An adsorption capacity of 35.52 mg/g was reported for Cd(II). They used the same adsorbent for Cr(VI) removal and reported an adsorption capacity of 131.56 mg/g in case of Cr(VI) [170]. The potential of banana peel as a sorbent for removing phenolic compounds from olive mill wastewaters has also been investigated by Achak et al. [171]. Banana peel showed high adsorption capacity for phenolic compounds (689 mg/g), exhibiting that banana peel could be employed as a promising adsorbent for phenolic compounds adsorption. Low-cost banana and orange peels were used as adsorbents for the adsorption of dyes from aqueous solutions by Annadurai et al. [172]. The adsorption capacities for both peels decreased in the order methyl orange > methylene blue > rhodamine B > congo red > methyl violet > amido black 10B. Based on the adsorption capacity, it was concluded that banana peels were more effective than orange peels for dyes removal.

Thermally treated watermelon peels (TWMP) have been utilized for the removal of methyl parathion (MP) pesticide from water by Memon et al. [173]. Maximum adsorption (99 ± 1%) was achieved for $(0.38–3.80) \times 10^{-4}$ mol/dm^3 of MP solution, using 0.1 g of adsorbent in 20 mL of solution for 60 min agitation time at pH 6. Activated carbons prepared from waste cassava peel (an agricultural waste from the food processing industry) employing physical and chemical methods have been investigated for their efficiency in the removal of dyes and metal ions from aqueous solutions [174]. The adsorbent was found to be effective for model pollutants removal from water. The ability of pomelo peel to remove Cd(II) from aqueous solution by Saikaew et al. was investigated [175]. Cadmium removal was found to increase significantly by increasing the pH of the solution from 1 to 5. At pH 5, the cadmium removal reached a maximum value. The equilibrium process was described well by the Langmuir isotherm model, with a maximum sorption capacity of 21.83 mg/g. The sorption was relatively quick (approximately 20 min) and adsorption kinetics followed pseudo-second-order model.

Mango peel waste (MPW) was evaluated as a sorbent for the removal of Cd^{2+} and Pb^{2+} from aqueous solution [176]. The maximum sorption capacity of Cd^{2+} and Pb^{2+} was found to be 68.92 and 99.05 mg/g, respectively. The kinetics of sorption of both metals was fast, reaching equilibrium in 60 min. Sorption kinetics and equilibria followed pseudo-second-order and Langmuir adsorption isotherm models. FTIR analysis revealed that carboxyl and hydroxyl functional groups were mainly responsible for the sorption of Cd^{2+} and Pb^{2+}. Chemical modification of MPW for blocking of carboxyl and hydroxyl groups showed that 72.46% and 76.26% removal of Cd^{2+} and Pb^{2+}, respectively, was achieved due to the involvement of carboxylic group, whereas 26.64% and 23.74% was due to the hydroxyl group. EDX analysis of MPW before and after metal sorption and release of cations (Ca^{2+}, Mg^{2+}, Na^+, K^+) and proton from MPW with the corresponding uptake of Cd^{2+} and Pb^{2+} revealed that the main mechanism of sorption was ion exchange. The regeneration experiments showed that the MPW could be reused for five cycles without significant loss in its initial sorption capacity. The same researchers also studied the potential of mango peel for the removal of Cu^{2+}, Ni^{2+}, and Zn^{2+} from constituted metal solutions and genuine electroplating industry wastewater [177].

The potential of garlic peel to remove methylene blue from aqueous solution was evaluated in a batch process [178]. Experiments were carried out as function of contact time, initial concentration (25–200 mg/L), pH (4–12) and temperature (303, 313 and 323 K). The maximum monolayer adsorption capacities were found to be 82.64, 123.45, and 142.86 mg/g at 303, 313, and 323 K, respectively. Besides these, various other peels from different sources have been explored as adsorbents for the removal of pollutants [179–187].

4.1.6. *Adsorbents from shells of different agricultural waste*

Shells of various agricultural products have also been investigated as adsorbents by different researchers for the removal of toxic pollutants from water. Bael fruit shell was used to prepare activated carbon and used as an efficient low-cost adsorbent to remove Cr(VI) from aqueous phase [188]. Maximum chromium removal was found at pH 2.0 in an equilibrium time of 240 min. The Langmuir monolayer sorption capacity was found to be 17.27 mg/g.

Acid formaldehyde pretreated chestnut shell was as adsorbent and the influence of initial cation concentration, temperature and pH was investigated to optimize Pb^{2+}, Cu^{2+} and Zn^{2+} removal from aqueous solutions [189]. The maximum adsorption capacity was obtained as 8.5 mg/g for Pb^{2+} ions and the order of cation affinity was $Pb^{2+} > Cu^{2+} > Zn^{2+}$. The sorption of Cu(II) onto chestnut shell in a batch adsorber has been studied [190]. The adsorption capacity of chestnut shell for Cu(II) was found to be 12.56 mg/g at 293 K. The kinetic data were found to follow the pseudo-second-order model.

Raw Brazil nut shells were explored as adsorbents for the adsorption of methylene blue and Indigo Carmine dyes [191]. The maximal uptakes were 7.81 mg/g for methylene blue, and 1.09 mg/g for Indigo Carmine, at room temperature. Thermodynamic studies showed that the adsorption of both dyes was spontaneous and exothermic occurring with entropy decrease. Carbonaceous adsorbents namely, activated (AC-PW) and non-activated (C-PW) were prepared from the Brazilian pine-fruit-shell (*Araucaria angustifolia*) and tested as adsorbents for the removal of Procion Red MX 3B dye (PR-3B) from aqueous effluents [192]. The activation process increased the specific surface area, average porous volume, and average porous diameter of the adsorbent AC-PW, when compared with C-PW. The effects of shaking time, adsorbent dosage and pH on adsorption capacity were studied. PR-3B uptake was favorable at pHs 2.0–3.0 for C-PW and 2.0–7.0 for AC-PW. The contact time required to obtain the equilibrium using C-PW and AC-PW as adsorbents was 6 and 4 h at 298 K, respectively.

The adsorption of two basic dyes, methylene blue and crystal violet on wood apple shell (WAS) was investigated using a batch adsorption technique [193]. It was observed that the WAS adsorbent showed higher adsorption capacity for crystal violet (130 mg/g) than methylene blue (95.2 mg/g). The FTIR studies indicate that the interaction of dye and WAS surface was found to occur via the nitrogen atoms of the adsorbate and oxygen groups of the adsorbent. The adsorption of dyes onto WAS proceeds according to a pseudo-second-order model.

The potential of shells of hazelnut (HNS) (*Corylus avellana*) and almond (AS) (*Prunus dulcis*) to remove Pb^{2+} from aqueous solutions was investigated by batch experiments [194]. The influences of initial Pb^{2+} ion concentration (0.1–1.0 mM), pH (2–9), contact time (10–240 min) and adsorbent amount (0.1–1.0 g) have been investigated. The equilibrium sorption capacities of HNS and AS were 28.18 and 8.08 mg/g, respectively, after equilibrium time of 2 h. The experimental result inferred that adsorption, chelation and ion exchange were major adsorption mechanisms for binding the Pb^{2+} ion onto the sorbents.

Hard shell of apricot stones was selected from agricultural solid wastes to prepare effective and low-cost adsorbent for gold separation from gold-plating wastewater [195]. The results showed that under the optimum operating conditions, more than 98% of gold was adsorbed onto activated carbon after only 3 h.

The feasibility of coconut shell charcoal (CSC) and commercial activated carbon (CAC) for Cr(VI) removal was investigated in batch mode using synthetic electroplating wastewater by Babel and Kurniawan [196]. Surface modifications of CSC and CAC with chitosan and/or oxidizing agents, such as sulfuric acid and nitric acid, respectively, were also conducted to improve removal efficiency. It was reported that both CSC and CAC, which were oxidized with nitric acid, showed higher Cr(VI) adsorption capacities (CSC: 10.88, CAC: 15.47 mg/g) than those oxidized with sulfuric acid (CSC: 4.05, CAC: 8.94 mg/g) and non-treated CSC coated with chitosan (CSCCC: 3.65 mg/g). It was suggested by them that surface modification of carbon adsorbent with a strong oxidizing agent generates more adsorption sites on their solid surface for metal adsorption.

Activated carbon was prepared from hazelnut shells using two steps of carbonization followed by steam activation by Khalkhali and Omidvari [197] and investigated for the adsorption of mercuric ion. It was reported that pretreatment or impregnation of prepared adsorbent with chemicals such as permanganate, dichromate, EDTA and dithizone (chelating agents), methylene blue dye, and Na_2S increased the adsorption efficiency of mercuric ion from aqueous solutions. Walnut shells were used for the production of powdered activated carbon by chemical activation method using $ZnCl_2$ as an activating agent and further investigated for Hg(II) removal from water by Zabihi et al. [198]. They reported an adsorption capacity of 151.5 mg/g for Hg(II) by prepared adsorbent. Besides these, shells from different agricultural wastes have also been explored as adsorbents for the removal of various pollutants [199–201].

4.1.7. Adsorbents from seed, seed coat, stem and stalk of different agricultural products

Many researchers also examined the suitability of seed, seed coat, stem and stalk of various agricultural products as inexpensive adsorbents for the removal of toxic pollutants from water.

The feasibility of papaya seeds (PS), for the methylene blue adsorption has been investigated [202]. Batch adsorption studies were conducted to study the effects of contact time, initial concentration (50–360 mg/L), pH (3–10) and adsorbent dose (0.05–1.00 g) on the removal of methylene blue (MB) at 30 °C. The data fitted well with the Langmuir model with a maximum adsorption capacity of 555.55 mg/g. The pseudo-second-order kinetics fitted well for the adsorption of MB by PS with good correlation.

Tamarind seeds were used as adsorbent after activation for the removal of Cr(VI) from aqueous solutions [203]. The tamarind seeds were activated by treating with concentrated sulfuric acid (98%, w/w) at 150 °C. The adsorption of Cr(VI) was found to be maximum at low initial pH in the range of 1–3. The maximum adsorption capacity of 29.7 mg/g was observed at an equilibrium pH ranging from 1.12 to 1.46. The adsorption process followed second-order kinetics The regenerated activated tamarind seeds showed more than 95% Cr(VI) removal of that obtained using the fresh activated tamarind seeds. A feasible solution was proposed for the disposal of the contaminants (acid and base solutions) containing high concentrations of Cr(VI) obtained during the regeneration (desorption) process. Guava and mango seeds have also been used as adsorbents for the removal of dyes and phenols from aqueous solutions [204–206].

The potential of palm seed coat for the adsorption of *o*-cresol was tested by Rengaraj et al. [207] and an adsorption capacity of 19.58 mg/g was observed with film diffusion as the rate limiting step during the process. Rubber seed coat was used as activated carbon for the removal of phenols using batch and column operations [208]. It was suggested that the adsorbed phenol could be desorbed by sodium hydroxide. The adsorption process was found to follow first-order kinetics and the isotherm fitted to both Freundlich and Langmuir models. Activated carbon prepared from rubber (*Hevea brasiliensis*) seed coat was used to remove basic blue 3 (BB3) from aqueous solutions [209]. Batch adsorption studies were conducted to evaluate the effect of contact time, and initial concentration (50–500 mg/L) on the removal of BB3. The monolayer adsorption capacity was observed as 227.27 mg/g at 30 °C. The adsorption kinetics was well described by the pseudo-second-order kinetic model.

Pineapple stem waste, an agricultural waste, was investigated as low-cost adsorbent to remove methylene blue from aqueous solutions [210]. The monolayer adsorption capacity of 119.05 mg/g was reported in this study. Grape stalks waste, generated in the wine production process, have been used as an adsorbent for the removal of copper and nickel ions from aqueous solutions by Villaescusa et al. [211]. Maximum uptake of 1.59×10^{-4} mol of copper and 1.81×10^{-4} mol of nickel per gram of dry sorbent was reported. The workers suggested that lignin C–O bond might be involved in metal uptake based on Fourier transform infrared spectrometry analysis.

Sunflower stalks as adsorbents were used for the removal of two basic dyes (methylene blue and basic red 9) and two direct dyes (congo red and direct blue 71) from aqueous solutions by Sun and Xu [212]. The maximum adsorption of two basic dyes on sunflower stalks was found to be 205 and 317 mg/g for methylene blue and basic red 9, respectively. They also reported relatively lower adsorption for two direct dyes on sunflower stalks. The removal of metal ions such as copper, cadmium, zinc, and chromium ions from aqueous solutions has also been studied using sunflower stalks as adsorbents [213]. The maximum adsorption of four heavy metals was 29.3 mg/g (Cu^{2+}), 30.73 mg/g (Zn^{2+}), 42.18 mg/g (Cd^{2+}), and 25.07 mg/g (Cr^{3+}).

The Cr(VI) removal from aqueous system under different process conditions by sunflower waste was examined by Jain et al. [214]. Two adsorbents were prepared by pretreating the sunflower stem waste. One adsorbent was prepared by boiling it and second adsorbent was prepared by treating it with formaldehyde. It was found that the removal of chromium was dependent on the physico-chemical characteristics of the adsorbent, adsorbate concentration and other studied process parameters. Maximum metal removal was observed at pH 2.0. The efficiencies of boiled sunflower stem adsorbent and formaldehyde-treated sunflower stem adsorbent for the removal of Cr(VI) were 81.7% and 76.5%, respectively for dilute solutions at 4.0 g/L adsorbent dose.

4.1.8. Adsorbents developed from miscellaneous agricultural wastes

Besides the various agricultural by-products mentioned above, different miscellaneous agricultural wastes have also been investigated as low-cost adsorbents. Some of the important ones have been briefly summarized here. Hasar [215] utilized almond husk as a potential adsorbent for the removal of Ni(II) ions. Almond husk samples were carbonized at different temperatures ranging from 300 to 1000 °C under nitrogen atmosphere in an oven for 1 h. They were activated by carbon dioxide gas for 1 h. Adsorbent obtained by this procedure is termed as '*Modified Activated Carbon-I*, *MAC-I*'. In second experimental series, a portion of the powdered material was chemically activated by washing 10% H_2SO_4 (in weight 1:1) for 6 h. Then, this material was carbonated at different temperatures under nitrogen atmosphere for 1 h, and it was again activated by carbon dioxide gas at 1 h. Adsorbent obtained by second experimental series is termed as '*Modified Activated Carbon-II*, *MAC-II*'. This study proved that MAC-II performed better for Ni(II) removal.

Utilization of grape bagasse as an adsorbent to remove Cd(II) and Pb(II) ions has been investigated by Farinella et al. [216]. Adsorption capacity was found to be 0.774 and 0.428 mmol/g for Cd(II) and Pb(II), respectively. The adsorption of iron and manganese either to remove each metal ion singly or both metals as a multi-component system was studied using maize cob and palm fruit bunch [217]. The adsorption capacities for iron on palm fruit bunch and maize cob were in the range of 57–80% and for the manganese in the 50–79% range for initial concentrations between 1 and 10 mg/L. In case of both metal ions present in a mixture, removal of iron from the mixture was in the 54–79% range and for the manganese in the 54–76% range.

The adsorption potential of black gram husk (BGH) was tested for the removal of different metal ions from water [218]. The maximum amount of heavy metals adsorbed at equilibrium was 49.97, 39.99, 33.81, 25.73 and 19.56 mg/g on BGH for Pb, Cd, Zn, Cu and Ni, respectively. The sorption capacities were found to be pH dependent and the maximum adsorption occurred at the solution pH 5. Dried sugar beet pulp, was used as sorbent for the removal of copper(II) from aqueous solution [219]. The results indicated that at 250 mg/L initial copper(II) concentration, dried sugar beet pulp exhibited the highest copper(II) uptake capacity of 28.5 mg/g at 25 °C and at an initial pH value of 4.0. The intraparticle diffusion played an important role in the sorption mechanisms of copper(II), and sorption kinetics followed pseudo-first- and pseudo-second-order kinetic models.

Sugarcane bagasse was converted into a carbonaceous adsorbent and used for the removal of cadmium and zinc from wastewater [220]. It was found that adsorption of Cd(II) and Zn(II) ions increased with an increase in temperature and solution pH. The adsorption capacity of bagasse carbon for Cd(II) and Zn(II) was 38.03 and 31.11 mg/g. Bagasse pith, a waste product from sugarcane industry has been studied by McKay et al. [221] without any pretreatment, for the removal of two basic dyes and two acidic dyes from aqueous solutions. High adsorptive capacity was observed for the adsorption of basic dyes (158 mg/g for basic blue 69 and 177 mg/g for basic red 22), while lower capacity of 23 and 22 mg/g was observed for acid red 114 and acid blue 25, respectively.

The removal of Cu^{2+} and Pb^{2+} from aqueous solutions using barley straw (BS) was investigated [222]. The influences of initial Cu^{2+} and Pb^{2+} concentration (0.1–1 mM), pH (2–9), contact time (10–240 min) and adsorbent amount (0.1–1.0 g) were investigated. Adsorption of Cu^{2+} and Pb^{2+} was found to be pH dependent in all cases showing maximum equilibrium adsorption at pH 6.0. The equilibrium sorption capacity of BS for Cu^{2+} and Pb^{2+} after 2 h were 4.64 and 23.20 mg/g, respectively. Carbon prepared from silk cotton hull was used to remove a textile dye (reactive blue MR) from aqueous solution [223]. Adsorption process was found to be dependent on solution pH, dye concentration, carbon concentration and contact time. Equilibrium was attained within 60 min. Adsorption followed both Langmuir and Freundlich isotherm models. The adsorption capacity was reported as 12.9 mg/g at an initial pH of 2 ± 0.2 for the particle size of 125–250 μm at room temperature (30 ± 2 °C).

The potential of pearl millet husk (PMHC) as an adsorbent was explored by Inbaraj et al. [224]. An adsorption capacity of 82.37 mg/g of this adsorbent for methylene blue at pH 6.0 was reported. They further reported that methylene blue adsorption on PMHC was a chemisorption process and formic acid could be used to remove the adsorbed dye. The potential of cotton plant wastes – stalk (CS) and hull (CH) – as sorbents for the removal of Remazol Black B (RB5) was investigated [225]. The results indicated that adsorption was strongly pH dependent but slightly temperature-dependent for each sorbent–dye system. CS and CH sorbents exhibited the highest RB5 dye uptake capacities of 35.7 and 50.9 mg/g, respectively, at an initial pH value of 1.0. It was found that both, external mass transfer and intraparticle diffusion, played an important role in the adsorption mechanisms of dye, and adsorption kinetics followed the pseudo-second-order kinetic model for each sorbent.

The adsorption potential of thermally activated carbon developed from maize cob for the removal of 2,4-dichlorophenol (2,4-DCP) from aqueous solutions has been investigated by Sathishkumar et al. [226]. The maximum adsorption capacity was reported as 17.94 mg/g for the particle size of 250–500 μm. Chemically modified maize cob carbon was prepared by pyrolysis at 300 and 400 °C for 35 min followed by steeping in saturated ammonium chloride [227]. The prepared carbon was tested for the removal of three metal ions. The effectiveness of the modified maize cobs in removing the metal ions from solution was found to be Zn > Ni > Cd. The removal efficiency of the metal ions was dependent on the metal-ion concentration and carbonization temperature. Besides these, several other researchers also examined the potential of maize and corn cob for the removal of aquatic pollutants [228,229].

Bark is one of the waste product from the timber industry. Edgehill and Lu [230] investigated the potential of carbonized slash pine bark for the removal of phenol. The adsorption capacities for phenol and pentachlorophenol (PCP) at pH 2 and 8 were evaluated. The calculated Freundlich constants, $K = 0.41–0.58\ \text{mmol/g/(mmol dm}^{-3})^{1/n}$ and $1/n = 0.30–0.41$, were reported. Various other agricultural waste materials have also been investigated by different researchers to test their adsorption potential for different aquatic pollutants [231–238]. A comparison of various low-cost adsorbents derived from different agricultural wastes for the removal of diverse types of aquatic pollutants is summarized in Table 1.

4.2. Industrial and municipal wastes as low-cost adsorbents

Widespread industrial activities generate huge amount of solid waste materials as by-products. Some of this material is being put to use while others find no proper utilization and are dumped elsewhere. The industrial waste material is available almost free of cost and causes major disposal problem. If the solid wastes could be used as low-cost adsorbents, it will provide a two-fold advantage to environmental pollution. Firstly, the volume of waste materials could be partly reduced and secondly the low-cost adsorbent if developed can reduce the pollution of wastewaters at a reasonable cost. In view of the low cost of such adsorbents, it would not be necessary to regenerate the spent materials. Thus, a number of industrial wastes have been investigated with or without treatment as adsorbents for the removal of pollutants from wastewaters. Some of them are discussed here.

Table 1
Adsorption capacities of different agricultural wastes as adsorbents for the removal of various pollutants from water.

S. No.	Adsorbent	Adsorbate	Adsorption capacity	Reference
1.	Raw rice husk	Cd(II)	8.58 mg/g	[96]
2.	Epichlorohydrin treated rice husk	Cd(II)	11.12 mg/g	[96]
3.	NaOH treated rice husk	Cd(II)	20.24 mg/g	[96]
4.	$NaHCO_3$ treated rice husk	Cd(II)	16.18 mg/g	[96]
5.	H_3PO_4-treated rice bran	Ni(II)	102 mg/g	[98]
6.	Rice husk	Safranine	838 mg/g	[99]
7.	Rice husk	Methylene blue	312 mg/g	[99]
8.	Rice husk ash	Acid violet 54, acid violet 17, acid blue 15, acid violet 49 and acid red 119	99.4–155 mg/g	[100]
9.	Rice husk ash	Methylene blue dye	690 mg/g	[101]
10.	Rice husk ash	Indigo Carmine	29.3–65.9 mg/g	[102]
11.	Rice husk carbon	Cr(VI)	45.6 mg/g	[105]
12.	Rice husk	Basic blue 9	19.83 mg/g	[106]
13.	Rice husk	Acid yellow 36	86.9 mg/g	[107]
14.	Sulfuric acid and zinc chloride activated rice husk carbon	Crystal violet	64.87 and 61.57 mg/g	[108]
15.	Rice husk	*p*-Chlorophenol	14.36 mg/g	[109]
16.	Rice husk	*p*-Nitrophenol	15.31 mg/g	[109]
17.	Wheat bran	Pb(II)	69.0–87.0 mg/g	[113]
18.	Wheat bran	Reactive blue 19, reactive red 195 and reactive yellow 145	117.6–196.1 mg/g	[116]
19.	Tea waste	Cu(II) and Pb(II)	48–65	[118]
20.	Turkish tea waste	Cu(II) and Cd(II)	6.65–8.64 mg/g (Cu) 2.59–11.29 mg/g (Cd)	[120]
21.	Tea factory waste	Zn(II)	8.9 mg/g	[121]
22.	Spent tea leaves	Methylene blue	300.05 mg/g	[122]
23.	Tea waste	Methylene blue	86.16 mg/g	[123]
24.	Blend coffee	Cu(II) and Cd(II)	2.0 mg/g	[126]
25.	Coffee residue	Pb(II)	63 mg/g	[127]
26.	Coir pith waste	Pb(II)	263 mg/g	[135]
27.	Coir pith waste	Co(II), Cr(III) and Ni(II)	12.82, 11.56 and 15.95 mg/g	[136]
28.	Coir pith waste	Rhodamine B and acid violet	2.56 and 8.06 mg/g	[137]
29.	Coir pith	Congo red	6.72 mg/g	[138]
30.	Coconut bunch waste	Methylene blue	70.92 mg/g	[139]
31.	Coconut activated carbon	2,4,6-Trichlorophenol	716.10 mg/g	[140]
32.	Coconut husk activated carbon	Methylene blue	434.78 mg/g	[141]
33.	Functionalized coconut fibers	Cd(II)	0.2–5 mmol/g	[142]
34.	Coconut copra meal	Cd(II)	4.99 mg/g	[144]
35.	Male flower of coconut treated with H_2SO_4 and H_3PO_4	Crystal violet	60.42–85.84 mg/g	[145]
36.	EDTA-modified groundnut husk	Cd(II) Pb(II)	0.36 mmol/g 0.19 mmol/g	[150]
37.	Peanut hull	Cu(II) Pb(II)	0.21 mmol/g 0.18 mmol/g	[155]
38.	Peanut hull	Cu(II)	21.25 mg/g	[156]
39.	Peanut husk	Neutral Red	37.5 mg/g	[158]
40.	Orange peel	Ni(II)	158 mg/g	[164]
41.	Orange peel	Acid violet 17	19.88 mg/g	[166]
42.	Orange peel	Direct Red 23 and Direct Red 80	10.72 and 21.05 mg/g	[168]
43.	Banana peel	Cd(II)	35.52 mg/g	[169]
44.	Banana peel	Cr(VI)	131.56 mg/g	[170]
45.	Banana peel	Phenolic compounds	689 mg/g	[171]
46.	Banana peel	Methyl orange Methylene blue Rhodamine B Congo red Methyl violet Amido black 10B	17.2 mg/g 15.9 mg/g 13.2 mg/g 11.2 mg/g 7.9 mg/g 7.9 mg/g	[172]
47.	Orange peel	Methyl orange Methylene blue Rhodamine B Congo red Methyl violet Amido black 10B	15.8 mg/g 13.9 mg/g 9.1 mg/g 7.9 mg/g 6.1 mg/g 3.8 mg/g	[172]
48.	Pomelo peel	Cd(II)	21.83 mg/g	[175]
49.	Mango peel	Cd(II) Pb(II)	68.92 mg/g 99.05 mg/g	[176]

Table 1 (*Continued*)

S. No.	Adsorbent	Adsorbate	Adsorption capacity	Reference
50.	Mango peel	Cu(II) Ni(II) Zn(II)	46.09 mg/g 39.75 mg/g 28.21 mg/g	[177]
51.	Garlic peel	Methylene blue	82.64–142.86 mg/g	[178]
52.	Waste jack fruit peel carbon	Malachite green	166.37 mg/g	[179]
53.	Bael fruit shell	Cr(VI)	17.27 mg/g	[188]
54.	Chestnut shell	Cu(II)	12.56 mg/g	[190]
55.	Brazil nut shell	Methylene blue, Indigo caramine dye	7.81 mg/g 1.09 mg/g	[191]
56.	Wood apple shell	Methylene blue Crystal violet	95.2 mg/g 130 mg/g	[193]
57.	Hazelnut shell Almond shell	Pb(II)	28.18 mg/g 8.08 mg/g	[194]
58.	$ZnCl_2$-modified walnut shells	Hg(II)	151.5 mg/g	[198]
59.	Hazelnut shell	Basic blue 9	8.82 mg/g	[199]
60.	Papaya seed	Methylene blue	555.55 mg/g	[202]
61.	Tamarind seed	Cr(VI)	29.7 mg/g	[203]
62.	Palm seed coat	*o*-Cresol	19.58 mg/g	[207]
63.	Rubber seed coat	Basic blue 3	227.27 mg/g	[209]
64.	Pineapple stem waste	Methylene blue	119.05 mg/g	[210]
65.	Sunflower stalks	Methylene blue and basic red 9	205 and 317 mg/g	[212]
66.	Sunflower stalks	Copper, cadmium, zinc, and chromium	29.3 mg/g (Cu^{2+}), 30.73 mg/g (Zn^{2+}), 42.18 mg/g (Cd^{2+}), 25.07 mg/g (Cr^{3+})	[213]
67.	Grape bagasse	Cd(II) Pb(II)	0.774 mmol/g 0.428 mmol/g	[216]
68.	Black gram husk	Pb, Cd, Zn, Cu and Ni	19.56–49.97 mg/g	[218]
69.	Sugar beet pulp	Cu(II)	28.5 mg/g	[219]
70.	Sugarcane bagasse	Cd(II) Zn(II)	38.03 mg/g 31.11 mg/g	[220]
71.	Bagasse pith	Basic blue 69 and basic red 22	158 and 177 mg/g	[221]
72.	Bagasse pith	Acid red 114 and acid blue 25	23 and 22 mg/g	[221]
73.	Barley straw	Cu(II) Pb(II)	4.64 mg/g 23.2 mg/g	[222]
74.	Silk cotton hull	Reactive blue MR	12.9 mg/g	[223]
75.	Pearl millet husk	Methylene blue	82.37 mg/g	[224]
76.	Cotton stalk Cotton hull	Remazol Black B	35.7 mg/g 50.9 mg/g	[225]
77.	Maize cob	2,4-Dichlorophenol	17.94 mg/g	[226]
78.	Date pits	Basic blue 9	17.3 mg/g	[231]
79.	Palm pith carbon	2,4-Dichlorophenol	19.16 mg/g	[232]
80.	Maize cob	Basic dyes (Astrazone blue and maxilon red)	160 and 94.5 mg/g	[233]
81.	Maize cob	Acid dyes (telon blue and erinoyl red)	47.7 and 41.4 mg/g	[233]
82.	Pinewood	Acid blue 264	1176 mg/g	[235]
83.	Pinewood	Basic blue 69	1119 mg/g	[235]
84.	Pinewood	Basic blue 9	556 mg/g	[235]

4.2.1. *Fly ash*

The major solid waste by-product of thermal power plants based on coal burning is fly ash. The main uses of fly ash include construction of roads, bricks, cement, etc. The high percentage of silica and alumina in fly ash make it a good candidate for utilization as an inexpensive adsorbent for bulk use. Panday et al. [239] used fly ash without any pretreatment for the removal of Cu^{2+} and found that the adsorption data conforms to Langmuir model. Sen and Arnab [240] investigated the potential of fly ash for Hg(II) removal and an adsorption capacity of 2.82 mg/g was reported by them. The removal of lead and copper from aqueous solution by fly ash was investigated under various experimental conditions, e.g. contact time, pH and temperature [241]. The level of uptake of Pb^{2+} and Cu^{2+} by the fly ash generally increased, but not in a progressive manner, at higher pH values. The effect of temperature on the uptake of Pb^{2+} and Cu^{2+} revealed that the adsorption was enhanced on lowering the temperature. The experimental results emphasized the potential of fly ash for the recovery of metal ions from wastewater. The main mechanisms involved in the removal of heavy metal ions from solution were adsorption at the surface of the fly ash and precipitation.

A special iron-abundant fly ash was used to develop a novel adsorbent for arsenic(V) removal from wastewater through simple chemical processes [242]. In the synthesis process, the inherent iron in the fly ash was rearranged and loaded on the surface of the fly ash by dissolution and precipitation processes. The results showed that porous amorphous FeOOH was loaded on the surface of the adsorbent (HIOFAA) successfully. The adsorption capacity for arsenic removal was found to be 19.46 mg/g.

The use of fly ash for phenol removal was reported by Khanna and Malhotra [243]. The kinetics and mechanism of phenol removal were investigated. They also provided useful data for the design of phenol–fly ash adsorption systems. Fly ash was also used by Haribabu et al. [244] for the removal of phenol and chlorophenols and the process was found to be endothermic and followed first-order kinetics. Akgerman and Zardkoohi [245] also investigated the potential of fly ash as an adsorbent for the removal of phenolic compounds and found it to have an adsorption capacity of 67, 20 and 22 mg/g for phenol, 3-chlorophenol and 2,4-dichlorophenol, respectively. Aksu and Yener [246] investigated the potential of fly ash as a substitute for activated carbon for phenol adsorption. The maximum phenol adsorption capacity was found to be 27.9 mg/g for fly ash and 108.0 mg/g for granular activated carbon at initial phenol concentration of 100 mg/L. They further investigated the removal of *o*-chlorophenol and *p*-chlorophenol and reported that the pollutant binding capacity of the adsorbent was influenced by the substituted groups, initial pH and initial mono-chlorinated phenol concentration [247]. Fly ash was also utilized for the removal of 2-chlorophenol (2-CP) and 2,4-dichlorophenol (2,4-DCP) by Kao et al. [248]. It was reported that the adsorption of 2-CP and 2,4-DCP was affected by the solution pH.

Sarkar et al. [249] investigated the kinetic aspects of adsorption of some priority organic pollutants, viz. phenol (hydroxybenzene), *o*-hydroxyphenol (1,2-dihydroxybenzene), *m*-hydroxyphenol (1,3-dihydroxybenzene), and 4-nitrophenol (1-hydroxy-4-nitrobenzene), on fly ash. The process was found to be of complex nature controlled by both, surface adsorption and pore diffusion, the extent being estimated from the diffusion coefficient value. Efficacy of fly ash (FA) and impregnated fly ash (IFA) was also examined in removing the phenols from aqueous solution [250]. Impregnated fly ash has shown better efficiency of phenol removal than fly ash. It was observed that polar substituted phenols having less steric hindrance were better adsorbed than others. Substituted phenols with hindered group were less adsorbed than phenol (*m*-nitrophenol > *o*-nitrophenol > phenol > *m*-cresol > *o*-cresol). The observed trend was explained due to the presence of various substituents in the phenolic compounds. The removal mechanism of phenols was explained due to chemical coagulation with metallic oxides. Singh et al. [251,252] investigated the adsorption of 2,4-dichlorophenol and tetrachlorocatechol by bagasse fly ash (BFA), rice husk fly ash (RHFA) and activated carbon (AC). The removal kinetics of 2,4-dichlorophenol showed first-order rate expression and equilibrium adsorption data suited well for both Langmuir and Freundlich isotherms. Fly ash has also been examined by other researchers for phenols removal [253,254]. Fly ash was also studied by Viraraghavan and Ramakrishna [255] for the removal of cationic and anionic dyes. The process was found to follow first-order kinetics and the isotherms conformed both Freundlich and Langmuir models. Few other researchers also examined the use of fly ash for dyes removal [256,257].

Chemically modified fly ash was investigated for the removal of fluoride from drinking water by Goswami and Das [258]. It was reported by them that the potential of the modified fly ash bed was satisfactory for fluoride removal. The effectiveness of fly ash in removing phosphate from aqueous solution and its related removal mechanism was studied by Lu et al. [259]. Results showed that the removal of phosphate by fly ash was rapid and the removal percentage of phosphate in the first 5 min reached 68–96% of the maximum removal of phosphate by fly ash. The removal processes of phosphate by fly ash included fast kinetics and high removal representing precipitation, then a slower and longer removal due to adsorption. The pH and Ca^{2+} concentration of fly ash suspension were decreased with the addition of phosphate, which suggested that calcium phosphate precipitation was a major mechanism of the phosphate removal. Comparison of the relative contribution of the adsorption and precipitation to the total removal of phosphate by fly ash showed that the adsorption accounted for 30–34% of the total removal of phosphate, dependent on the content of CaO in fly ash. XRD patterns of the fly ash before and after phosphate adsorption revealed that phosphate salt ($CaHPO_4{\cdot}2H_2O$) was formed in the adsorption process. Therefore, the removal of phosphate by fly ash was attributed to the formation of phosphate precipitation as a brushite and the adsorption on hydroxylated oxides. The results suggested that the use of fly ash could be a promising solution for the removal of phosphate in the wastewater treatment and pollution control. Environmental-benign utilization of fly ash as low-cost adsorbents has been reviewed by Wang and Wu [260].

4.2.2. Steel industry wastes (blast furnace slag, sludge and dust)

The steel industry also produces a number of wastes in large quantities such as blast furnace slag, dust and sludge, etc. and these have been investigated as adsorbents. Dimitrova [261] investigated ungranulated blast furnace slag for the removal of Cu^{2+}, Ni^{2+}, and Zn^{2+} ions from water and reported that slag alkalizing activity creates conditions for adsorption through hydroxocomplex formation and colloidal particles of silicic acid. Slag columns were utilized by the same worker [262] for lead removal. Kanel et al. [263] used blast furnace slag for As(III) remediation. The maximum As(III) adsorption by slag was reported as 1.40 mg As(III)/g of slag at 1 mg/L As(III) initial concentration. Several other researchers also tested the potential of slag for different metal ions removal [264–268].

Many researchers also investigated the potential of slag for phosphate removal from water [269–273]. Yamada et al. [269] studied phosphate removal using soft and hard granulated slag and observed that phosphate adsorbed well on soft granulated slag than on the hard granulated slag and explained it on the basis of porosity of the adsorbent. Kinetic studies on phosphorus (P as orthophosphate) sorption onto sandy loam soil and slag were conducted by Lee et al. [270] which revealed that more than 90% of P was adsorbed within 70 and 12 h for the soil and slag, respectively. The pH of the P solution played a critical role in the rate of P removal. Removal was minimum at pH 2. Dominant removal mechanism of P at pH less than 8 was physical sorption, while it was chemical precipitation at pH greater than 10. Adsorptive capacity of the slag was 225 and 53 times of that of the soil for the static and dynamic systems, respectively. Breakthrough curves obtained from the column experiments were S-shaped and more spreaded with the decrease of influent P concentration. Mobility of P was restricted by the adsorbent due to its high sorption capacity of P.

Blast furnace slag was also used by Oguz [271] to remove phosphate from aqueous solutions. The mechanisms of phosphate removal were explained on the basis of the results of X-ray spectroscopy, measurements of zeta potential of particles, specific surface area, and images of scanning electron microscopy (SEM) of the particles before and after adsorption. It was proposed that the removal of phosphate predominantly occurred by a precipitation mechanism and weak physical interactions between the surface of adsorbent and the metallic salts of phosphate. Heated samples of acid-treated slag and mixtures of slag with kaolinite and $Al(OH)_3$ were examined by Jha et al. [273] to determine their uptake capacities for NH_4^+ and PO_4^{3-} from aqueous solutions. The highest PO_4^{3-} uptake capacity was obtained with the slag–$Al(OH)_3$ mixture heated at 900 °C whereas the highest uptake of NH_4^+ was found in the slag–$Al(OH)_3$ mixture heated at 800 °C. It was shown that heating slag composites prior to incorporation into wastewater treatment filters resulted in an increased degree of NH_4^+ and PO_4^{3-} uptake.

Slag has also been used for dyes removal [274,275]. The utilization of treated basic oxygen furnace slag (BOF slag) was successfully carried out to remove three synthetic textile dyes (reactive blue 19 (RB19), reactive black 5 (RB5) and reactive red 120 (RR120)) from aqueous solutions by Xue et al. [275]. In the batch system, the maximum dye uptake on acid-treated BOF slag adsorbent was observed at pH 2.0, and the maximum RB5, RB19 and RR120 uptake capacities (at 500 mg/L dye concentration) were 76, 60 and 55 mg/g, respectively. Adsorption kinetic process followed first-order kinetic model.

Other waste materials generated in steel industry are blast furnace sludge and blast furnace flue dust, which have also been explored as possible adsorbents. Jallan and Pandey [276] reported the use of blast furnace sludge as adsorbent without any treatment for the removal of some toxic ions, viz. Pb^{2+}, Ni^{2+}, Cd^{2+}, Cu^{2+}, Zn^{2+} and CN^-. It was found by them that sludge has a good adsorptive capacity for metal ions as well as cyanide but the maximum adsorption (9 mg/g) was poor in the case of Zn^{2+}. Sludge was also tested as adsorbent by López-Delgado et al. [277] for the removal of some heavy metal ions who reported that metal ions were adsorbed in the order, $Pb^{2+} > Ni^{2+} > Cd^{2+} > Cu^{2+} > Zn^{2+}$. The process was found to be endothermic and the adsorption data fitted both to Freundlich and Langmuir isotherms. Patnaik and Das [278] investigated the use of blast furnace flue dust as adsorbent for the removal of hexavalent chromium and found the applicability of first-order kinetics for the adsorption process.

4.2.3. Aluminium industry waste (red mud)

Red mud, a solid waste product of aluminium industry produced during bauxite processing, was explored as adsorbent by López et al. [279] for wastewater treatment. The maximum adsorption capacities for Cu^{2+}, Zn^{2+}, Ni^{2+} and Cd^{2+} were found to be 19.72, 12.59, 10.95 and 10.57 mg/g, respectively for a contact time of 48 h. Red mud has also been explored as an adsorbent for the removal of lead and chromium by Gupta et al. [280]. Freundlich and Langmuir models were found to fit the sorption data. The maximum adsorption capacity for Pb(II) by red mud was reported 64.79 mg/g (batch capacity) and 88.20 mg/g (column capacity). The maximum adsorption capacity for Cr(VI) was 35.66 mg/g (batch capacity) and 75.00 mg/g (column capacity). Red mud has been explored as an adsorbent for arsenic removal from water [281,282]. It was found by the researchers that an alkaline aqueous medium (pH 9.5) favored As(III) removal, whereas the acidic pH range (1.1–3.2) was effective for As(V) removal. The capacities were reported as 4.31 μmol/g at pH 9.5 for As(III) and 5.07 μmol/g at pH 3.2 for As(V). The workers reported that heat and acid treatments on red mud increased its adsorptive capacity. Arsenic adsorption on acid and heat treated red mud was also found pH dependent, with an optimum range of 5.8–7.5 for As(III) and 1.8–3.5 for As(V).

Red mud was also used for the removal of fluoride from aqueous solution by Çengeloğlu et al. [283]. They used both the original and activated red mud forms in batch mode and found that the adsorption capacity of activated form for fluoride was higher than that of original form. They further reported that the maximum fluoride removal occurred at pH 5.5. Red mud has also been used to remove phosphate (P) from aqueous solutions by Huang et al. [284]. Acid and acid-thermal treatments were employed to treat the raw red mud. It was observed that the red mud with HCl treatment showed the highest adsorption capacity among all the red mud samples, giving adsorption capacity of 0.58 mg/g at pH 5.5 and 40 °C.

Fly ash and red mud have been employed as adsorbents for the removal of methylene blue dye from aqueous solutions by Wang et al. [285]. It was found that fly ash generally showed higher adsorption capacity than red mud. The adsorption capacity of 1.4×10^{-5} and 7.8×10^{-6} mol/g, for raw fly ash and red mud, respectively, have been reported. Red mud was explored as low-cost adsorbent for the removal of phenol, 2-chlorophenol, 4-chlorophenol, and 2,4-dichlorophenol from wastewater by Gupta et al. [286]. It was reported that 2,4-dichlorophenol and 4-chlorophenol were adsorbed by the developed adsorbent up to 94–97%, while the removal of 2-chlorophenol and phenol was up to 50–81%. The order of removal was 2,4-dichlorophenol > 4-chlorophenol > 2-chlorophenol > phenol. It was suggested by them that the removal of phenols took place through a particle diffusion mechanism. The adsorption was found to be endothermic in nature and followed both Langmuir and Freundlich models. Neutralized red mud was used for phenol removal from aqueous solution [287]. The experiments demonstrated that maximum phenol removal was obtained in a wide pH range of 1–9 and it took 10 h to attain equilibrium. Namasivayam and Arasi [288] investigated the adsorption potential of red mud for congo red dye removal from aqueous solution. The removal capacity of the red mud for the dye was 4.05 mg/g. Wang et al. [289] have discussed the novel applications of red mud as coagulant, adsorbent and catalyst for environmentally benign processes in their comprehensive review.

4.2.4. Fertilizer industry waste

Fertilizer industry also produces a number of by-products in large quantities which create serious disposal problems and degrade the surrounding environment. Fe(III)/Cr(III) hydroxide, a waste material from the fertilizer industry has been used by Namasivayam and Ranganthan [290] for the adsorption of Cr(VI) from aqueous solution. The adsorption data fitted with both Freundlich and Langmuir models. Namasivayam and Senthilkumar [291] investigated the potential of Fe(III)/Cr(III) hydroxide waste for the removal of As(V). Adsorption was explained by considering zero-point of charge (ZPC) of the adsorbent material. The pHzpc of waste Fe(III)/Cr(III) hydroxide was 8.3. In the pH range 3–7, $H_2AsO_4^-$ was predominantly adsorbed. The adsorbent acquired positive charge in this pH range and adsorption was facilitated by coulombic interactions. In the pH range 7–10, specific interactions occurred, since dissociation of arsenic acid is expected. The transfer of a proton to the hydroxyl groups of the Fe(III)/Cr(III) hydroxide surface is also possible. A surface complexation model was proposed for As(V) adsorption on Fe(III)/Cr(VI) waste.

Waste Fe(III)/Cr(III) hydroxide has also been investigated for phosphate removal from water [292] and an adsorption capacity of 6.5 mg/g was reported. The potential use of Fe(III)/Cr(III) hydroxide as an adsorbent for the removal of bisphenol A and 2-aminophenol from aqueous solution has also been investigated [293]. The Langmuir adsorption capacity of untreated and pretreated adsorbent was found to be 3.47 and 3.67 mg/g, respectively, for bisphenol A and 2.94 and 6.03 mg/g for 2-aminophenol.

The waste carbon slurry, another fertilizer industry waste, was tested to remove Cu^{2+}, Cr^{6+}, Hg^{2+}, and Pb^{2+} from aqueous solutions by Srivastava et al. [294] and it showed excellent adsorption capacities for the studied metal ions. Carbon slurry waste has also been investigated for the removal of dyes, phenols, anions, pesticides, etc. from water [295–298] and found to be promising adsorbent.

4.2.5. Leather industry waste

The feasibility of leather industry solid waste was evaluated as an adsorbent for removal of Cr(VI) and As(V) from aqueous media [299]. The high amounts of Cr(VI)-133 mg/g and As(V)-26 mg/g adsorbed demonstrated the great potential for using this solid waste from the leather industry as a low-cost alternative to the traditionally used adsorbent materials. Fleshing from animal hides/skins, another waste from leather industry, is high in protein content. Raw fleshing has been complexed with iron and is used for the removal of chromium(VI) [300]. Iron treatment greatly improved adsorption of the fleshing for hexavalent chromium. The ultimate adsorption capacity of iron treated fleshing was 51 mg of chromium(VI) per gram of fleshing. The sorption capacity of untreated fleshing was 9 mg/g indicating that iron treatment significantly increased the adsorption capacity of fleshing. The adsorption kinetics was well described by pseudo-second-order kinetic model. X-ray photoelectron spectroscopic (XPS) studies showed that the iron was incorporated into the protein matrix. Shifts in XPS spectra suggested that dichromate binding occurred with iron at active adsorption sites and that iron treated fleshing removed chromium(VI) without reducing it to chromium(III).

Sekaran et al. [301] used buffing dust, waste generated from leather industry, for the removal of dyes. The adsorption capacity of buffing dust was found to be 6.24 mg/g at pH 3.5 and 30 °C for acid brown dye. The removal of methylene blue from aqueous solutions by adsorption on tanned solid wastes was studied by Tahiri et al. [302]. The maximum adsorption capacity was found ca. 84 mg of methylene blue for each gram of buffing dust.

4.2.6. Paper industry wastes

Paper industry also produces a number of by-products in large quantities which create serious disposal problems and degrade the surrounding environment. Black liquor, a waste product originated from paper industry, was examined for the adsorption of Pb^{2+} and Zn^{2+} by Srivastava et al. [303]. An adsorption capacity of 1865 and 95 mg/g was reported for Pb^{2+} and Zn^{2+}, respectively, at 40 °C. The adsorption of phenols on papermill sludges was studied by Calce et al. [304]. The retention capacity of papermill sludges was found in the order: 2-nitrophenol = 4-nitrophenol $\ll$ 2-chlorophenol < phenol < 4-chlorophenol $\leq$ 3–chlorophenol < 2,4-dichlorophenol < 3,4-dichlorophenol = 2,4,5-trichlorophenol < 3,5-dichlorophenol. Papermill sludge was also investigated for the removal of orange G dye (an anionic dye) from aqueous solutions [305]. The adsorption capacity of papermill sludge adsorbent for orange G dye was found to be 62.3 mg/g at 25 °C.

Shimada et al. [306] used waste newspaper as raw material for the production of activated carbon. The developed adsorbent showed good sorption capacity as evident by iodine (1310 mg/g) and methylene blue numbers (326 mg/g). The removal of Cu^{2+} from water was investigated using adsorbents prepared from paper industry waste (de-inking paper sludge and sludge from virgin pulp mill) [307]. Experimental results showed that de-inking paper sludge lead to mesoporous materials whereas the sludge from virgin pulp mill produced high microporous adsorbents. Adsorbent materials were then used for Cu^{2+} removal from water at acid pH. During water treatment, heavy metals lixiviation from adsorbent materials was not produced. However, Ca and Mg leaching was observed. Final pH significantly increased after treatment of water with adsorbent materials probably due to their elevated $CaCO_3$ content. In general, highest Cu^{2+} removal was obtained using adsorbent materials from de-inking paper sludge. This result could be due to their higher content in oxygenated surface groups, high average pore diameter, elevated superficial charge density, high $CaCO_3$ amount and high Ca and Mg exchange content.

4.2.7. Different types of wastes sludges as adsorbent

Different industries produce sludge as by-products which have been examined as adsorbents by various researchers. Chrome sludge, a solid waste material from an electroplating industry, was used as an adsorbent for As(V) removal by Lee and Lee [308]. The maximum sorption capacity of chrome sludge for As(V) was reported as 21 mg/g. Chrome sludge was also tested to remove dyes from aqueous solutions [309] and the results indicated that the sludge had a better affinity for acid dyes than basic dyes.

Color removal of an aqueous solution containing water-based inks, a mixture of resins, pigments, and dyes, was investigated by using metal hydroxide sludge, which is a waste from the electroplating industry by Netpradit et al. [310]. The maximum adsorption capacity for the red ink was reported as 34.48 mg/g of metal hydroxide sludge. Waste metal hydroxide sludge was used as low-cost adsorbent for removing a reactive textile dye (*Remazol Brilliant Blue*) by Santos et al. [311]. The maximum adsorption capacity of 91.0 mg/g was found at 25 °C and pH 7.

Sewage sludge has also been used to develop an effective adsorbent. This adsorbent was employed for the removal of azo dye such as Direct Dark Brown M and Acid Mordant Brown RH by Chen et al. [312]. The equilibrium adsorption capacity of a carbonaceous adsorbent prepared using the sludge obtained from city wastewater treatment plant was 502 and 329.7 mg/g for Direct Dark Brown M and Acid Mordant Brown RH, respectively. Sewage sludge was converted into carbonaceous materials by chemical activation with sulfuric acid by Rio et al. [313]. The developed adsorbents were tested for copper ion, phenol and dyes (acid red 18 and basic violet 4) removal from aqueous solution as well as volatile organic compounds (VOC) removal from gas phase. An adsorption capacity of 77–83 mg/g for copper, 41–53 mg/g for phenol and VOC adsorption capacities (acetone and toluene) ranging between 12 and 54 mg/g were reported. Thawornchaisit and Pakulanon [314] investigated the potential of dried sewage sludge for the removal of phenol from aqueous solution. The adsorption capacity decreased from 94 to 5 mg/g when adsorbent concentration was increased from 0.5 to 10 g/L, suggesting a possible competitive effect of leachable heavy metals from the sludge.

Dried activated sludge has been explored for the sorption of phenol, *o*-chlorophenol and *p*-chlorophenol from aqueous solutions [315]. The maximum sorption capacity of dried activated sludge was found to be 86.1 mg/g for phenol, 102.4 mg/g for *o*-chlorophenol and 116.3 mg/g for *p*-chlorophenol at 100 mg/L initial pollutant concentration. Selvaraj et al. [316] investigated the potential of distillery sludge for Cr(VI) removal and reported the Langmuir adsorption capacity of 5.7 mg/g for Cr(VI). Li et al. [317] utilized wine processing waste sludge as an effective adsorbent for Cr(III) removal. Adsorption dynamics had been successfully described by the Lagergren model and intraparticle diffusion model.

Biological sludge after physical activation with steam was studied for industrial effluent treatment [318]. It was shown that after activation, there was an improvement of pore and adsorption characteristics of the adsorbent, with specific surface area reaching 230 m^2/g and equilibrium adsorption capacities of 50 mg/g for phenol.

4.2.8. Miscellaneous industrial wastes as adsorbents

Namasivayam and Yamuna [319] explored waste biogas residual slurry for Cr(III) removal from aqueous solutions. The maximum adsorption potential of biogas residual slurry for Cr(III) was found 7.8 mg/g at pH 2.5.

Solid wastes from the distiller waste (DW), which is the by-product of the ammonia-soda process for the production of soda ash, has been used as an alternative adsorbent for removing the anionic dyes from aqueous medium [320]. The results indicated that adsorption was strongly pH dependent. The typical dependence of dye uptake on temperature and the kinetics of adsorption indicated the process to be chemisorption. The results showed that as the pH increased, extent of dye uptake increased and $Ca(OH)_2$ particles precipitated at higher pHs were mainly responsible for the removal of anionic Procion Crimson H-EXL (reactive red 231) dye. The required time to reach the equilibrium was found to be less than 2 min. Dye uptake process followed the pseudo-second-order rate expression. The Freundlich isotherm best fitted for the adsorption of the dye.

Battery industry waste has been investigated for the removal of some metal ions (Pb, Cu, Cr and Zn) from aqueous solution by Bhatnagar et al. [321] and an appreciable sorption potential (33–64 mg/g) of the prepared adsorbent was observed for the metal ions. The adsorption characteristics of basic yellow 28 (BY 28) and basic red 46 (BR 46) onto boron waste (BW), a waste produced from boron processing plant were investigated [322]. Kinetic studies indicated that the kinetics of the adsorption of BY 28 and BR 46 onto BW followed a pseudo-second-order model. The maximum adsorption capacities for BY 28 and BR 46 are reported as 75.00

Table 2
Adsorption capacities of different industrial wastes as adsorbents for the removal of various pollutants from water.

S. No.	Adsorbent	Adsorbate	Adsorption capacity (mg/g)	Reference
1.	Fly ash	Cu(II)	1.39	[239]
2.	Fly ash	Hg(II)	2.82	[240]
3.	Fly ash	As(V)	19.46	[242]
4.	Fly ash	Phenol	5.58–6.48	[244]
		4-Chlorophenol	8.62–10.0	
		2,4-Dichlorophenol	8.16–8.72	
5.	Fly ash	Phenol, 3-chlorophenol and 2,4-dichlorophenol	67, 20 and 22	[245]
6.	Fly ash	Phenol	27.9	[246]
7.	Fly ash and impregnated fly ash	Phenol	3.8–6.3	[253]
		o-Cresol	3.1–4.7	
		m-Cresol	3.5–5.5	
		p-Cresol	4.6–6.7	
		o-Nitrophenol	5.8–6.9	
		m-Nitrophenol	6.5–8.3	
		p-Nitrophenol	7.8–9.6	
8.	Fly ash	2,4-Dimethylphenol	1.39	[254]
9.	Fly ash	Alizarin sulfonic	11.21	[256]
10.	Fly ash	Basic blue 9	75.52	[257]
11.	Blast furnace slag	As(III)	1.40	[263]
12.	Blast furnace slag	Pb(II)	40	[264]
		Cr(VI)	7.5	
13.	Blast furnace slag	Zn, Cd	17.66, 18.72	[265]
14.	Activated slag	Cu, Ni	30.0, 29.35	[266]
15.	Modified basic oxygen furnace slag	Reactive blue 19	76	[275]
		Reactive black 5	60	
		Reactive red 120	55	
16.	Blast furnace sludge	Pb(II)	64.17–79.87	[277]
		Cu(II)	16.07–23.66	
		Cr(III)	9.55–16.05	
		Cd(II)	6.74–10.15	
		Zn(II)	4.25–9.65	
17.	Red mud	Cu^{2+}, Zn^{2+}, Ni^{2+} and Cd^{2+}	19.72, 12.59, 10.95 and 10.57	[279]
18.	Red mud	Pb(II) and Cr(VI)	64.79 and 35.66	[280]
19.	Red mud	Phosphate	0.58	[284]
20.	Neutralized red mud	Phenol	4.12	[287]
21.	Red mud	Congo red dye	4.05	[288]
22.	Waste Fe(III)/Cr(III) hydroxide	Phosphate	6.5	[292]
23.	Untreated and pretreated waste Fe(III)/Cr(III) hydroxide	Bisphenol A	3.47 and 3.67	[293]
24.	Untreated and pretreated waste Fe(III)/Cr(III) hydroxide	2-Aminophenol	2.94 and 6.03	[293]
25.	Solid waste from leather industry	As(V) and Cr(VI)	26 and 133	[299]
26.	Iron complexed leather industry waste	Cr(VI)	51	[300]
27.	Buffing dust	Acid brown dye	6.24	[301]
28.	Tanned solid wastes	Methylene blue	84	[302]
29.	Black liquor	Pb^{2+} and Zn^{2+}	1865 and 95	[303]
30.	Biogas residual slurry	Cr(III)	7.8	[319]
31.	Boron industry waste	Basic yellow 28 and basic red 46	75.00 and 74.73	[322]

and 74.73 mg/g, respectively. The dye adsorption was found to be dependent on the initial pH of the solution with maximum uptake occurring at about pH 9.

A comparison of various low-cost adsorbents derived from industrial and municipal wastes for the removal of diverse types of aquatic pollutants is summarized in Tables 2 and 3, respectively.

5. Conclusions and future perspectives

In this review, an attempt has been made to focus on the recent developments related to the detoxification of water and wastewater by low-cost adsorbents utilizing agro-industrial and municipal wastes. It is important to note here that the maximum adsorption capacities reported in this paper provide some idea of

Table 3
Adsorption capacities of different types of sludges as adsorbents for the removal of various pollutants from water.

S. No.	Adsorbent	Adsorbate	Adsorption capacity (mg/g)	Reference
1.	Metal hydroxide sludge	Red ink	34.48	[310]
2.	Metal hydroxide sludge	Remazol Brilliant Blue	91.0	[311]
3.	Sewage sludge	Direct Dark Brown M, Acid Mordant Brown RH	502 and 329.7	[312]
4.	Sewage sludge	Copper	77–83	[313]
5.	Sewage sludge	Phenol	41–53	[313]
6.	Dried activated sludge	Phenol, *o*-chlorophenol and *p*-chlorophenol	86.1, 102.4 and 116.3	[315]
7.	Distillery sludge	Cr(VI)	5.7	[316]
8.	Biological sludge	Phenol	50	[318]

adsorbent's effectiveness for each type of pollutant, and mainly depends on experimental conditions. The use of waste materials as low-cost adsorbents for removing various pollutants from water and wastewater presents many attractive features especially their contribution in the reduction of costs for waste disposal, therefore contributing to environmental protection. Although the amount of available literature data on the use of low-cost adsorbents in water and wastewater treatment is increasing at a tremendous pace, there are still several gaps which need to be filled. Some of the important issues can be summarized below:

(1) Selection and identification of an appropriate low-cost adsorbent is one of the key issues to achieve the maximum removal/adsorption of specific type of pollutant depending upon the adsorbent–adsorbate characteristics.
(2) The conditions for the production of low-cost adsorbents after surface modification for higher uptake of pollutants need to be optimized.
(3) Cost factor should not be ignored. Low production cost with higher removal efficiency of adsorbents would make the process economical and efficient.
(4) Mechanistic studies need to be performed in detail to propose a correct binding mechanism of aquatic pollutants with low-cost adsorbents.
(5) Regeneration studies need to be performed in detail with the pollutants-laden adsorbent to recover the adsorbate as well as adsorbent. It will enhance the economic feasibility of the process.
(6) The potential of low-cost adsorbents under multi-component pollutants needs to be assessed. This would make a significant impact on the potential commercial application of low-cost adsorbents to industrial systems.
(7) There is scarce data available for the competitive adsorption of pollutants (metal ions adsorption in presence of phenols, dyes and other contaminants and vice-versa). Therefore, more research should be conducted in this direction.
(8) It is further suggested that the research should not limit to only lab scale batch studies, but pilot-plant studies should also be conducted utilizing low-cost adsorbents to check their feasibility on commercial scale.
(9) The effectiveness of the treatment depends not only on the properties of the adsorbent and adsorbate, but also on various environmental conditions and variables used for the adsorption process, e.g. pH, ionic strength, temperature, existence of competing organic or inorganic compounds in solution, initial adsorbate/adsorbent concentration, contact time and speed of rotation, particle size of adsorbent, etc. These parameters should also be taken into account while examining the potential of low-cost adsorbents.
(10) The development in the field of adsorption process using low-cost adsorbents essentially requires further investigation of testing these materials with real industrial effluents.
(11) Last but not the least, environmentally safe disposal of pollutants-laden adsorbents is another important topic of concern which should not be overlooked.

If it is possible to develop such adsorbents having all the above-mentioned characteristics, then these adsorbents may offer significant advantages over currently available expensive commercially activated carbons and, in addition contribute to an overall waste minimization strategy.

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