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Removal of toxic pollutants from aqueous medium through adsorption: a review

Asma Shafique

Department of Chemistry, Government College University, Lahore-54000, Pakistan, email: asmashafiqe246@gmail.com

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A B S T R A C T

Water pollution is increasing day by day due to the wastewater discharge from various industries. Different pollutants like pesticides, insecticides, bromate ions, heavy metals such as copper, zinc, antimony, lead, nickel, cadmium, mercury, etc. create various health and environmental problems due to their toxicity. Overall, this review paper intends to provide literature concerning the removal of various pollutants from aqueous medium by adsorption with two objectives. First, it offers various health effects associated with different pollutants in wastewater followed by various adsorbent materials and applications, adsorption capacities, and certain characteristics of these adsorbents to remove pollutants from aqueous solution.

Keywords: Wastewater; Pollutants; Adsorption; Dyes, Metals; Organic pollutants

1. Introduction

Water is recognized as a precious resource for human civilization and all kinds of life on earth, so its demand is increasing with the improvement of living standards and the rapidly growing global population [1–3]. Due to global climate change and population growth, the current water supply is facing enormous challenges. Millions of people and animals are suffering a water crisis with a shortage of fresh and clean water throughout the world due to the release of pollutants in freshwater supplies [4–6]. According to the WHO report, 780 million people worldwide still lack access to clean water sources.

Rapid growth in industrialization, urbanization, inadequate sanitation, household wastes, agricultural practices, strenuous activities of human beings, unrestrained use and exploitation of natural water resources which have steadily increased over the last decades, are main and major sources of water pollution [4,7–10]. The disposal of highly polluted water is rising. It has become a serious issue and the worst enemy for humans and animals on planet earth. Reduction in economic status, environment, and human sustenance are the results of water pollution [11–15]. High levels of toxic and bio-degradable pollutants produce considerable adverse effects on the environment, plants, animals, and human beings [16,17]. In developing countries, almost 70–80% of the diseases [18,19] are mainly related to contaminated water. Accumulation of pollutants and their ions in water results in cerebral and nervous system damage, memory loss, an increase of allergies, blood pressure, poisoning, cancer, lungs, kidneys and other organs diseases and cell death which ultimately leads to death [7]. These pollutants and effluents also affect the nature of water by inhibiting the penetration of sunlight in water, which results in the reduction of photosynthesis reaction. Besides this, these pollutants are the main cause of serious ecological issues. They pose a great threatening to aquatic life, animals and plants due to their non-biodegradable and toxic nature [20–22]. So, the most basic humanitarian goal of the 21st century is to provide reliable access to clean and affordable water and to reduce, eliminate or treat the non-biodegradable, toxic, and life-threatening pollutants from the wastewater [23]. Therefore, researchers are executing and focusing on promoting feasible technologies for removing or converting the pollutants and metal ions present in the water into less harmful substances [24]. The present review article intends to present the relevant studies on the adsorption of various pollutants from aqueous medium using various adsorbents, particularly those that are economical and easily available, while also commenting on their efficiency.

2. Various types of pollutants in wastewater

Different types of pollutants such as pathogenic organisms, endocrine-disrupting chemicals, pharmaceutical products and personal care products (PCP), phenols [25,26], pesticides, bromate ions (BrO_3^-) [22], oils, metal-bearing contaminated effluents [27], dyes [28,29], and heavy metals, etc. [20,21,30–32] which are released from anthropogenic sources like metallurgical, galvanizing, metal finishing, power generation, electronic device manufacturing, metal plating, steel, fertilizers and paper industries, mining operations, pesticides, nuclear power plants, coal processing, battery manufacturing processes, paints and pigments production, glass operations, smelters metallurgy, pharmaceutical, and PCP, oil refineries, petrochemicals, textiles, leather tanning, municipal and stormwater runoff [16,17,33–36] are increasing day by day in the ground, industrial, surface and household water. Some common pollutants present in wastewater are discussed below:

2.1. Pesticides

Pesticides like organophosphorous compounds, dimethoate, methyl parathion, phosphamidon, fenitrothion, malathion, monocrotophos, and phorate [37] are the compounds used in domestic and agricultural activities that deter, incapacitate, kills or discourage pests. These are toxic and can harm more than just the "pests" at which they are targeted. These pesticides and their metabolites are also contaminating natural resources of water, and exposure to pesticides can cause a number of health effects, including kidney problems, immune and endocrine system, cancer, infertility, and behavioral and neurological disorders and are also affecting animals, fish, and birds population [38–40]. Malathion is an example of toxic dyes that may cause vomiting, chest infection, diarrhea, vision problems, diseases of eyes, dizziness, sweating, headache, unconsciousness, difficulty in breathing, and even death [37].

2.2. Insecticides

Insecticides, which are very active and toxic compounds formulated to kill or harm different species of insects, are mainly used in the agriculture and medicine industries. These are of mainly two types, including ovicides and larvicides used against insect eggs and larvae, respectively. These are available in different forms like gels, baits, dusts and sprays, having the potential to alter ecosystems and can pose different levels of risk to non-target insects, people, pets, and the environment and their exposure can affect the liver, respiratory system in humans and can cause severe headache, influenza, nausea, abdominal pain, cancer, paralysis, diarrhea, and sweating [41].

2.3. Dyes

Dyes discharged from printing, textile, food, and leather industries are toxic and carcinogenic pollutants [42–44]. These are of various types like acidic, dispersive, reactive, and basic, but two main types are cationic (basic) and anionic (acidic), which are commonly used in these industries. More than 7×10^5 metric tons of dyes are produced annually throughout the world, and at the time of manufacturing, almost 12% of these dyes are misplaced, from which 20% move in water bodies, which causes water pollution [45]. These dyes inhibit the penetration of sunlight in water reducing the reaction of photosynthesis and affect the nature of water and aquatic life [4,22] and also cause adverse effects on human health by causing skin, eyes, kidney diseases, allergic dermatitis and cancer etc.

2.4. Phenolic compounds

Phenolic compounds released into water resources by the industries of steel, pharmaceuticals, chemical, fertilizers, dyes, phenolic resins, petrochemical, and petroleum refineries are considered toxic and dangerous [46,47]. The content of phenolic compounds in industrial wastewater should be less than 0.5 mg/L according to the standard limits but in industrial wastewater, their amount is almost 200–2,000 mg/L, which is very high. Short-term exposure to high levels of phenol has caused irritation of the respiratory tract and muscle twitching in animals; anorexia, progressive weight loss, diarrhea, vertigo, salivation, dark coloration of the urine have been reported in humans due to long term exposure [48].

2.5. Bromine

Bromate ion (BrO_3^-) is a bromine-based oxoanion which is formed when ozone used to disinfect drinking water reacts with naturally occurring bromide found in water. It is a human carcinogen that causes nausea, cancer, diarrhea, vomiting, anuresis, anemia, pulmonary, and central nervous system diseases, depression, and abdominal pain [34].

2.6. Heavy metals

The word 'heavy metal' refers to the element having a higher density, specific gravity greater than 5.0, atomic weight between 63.5 and 200.6 [49], and toxic even at very low concentration [50,51]. Unlike most organic pollutants, heavy metals are generally refractory, notorious, highly toxic, non-thermo degradable, carcinogenic and non-biodegradable elements, so there is a risk that the surrounding soil, as well as the ground and surface water, is contaminated by metal-containing effluents, then these effluents accumulate in the body of living organisms and cause a variety of diseases. In addition, if these heavy metals are present in the water above the permissible limit, they cause incurable toxicity, which threatened the growth of flora and fauna [52,53]. Main pervasive and hazardous heavy metals present in wastewater and induce dangerous and toxic effects to the living environment are lead, copper, cadmium, chromium, cobalt, mercury, nickel, phosphorus, arsenic, zinc, and antimony etc. [14,49,54].

2.6.1. Lead

Lead is among the very toxic heavy metal ions even at low concentrations when found in the environment. It usually exists in sulfide, galena, and cerussite forms. Its main cause is effluents from lead-acid batteries, electroplating, steel industries. It enters the environment through air, soil, and water and finally enters the food chain [7]. It causes a decrease in intelligent quotient (IQ) score, physical growth retardation and impaired functions of hearing, decreased attention, and performance in learning in children and mainly affects the central nervous system, brain, reproductive parts, kidney, and liver. It may also cause diseases such as encephalopathy, anemia, high blood pressure, hepatitis, and nephritic syndrome in males and females of middle and old age [55,56]. Its availability is of high risk to plants and animals' life also. The maximum permitted level of lead in drinking water is 0.5 mg/L. Hence, the removal of lead ions from water and wastewater is essential for the protection of the environment and public health [57,58].

2.6.2. Copper

Copper is an essential element that humans need for the development of bones and enzyme synthesis. It usually presents in different forms like elemental, cupric, or cuprous ions, and cupric is considered the most toxic form present in the environment. Copper and its compounds are frequently discharged from municipal water, mining, metallurgy, steel and fertilizer industries, pigments, paints, and electroplating runoff. The maximum permissible concentration of copper is 1.3 and 1.0 mg/dm^3, respectively by World Health Organization (WHO) and the United States Public Health Service (USPHS) in drinking water [59,60] and heavy doses of copper beyond this limit lead to depression, liver and gastrointestinal diseases, renal failure, hair fall, anemia, central nervous system retardation and mucosal irritation [61,62]. It may accumulate in the brain, liver, and pancreas, which lead to death. Therefore, it is needed to recover the copper from industrial effluents and aquatic systems.

2.6.3. Cadmium

Cadmium and its derivatives are another types of water pollutants that have been listed as a carcinogen by the Environmental Protection Agency (EPA) [63]. Even at low concentrations, its compounds get concentrated in the ecosystem and are harmful. These are discharged into water from the industrial wastewater, especially from pharmaceutical, paint, paper, nickel-cadmium batteries, electroplating, coal processing, and pulp industries, and refineries are hazardous to all types of life. When ingested, these replace the Zn^{2+} at key enzymatic sites, which results in the metabolic, bone marrow, and kidney disorders, digestive system diseases, renal failure, hypertension, cardiovascular system damage, cancer, choking, and anemia Moreover, these also create negative effects on eyes, lungs, skin, liver and unborn babies and are proved to be very toxic and objectionable even at trace level to aquatic life. According to WHO, 0.005 mg/L is the maximum permitted level of Cd^{2+} in drinking water [64]. So, it is needed to remove the cadmium and its derivatives from wastewater.

2.6.4. Chromium

Chromium is the seventh-largest element in the earth's crust, where it is found in the form of ores. It usually exists in oxidation states from +2 to +6, but only Cr(III) and Cr(VI) are of significance in water and according to EPA Cr(VI) is highly toxic to aquatic life even at very low concentrations and usually causes cancer and other diseases in humans. It also causes skin diseases, pulmonary, and digestives system failure [65,66]. Its compounds usually enter into aquatic systems by discharging concentrated effluents mainly from plastic coatings, tanning, electroplating of metals, paint, and textile industries. The legal discharge limit of chromium in water varies from 0.5 mg/L (surface water) to 2.0 mg/L (in sewer) [67] and according to the WHO and EPA 0.05 mg/L is the maximum permitted level of Cd^{2+} in drinking water [68–70]. Due to these harmful effects, it should be removed or modified into less toxic ions before its discharge into to water bodies.

2.6.5. Mercury

Mercury (Hg^{2+}) usually exists in elemental, mercuric or mercurous ions and is the highly toxic metal that is present in wastewater. It is released in different forms in the aquatic system from various sources like paper and pulp manufacturing, oil refineries, chloralkali wastewater, power generation plants, and fertilizers industries and has no positive impacts on human life [71,72] and Overexposure of mercury causes ill effects on the human nervous system, kidneys, reproductive and respiratory system and other body systems, like one of its compounds named methyl mercury damaged enzymes sites and affects the protein synthesis system. Minamata convention was designed to preserve the environment and health of humans from mercury. The prescribed limit for mercury is 0.001 ppm, which is the lowest among all heavy metal ions. The maximum permitted level of mercury in drinking water is 0.002 mg/L [20]. Because of its harmful effects, its removal from industrial wastes has gained special attention.

2.6.6. Arsenic

Arsenic, which is toxic heavy metal for humans, animals, and plants, exists in the different oxidation states like −3, 0, +3, and +5 but arsenite (AsO_3^{3-}) and arsenate (AsO_4^{3-}) known as arsenic(III) and arsenic(V) are common forms present in wastewater from which arsenite is more mobile and toxic [73]. The main sources of arsenic are natural [70,74], in addition, many factors like biological and geological actions, mining activities, anthropogenic activities, mining activities, fossil fuels burning, arsenic pesticides, and disposal of fly ash elevate the concentration of arsenic in soil and water. The maximum permitted level of chromium in drinking water is 0.05 mg/L, and due to its toxicity, drinking water contaminated with arsenic beyond its permitted level causes severe health problems, including anemia, cardiovascular and skin diseases, lungs, liver, bladder, and kidney cancer [75,76]. Table 1 shows various types of pollutants, their permitted limits, sources and toxic effects.

Table 1
Various types of pollutants, their permitted limit, sources and toxic effects

Pollutants	Permitted level	Sources	Diseases	References
Pesticides	–	Used in domestic and agricultural activities	Diseases of kidney, immune and endocrine system, cancer, behavioral and neurological disorders, and infertility	[38–40]
Insecticides	–	Agriculture and medicine industries	Liver and respiratory system disorders, severe headache, influenza, nausea, abdominal pain, cancer, paralysis, diarrhea, and sweating	[41]
Dyes	–	Printing, textile, food, and leather industries	Vomiting, chest infection, diarrhea, problems of vision, diseases of eyes, dizziness, sweating, headache, unconsciousness, difficulty in breathing, and even death	[37]
Phenolic compounds	–	Industries of steel, pharmaceuticals, chemical, fertilizers, dyes, phenolic resins, refineries of petroleum	Respiratory tract irritation and twitching of muscles in animals, weight loss, diarrhea, dark coloration of the urine, anorexia, vertigo, salivation, liver and blood disorders in humans	[48]
Bromate Ions	–	Reaction naturally occurring bromide with Ozone which disinfects drinking water	Nausea, cancer, diarrhea, vomiting, anuresis, anemia, pulmonary and central nervous system diseases, depression, abdominal pain	[34]
Lead	0.5 mg/L	–	Effects the central nervous system, brain, reproductive parts, kidney and liver, diseases such as encephalopathy, anemia, high blood pressure, hepatitis and nephritic syndrome in males and females of middle and old age and ultimately causes death	[55]
Copper	1.3 mg/dm^3	Municipal water, fertilizer, pigments, paints, and electroplating industries runoff	Depression, liver and gastrointestinal diseases, renal failure, central nervous system retardation, and mucosal irritation	[61]
Cadmium	0.005 mg/L	Pharmaceutical, paint, paper, nickel-cadmium batteries, electroplating, coal processing, and pulp industries	Metabolic, bone marrow and kidney disorders, digestive system diseases, renal failure, hypertension, cardiovascular system damage, cancer, choking and anemia; moreover, it also creates negative effects on eyes, lungs, skin, liver, and unborn babies	[178]
Chromium	0.05 mg/L	Plastic coatings, tanning, electroplating of metals, textile and paint industry	Cancer	[70]
Mercury	0.002 mg/L	Paper manufacturing, power generating plants, refineries of oil, fertilizer industry	Nervous system disorders	[20]
Arsenic	0.05 mg/L	Natural sources, biological and geological, anthropogenic and mining activities, use of arsenic pesticides and herbicides, by the burning of fossil fuels and disposal of fly ash	Cardiovascular and skin diseases, lungs, liver, bladder, and kidney cancer and anemia	[76]

3. Sequestration of water pollutants

To control the water pollution, several technologies and conventional treatment methods have been developed. The selection of water treatment process (physical, chemical, and biological processes) is a complex task which involves the consideration of many factors like, available space for the construction of treatment unit, reliability of equipment used in the treatment process, waste disposal limits, quality of desired water and operating costs. Various technologies like adsorption [77,78], activated sludge, aerobic and anaerobic treatment, biological removal, catalytic reduction, coagulation/flocculation [22], photocatalysis, chromatographic techniques, electrodialysis [7] electrolysis, foam or froth flotation, electrochemical technology [23], Fenton degradation, membrane filtration [56,79], ion exchange method [80], reverse osmosis [14,30], microbial reduction, oxidation processes, precipitation, photosonochemical method, photodegradation [81–86], phytoextraction, ultra-filtration [21], sonochemical and sonoelectrochemical technology, solvent extraction and UV irradiation etc. either independent or in conjunction are available with promising results to control water pollution and for the removal of toxic substances. Fig. 1 shows various available technologies for the removal of different types of pollutants from wastewater.

3.1. Adsorption method

For the removal, determination, and treatment of hazardous pollutants which are discharged from various industries and to control water pollution, currently, adsorption has been proven and believed to be main and simple, most attractive and effective, widely adopted and economically feasible advance process due to the better mechanical, chemical, thermal, and radiation stabilities, easy operation, simple design, high efficiency, economic convenience and low cost [16,24,48,87,88].

The term adsorption is defined as accumulating a substance at the interphase between two phases like 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 known as "adsorbent". The efficiency of adsorbents is characterized by surface area, effectiveness, number of active sites, the selectivity of adsorbents, and adsorbent kinetics [53,89]. Different adsorbents including organic as well as inorganic ion-exchange materials [90], composite ion-exchangers [91–93], activated carbon [94], clay minerals [4], silica, alumina and some industrial solid wastes [95], zeolites [96], polymeric adsorbents [97–99], fibers [100], hydrogels [101], agriculture materials, biomaterials [102], peat and bentonite [103] have gained appreciable status in

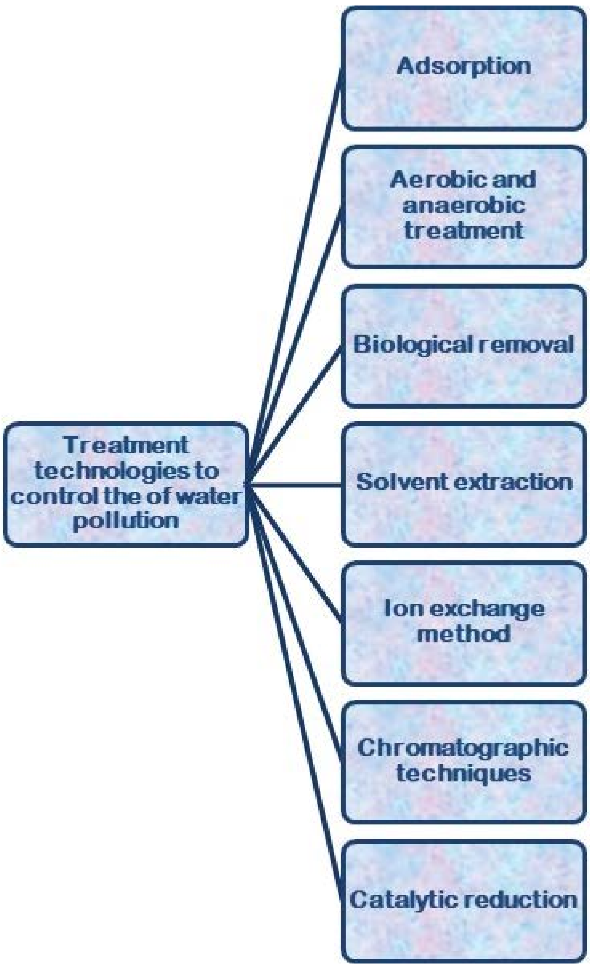

Fig. 1. Different technologies for removal of pollutants from aqueous medium.

current research due to high ion-exchange capacity, chemical stability, better regeneration, high thermal and radiation stability, rigid structure and negligible swelling characteristics [12,56]. Fig. 2 represents the main type of adsorbents for the removal of pollutants from an aqueous solution. In this review paper, different adsorbent materials such as composite-ion exchange materials, agricultural materials, activated carbon, and resins have been discussed.

3.2. Composite ion-exchange materials

Organic exchangers decompose at elevated temperature, and high ionization radiation and inorganic exchangers hydrolyze with aqueous solvents. Due to these certain limitations encountered with pure organic and inorganic exchangers, efforts have been made to overcome these barriers [104,105]. Large numbers of different types of composite cation-exchange and anion-exchange materials have been synthesized by incorporating organic polymer into the matrix of inorganic precipitates by a well-known sol-gel mixing method. These composite materials have well established their position. They have shown valuable applications for the adsorption and removal of metal ions and other pollutants from the contaminated water due to their characteristic properties like excellent adsorption capacity, better functionality, chemical, mechanical, radiation, and thermal stability and good selectivity [106]. Composite cation- exchange materials have the property of regeneration, by which these exchangers can be used for several cycles in an economic way [27,41,107,108].

AL-Othman et al. [21] synthesized polyaniline Sn(IV) molybdate (crystalline organic–inorganic composite cation exchanger) having good ion exchange capacity, high chemical and thermal stabilities. It was analyzed by applying Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis-differential thermal analysis (TGA-DTA), scanning electron microscopy (SEM) and X-ray analysis. 0.1 M nitric acid was used for the elution of metal ions as an eluting reagent, in the presence of which 90%–99% metal ions were adsorbed. The optimum temperature for the maximum adsorption of metal ions was 45°C. Based on distribution studied it was observed that adsorption of Pb^{2+} was highest among all metal ions and equilibrium was attained after 30 min at optimum pH (8.0). So, it is clear from the results that the quantitative and efficient separations of various metal ions are feasible on polyaniline Sn(IV) molybdate columns. This cation exchange material is a potential candidate for removing heavy metals from contaminated water to control pollution [21]. Similarly, Al-Othman et al. [109] studied the adsorption capacity of composite cation exchanger polypyrrole Th(IV) phosphate (prepared by sol–gel method) for the adsorption of trichloroacetic acid (TCA), from aqueous solutions. TCA, a very toxic water contaminant even at very low concentration, is present in a very toxic

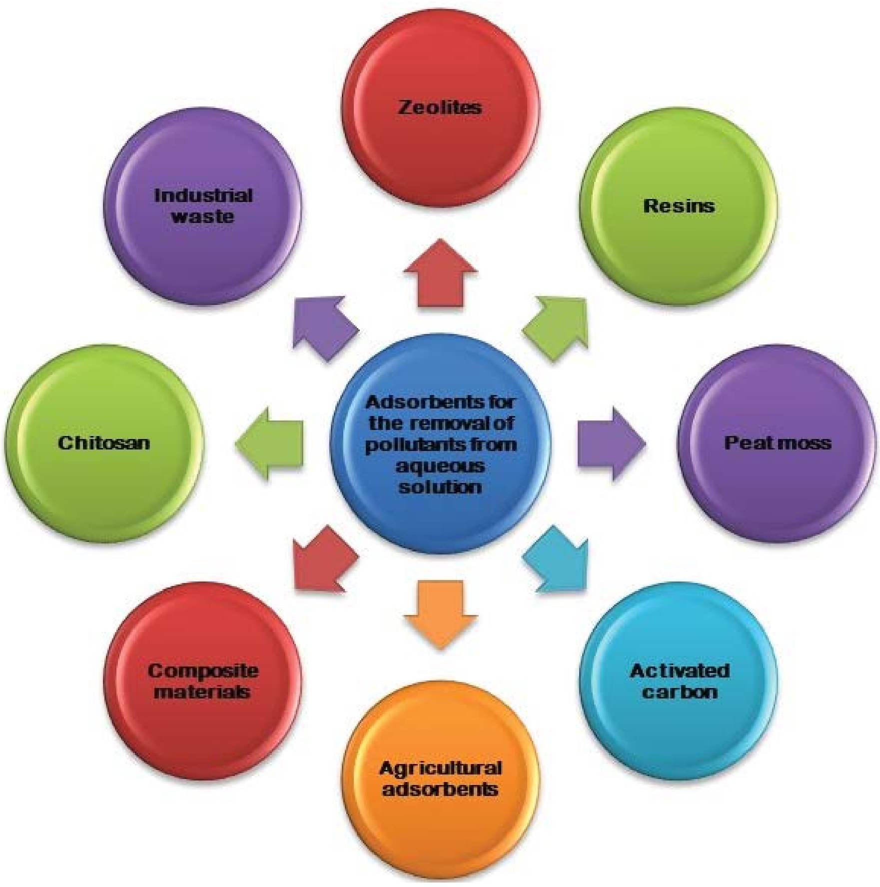

Fig. 2. The main type of adsorbents for the removal of pollutants from an aqueous solution.

water contaminant even at very low concentration, present in the aquatic environment. Investigations showed an ion exchange capacity of 1.56 meq dry g^{-1} and equilibrium was established within 72 min, and 98% adsorption was achieved at optimal conditions. So, it was concluded that this composite ion exchanger was very good for removing pollutants (TCA) from the aqueous solutions.

Naushad et al. [17] synthesized an organic–inorganic based composite cation exchanger polyaniline Sn(IV) silicate. The physicochemical properties of that cation exchanger were studies by applying different analytical techniques like X-ray diffraction (XRD), SEM, energy-dispersive X-ray spectroscopy (EDX), FTIR, elemental analysis and thermogravimetry analysis etc. Adsorption properties of this ion exchanger were investigated. The adsorption was very fast, and equilibrium was established within 40 min which showed that polyaniline Sn(IV) silicate is a very excellent ion-exchanger for the removal of metal ions and some binary separations showed that it has the highest adsorption capacity for Cd^{2+}. It was highly thermally and chemically stable. The feasibility of regeneration by diluted HNO_3 makes it perfect candidate for removing heavy metals from water to control water pollution. Similarly, sodium dodecyl sulfate acrylamide Zr(IV) selenite (SDS-AZS) was developed by sol–gel method, and this composite material was used for the removal of highly toxic lead ion (Pb^{+2}) from aqueous medium. The physicochemical properties of SDS-AZS were studied using EDX, TGA, transmission electron microscope (TEM), SEM, FTIR, and XRD. The results showed that SDS-AZS can be used in water pollution control [7]. Bushra et al. [30] synthesized an ion exchanger, Polyaniline Sn(IV) tungstomolybdate nanocomposite (PSTM) by sol-gel method for the efficient removal of Pb^{2+}. The properties (physico-chemical) of PSTM were characterized by using techniques like TEM, FTIR, XRD, and SEM. The results showed that 85% adsorption of Pb^{2+} was obtained at the optimal pH (6.0), and equilibrium was achieved after 50 min. From these results, it was concluded that PSTM cation exchanger is the best option and excellent ion-exchanger for the removal of lead ions from the aquatic system. AL-Othman et al. [21] synthesized crystalline composite cation exchanger polyaniline Sn(IV) molybdate to remove Pb^{2+}. This composite ion exchanger's practical usefulness was studied by carrying out the selective separation of Pb^{2+} on this newly synthesized material. It was observed that 99% adsorption was achieved within 30 min at pH 8.0. Bushra et al. [79] synthesized a composite material polyaniline-Zr(IV) phosphoborate (PZPB) via sol–gel method by combining Zr(IV) phosphoborate and polyaniline, which was selective for the removal of Hg^{2+} by adsorption method. It was characterized by various analytical techniques. X-ray diffractogram of this material showed that it was semi-crystalline in nature. Table 2 shows different types of composite materials and their applications to remove pollutants at optimum parameters.

3.3. Agricultural and food waste-derived adsorbents

Scientists and Investigators have discovered low-cost, eco-friendly, and economical agricultural adsorbents materials to remove pollutants from wastewater, which have shown potential sorption properties due to their composition, renewable nature, and availability. Agriculture wastes usually contain renewable organic matters, including plant materials like seeds, fruit components and pulp, food processing, and forestry by-products [67]. These materials usually contain high levels of cellulose, hemicellulose, lignin, proteins, lipids, water, sugars, hydrocarbons and starch [53,110]. These materials require little processing and are used in natural form or in modified form. In natural form, these materials are washed, crushed, ground, cut into small pieces, sieved, then washed with water and weak acid solutions, while modified form product is subjected to modification techniques [111].

Various researchers have demonstrated good adsorption potential in agriculture materials like *Acacia leucocephala* bark [112], apricot seeds, cotton stalks [113], avocado, melon and dragon fruit peels [114], Bael fruit (*Aegle marmelos correa*) shell [115], banana peels [116,117], coconut coir pith, [118], coffee grounds [119], mango peel [120], *Moringa oleifera* bark [121,122], orange peel [123], Peanut hull [124], peanut husk [125], pomegranate peel [126], red pepper [127], rice bran [128], sago [129], *Salsola vermiculata* [130], Swede rape straw (*Brassica napus* L.) [131], turmeric [67], walnut shell [132], wheat bran [133], soybean hulls, grapes stalks, sugar beet pulp, sunflower stalks, coffee beans [134], pine bark, cone and sugar cane, due to low cost, availability and eco-friendly nature and high efficiency [135,136].

Sha et al. [123] described copper and cadmium's adsorption behavior on acetic acid modified orange peel. The basic characteristics of this adsorbent were studied by infrared spectrophotometer. It was found that adsorption of Cu^{2+} and Cd^{2+} was dependent on pH, contact time, and initial metal ion concentration. Results showed that maximum adsorption capacities of Cu^{2+} and Cd^{2+} on the modified orange peel were 70.67 and 136.05 mg/g, respectively. Moreover, this material can be used up to five times due to regeneration ability. It was concluded from this study that the orange peels, after simple modification could be used as potential adsorbent for the removal of heavy metals.

Ajmal et al. [135] studied the ability of orange fruit peel to remove heavy metals like nickel from aqueous solution, and it was found that removal extent was dependent on pH, temperature, and initial concentration. The process was endothermic, and the maximum adsorption percentage was 96% at 50°C and pH 6. Moreover, the adsorbent can be regenerated and recycled thrice. From these results, it was concluded that peels of orange can be used as an effective adsorbent for the removal of nickel from wastewater.

Mallampati et al. [114] selected three natural, simple, and renewable materials such as avocado, melon and dragon fruit peels for water purification, and different analytical techniques were applied to characterize the morphologies of the peels. Results showed that the extraction efficiency of dragon fruit peels was highest for alcian blue dye (71.85 mg/g) and methylene blue dye (62.58 mg/g), and extraction capacity of melon peels and avocado peels was (7.89 and 9.82 mg/g) for Pb^{2+} and (9.45 and 4.93 mg/g) for Ni^{2+}. The results also revealed that these materials can be regenerated and reused for many cycles.

Table 2
Various types of composite materials and their applications for the removal of pollutants at optimum parameters

Composite ion	Type	Pollutants	Analytical technique	Temp.	Time for equilibrium	pH	Adsorption	Ref.
Polyaniline Sn(IV) molybdate	Crystalline organic–inorganic composite cation exchanger	Lead	FTIR, TGA-DTA, SEM and X-ray analysis	45°C	30 min	8.0	99%	[21]
Polypyrrole Th(IV) phosphate	Composite cation exchanger	TCA	–	–	72 min	–	98%	[109]
Sodium dodecyl sulfate acrylamide Zr(IV) selenite (SDS-AZS)	Nanocomposite cation exchanger	Lead	EDX, TGA, TEM, SEM, FTIR and XRD	55°C	40 min	6.0	90%	[7]
Polyaniline Sn(IV) molybdate	Organic–inorganic composite cation exchanger	Lead	FTIR, TGA-DTA, X-ray, SEM	200°C	–	2.8	98.5%	[21]
Polyaniline Sn(IV) tungstomolybdate nanocomposite (PSTM)	Ion exchanger	Lead	TEM, FTIR, XRD and SEM	50°C	50 min	6.0	85%	[30]
Polyaniline-Zr(IV) phosphoborate (PZPB)	Composite material	Mercury	X-ray diffractogram	37°C	180 min	6.0	86.3%	[79]
Polyaniline Sn(IV) silicate	Organic–inorganic based composite cation exchanger	Cadmium	XRD, SEM and TGA	45°C	40 min	9.0	92%	[17]
Polyaniline Th(IV) tungstomolybdophosphate (PANI/TWMP)	Nanocomposite ion exchanger	Copper	FTIR, XRD, SEM and TEM	400°C	–	–	96%	[14]
Fe_3O_4–TSC nanocomposite	Nanocomposite ion exchanger	Malachite green (MG) dye	FTIR	25°C	40 min	7.0	98.5%	[45]
Polypyrrole Th(IV) phosphate composite cation-exchanger	Composite cation-exchanger	Trichloroacetic acid herbicide	–	25°C	70 min	6.0	98%	[179]

Wang et al. [124] used peanut hull (an agricultural by-product abundant in China) as adsorbent for removing Cu(II) from aqueous solution. Adsorption extent was investigated as a function of contact time, pH, concentration, and temperature, and it was found that maximum removal was reached at pH 5.5, and equilibrium was obtained within 2 h. The adsorption capacity of Cu(II) was 21.25 mg/g at 30°C, so, from obtained data, it was concluded that peanut hull is an excellent element for the removal of heavy metals from wastewater.

Bhatnagar and Minocha [126] studied the feasibility of pomegranate peel waste for the removal of nickel from water, and it was found that adsorption was endothermic and maximum adsorption capacity of pomegranate peel for nickel removal was 52–72 mg/g with the variation of temperature, moreover, it was seen uptake rate of nickel was initially rapid, and 50% adsorption was obtained within 2 h while equilibrium was achieved within 7h, so, it was concluded from the present study that pomegranate peel waste could be used beneficially for the removal of nickel from wastewater.

Jain et al. [23] used the rice husk to remove the Rhodamine B (toxic and hazardous dye of xanthene class, used in textiles and foodstuffs as a colorant) from the wastewater. The results showed that 90% adsorption was obtained at 2.1 pH, and equilibrium was achieved within 2 h. Chemical oxygen demand (COD) measurements were carried out to observe the quality of water before and after the treatments and it was observed that COD value of water decreased from 1,608 to 276 mg/L after the treatment, so it was concluded that rice husk is a feasible and good adsorbent for the wastewater treatment.

Özcan et al. [127] investigated the adsorption of copper ions on *Capsicum annuum* (red pepper) seeds. The nature of adsorption and interaction of metal ions was examined by the application of different techniques with the variation in the parameters of temperature, initial concentration, time, adsorbent dose, and pH. The results showed that equilibrium was attained within 60 min at 50°C, and 11.53 mg/g copper was adsorbed on the red pepper seeds. These results concluded that red pepper can be further used as a copper adsorbent from wastewater.

Bhanger et al. [137] utilized the water melon peels for the removal of methyl parathion (pesticide) after their chemical and thermal treatment. For the characterization of water, melon peels, different analytical techniques like SEM and BET (Brunauer–Emmett–Teller) were carried out, and the effect of pH, shaking speed, contact time, adsorbent dose, initial concentration and temperature was measured by variation of these parameters. By using 0.1g of adsorbent at 6.0pH maximum adsorption was achieved within 60 min and this method was employed for the removal of methyl parathion from surface water with 99% ± 1% removal.

Rao and Rehman [138] studied the adsorption of Cr(VI) by using the fruits of *Ficus glomerata* in the batch system. They investigated the effect of the initial concentration of chromium, pH, temperature and time while surface morphology and active functional groups present on the adsorbent surface were investigated by using the SEM and FTIR. The applicability of the adsorbent was demonstrated by removing the Cr(VI) from electroplating wastewater, the percentage adsorption of Cr(VI) was 98% at 50°C and 2.0 pH while the adsorbent dose was ranging from 0.2 to 0.1 g/L, and equilibrium was obtained within 40 min. This study indicated that *Ficus glomerata* is the best adsorbent for the removal of Cr(VI) from aqueous solution.

Table 3 shows the main characteristics of agricultural-based adsorbents with their adsorption capacities for the removal of various pollutants from aqueous solutions.

3.4. Activated carbons (AC)

Among many adsorbent, activated carbon (Non-graphite class of carbons) has gained widespread importance due to its use for [56,89] the treatment of pollutants released from textile industries [139,140]. It can be defined as "solid carbon, which is thermally produced having porous structure [141], large internal surface area (500–3000 m^2/g, high mechanical strength, highly active [142] and suitable surface for the reactivity". Generally, it may be macro-porous or meso-porous and micro-porous used to remove large molecules and small molecules, respectively.

Activated carbon contains heteroatoms like hydrogen and oxygen and has many carbon-carbon incomplete bonds that create chemically active sites responsible for specific chemical and physical interactions [143]. It is one of the most common, versatile and effective media for removal, separation and purification of a wide range of pollutants released from industrial and municipal wastewaters [111] throughout the world due to having a high capacity for the adsorption of metal ions, phenols, dyes and other pollutants [141–145]. The activated carbon is produced from various raw materials, so, it exhibits different properties, moreover, agriculture and waste food materials can be turned into activated carbons by some pretreatments, which include carbonization. After carbonization, this carbonized biomass (charcoal) is activated by activation process (single or many steps). This activation process, either physical or physical/chemical must be at low temperature (400°C–600°C) or at high temperature (700°C–1,000°C) [67,141,145]. These pretreatments are done to enhance and reinforce the functional group potential and consequently increase the number of active sites. These raw materials can be modified into activated carbon for the development of desirable properties for adsorption like pore size, volume, surface area and functional groups. The modification of techniques can be classified into three main groups: (i) modification with biological compounds; (ii) modification with physical compounds and; (iii) modification with chemical compounds. Modification with chemical compounds has been more frequently used to increase the adsorption process.

Many cheap and easily available agricultural materials such as apple pulp [146], apricot stone [147], avocado kernels [148], carrot residue [149], bagasse pith [150], babbool wood [151], citrus fruit peel [152], coconut shell fibers [67], cocoa shells and siriguela seeds [153], corncob [154], date stems [130], 2-amino-5-guanidinopentanoic acid modified activated carbon (AGDPA@AC) [155], grape seed [156], guava seeds [157], jack fruit peel [158], jatropha husk [159], longan seed [160], lotus stalks [139], mango seed [161], olive fruit stone [162], orange peel [163], palm seed coat [164], pea shell [165], peanut shell [166], pomelo skin [167], Potato peel [168,169], pumpkin seed hull [170], rambutan (*Nephelium lappaceum*) peel [171], reedy grass leaves [139], rice husk, cotton [172], sago waste, maize cob, banana pith [173], soybean, cotton seed hulls, rice straw, sugarcane bagasse [174], sugar cane bagasse [130] and Sunflower seed oil [175] have been used as a source for the production of activated carbon for the removal of pollutants and heavy metals from the wastewater, due to their low cost and abundance (Fig. 3) [155].

Mohan et al. [67] prepared a low-cost activated carbon from coconut shell fibers and utilized it in the adsorption process for the decontamination of chromium Cr(III) from tanneries effulents/ wastewater. The maximum adsorption capacity was 12.2 mg/g at 25°C and 16.10 mg/g at 40°C. The adsorption of Cr(III) was almost 100% at pH 5.0, and equilibrium was established within 48 h. It was concluded that adsorption was rapid within the first hour of contact, and 40%–50% adsorption occurred within the first hour of contact.

Mohan et al. [67] prepared a low-cost activated carbon from the fibers of coconut shell (agriculture waste) and utilized it to treat trivalent chromium Cr(III) from wastewater. It was seen that the maximum adsorption capacity at 25°C was 12.2 mg/g, and at 40°C, it was 16.10 mg/g.

Jain et al. [23] used the activated carbon to remove the Rhodamine B (toxic and hazardous dye of xanthene class, used in textiles and foodstuffs as a colorant) from the wastewater. The results showed that 93.2% adsorption was obtained at 2.1 pH. COD measurements were also carried out to observe the quality of water before and after the treatments and it was observed that COD value of water was decreased from 1,608 to 267 mg/L after the treatment. It was concluded that AC is feasible and good adsorbent for the wastewater treatment.

Wang et al. [56] synthesized AC with the surface area of about 1,400 m^2/g from *Polygonum orientale* Linn. (PL) by phosphoric acid activation for the adsorption of Pb(II) ions from aqueous solutions. Morphological features and ability of this low cost activated carbon was investigated

Table 3
Main characteristics of agricultural-based adsorbents with their adsorption capacities for the removal of various pollutants from aqueous solutions

Agriculture adsorbents	Pollutants to be removed	Adsorbent dose	Temperature	Contact time	pH	Initial pollutant concentration range	Amount adsorbed (q_e)	Ref.
Rice husk	Phosphate	3 g/L	24.85°C	180 min	6.0–8.0	10–30 mg/L	68 mg/g	[180]
Coconut shells	Urea	8 g/L	30°C	200 min	–	4959 mg/L; 19,800 mg/L	60 mg/g; 256.41 mg/g	[181]
Branches of pomegranates trees	Anionic herbicide bentazon	1.5 g/L	30°C	8–10 h	–	200–250 mg/L	80 mg/g	[182]
Grapefruit peel	U(VI)	2 g/L	24.85°C	60–80 min	4.0–6.0	50–500 mg/L	140.79 mg/g	[183]
Lentil shell (raw)	Cu(II)	10 g/L	39.8°C	180 min	6.0	25–500 mg/L	9.59 mg/g	[184]
Mango peel waste	Cu(II)	5 g/L	25°C	60 min	5.0–6.0	10–500 mg/L	46.09 mg/g	[185]
Garlic peel	Pb(II)	–	25°C	60–120 min	4.0–7.0	1–200 mg/L	109.05 mg/g	[186]
Orange peel	Cu(II)	5 g/L	25°C	120 min	5.0–5.5	10–300 mg/L	40.37 mg/g	[187]
Melon peel	Pb(II)	5 g/L	25°C	120 min	4.5	1–5 mmol/L	167.8 mg/g	[188]
Grapefruit peel	Pb(II)	10 g/L	30°C	90 min	5.3–6.5	100 mg/L	12.73 mg/g	[189]
Cucumis sativus peel	Cd(II)	1 g/L	24.85°C	60 min	5.0	5–150 mg/L	58.14 mg/g	[190]
Sunflower hull	Cu(II)	2 g/L	30°C	180 min	5.0	25–200 mg/L	57.14 mg/g	[191]
Sugarcane bagasse	Hg(I)	5 g/L	30°C	60 min	4.0	76 mg/L	35.71 mg/g	[192]
Pomegranate peel	Ni(II)	10 g/L	25°C	120 min	5.5–6.5	–	52 mg/g	[193]
Mosambi (*Citrus limetta*) peel	Cr(VI)	5–50 g/L	40°C	120 min	2.0	200–300 mg/L	250 mg/g	[194]
Rice straw	Cd(II)	–	25°C	180 min	2.0–6.0	25–350 mg/mL	13.84 mg/g	[195]
Cashew nut shell (raw)	Ni(II)	3 g/L	30°C	30 min	5.0	10–50 mg/L	18.86 mg/g	[196]
Potato peels	Cu(II)	10 g/L	29.85°C	20 min	6.0	150–400 mg/L	0.3877 mg/g	[197]
Olive stone	Cu(II)	–	20°C	60 min	5.5–6.0	3.0×10^{-4} to 0.15 mol/L	3.19×10^{-5} mol/g	[198]
Garden grass	Pb(II)	0.5 g/L	29.85°C	–	–	1–500 mg/L	58.34 mg/g	[199]
Gourd peel powder	Cr(VI)	6 g/L	28°C	40–60 min	1.0	75–350 mg/L	18.7 mg/g	[200]
Pomelo peel	Cu(II)	5 g/L	25°C	60 min	4.0	25–125 mg/L	19.7 mg/g	[201]
Barley straw	Cu(II)	1 g/L	25°C	120 min	6.0–7.0	0.0001–0.001 mol/L	4.64 mg/g	[202]
Egyptian mandarin peel (raw)	Hg(II)	5 g/L	19.85°C	24h	6.2	50–200 mg/L	19.01 mg/g	[203]
Banana peel	Cd(II)	30 g/L	25°C	20 min	3.0	30–80 mg/L	5.71 mg/g	[204]
Pear millet husk carbon	Methylene blue dye	100 mg/L	32°C	–	6.0	10–60 mg/L	82.37 mg/g	[205]
Mango seed powder	Methylene blue	6.67 mg/L	30°C	140 min	8.0	20–100 mg/dm^3	142.86 mg/g	[161]
Ficus glomerata	Cr(VI)	0.2–1.0 g/L	50°C	40 min	2.0	10–100 mg/L	98%	[138]
Capsicum annuum (red pepper) seeds	Copper(II)	–	50°C	60 min	5.0	1.2 g/dm^3 2.0 g/dm^3	5.55–11.53 mg/ g	[127]
Watermelon peels	Methyl parathion pesticide	–	30°C	60min	6.0	(0.38–3.80) × 10^{-3} mol/dm^3	99%	[137]
Carica papaya seeds	Methylene blue dye	0.05 mg/L	30°C	120 min	7.0	10–400 mg/L	99%	[206]

by different techniques like XRD, BET, FTIR and SEM analysis. About 69.5% Pb(II) was taken up by *Polygonum orientale* Linn activated carbon (PLAC) and equilibrium was established after 30 min. It was concluded that carboxyl groups play an important role in Pb(II) adsorption and adsorption of Pb(II) on PLAC was strongly influenced by ionic strength and pH. It was also studied that PLAC has the ability to regenerate after desorption with HCl. Therefore, it has great potential to be used as an economically efficient adsorbent.

Naushad et al. [149] used a bio-adsorbent carrot residue (CR) (*Daucus carota* L.) for the production of AC by using HNO_3 as a chemical activator for the removal of BrO_3^-. Several analytical techniques were applied for the characterization of this chemically AC. Ultra-performance liquid chromatography mass spectrometry was used to assess the concentration of BrO_3^-. Equilibrium was achieved within 40 min and 93.7% BrO_3^- was adsorbed at the most suitable pH (3.5) and temperature. It was concluded that AC, which was derived from carrot residue is excellent and feasible candidate for the removal of BrO_3^-, which is a very toxic water pollutant.

Hai et al. [176] developed the date-based bio-char through the pyrolysis technique. The developed bio-char was modified into porous activated carbon using KOH. Finally, AC based electrodes were fabricated as high performed CDI electrodes for NaCl removal from saline solution as clearly shown in Fig. 4.

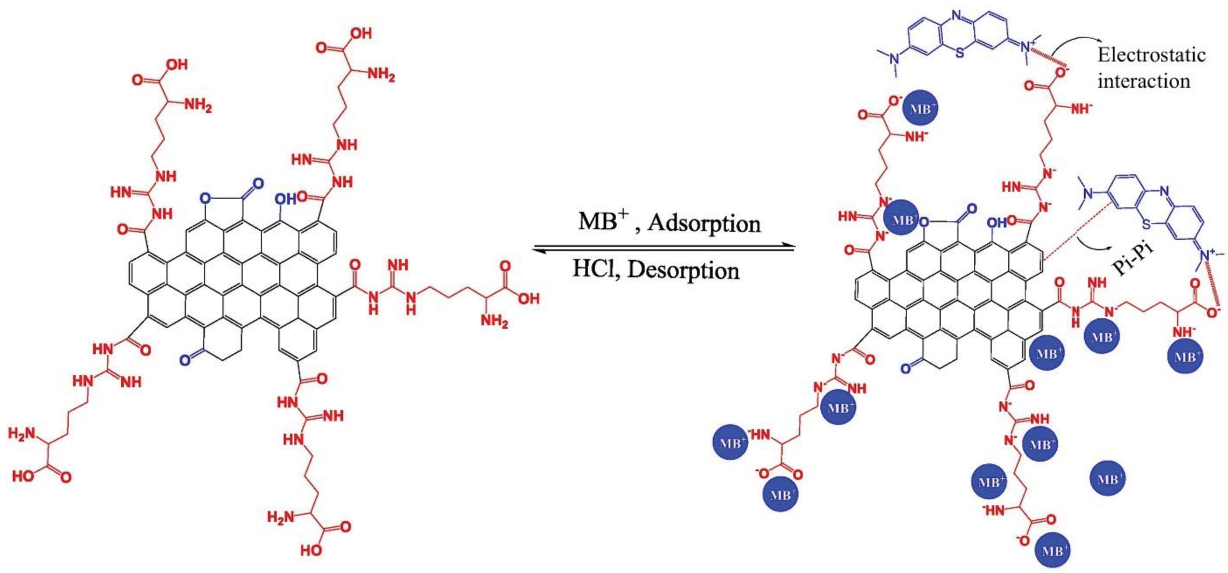

Fig. 3. Adsorption/desorption mechanism for the MB dye onto AGDPA@AC composite adapted with permission from the study of Naushad et al. [155].

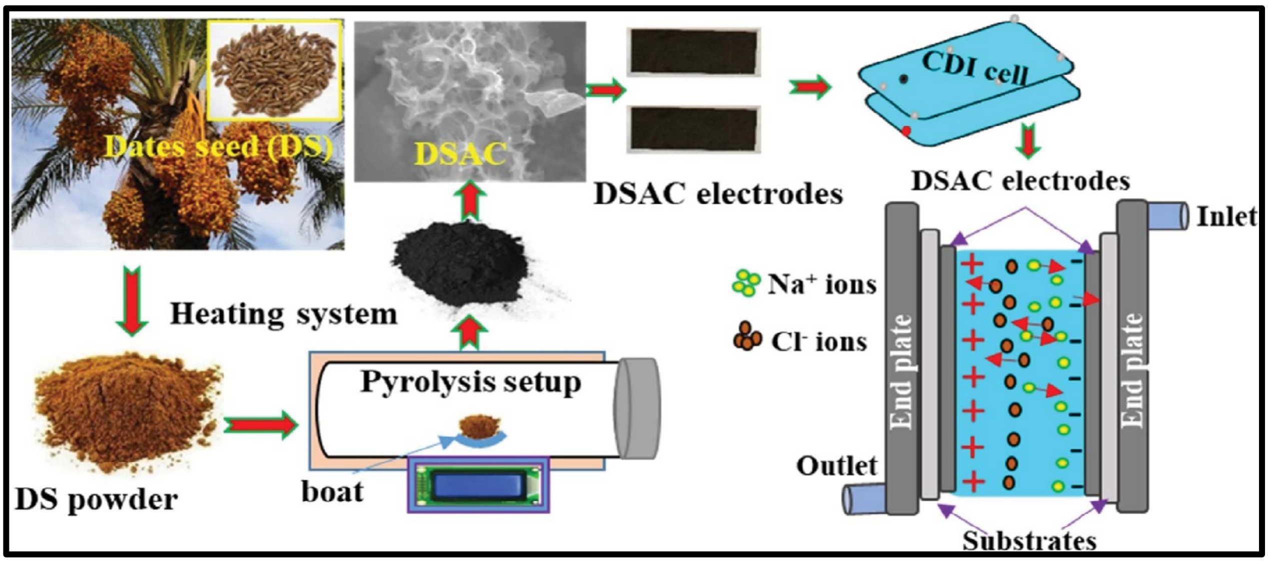

Fig. 4. Preparation of date seed-derived highly mesoporous activated carbon for high electrochemical performances of capacitive deionization electrodes for NaCl removal. Adapted with permission from the study of Hai et al. [176].

Rambabu et al. [177] developed the watermelon-based activated carbon (WMAC) and its $MnFe_2O_4$ composite (WMAC/$MnFe_2O_4$) for NaCl desalination via single-step low-temperature pyrolysis by H_3PO_4 activation. NaCl removal studies using WMAC/$MnFe_2O_4$ showed an electrosorption capacity of 29 mg/g for the nanocomposite, with rapid desalination and good recyclability. Fig. 5 shows the preparation of WMAC/$MnFe_2O_4$ composite material and SEM images for WMAC and WMAC/$MnFe_2O_4$ composite.

Table 4 shows the main characteristics of activated carbon adsorbents as well as their adsorption capacities for the removal of various pollutants from aqueous solutions.

4. Advantages, disadvantages and future prospects

The presence of different pollutants and heavy metal ions in water at higher concentrations is a severe matter. These pollutants induce adverse health effects and environmental problems; therefore, there is a requirement for detailed studies on the presence of these pollutants in water, their health effects, and their treatment by using different adsorbents. This review paper discusses various health problems associated with these pollutants and other adsorption materials for the removal of various pollutants from aqueous medium. Generally, adsorption is one of the best water treatment technologies suitable for the adsorption of various pollutants from waste and natural water sources. Different adsorbent materials cited in this paper are very effective materials for the treatment of pollutants. Cited composite materials, agriculture materials, and activated carbon have been discussed due to their specific characteristics like low cost, abundant availability, fast and high removal efficacy.

5. Conclusion

Nowadays, the removal of different pollutants and heavy metal ions from water is essential due to their adverse effects on the environment and living species. In this review article, a detailed discussion on the adverse effects of the different pollutants and heavy metals and their toxicity has been presented. This review article also shows the removal of different pollutants from water. It was concluded that composite ion exchange materials,

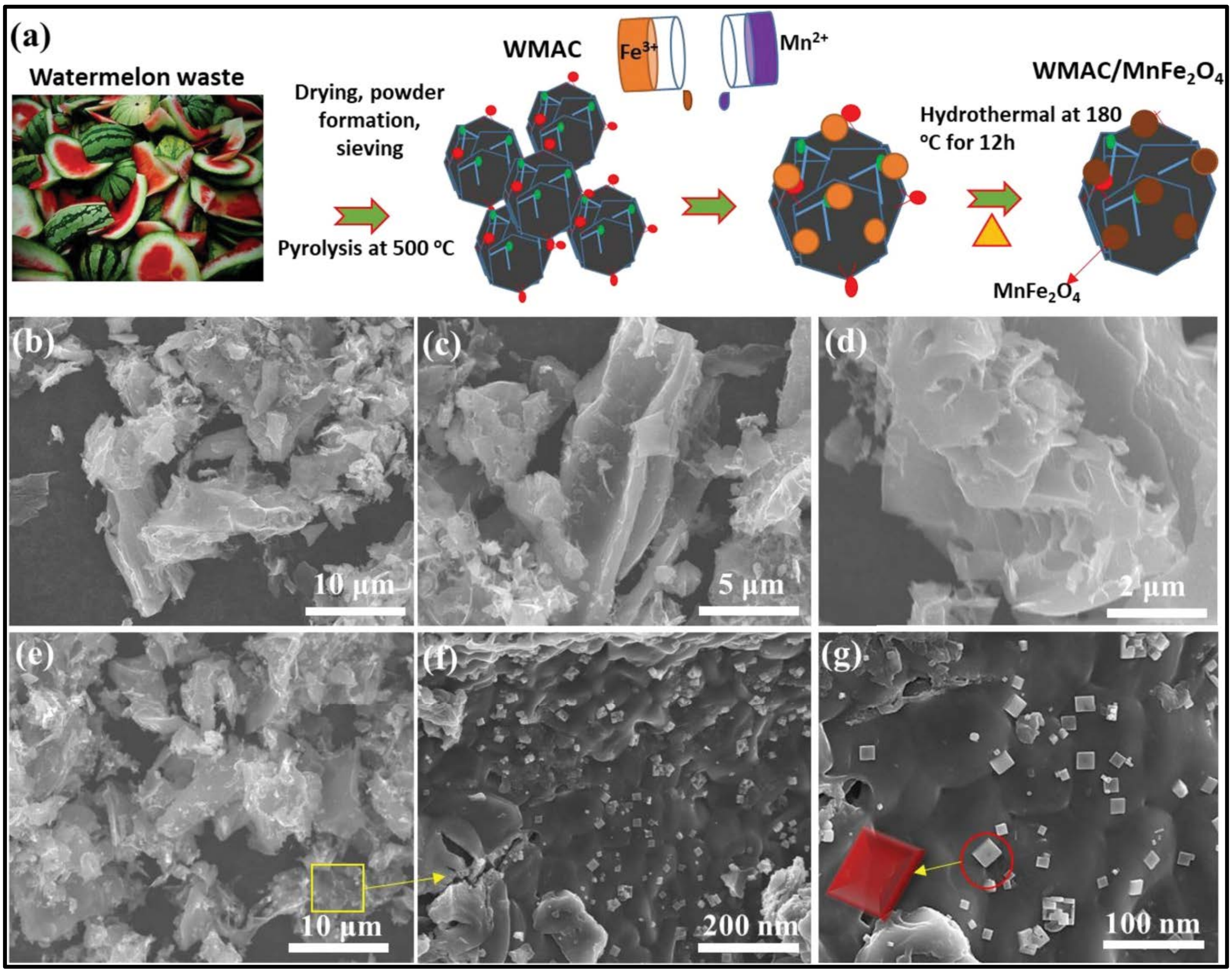

Fig. 5. (a) Scheme for the development of WMAC/$MnFe_2O_4$, (b–d) FE-SEM images of WMAC, (e–g) FE-SEM images of WMAC/$MnFe_2O_4$ nanocomposite. Adapted with permission from the study of Rambabu et al. [177].

Table 4
Main characteristics of activated carbon adsorbents as well as their adsorption capacities for the removal of various pollutants from aqueous solutions

Source of activated carbon	Pollutants to be removed	Adsorbent dose	pH	Contact time	Temperature	Initial pollutant concentration range	Amount adsorbed (q_e)	Ref.
Silk cotton hull	Methylene blue, Mercury(II) Nickel(II)	5.0 g/L	4.0–5.0	–	31°C	25 mg/dm^3	4.7 mg/g	[173]
Cassava peels	Cu(II)	10 g/L	8.0	20–120 min	39.85°C	4 mg/L	8.0 mg/g	[207]
Date seed	Anionic herbicide bentazon	0.8 g/L	5.5	24 h	30°C	25–250 mg/L	86.26 mg/g	[208]
Rice husk	Phosphate	3.0 g/L	6.0–8.0	180 min	24.85°C	10–30 mg/L	97.0%	[180]
Coconut tree	Mercury(II) Nickel(II) methylene blue	5 g/dm3	4.0–6.0	–	31°C	25 mg/dm^3	4.70 mg/g	[173]
Pomegranates trees	Anionic herbicide bentazon	1.5 g/L	–	8–10 h	30°C	200–250 mg/L	80.0 mg/g	[182]
Sago waste	Methylene blue, Mercury(II) Nickel(II)	5.0 g/L	3.0–4.0	–	31°C	25 mg/dm^3	4.51 mg/g	[173]
Fruit juice residue	Phosphate	3.0 g/L	6.0–8.0	180 min	24.85°C	10–30 mg/L	99.9%	[180]
Apricot waste	Methylene blue	2.0 g/L	–	–	50°C	100–400 mg/dm^3	136.98 mg/g	[209]
Almond shell	Methylene blue	–	–	24 h	–	–	1.33 mg/g	[210]
Maize cob	Methylene blue, Rhodamine, Mercury(II) Nickel(II)	5.0g/L	4.0	–	31°C	25 mg/dm^3	5.0 mg/g	[173]
Banana stalks	Anionic herbicide bentazon	1.2 g/L	–	1 d	30°C	250 mg/L	100.95 mg/g	[211]
Sugar cane bagasse	Acid blue 80	0.01 g/L	7.4	21 h	–	20–1,050 mg/dm^3	391 mg/g	[212]
Date pits	Methylene blue	5.0 g/L	–	–	25°C	20–400 mg/dm^3	123.1 mg/g	[213]
Sky fruit husk	Anionic herbicide bentazon	0.8 g/L	5.0	20 h	30°C	250 mg/L	131.11 mg/g	[214]
Longan seed	Cr(VI)	0.5 g/L	3.0	1.5 h		50–500 mg/L	28.74 mg/g	[160]
Euphorbia rigida	Disperse orange dye	2 g/L	7.0	–	20°C	–	118.93 mg/g	[215]
Banana pith	Methylene blue, Mercury(II) Nickel(II), Rhodamine	5.0 g/L	4.0	–	31°C	25 mg/dm^3	4.67 mg/g	[173]
Palm seed coat	Phenol	2 g/L	4.0–9.0	3 h	-	10–60 mg/L	96%	[164]
Coconut	Methylene blue	0.25 g/L	8.0	–	25°C	1.00 × 10–5 mg/dm^3	15.25 mg/g	[216]
Rambutan (*Nephelium lappaceum*) peel	Acid yellow dye	–	6.8	4 h	30°C	50–400 mg/L	218 mg/g	[171]
Jute fiber	Methylene blue	0.4–4.0 g/L	5.0–10.0	250 min	28°C	50–200 mg/dm^3	225.64 mg/g	[217]
Corncob	Acid blue dye	1 g/L	6.1	–	30°C	–	1,060 mg/g	[218]
Coconut shell fibers	Cr(III)	2 g/L	5.0	48 h	25°C	25–100 mg/L	12.2 mg/g	[67]
Palm kernel shell	Methylene blue dye	1g/L	7.0	40 min	28°C	100–700 mg/dm^3	311.72 mg/g	[219]

(Continued)

Cotton seed shell	Acid red blue	1 g/L	3.0	240 min	–	100 mg/L	153.85 mg/g	[220]
Apricot shell	Basic blue dye	–	–	24 h	–	–	4.11 mg/g	[210]
Avocado kernel seeds	Phenol	0.1 g/L	4.0–8.5	60 min	25°C	100–600 mg/dm^3	17.17 mg/g	[148]
Tapioca peel	Magenta MB cold brand reactive dye	1 g/L	7.0	120 min	–	–	7%	[221]
Euphorbia rigida	Methylene blue dye	–	6.0	60 min	40°C	200 mg/dm^3	114.45 mg/g	[222]
Siris seed pods	Nitroimidazole anti-biotic, metronidazole (MNZ)	0.5 g/L	7.0	30 min	500°C	100 mg/L	169.38 mg/g	[223]
Grape seeds	Diuron	–	–	8 h	25°C	65.7 μmol/L	61.2 μmol/L	[224]

activated carbon, and agricultural materials are very effective adsorbent materials for different pollutants from water due to their availability, low-cost and high removal efficacy. The novel literature on water treatment applications of these adsorbents has been thoroughly discussed. The main characteristic and adsorption capacities of these adsorbents have also been discussed. Therefore, the present study in the form of a literature review would be beneficial for further research in the field of water treatment.

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