Recovery, regeneration and sustainable management of spent adsorbents from wastewater treatment streams: A review

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Recovery, regeneration and sustainable management of spent adsorbents from wastewater treatment streams: A review

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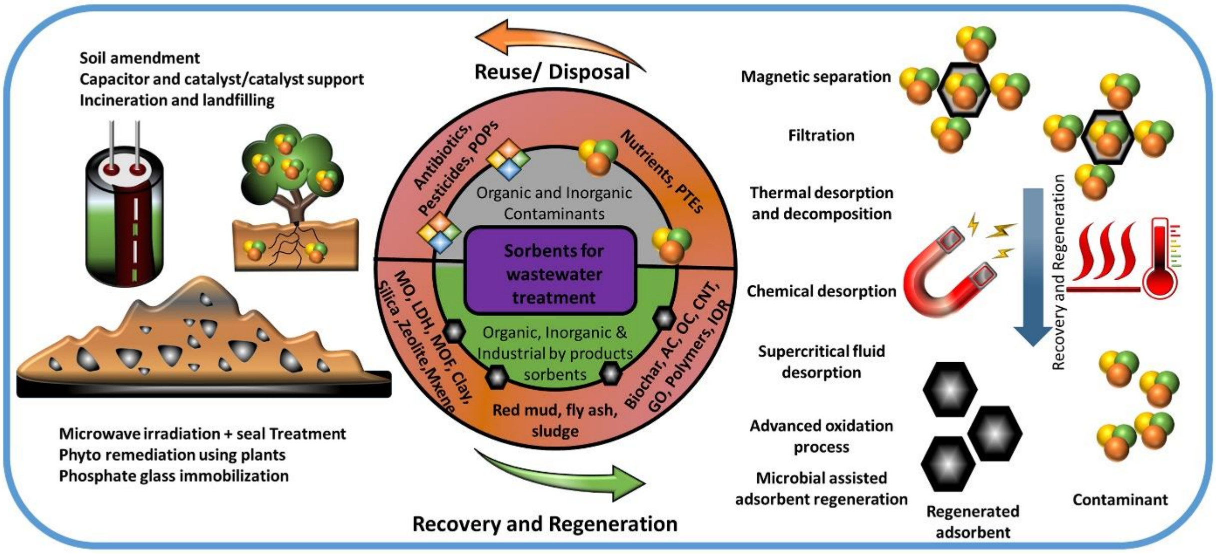
Graphical abstract



Abstract

Adsorption is the most widely adopted, effective, and reliable treatment process for the removal of inorganic and organic contaminants from wastewater. One of the major issues with the adsorption-treatment process for the removal of contaminants from wastewater streams is the recovery and sustainable management of spent adsorbents. This review focuses on the effectiveness of emerging adsorbents and how the spent adsorbents could be recovered, regenerated, and further managed through reuse or safe disposal. The critical analysis of both conventional and emerging adsorbents on organic and inorganic contaminants in wastewater systems are evaluated. The various recovery and regeneration techniques of spent adsorbents including magnetic separation, filtration, thermal desorption and decomposition, chemical desorption, supercritical fluid desorption, advanced oxidation process and microbial assisted adsorbent regeneration are discussed in detail. The current challenges for the recovery and regeneration of adsorbents and the methodologies used for solving those problems are covered. The spent adsorbents are managed through regeneration for reuse (such as soil amendment, capacitor, catalyst/catalyst support) or safe disposal involving incineration and landfilling. Sustainable management of spent adsorbents, including processes involved in the recovery and regeneration of adsorbents for reuse, is examined in the context of resource recovery and circular economy. Finally, the review ends with the current drawbacks in the recovery and management of the spent adsorbents and the future directions for the economic and environmental feasibility of the system for industrial-scale application.

Keywords: Adsorbents, Wastewater treatment, Recovery and regeneration, Disposal, Reuse

Abbreviations	Full form
4-CBA	4-carboxybenzaldehyde
AAP	Acetaminophen
AC	Activated carbon
AC-NCS	Activated carbon loaded with Ni-Co-S nanoparticles
AC-$ZnCl_2$	Activated carbon with $ZnCl_2$ activation
AMD	Acid mine drainage
AO	Acid orange
AOPs	Advanced oxidation processes
AOX	Adsorbable organic halogens
BA	Benzoic acid
BC	Black carbon
BMDCs	Bio-metalorganic framework -derived carbons
BPA	Bisphenol A
BTP-FA	Fly ash from biothermal power plant
CBZ	Carbamazepine
CFHC	Carboxylate-functionalized hydrochar
CNTs	Carbon nanotubes
CMK	Carbon Mesostructured by KAIST
CS-GTU	Chitosan-based adsorbent from guanylthiourea
CTP-FA	Fly ash from coal thermal power plant
CV	Crystal violet
CVD	Chemical vapor deposition
DEHP	Di-ethylhexylphthalate
DES	Deep EutecticSolvents
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
DQ	Diquat dibromide
DS	Diclofenac sodium
EDCs	Endocrine disturbing chemicals

EDTA	Ethylenediaminetetraacetic acid
GAC	Granular activated carbon
GIC	Graphite Intercalation Compound
GO	Graphene oxide
HDTMA	Hexadecyltrimethylammonium
HTC	Hydrothermal carbonization
HM	Heavy-metal
HMB900	Hierarchically microporous biochar
IBU	Ibuprofen
MAF	Metal azolate framework
MB	Methylene blue
MCM	Mobil Composition of Matter
MIL	Matériaux de l'Institut Lavoisier
MO	Methyl orange
MOFs	Metal organic frameworks
MOF-MA	Mercaptosuccinic anchored metal organic framework,
MP	Methyl paraben
MPHAC	Magnetized activated carbon pomegranate husk
MTBE	Methyl tert-butyl ether
MWCNTs	Multi walled carbon nano tubes
OCFGs	Oxygen containing functional groups
OM	Organic matter
p-Tol	p-toluic acid
PA	phthalic acid
PAHs	Polycyclic aromatic hydrocarbons
PAMAM	Poly(amidoamine)
PANI	Polyaniline
PCDDs	Polychlorinated dibenzo dioxins
PCDFs	Polychlorinated dibenzo furans
PDDA	Polydiallyldimethylammonium chloride
PEDOT	Poly(3,4-ethylenedioxythiophene)

PFAS	Polyfluoroalkyl substances
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PH	Peanut-husk
POPs	Persistent organic pollutants
PPCPs	Pharmaceutical and personal care products
PPY	Polypyrrole
PS	Persulfate
PTEs	Potentially toxic element
rGO	Reduced graphene oxide
SAS	Steam activated sawdust
SBA	Santa Barbara Amorphous
SCF	Supercritical fluid
SMX	Sulfamethoxazole
StAM-Arg	Corn starch modified with polyacrylamide and arginine
TA	Terephthalic acid
TC	Tetracycline
TCS	Triclosan
TRPO/SiO_2-P	Silica-polymer based adsorbent
WWT	Wastewater treatment
ZFA	Zeolite from fly ash

1. Introduction

Water is considered as one of the most precious natural resources in the 21st century for supporting human civilisation on earth. The increase in global human population that is happening simultaneously with rapid agricultural, industrial, and urban expansion has created a long-lasting imbalance between the availability of usable water and its demand at numerous places around the world (Boretti and Rosa, 2019, Singh, 2021). The impact of climate change, which is causing uncertainty in global and regional rainfall patterns, has been increasingly realised as a cause for concern in recent decades, and it has aggravated the demand-supply imbalance of freshwater. The need for freshwater has resulted in an overuse of groundwater causing secondary environmental degradation issues, such as mass scale contamination of water with potentially toxic elements (PTEs) due to chemical alterations of underground rocks and minerals [e.g., arsenic (As) contamination in the Indo-Gangetic plains] (Sarkar et al., 2021, Bolan et al., 2014). Furthermore, degradation of water resources has occurred due to point-source and diffuse-source pollution from industrial and agricultural activities (Alygizakis et al., 2020, Hube & Wu, 2021, Shi et al., 2021). These issues call for reclamation and recycling of wastewaters generated due to human activities, wherever possible, and also for the need for environmental sustainability. The use of reclaimed or treated wastewaters should be carried out in the agricultural, industrial, and public-use sectors. Treatments must be done according to the type of wastewater concerned and its degree of pollution.

Pressure is being placed on stakeholders to consider eco-sustainable water supplies for agricultural irrigation, because agriculture is one of the key sectors that contributes to the demand-supply imbalance of water resources (Dery et al., 2019). As an alternative source for irrigation water, wastewaters, which are derived from various sources, including domestic sewage (municipal wastewater), agricultural and industrial effluents, and stormwater, have been increasingly used following treatment (Jaramillo & Restrepo, 2017, Poustie et al., 2020, Singh, 2021). Wastewater irrigation of agricultural land has benefits, such as supplying essential nutrients to plants (Chojnacka et al., 2020, Perulli et al., 2019) and recharging groundwater (El Sheikh & Hamdan, 2020). There are, however, some detrimental effects, such as build-up of salts (Chaganti et al., 2020, Mukhopadhyay et al., 2020), pesticides (Westlund & Yargeau, 2017), PTEs such as arsenic (As), cadmium (Cd), lead (Pb) and mercury (Hg) (Shaheen et al., 2017), and persistent organic pollutants (POPs) such as per- and polyfluoroalkyl substances (PFAS) and polycyclic aromatic hydrocarbons (PAHs) (Bolan et al., 2021a, Kah et al., 2020, Lenka et al., 2021, Sun et al., 2018). In agricultural lands irrigated with wastewater, mobilization and transport of these contaminants into groundwater have been noted, as well as their enhanced bioavailability to soil biota and higher plants (Ali & Khan, 2019, Ofori et al., 2020). For example, dissolved organic matter (DOM) present in wastewater has been shown to facilitate the transport and mobility of both PTEs and POPS (Kunhikrishnan et al., 2017, Peña et al., 2020).

Large volumes of wastewater are produced from mining activities and industrial operations. For example, in mining operations, when mineral ores and tailings containing sulphide minerals are exposed to air and water, they get oxidised, thereby releasing sulphuric acid. The acid is leached out of mine site by rainwater or surface drainage and deposited into streams and groundwater, thereby generating acid mine drainage (AMD). AMD is one of the major global environmental issues that severely degrades water quality, kills aquatic life, and makes water unusable, not only due to the high acidity of the water but also because of the enormous load of PTEs leaching out from the rocks (Gurung et al., 2019).

Agricultural, manufacturing, and processing industries also generate large volumes of wastewater streams. Wastewater streams from agricultural industries include dairy and piggery farm effluents and abattoir effluents, which often contain high loadings of the following: nutrient elements that can potentially cause eutrophication, PTEs (e.g., Zn and Cu that are intentionally added to animal feed), various agrochemicals (e.g., pesticides), and veterinary pharmaceutical residues (e.g., antibiotics, hormones) (Hilares et al., 2021, Varma et al., 2021). Wastewater streams from manufacturing and processing industries include paper and pulp effluent (persistent organic pollutants, chemical solvents, PTEs) (Singh & Chandra, 2019), metal processing wastewater (PTEs) (Shrestha et al., 2021), tannery effluent (PTEs, specially Cr) (Lofrano et al., 2013), textile and dye wastewater (various colourants) (Kishor et al., 2021), petroleum refinery and petrochemical plant wastewater (various hydrocarbon contaminants) (Jain et al., 2020), and pharmaceutical wastewater (antibiotics, hormones, drug residues) (Khasawneh & Palaniandy, 2021). Additionally, municipal wastewater coming from households may contain a wide range of organic and inorganic contaminants originating from activities of everyday life and use of various essential commodities and products. Hence, the removal of wastewater-borne contaminants before reusing wastewater streams for any purpose is necessary.

Amongst various technologies deployed to remove contaminants during wastewater treatment, adsorption is considered to be an effective and eco-friendly approach (Loganathan et al., 2014., Burakov et al., 2018., Crini et al., 2019). Both inorganic and organic adsorbents have been found to be effective in the capture and removal of these contaminants (Mo et al., 2018, Pandey, 2017, Rasheed et al., 2020). Recently, there have been increasing efforts in designing engineered adsorbents with enhanced adsorption capacity and specific removal of contaminants (Dutt et al., 2020, Vithanage et al., 2017). One of the major practical challenges in the context of resource recovery is the sustainable management of spent adsorbents (Hossain et al., 2020). Various technologies, including sedimentation, filtration, centrifugation, and magnetic separation techniques, are used to separate and recover spent adsorbents during wastewater treatment (Hassan et al., 2020b, Vakili et al., 2019). The spent adsorbents are subsequently either regenerated for reuse or safely disposed through incineration and landfilling (Kozyatnyk et al., 2020). A number of techniques involving desorption, photodegradation, and biodegradation of sorbed contaminants have been examined to regenerate and reuse the spent adsorbents (Lata et al., 2015, Vakili et al., 2019).

Several reviews have demonstrated the potential value of a wide range of adsorbents used in wastewater treatment (De Gisi et al., 2016, Kah et al., 2020, Mehta et al., 2015). In addition, the regeneration of spent adsorbents loaded with contaminants using specific methods for their reuse has also been mentioned (Hassan et al., 2020b, Lata et al., 2015). Despite these reviews, a comprehensive understanding of sustainable management of spent adsorbents loaded with contaminants is still lacking. The present review aims to provide a critical analysis of the following: (i) the effectiveness of emerging adsorbents in removing contaminants from wastewater streams; and (ii) sustainable management of spent adsorbents involving the regeneration for reuse. This study will help the readers to understand the current regeneration techniques and how to regenerate the adsorbent without much loss in adsorption capacity even after many regeneration cycles. At the same time, using regeneration, the secondary pollution is minimised whereas the reuse in other applications results in cost-effectiveness and resource recovery.

2. Sources of contaminants in wastewater streams

The composition and concentrations of nutrients and other pollutants in wastewater are mainly dependent on the sources and installations where the water is drawn (Eriksson et al., 2002). Contaminants in the wastewater streams include inorganic chemicals, such as nutrients (nitrate and phosphate), PTEs (As, Cd, Pb, and Hg), and organic contaminants, such as persistent organic pollutants (POPs) (pesticides, PAH, and PFAS) (Müller et al., 2007) (Table 1). An in-depth understanding of wastewater stream characteristics is necessary so that suitable technologies can be developed and deployed for wastewater treatment.

2.1 Inorganic contaminants

Inorganic contaminants in wastewater include mostly nutrients and PTEs. The major nutrient elements that can lead to contamination of waterways include N and P (Cai et al., 2013, Ye et al., 2017). They are often present in high concentrations in domestic wastewater and most farm effluents (e.g., dairy, piggery). For example, N and P enter the stormwater system predominantly through soil organic matter (OM), inorganic and organic fertilizers, kitchen wastes (including detergents), animal faeces, poorly maintained sewage infrastructure, and gaseous N (nitric and nitrous oxides) produced from vehicle exhausts and ash from bushfires (Powley et al., 2016, Taylor et al., 2005). These nutrients are primarily derived from mineral fertilizer and manure applications to managed farmlands as a nutrient source (Zak et al., 2018). Loss of these nutrients through leaching, erosion, and gaseous emissions contributes to nitrate toxicity in potable water, eutrophication of waterways, and greenhouse gas emissions (Dalu et al., 2019). Elevated nutrient levels in waterways can encourage algal blooms, causing hypoxia and biodiversity loss in aquatic environments (Padedda et al., 2017).

Although pedogenic processes can release PTEs into aquatic environments, anthropogenic activities are considered the primary entry of PTEs in wastewater (Shaheen et al., 2019). Industrial manufacturing, mining activities, and the disposal of domestic and industrial wastes (both liquid and solid) are the major sources of PTE enrichment in aquatic environments (Vareda et al., 2019). While sewage effluents derived from domestic wastewater treatment plants are enriched with biologically essential PTEs such as copper (Cu), zinc (Zn), and iron (Fe), most of the industrial effluents are enriched with biologically non-essential PTEs such as Cd and Hg (Atamaleki et al., 2019, Attari et al., 2017, Muhammad et al., 2021). Stormwater is often found to be enriched with PTEs, such as Cr, Zn, and nickel (Ni) derived from wear of vehicle tire and brake pads and corrosion of roofing and building materials (Behbahani et al., 2020).

2.2 Organic contaminants

The major organic contaminants in wastewater streams include endocrine disturbing chemicals (EDCs) (e.g., antibiotics, pesticides) and POPs (e.g., PAHs, PFAS) (Trojanowicz, 2020, Zhang et al., 2020f). Endocrine disrupting chemicals comprise a variety of substances that adversely affect hormonal and other regulatory systems of animals and humans causes a range of human disorders such as prostate cancer and changes in thyroid and cardiovascular endocrinology (Diamanti-Kandarakis et al., 2009). Of these micropollutants, polychlorinated dibenzo dioxins (PCDDs) and furans (PCDFs), adsorbable organic halogens (AOX), and di-ethylhexylphthalate (DEHP) are frequently reported in wastewater streams (Hwang et al., 2012, Zolfaghari et al., 2014). These compounds enter sewerage systems through various sources, such as domestic sewage discharges, stormwater and agricultural runoff, livestock wastes, and industrial effluents (Xu et al., 2020).

PAHs, which originate from both natural (e.g., volcanos, bush fires) and anthropogenic (e.g., burning coal, petroleum refineries, motor vehicle exhaust) sources, are also present in significant amounts in wastewater. These compounds are of ecological and health concerns owing to their carcinogenic, teratogenic, and mutagenic characteristics (Hu et al., 2014). PAHs mainly enter the wastewater systems from stormwater runoff, domestic discharges, and industrial waste effluents (Gaurav et al., 2020, Huang et al., 2020b).

PFAS are one of the emergent contaminants reaching wastewater treatment plants (Bolan et al., 2021b). PFAS are a group of manufactured, fluorinated organic chemicals that contain one or more C atoms on which all or most of the H substituents have been replaced by F atoms. Due to their high resistance to heat, oil, water, and grease, the compounds have been widely utilized in a variety of applications (e.g., fire-fighting foam, water-repellent fabrics, non-stick cookware) (Buck et al., 2011). PFAS exist in wastewater streams through a variety of sources, such as agricultural runoff (especially where biosolids are applied), stormwater, industrial effluents, and disposal of by-products.

3. Adsorbents for the removal of contaminants

Adsorbents for the removal of contaminants in wastewater can be classified into three main categories including inorganic, organic, and industrial by-products. Low cost and high efficiency are the two main parameters that determine the effectiveness of adsorbents to be used under realistic wastewater systems. The adsorbents used for the removal of contaminants are numerous and will be discussed in detail in the forthcoming discussions. The removal efficiency (percentage of contaminants removed by adsorbents, calculated from concentrations of contaminants) and adsorption capacity (quantity of contaminants adsorbed by unit mass of adsorbent) are dependent upon several factors, including the type of material, porous features, such as specific surface area, pore morphology, and pore size, and structural features, such as mechanical and chemical stability, and the type and density of surface functional groups (Xia et al., 2019). The adsorbents with large specific surface areas generally offer a large number of adsorption sites for chemical and/or physical entrapment of the contaminants present in wastewater. In terms of industrial application, the key features that need to be addressed include adsorbent stability, shearing during process flows, mechanical integrity on load and recyclability. Often, the wastewater contains different pollutants both organic and inorganic pollutants coexisting in them. Hence, studies on selective adsorption of contaminants where different pollutants coexist need more focus. Understanding the adsorption mechanism of the adsorbent used can be useful for tuning the adsorbent for enhanced adsorption capacity. Figure 1. shows how wastewater can be remediated by using adsorbents and their recovery for reuse in adsorption. The experimental parameters affecting the adsorption performance of an adsorbent involve solution pH, the concentration of adsorbates, the amount of adsorbents, solution temperature, contact time, and coexistence of other pollutants (Akhtar et al., 2016). Table 2 shows a summary of the different types of adsorbents, their properties, and adsorption capacities for the removal of different types of contaminants in wastewater.

3.1 Inorganic adsorbents

Inorganic adsorbents that are commonly used for wastewater treatment involve metal oxides (Wang et al., 2020d), layered double hydroxides (LDHs) (Zubair et al., 2021), silica (Vunain et al., 2016), clays (Thiebault 2020), zeolite (Irannajad and Haghighi 2021), MXenes (Jeon et al. 2020) and MOFs (Metal organic frameworks) (Huang et al., 2021). More materials are available for wastewater treatment including pre-treated (Low et al., 2018, Saadat et al., 2016, Bansal et al. 2016) and various chemically modified adsorbents (Zubair et al. 2017, Rivas et al., 2018) for enhanced adsorption capacity.

The metal/metal oxide-based adsorbents both in bulk and nanostructured form are effective adsorbents because of the availability of a large number of surface-active sites, high mechanical stability, tunable particle and pore size, adjustable morphology, and high chemical stability (Wang et al., 2020d). With the introduction of porosity in metal oxides, nanostructures with a high specific surface area can be realised that exhibit higher activity towards the removal of contaminants. Due to their small size, some nanoparticles are prone to agglomeration, which can be avoided by using porous supports such as porous clays, carbon (Mahvi et al., 2021), silica (Wang et al. 2019b), or biochar (Zhang et al., 2021). Commonly used metal oxides for the removal of contaminants from wastewater include Fe oxides (Fe_2O_3 and Fe_3O_4), Al_2O_3, MnO_2, TiO_2, ZnO, MgO, and ZrO_2 (Wang et al., 2020d). Simultaneous introduction of porosity and phosphorous doping into a TiO_2 matrix enabled high adsorption of Cr^{3+} (92 mg/g) at 0.1 g/L adsorbent dosage and 0.5 mmol/L Cr^{3+} concentration (Wang et al. 2020e). Magnetic oxides such as Fe_3O_4 are appealing for the removal of contaminants from wastewater due to their high specific surface charge and redox characteristics and easy recovery using magnetic separation (Maksoud et al., 2020). For instance, a hybrid of Fe_3O_4 (kaolin/Fe_3O_4) composite was found to be effective in removing naphthalene from aqueous solution with a removal efficiency of 97% at pH 6.5 at 4.8 g/L adsorbent dosage and 10 mg/L pollutant concentration (Arizavi et al., 2020). Interestingly, their reusability studies show that the recovered adsorbent treated with methanol (88%) and DI water (75%) exhibited better adsorption efficiency than the adsorbent without any treatment (~74%) after 4 cycles. Layered double hydroxides, such as hydrotalcite (Liu et al., 2019b) and various synthetic LDHs (Wang et al., 2020c), possess a 2D lamellar structure, high specific surface area, high ion exchange capacity with positively charged layers of metal hydroxides, which make them suitable for the treatment of contaminants in wastewater (Zubair et al., 2021). Similar to the metal oxides, the LDHs can be used as stand-alone or in combination with porous supports, such as biochar, for improved capacity and fast adsorption (Zubair et al., 2021). For example, MgAl-LDH supported on pinewood biochar removed Pb^{2+} (591 mg/g) and CrO_4^{2-} (331 mg/g) from simulated wastewater (Wang et al., 2020c).

Silica based materials, such as MCM-41, MCM-48, SBA-1, SBA-15, and SBA-16 (MCM stands for Mobil Composition of Matter and SBA stands for Santa Barbara Amorphous), are also strong candidates for wastewater remediation due to their large specific surface area, good thermal, mechanical, and water stability, and non-toxic nature (Vunain et al., 2016, Vinu et al., 2005). Moreover, with the conversion of silanol groups on the surface of these materials into siloxane groups, the hydrophobic weak basic sites can capture a wide range of contaminants from wastewater under acidic conditions (de Paula et al., 2021). Using this concept, mesoporous silica with a specific surface area of 348 m^2/g was developed, which efficiently adsorbed methylene blue dye in an aqueous solution to the amount of 61 mg/g at pH 0.5 (de Paula et al., 2021).

The abundant availability, low cost, small particle size, high electrostatic repulsion, and excellent cation exchange capacity of clay minerals, including kaolinite, halloysite, and montmorillonite, make them one of the best materials for adsorption (Thiebault 2020). Among the different clay minerals, halloysite has a moderate specific surface area with an inherent nanotube structure, small pores, and abundant hydroxyl groups (Ramadass et al., 2019). Halloysite functionalized with chitosan showed an adsorption capacity of 238 mg/g for malachite green dye at 2.5 g/L dosage of sorbent and 750 mg/L pollutant concentration (Peng et al., 2015). The separation of the adsorbent after the adsorption process was much easier in the case of the halloysite and chitosan composite as compared to the pristine halloysite. Attapulgite is a microporous phyllosilicate clay mineral with a unique layer-chain crystal structure and relatively high specific surface area. Functionalized attapulgite with polyaniline and magnetite demonstrated an excellent adsorption capacity of 270, 189, and 143 mg/g for commonly-encountered Pb^{2+}, Cu^{2+}, and Ni^{2+} in wastewater, respectively (Sun et al., 2021). Palygorskite nanoparticles and palygorskite microparticles achieved maximal adsorption capacities of 238 and 64 mg/g for Cr^{6+}, respectively, mainly via film diffusion and pore diffusion processes (Rouhaninezhad et al., 2020). Among various clay minerals, the smectite group of minerals (for example, montmorillonite) is the most commonly used one for removing contaminants through adsorption. Owing to its higher charge density, cation exchange capacity, and specific surface area than most other clay minerals, montmorillonite, with or without modification, has seen enormous applications in wastewater-treatment studies (Sarkar et al., 2019).

Natural zeolites are low-cost adsorbents, and their primary mechanism of adsorption is ion-exchange interaction, which can be enhanced by chemical modification with different metals, such as Mn, Fe, Na, Ag, and TiO_2, to improve trapping of contaminants (Irannajad and Haghighi 2021). Zeolite synthesised from fly ash was used as an adsorbent for ammonium ions from swine wastewater with an adsorption capacity of 32 mg/g at 10 g/L adsorbent dose, 100 mg/L pollutant concentration, and under room temperature and neutral pH (Tang et al., 2020). An Fe-zeolite was tested as a sorbent for different phenolic compounds in a wastewater system, and it showed adsorption capacities of 139 mg/g (phenol), 159 (2-chlorophenol), and 171.2 mg/g (2-nitrophenol) (Tri et al., 2020) [30].

MXenes are another rapidly evolving class of materials based on transition metal (such as Ti, Nb, and V) carbides or nitrides that are used for different applications. Along with the properties of having large specific surface area and chemical stability, MXenes possess a large number of active adsorption sites arising from surface terminal groups, such as such as O, F, and OH (Jeon et al., 2020). Owing to oppositely charged sorbate and sorbent, the electrostatic interactions on the surface of MXenes make them excellent candidates for metal ion and radionuclide adsorption (Jeon et al., 2020). For example, $Ti_3C_2T_x$ (where T is the surface terminal groups such as OH, O, F) adsorbed 180 and 225 mg/g of Ba^{2+} and Sr^{2+} in model fracking wastewater due to high negative surface charge and a stacked-sheet-like structure containing a large number of active sites (Jun et al., 2020).

MOFs are inorganic-organic hybrid-type crystalline materials. They possess extremely high specific surface areas of up to ~6500 m^2/g (Wang et al., 2015c), a tunable pore size, and a spatial topology with an ordered porous structure originating from metal cations, metal clusters, and organic linkages, which make them strong candidates for adsorption of contaminants (Huang et al., 2021). MOFs suffer from their instability in wet conditions, however, this can be improved by modifying the surface through suitable functionalisation (Huang et al., 2021). For instance, a high specific surface area (1288 m^2/g) MOF MIL-53 (MIL stands for Matériaux de l'Institut Lavoisier) was employed as a water-stable MOF with a large adsorption capacity of ~505 mg/g for the antibiotic amoxicillin, which had an initial concentration of 150 mg/L, and a dosage 0.1 g/L MOF was used (Imanipoor et al., 2021). In another report, potentially toxic metals in wastewater were adsorbed by a mercaptosuccinic-functionalised, Zr-based MOF with an adsorption capacity of 1080 mg/g for Hg^{2+} and 510 mg/g for Pb^{2+} at pH 4.0 (Wang et al., 2020a).

3.2 Organic Adsorbents

Among organic adsorbents, activated carbon (AC) (Yu et al., 2016) biochar (Almanassra et al., 2021), biomass-derived polysaccharides (Nasrollahzadeh et al. 2021), ordered carbon (Zhang et al., 2020a), graphene (Baig et al., 2019), CNT (carbon nanotubes) (Mashkoor et al., 2020), polymers (Zhao et al., 2018) and ion exchange resins (Ahmed et al., 2015) are commonly studied. Various types of hybrid materials formed by the combination of these materials are also frequently reported.

Activated carbon is one of the most common adsorbents due to its low cost and high efficiency for the removal of contaminants. It can be prepared by using either physical or chemical activation of carbon-containing precursors at high temperature. The ACs derived from biomass are some of the most widely used materials for the removal of contaminants from aqueous media due to their properties, such as low cost, large specific surface area, favourable surface chemistry, strong adsorption ability, and renewability (Yu et al. 2016, Joseph et al. 2021). For example, AC produced from coconut shells using KOH activation showed high adsorption of fluorooctanoic acid (1269 mg/g) at 0.2 g/L adsorbent dosage and 100 mg/L pollutant concentration (Zhou et al., 2021). Biochar differs from ACs in terms of having a reduced porosity; however, it contains an ample amount of surface, functional groups and is generally synthesized without the aid of any activation at relatively low temperatures and with higher yields (Almanassra et al., 2021). The high adsorption capacity, low cost, large biomass-feedstock options, and ease of functionalisation make biochar and its hybrids ideal candidates for contaminant immobilisation in wastewater (Zhang et al. 2020d). For example, Zn-loaded biochar derived from *Fraxinus pennsylvanica* (green ash) marsh leaves showed good adsorption (160 mg/g) for tetracycline and reusability (86 mg/g after 5 cycles) (Wang et al., 2021b).

In addition to activated carbon and biochar, polysaccharides, such as chitosan, cellulose, starch, chitin, pectin and alginate, are also viable options for wastewater treatment due to their abundant availability, low cost, and the presence of naturally occurring functional groups (Nasrollahzadeh et al., 2021). Among these, chitosan-based adsorbents offer a large number of adsorption sites and deliver large adsorption capacity for effective removal of metal ions (Yong et al., 2014; Ahmad et al., 2019b., Manzoor et al., 2019a., Manzoor et al., 2019b). For instance, Ahmad et al. (2019b) showed an adsorption capacity of 185 mg/g for removal of Cu^{2+} within 30 minutes contact time in pH 6 environment. A highly selective, chitosan-based adsorbent with functional groups derived from guanylthiourea modification showed an adsorption capacity of 696 mg/g for Au^{3+} at pH 5 and 30°C (Zhao et al., 2021). Moreover, these prepared materials could be reused with high efficiency of 87 % over 5 cycles, which demonstrates their economic value.

Ordered carbon materials produced using hard and soft templating methods possess a regular morphology, a tunable pore structure, and high specific surface area, which make them effective candidates to adsorb pollutants from wastewater (Zhang et al., 2020c, Benzigar et al., 2018). A functional ordered carbon CMK-1/PDDA proved to be an efficient adsorbent for capturing different acidic compounds, such as p-toluic acid (p-Tol), benzoic acid (BA), 4-carboxybenzaldehyde (4-CBA), phthalic acid (PA), and terephthalic acid (TA). (CMK stands for

Carbon Mesostructured by KAIST, and PDDA stands for polydiallyldimethylammonium chloride.) The adsorption capacities were large, and the selectivity was high, mainly due to the strong electrostatic attraction between the sorbate and the sorbent (Anbia and Salehi 2012). Taking into account parameters like pollutant concentration, adsorbent dosage, pH, and temperature, the adsorption capacity of CMK-1/PDDA for different pollutants varies as reflected in Figure 2.

Among the various carbon nanostructures, graphene and its derivatives, graphene oxide (GO) and reduced graphene oxide (rGO), which have sheet-like structures, show a large specific surface area, high thermal stability, mechanical stability, and surface functionalities (Thakur and Kandasubramanian 2019). Although such materials have good adsorption performance, their tendency to form aggregates is a major issue that renders some of the active sites unavailable for pollutant adsorption. The addition of functional groups, such as those containing oxygen, or the inclusion of spacer materials among graphene layers are methods used to address aggregation (Baig et al. 2019). For example, a silica gel/GO based adsorbent obtained using an ion imprinted technique exhibited an adsorption capacity of 147 mg/g for In^{3+} ions and showed effective reusability, which was evident from the regeneration results of fixed-bed adsorption (Li et al., 2021a). Structural intactness, susceptible to experimental conditions such as heat, irradiation, and acid/base conditions, is an important property that determines electrical properties and oxygen content of GO and/or rGO and interactions with contaminants. A controllable change of oxygen content of GO was obtained by swift, heavy-ion-beam and electron-beam irradiation and showed that the removal capacity of Pb^{2+} increased with irradiation doses but a reversed trend occurred for Cr^{6+} (Bai et al., 2016, Yang et al., 2021). Another class of carbon nanomaterials is carbon nanotubes, including functionalised CNTs. Carbon nanotubes have interesting features, such as high thermal stability, high chemical stability, nanostructure, the curvature of sidewalls, uniform pore size, large specific surface area, ease of functionalisation, and tubular structure, all of which generate large numbers of adsorption sites for adsorption of pollutants such as metal ions and dyes (Mashkoor et al., 2020, Sarkar et al., 2018). A diatomite-CNT prepared by acid treatment and chemical vapor deposition (CVD) with a moderate specific surface area of 50 m^2/g showed adsorption capacities for two phenolic compounds of 8 mg/g for phenol and 17 mg/g for p-cresol at 2 g/L sorbent dosage and 50 mg/L pollutant concentration (Wang et al., 2019a). Graphene-oxide-type and CNT adsorbents also showed appreciable affinity towards perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) (Liu et al., 2020a).

Organic polymers, such as polyaniline, polypyrole, polyacrylamide, and PEDOT [Poly(3,4-ethylenedioxythiophene)], can also be used for wastewater-pollutant adsorbents due to their properties, such as ease of synthesis, effectively degrading natural materials, mechanical stability, chemical stability, and enhanced performance on doping or hybridising with other materials (Zhao et al., 2018). Using the polymers PPY (polypyrrole) and PANI (polyaniline), two different materials were prepared by making a composite with peanut-husk (PH) biomass (Ishtiaq et al. (2020). The resultant polymer, composite materials registered high adsorption capacities (~ 7 mg/g for PPY/PH and ~9 mg/g for PANI/PH) when compared to pristine peanut-husk biomass (2 mg/g) for imidacloprid as the adsorbate (initial concentration of 25 mg/L) at pH 3 with sorbent concentration of 0.2 g/L. Ion exchange resins in both cationic and anionic forms can adsorb a wide range of pollutants with high capacity, while offering low cost compared to materials such as CNT (Ahmed et al., 2015). Arginine-modified starch resin was used as an effective adsorbent for three different dyes and exhibited an adsorption capacity of ~ 25 mg/g at pH 3, which was much higher than zeolite, diatomite, and active carbon (Zhang et al., 2020b).

In addition to the materials discussed above, carbon-based materials such as carbon aerogels (Kalotra and Mehta., 2021), carbon hydrogels (Yang et al., 2020b), and carbon xerogels (Girgis et al., 2012) also have been reported for pollutant removal. Although these materials are prepared using different synthesis techniques, these materials share common properties, such as a large specific surface area, porous structure, and ordered pores. Furthermore, materials like carbon nitride (Martins et al., 2021) and boron nitride (Chao et al. 2021), are some of the emerging materials for pollutant removal from wastewater. The unique structure and chemical properties of these materials make them ideal candidates for further research.

3.3 Industrial by-products

In general, the disposal of industrial by-products involves consumption of additional resources and financial cost, and, hence, the effective usage of these materials as adsorbents could not only solve the disposal issue but also benefit large-scale water treatment due to their low cost. In particular, industrial by-products, such as red mud (Joseph et al., 2020), fly ash (Ge et al., 2018), and sludge (Devi and Saroha 2017), could be used as an alternative to natural or large-scale synthesised adsorbents. However, care should be taken to select appropriate industrial by-product materials as contaminant adsorbents, because some of these materials may pose risk of secondary pollution, such as potentially toxic elements, depending on the source and type of the materials.

Red mud, a widely available industrial waste product resulting from the bauxite refining process for alumina extraction, can be chemically modified by various pre-treatment methods and effectively used for the capturing of metals, inorganic ions, dyes, and phenolic compounds (Joseph et al., 2020). For instance, red mud modified sawdust showed a high affinity towards PFOS with an adsorption capacity of 195 mg/g, greater than sawdust without red mud (179 mg/g) (Hassan et al., 2020a). Fly ash, an industrial waste from coal combustion, has good textural properties with an average particle size of 20 µm, large specific surface area, large porosity, and diverse chemical composition, and it is a low-cost material that makes it an efficient water-treatment adsorbent (Ge et al., 2018). Two types of fly-ash-based adsorbents, collected from a biomass thermal power plant and a coal thermal power plant, showed effective trapping of phosphate, delivering adsorption capacities of 62 mg/g and 4 mg/g, respectively (Park et al., 2021).

Sludges resulting from industrial and municipal wastewater treatment can either be used with simple processing (Maqbool et al., 2016) or by using modifications, such as carbonisation/activation (Sanz-Santos et al., 2021), for usage in pollutant removal from aqueous sources (Devi and Saroha 2017). Pharmaceutical industry sludge, activated with various activating agents [$ZnCl_2$, $FeCl_3.6H_2O$, $Fe(NO_3)_3.9H_2O$, and $Fe(SO_4)_3.H_2O$,] was used for trapping 3 different pesticides, and it showed adsorption capacities of 129 mg/g (acetamiprid), 127 mg/g (thiamethoxam), and 166 mg/g (imidacloprid) with 1.5 g/L dosage and 50 mg/L pollutant concentration for $ZnCl_2$-activated sludge (Sanz-Santos et al., 2021).

In summary, the recent research has led to a significant growth in the development of metals, carbon, silica, clay minerals, MOF, MXenes, and polymer-based adsorbents. These adsorbents can be designed into suitable nanostructures with a large porosity, which helps in delivering better performance towards the adsorptive removal of various contaminants. Some of these materials are low-cost and could be produced in quantities sufficient for large-scale industrial usage. Other materials involving a higher cost could be dealt with by the development of innovative and low-cost technologies and synthesis methods. Particle size, porosity, morphology, and surface functionalisation are critical factors in determining the effectiveness of adsorbent materials. Surface functionalisation or modification of adsorbents with heteroatoms, hydroxyl groups, metals, and carbon nanostructures tend to enhance sorbent-sorbate interactions. The use of inexpensive industrial by-products or wastes, biomass wastes, and natural materials, like clay minerals and zeolites, as adsorbents is a practical approach for large-scale removal of wastewater contaminants. Toxicity and disposal are two vital factors that need to be carefully considered while designing novel materials, so that new materials do not pose any direct or indirect effect on human health and the ecosystem. Because wastewater contains a variety of contaminants in different concentrations, it would be worthwhile to explore novel materials in the form of hybrids and composites that can simultaneously remove multiple contaminants.

4. Recovery and regeneration of spent adsorbents

Adsorbents having high aquatic stability can easily be separated from wastewater streams after the removal of contaminants. Recovery, decontamination, and regeneration potency of spent adsorbents will determine their reusability (Yang et al., 2020a). A good sorbent displays reuse and recovery ability for commercial and industrial applications and may significantly minimize the associated cost of fabrication of adsorbents (Gupta et al., 2020). The regeneration process of spent adsorbents can be repeated several times, however, the regenerated adsorbent exhibits reduced adsorption capacity in comparison to fresh adsorbents (Reddy et al., 2017) (Table 3). Choosing the right regeneration technique is vital for improving the desorption efficiency of the contaminant. Factors such as type of adsorbent, contaminant, stability of the adsorbent, toxicity of the spent adsorbents, the cost and energy requirement of the regeneration process are important for the feasibility of the industrial-scale application. There are several approaches applied to recover and regenerate spent adsorbents, such as magnetic separation (Tamjidi et al., 2019), filtration (Da'na and Awad, 2017), thermal desorption (Hwang et al., 2020), solvent regeneration (Jiang et al., 2018), microwave irradiation (Zhang et al., 2014), supercritical fluid regeneration (Shahadat and Isamil, 2018) advanced oxidation process (Acevedo-García et al., 2020) and microbial-assisted adsorbent regeneration (Abromaitis et al., 2016). A few times interconnected magnetic centrifugal sedimentation also has been applied to separate out Fe/Fe-oxide altered adsorbents (Matsuda et al., 2016). Each approach has its own pros and cons. Therefore, it is key to assess several recovery and regeneration approaches to understand the final reuse and disposal of spent adsorbents.

4.1 Magnetic separation

Metal impregnated adsorbents or magnetic adsorbents are tailored by introducing metal nanoparticles, and they display enhanced specific surface area, pore size, thermal stability, crystallinity, and surface functional groups, which results in improved adsorption efficiency and recovery rate (Gupta et al., 2020). Biomass pre-treated by applying salts of Fe, such as $K_2Fe_2O_4$ and $FeCl_2$/ $FeCl_3$, can be used to synthesized magnetic biochar and, therefore, it can be easily separated by a bar magnet (Zhang et al., 2019a., Li et al., 2016., Wang et al., 2015b). In contrast, Fe-rich biochar feedstocks, such as biosolids or plant biomass that previously has accumulated Fe, can be applied to synthesize magnetic biochar via direct pyrolysis (Ren et al., 2018). Zhang et al. (2019a) verified the magnetization value of adsorbent synthesized via $K_2Fe_2O_4$ pre-treatment, and the value altered from 57.9 electromagnetic unit/g (57.9 emu/g) to 45.2 emu/g after application, which displayed no substantial alteration in the value of magnetization; therefore, there was a similar rate of separation and recovery after use. Clay minerals also can be converted to superparamagnetic adsorbents by depositing nanoscale Fe oxides particles (magnetite) within the mineral structure. For example, an adsorbent prepared by depositing magnetite nanoparticles on palygorskite showed a magnetic susceptibility of 20.2 emu/g and removed 26.6 mg/g of Pb^{2+} from water, and there was an easy separation of the spent adsorbent by applying a simple bar magnet (Rusmin et al., 2017).

Rusmin et al. (2022) prepared magnetic chitosan-palygorskite for the adsorption of Pb^{2+} from the wastewater system showing a maximum adsorption capacity of 58.5 mg/g. The regeneration is done magnetically and showed 82% Pb^{2+} removal after 4 regeneration cycles. In another work by Liang et al. (2022), Co-CNT/N-doped porous carbon was prepared from Zn/Co-zeolitic imidazolate framework and subsequently used for ofloxacin antibiotic showing a high adsorption capacity of 118.3 mg/g. After magnetic regeneration for 4 cycles, the material showed an exceptional 97% adsorption capacity of the optimised sample. Magnetic chitosan microspheres are used to remove I^- from simulated nuclear wastewater system showing with adsorption capacity of 91% after 5 cycles of magnetic regeneration (Li et al., 2022).

Currently, nanoparticle-based technologies have been practiced to eliminate contaminants from wastewater streams, and they are used due to their high specific surface area that leads to improved adsorption capacities. But simultaneously, they impose challenges, like low rates of recovery and non-economical regeneration (Mukhopadhyay et al., 2021, Gupta et al., 2020). Hence, to resolve these issues, magnetic nanoparticles having magnetic properties have been fabricated, which show improved recovery and regeneration (Alqadami et al., 2018). Li et al. (2020a) synthesized a ball-milled magnetic nano-biochar and successfully applied it in the elimination of organic (tetracycline) and inorganic (Hg^{2+}) contaminants from liquid media. This finding revealed that the magnetic properties of nanobiochar enabled its recovery from a liquid stream and its further reuse. After accomplishment of the removal experiment, the nanobiochar was recovered by applying an external magnetic field. An external magnetic field is widely applied in laboratory experiments, but hardly applied at the commercial level. Nevertheless, an external magnetic force can be manually calibrated and applied for practical engineering applications (Ren et al., 2018).

4.2 Filtration

The application of carbonaceous materials, such as biochar and activated carbon, in wastewater treatment (WWT) has been effective (Skouteris et al., 2015). In the operation of WWT, biochar is typically applied as a filling agent in mixed matrix materials (Arrigo et al., 2019). Furthermore, the larger particle size of biochar compared to nanobiochar leads to easy separation through membranes. In this procedure, the recyclability of biochar and degree of separability of solid and liquid phases are improved by changing the dispersion ability of biochar. The use of biochar as a biofilter and immobilizing material in WWT indicates its wide applicability (Ulrich et al. 2017). The combined application of biochar and a membrane bioreactor showed good potential in reducing membrane fouling and an increased life of the bio-membrane was ensured (Tan et al., 2016). Biochar is becoming a part of wastewater treatment. Subsequent separation is not required after its application. However, a study on the mutagenic activity of biochar also shows that the choice of biomass feedstock, pyrolysis temperature and pyrolysis time influences the mutagenic potency (Piterina et al., 2017). Thus, optimal biomass processing is needed for the safe reuse of biochar in applications such as soil amendments and animal food additives. Hanandeh et al. (2017) used two different biochars prepared from the olive mill wastes as a filer amendment for the desorption of total phosphorus. Here the average removal efficiency of the total phosphorus using sand-course biochar (83.35) is better than sand-fine biochar (75.7%). Tejedor et al. (2020) used wood chips/peanut shells as a support matrix for the removal of organic matter from wastewater. Here the filtration systems used alongside the wood chips/peanut shell amendment are using microorganisms, plants and microorganisms, earthworms and microorganisms and all organisms (hybrid biofilters). The COD efficiency achieved for these biofilters with a support matrix is about 80%. Shazryenna et al. (2015) used coconut husk and loofah as support mediums on Candida tropicalis RETL-Cr1 for the adsorption of phenol. Although both coconut husk and loofah used as support medium delivered a similar biodegradation rate (0.0188 $gL^{-1}h^{-1}$), loofah showed enhanced yeast growth. Even though filtration is frequently applied in retrieval of spent sorbent from liquid media, it has a few constraints, such as the filtering agent needing backwashing. Also, nanosized adsorbents cannot be recovered using this method.

4.3 Thermal desorption and decomposition

Recovering metal from spent adsorbent via thermal desorption is an emerging technology. Thermal regeneration comprises heating a sorbent up to a certain temperature to disrupt the physical and chemical bonding between sorbate and sorbent (Shahadat and Isamil, 2018). This method is presently applied for the regeneration of activated carbon at industrial and commercial levels. Heating biochar in the presence of air at temperatures below 500 °C will eliminate the carbon matrix and its volatile components (Zhang et al., 2019a). Xu et al. (2017) removed Pb from liquid media using waste-art paper biochar having a high content of additives. Results of this study revealed a significant removal of Pb (1.5 g g^{-1}). Further, the spent biochar was heated at about 350 °C in a muffle furnace, which facilitated the capture of Pb^{2+} and its further conversion to nano-PbO on the surface of nano-biochar, and it had improved purity (>96 wt%). The end product was a high value product that can be used as an energy storage and conversion device (Yousefi et al., 2014). Up to now, few investigations have dealt with conversion of spent sorbents into value-added products via thermal desorption, and this process remains in its infancy. Nevertheless, the release of volatile components into the environment during the process could be a possible source of secondary pollution. The emission of PAHs and dioxin as by-products of the process shows potential environmental and health impacts. Therefore, the benefit of biochar in carbon immobilization is abolished. Tońki et al. (2021) successfully regenerated MWCNT and applied it for the removal of cyclophosphamide, ifosfamide and 5-fluorouracil with high adsorption capacity. The temperature and the time of thermal regeneration conditions are varied for the maximum recovery of MWCNT and the optimised conditions are found to be 300°C for 2 hours. Studies also show that even after 5 adsorption-desorption cycles, the adsorption capacity is not affected. In another work by Delkhosh et al. (2021), heat-treated gilsonite was used as an effective adsorbent for the removal of toluene from wastewater. For regeneration, 250°C and 20 minutes are applied with an adsorption efficiency of 62.12% after four thermal regeneration cycles. Notably, the thermal regeneration showed more toluene removal efficiency in comparison with acetone washing and ethanol washing.

Currently, microwave irradiation technology is applied as a substitute for thermal desorption due to its speediness, selectivity, and controlled heating (Falciglia et al., 2018). This method includes adsorption of microwave energy by adsorbent molecules and its further translation into heat energy at the molecular level (Falciglia et al., 2017). Microwave treatment heats the sorbent uniformly from the exterior surface to the interior. Dai et al. (2019) showed that the porous feature of the sorbent was not changed much, and, similarly, the properties of adsorbate were preserved during microwave heating compared to conventional thermal heating. The microwave irradiation technique exhibits a more effective controlled-heating method for regeneration of spent sorbent. Furthermore, the dielectric nature of activated carbon (sorbent) linked with the properties of the adsorbed organic pollutant (PFAS), like volatility, could permit PFAS-exhausted activated-carbon regeneration via interactions among delocalized π-electrons of the sorbent (activated carbon) and the microwave electrons. The industrial-scale application of microwave irradiation for thermal desorption of adsorbents is costly not only for setting up the plant but also energetically expensive as a sustainable process. Still, regeneration of spent sorbents via microwave treatment requires further investigation to make this technology economically viable (Gagliano et al., 2020).

Recently, contaminants loaded in spent adsorbents are decomposed via a thermal treatment giving rise to an adsorbent with a new porous structure and surface-chemical properties. The resulting adsorbent, following such thermal treatment of the spent adsorbent, has been reemployed for adsorbing the same contaminants with similar or slightly lower removal capacities. For example, Sonmez Baghirzade et al. (2021) suggested that an optimized thermal treatment could successfully regenerate PFAS-laden granular activated carbon (GAC) by mineralising the extremely persistent PFAS and could, thus, recover the spent GAC. PFAS compounds can be desorbed and volatilised at around 175 °C but can be mineralised at high temperatures (around 700 °C) (Xiao et al., 2020). In particular, high-temperature thermal desorption results in large energy requirements hindering its sustainability and industrial-scale production. The specific surface area and micropore volume of thermally-treated, spent GAC might increase with increasing temperature, but very high temperatures (>1200 ^{o}C) might destroy the pore structure permanently (Sonmez Baghirzade et al., 2021). Chang et al. (2021) employed a 600 °C treatment for 2 h to regenerate a montmorillonite adsorbent following the adsorption of an antidepressant-drug contaminant called amitriptyline. A change in the physico-chemical properties of the regenerated adsorbent was observed, displaying 71.7 mg/g amitriptyline removal, which was ~26% of the original montmorillonite. Therefore, in order to achieve successful adsorbent regeneration via the thermal decomposition method, appropriate temperature and treatment conditions (e.g., gaseous environment) are important. Thermal treatment conditions may vary depending on the type of adsorbents, contaminants, and purpose of subsequent use, which require future research for scientific advancement as well as for scaling up the process.

4.4 Chemical desorption

The main goals of applying organic and inorganic solvents for removing or eluting contaminants from adsorbents are to retain the sorbent properties and further their reuse. Table 4. covers the different adsorbents and the solvent used in recent research for regeneration. For regeneration (removal of pollutants) of spent magnetic bio-adsorbents, using a low concentration (0.1-0.2 M) of acids or bases is suggested. Acids and bases that have been used as regenerative solvents are HCl, HNO_3, H_2SO4, EDTA (ethylenediaminetetraacetic acid), $Ca(NO_3)_2$, NaOH, and $NaNO_3$ (Gupta et al., 2020, Yang et al., 2020a). Hassan et al. (2020b) and Baig et al. (2014) showed that As could be desorbed from magnetic sorbents and further magnetic adsorbents could be regenerated by applying 0.5 M NaOH. A significant desorption efficiency was observed when HCl, HNO_3, and H_2SO_4 were applied as regenerative solvents. A low pH induces the desorption of metals from the adsorbent surface and simultaneously improves the regeneration of adsorbents (Gupta et al., 2020). Addition of a strong acid generates competition among heavy-metal (HM) ions, the hydronium ion (H_3O^+), and the hydrogen ion (H^+) for active sites. For instance, Kołodyńska et al. (2017) found that 95% Cu desorption efficiency was achieved after application 3.5 M HNO_3 as eluent. However, simultaneously, a higher acidic environment can deform the adsorbent structure, which leads to decreased adsorption and desorption capacity. Hence, acid treatment should only be done if the sorbent has decent mechanical properties and steadiness. Khenniche et al. (2021) prepared ferromagnetic carbon from coffee residue and applied as an adsorbent for the removal of tetracycline and sulfamethazine in wastewater system. Further, using 0.1N NaOH, chemical regeneration was performed and adsorption capacity of ~72% and ~40% (for tetracycline) were delivered for fresh carbon and spent carbon respectively. In another work by Siciliano et al. (2021), thermo plasma expanded graphite is used as an adsorbent and regenerated using 1M HCl showing ~87% adsorption efficiency of MB dye after 5 cycles of regeneration.

Improved desorption rates could also be attained by applying chelators like ethylenediaminetetraacetic acid (Yang et al., 2020a). Chelators have a number of electron-donating functional groups, like carboxy (COOH) and amine, which show a great attraction towards HM ions; therefore, they can produce stable chelator-HM complexes. The adsorbed HM ions are desorbed from the adsorbents using chelators, and then they form complexes with these chelators. Hu and Shipley (2013) used EDTA and a mixed solution (NaCl, $NaNO_3$, $CaCl_2$, $NaHCO_3$, $MgSO_4$, $NaHPO_4$) of common ions to evaluate the capacity of nano-TiO_2 to desorb Pb^{2+}, Zn^{2+} and Cu^{2+}. The application of the solution of common ions resulted in nominal desorption efficiency, while the EDTA chelator showed 92% desorption efficiency. Improved regeneration cycles resulted, probably because of the robust chelating characteristics of EDTA. The possible reason for this improved adsorption capacity was the generation and activation of new adsorption sites in the adsorbent by EDTA-4Na.

Application of alkali eluents leads to a reduced degree of protonation of the sorbent surface, resulting in desorption of contaminants and regeneration of adsorbents. Likewise, HMs from chemical adsorbents, such as Mn-coated powder, Fe-coated powder, nZVI (ZVI stands for zero valent iron), oxides and hydroxides of Fe^{3+}, and magnetic biochar prepared from wheat straw were effective in removal when regenerated via alkalis (Lata et al., 2015). Wang et al. (2017a) applied 0.1 M NaOH and reported successful desorption of As up to 98.2% after 24 h from a spent sorbent. After achieving complete desorption, the adsorption sites on the spent sorbent are easily reactivated by adjusting the medium pH to neutral, using alkali or acid (Gupta et al., 2020). These laboratory-scale investigations have established the capability of chelators, acids, and alkalis to regenerate spent adsorbents, but still the viability of the whole procedure on a commercial scale remains uncertain.

4.5 Supercritical fluid desorption

A supercritical fluid (SCF) is produced when a substance has been heated above its critical temperature and compressed beyond its critical pressure (Shahadat and Isamil, 2018). The application of a SCF to regenerate spent adsorbents is extensively applied these days and contemplated as substitute for chemical-solvent and incineration processes (Efaq et al., 2015). In the soil matrix, the SCF behaves as a usual solvent and facilitates the process of desorption of pollutants. The contaminant is further condensed by decreasing the pressure and finally, it can be gathered into a small volume container. Carbon dioxide (CO_2) is the most preferable SCF that is used frequently due to its incombustibility and non-hazardous and economical nature (Noman et al., 2020). Also, it shows a high rate of mass transfer along with lower surface tension. In spite of its superiorities, CO_2 showed inferior regeneration capacity for phenol-loaded adsorbents (Humayun et al., 1998). To resolve this issue, Salvador et al. (2013) applied supercritical water instead of CO_2, which fully desorbed phenol from the phenol-loaded sorbent and attained nearly 100% efficiency. Application of supercritical water shows advantages and disadvantages, such as a very small process duration, which remarkably reduces the process cost, but simultaneously it needs high pressure, which enhances process cost and restricts its applications at the commercial scale. Therefore, supercritical water regeneration can only be applied at a small scale.

Using supercritical water regeneration, Zhang et al. (2019b) regenerated activated carbon while using H_2O_2 and alkali metal catalyst. Interestingly, the supercritical water regenerated samples exhibited enhanced specific surface area (813 m^2/g) in comparison to fresh sample (765 m^2/g) while exhibiting a regeneration efficiency of 107%. Moreover, the regeneration temperature (385°C, 405°C and 425°C), the concentration of H_2O_2 and alkali metal as a catalyst is found to vary the adsorption capacity of phenol as a contaminant. In another study by Carmona et al. (2014), granular activated carbon is regenerated using supercritical CO_2. Here, the desorption yield of the contaminants varied with respect to the pressure (6, 15, 20, 31 MPa) and temperature (45°C, 60°C). At 31 MPa and 45°C, desorption yield as high as 97.9%, 68.3%, 71.5% and 64.5% were obtained for phenol, 2-chlorophenol, 4-chlorophenol and 2,4-dichlorophenol respectively.

In place of applying only a SCF, SCF along with a co-solvent was also performed to improve solvent polarity and, subsequently, desorb contaminants from a spent sorbent. The desorption of 4-nitrophenol (4-NP) and phenol from organically functionalized smectite has been successfully carried out with ethanol (co-solvent) and without ethanol (Park and Yeo, 1999). Results of the study exhibited 73.6% desorption of the contaminant when there was no co-solvent applied in the reaction mixture but when 2.5% (v/v) ethanol was applied at 70 °C and 413.6 bar pressure, the percentage recovery was 90.8%. Similarly, Salgin et al. (2004) applied ethanol to eliminate salicylic acid from organically functionalized bentonite. The desorption efficiency was 76 wt% and 98 wt%, respectively, when there was no ethanol and when 10% (v/v) ethanol was applied. These findings reveal the potential role of SCF and application of a co-solvent in desorption of contaminants and regeneration of spent adsorbents. However, innovative approaches for reducing the cost of this process needs to be developed to enable its industrial-scale application.

4.6 Advanced oxidation processes

Recently, the application of advanced oxidation processes (AOPs) in the regeneration of spent adsorbents is gaining much recognition (Acevedo-García et al., 2020, Fdez-Sanromán et al., 2020). In recent studies, biochar was applied as a catalyst support to AOPs or the AOPs were used in the regeneration of spent biochar (Kumar et al., 2020a, Acevedo-García et al., 2020). Li et al. (2020c) fabricated an Fe and nitrogen (N) co-functionalized wheat straw biochar by applying urea and ferrous sulphate as chemical reagent, which activates persulfate (PS), for degradation of organic pollutants, such as acid orange (AO), methyl orange (MO), phenol, bisphenol A (BPA), and tetracycline hydrochloride. Mer et al. (2021) proposed a dual use of biochar. It was first used as a sorbent to remove Ni and Pb, and, subsequently, hydroxyl radicals assisted in situ degradation of phenol. Zhang et al. (2020a) determined that the defective surface structures and oxygen containing functional groups (OCFGs) of mesoporous biochar, prepared using bagasse as feedstock and further functionalized and activated via KOH and $CaCl_2$, played key roles in oxidative degradation of a contaminant (phenol). Moreover, during degradation of sulfamethoxazole (SMX) by PS activation, Lykoudi et al. (2020) found a strong, linear co-relation between concentration of the sodium persulfate and the spent sorbent (coffee biochar), and the AOP facilitated the surface adsorption of SMX by the spent sorbent. In the same way, Acevedo-García et al. (2020) established the improved adsorption of SMX and methylparaben by lime-fiber-synthesized biochar and its viable regeneration by various AOPs, such as the Fenton reaction, PS, electro-oxidation-H_2O_2, and an electro-Fenton reaction.

Dutta et al. (2009) used TiO_2 as an adsorbent for the removal of Reactive Red 198 dye from wastewater, where AOP is used for regenerating the adsorbent without the use of any chemicals. Gonzalez-Olmos et al. (2013) used Fe-zeolites as adsorbents for the removal of Methyl tert-butyl ether (MTBE) from wastewater. Subsequently, regeneration using AOP with H_2O_2 is performed where the MTBE concentration is reduced from 0.9–1.5mg/L to ≤0.1 mg/L within 2 to 3 days. In another work, Bach et al. (2009) used room temperature AOP for the regeneration of GAC using hydrogen peroxide (oxidant) and iron oxide (nanocatalysts). The regenerated samples show a negligible reduction in adsorption even after four cycles of regeneration.

4.7 Microbial-assisted adsorbent regeneration

Microbial-assisted regeneration of a spent adsorbent implies reviving the sorbent via microbial degradation of organic contaminants adsorbed by sorbent (Abromaitis et al., 2016). This process is generally performed by either a pure microbial culture or mixed microbial consortia, such as bacteria, fungi, and algae. Microbial degradation of organic contaminants can be accomplished via mixing microbes with saturated adsorbents in batch operations or it can be accomplished during biological wastewater treatment (Shahadat and Isamil, 2018). One pre-condition for applying microorganism-assisted regeneration is that the adsorbent surface should be non-toxic to the acting microorganisms and the material should support a microbial habitat and growth (Sarkar et al., 2012). In batch bio-regeneration of spent adsorbents, microorganisms and their carbon and nutrient sources are mixed along with organic-laded adsorbents, and these contaminants later are mineralized by microbial action and the adsorbents will be regenerated (Abromaitis et al., 2016)

Bio-regeneration of spent sorbents has been performed via two routes. The first route is desorption along a concentration gradient, in which the unconfined organic material is mineralized by microbial action, which decreases the concentration of the pollutants in the liquid media. Therefore, a concentration gradient is developed between the sorbent surface and the liquid media (Klimenko et al., 2002). The second route is enzymatic degradation of pollutants, in which extracellular enzymes released by microbes in liquid media diffuse into the pores of the sorbent and act on entrapped pollutants and hydrolyzed them. The bio-regeneration process of spent adsorbents depends on several factors, such as microbial diversity, their generation time, the microorganism-pollutant concentration ratio, nutrient availability, temperature, and dissolved oxygen level (Klimenko et al., 2003). Therefore, optimization of these parameters is key to achieve good bio-regeneration.

Bio-regeneration of clays or functionalized clays was investigated by Yang et al. (2003). They reported that the microbial regeneration of hexadecyltrimethylammonium (HDTMA)-functionalized montmorillonite was more effective than chemical regeneration. The yeast, *Pityrosporum* sp., was applied in the bio-regeneration of HDTMA-functionalized clay. To replace thermal granular activated carbon (GAC) reactivation, melamine is degraded by biomass with nitrification and denitrification step while methanol is used as an additional carbon source (Piai et al., 2021). Notably, the bio-regenerated GAC showed similar adsorption capacity during the first few hours when compared to fresh GAC; after 4.5 hours they showed a significant reduction in adsorption capacity. This shows that the bio-regeneration of GAC is partly successful to restore its adsorption active sites. Ren et al. (2013) successfully degraded phenol loaded on an animated hyper-cross-linked polymeric resin (NDA-802). Here the adsorption studies done after bio-regeneration showed less than 20% of adsorption capacity is lost even after 5 cycles when compared to NDA-802 before bio-regeneration. At the same time, the non-bioregenerated NDA-802 showed zero phenol adsorption after 3 cycles revealing the effectiveness of regeneration on phenol adsorption. In another interesting study, using Pseudomonasputida, formaldehyde is degraded from montmorillonite clay/polyethyleneimine/bacteria composite by self-regeneration process (Zvulonov et al., 2019). Notably, a high adsorption capacity of 62 mg/g and a high degradation rate of 600 $mg \cdot L^{-1} \cdot FA \cdot h^{-1}$ is obtained for this composite.

The primary disadvantage associated with bio-regeneration of spent sorbents is its slow regeneration rate. It requires further studies to make the process viable for commercial-scale application. Moreover, not all adsorbents are appropriate for biological regeneration. A few chemicals, such as cationic surfactants, which are frequently applied to improve the hydrophobicity and organic-contaminant adsorption capacity of adsorbents, can be harmful to microorganisms used in bio-regeneration (Sarkar et al., 2010, Zhu et al., 2009) and to ecological receptors such as earthworms (Sarkar et al., 2013).

5. Management, reuse, and disposal of spent adsorbents

Open dumping of spent adsorbents containing toxic organic pollutants poses environmental and social risks, particularly in developing countries where incineration and engineered landfilling facilities are scarce (Chaukura et al., 2016, Gwenzi et al., 2015). The sorbent applied in the removal of HMs may require disposal after desorption of the HMs or disposal even without desorption. In both cases, generation of secondary pollution is obvious (Tzou et al., 2007). Even then, sorbents loaded with HMs have toxic impacts on the health of humans and the environment. Henceforth, spent adsorbents should be discharged into the environment only after complete desorption of HMs or organic contaminants (Lata et al., 2015). Therefore, before commercializing production of adsorbents, appropriate consideration towards final management should be paid. Although disposing is a cost-effective process, the ecological feasibility and the long-term usage needs to be considered. On the other hand, reuse in other applications requires toxicity studies such as direct or indirect impact on human health. Overall, there are four approaches that have been employed in management, disposal, and reuse of spent sorbents. They are reuse (Haddad et al., 2018, Yang et al., 2020a), incineration (Fernández-González et al., 2019), landfilling (Dhillon et al., 2017), and other safe disposal techniques (Huang et al., 2020a, Ramrakhiani et al., 2017), each of which is discussed below. In the case of reuse, the spent adsorbents are used in applications such as soil amendment, capacitor and catalyst/catalyst support whereas incineration and landfilling are used as common safe disposal approaches.

5.1 Soil amendment

There is an increasing interest in rejuvenating low fertility soils to enhance crop yield and productivity using biochar that has been employed in the removal of nutrients from wastewater. Therefore, before the application of biochar for this purpose, proper selection of feedstocks and fabrication conditions must be evaluated according to types of soil and crops (El-Naggar et al., 2019). The re-utilization of nutrients recovered in biochar applied as a soil conditioner can be estimated by water extraction, which evaluates soil pore water and plant growth at the laboratory level (Shepherd et al., 2017). Justifications for using spent biochar include improving plant growth and enhancing soil CEC and organic matter (OM), which limit leaching of soil nutrients (Yu et al., 2019). Interaction mechanisms between phosphorus (P) and biochar frameworks are not sufficient to avert P release into the natural environment (Shepherd et al., 2016, Wang et al., 2014). Nevertheless, studies show that nutrient-enriched biochar could be a type of environmentally friendly fertilizer that can be used as substitute for synthetic fertilizer (Li et al., 2016, Shepherd, et al., 2017). Liu et al. (2019a) reported that spent biochar, which has a flower-like precipitate of $Ca_5(PO_4)_3(OH)$, can be applied as an inorganic fertilizer.

Wang et al. (2020b) observed that spent biochar applied in recovery of nutrients exhibited the capability to improve seed-germination rate and, simultaneously, enhance shoot length of the grass. Similarly, Xu et al. (2018) observed that the use of nutrient-rich, spent biochar enhanced the plant growth and biomass. Their study also demonstrated that there was no substantial difference obtained between nutrient-rich, spent biochar and synthetic fertilizer in improving the weight of the plant dry matter. Therefore, spent biochar can be applied as an inorganic fertilizer or soil conditioner (Haddad et al., 2018, Mosa et al., 2020). A relatively higher release of nutrients during an initial phase agrees with the growth curve of the plant (Shepherd et al., 2016). The application of biochar as a soil amendment for various types of soil was conducted by Yu et al. (2019). Choosing the right feedstock and synthesis techniques greatly affect the soil amendment properties of biochar. Further, by doping or making hybrids/composites with biochar using other appropriate materials, the needs of the particular soil can be met to enhance plant growth. Not only nitrogen

(N) and P, but also humate acid, are necessary as fertilizers and plant growth enhancers, and they can be successfully adsorbed from liquid media via biochar (Jing et al., 2019, Li et al., 2016). Spent biochar may also hasten the process of composting and also improve the quality of compost, which then can be used as an organic fertilizer in the field (Kumar et al., 2021a, Ye et al., 2019, El-Naggar et al., 2019). Nevertheless, owing to the diversity of feedstocks of biochar and its previous application, possible toxicity must be evaluated prior to its large-scale application (Shepherd et al., 2016). Depending on the biochar feedstock, the toxicity limits of PTE exceeded the International Biochar Initiative certification and European biochar certification guidelines in some cases. This signifies that the right biomass feedstock and biomass processing greatly determines the toxicity which in turn determines its ability to be used in soil amendment.

5.2 Capacitor and Catalyst/catalyst support

The electrochemical potency of several carbonaceous materials can be enhanced when impregnated with a certain quantity of a metal (Fu et al., 2019, Qin et al., 2019). Application of spent biochar, for use as an energy conversion and storage device or catalyst support/carrier, can be improved by re-treating spent biochar with microwave irradiation or pyrolysis (Wang et al., 2017b, 2018). During re-treatment, spent biochar and desorbed HMs react with each other, resulting in improved catalytic performance of spent biochar. In biorefinery and pollutant remediation, biochar plays various roles as a catalyst or as a support for catalysts (Kumar et al., 2020a, Xiong et al., 2017). Biochar can improve the transformation of tar during its pyrolysis. Also, biochar can transform high oxidation state metals to a lower oxidation state, which further improves catalytic performance. Metal-impregnated biochars can replace costlier synthetic carbon nanomaterials (carbon nanotubes), and they might be used as supercapacitors in the near future, as well as for tar removal during pyrolysis, gasification, and syngas purification (Tang et al.,2019, Wang et al., 2019c, Kumar et al., 2020a). A HM-loaded spent sorbent (biochar) can be used for the synthesis of supercapacitors. For instance, microwave oxidation has been performed with Ni^{2+}-loaded black carbon (BC), which decreased the carbon proportion and improved the oxygen proportion, resulting in improved capacitance along with efficiency and power density (Gupta et al., 2020).

5.3 Incineration and landfilling

Carbonaceous sorbents contain an abundant quantity of polymers such as cellulose, hemicellulose, and lignin (Kumar et al., 2020a, b). Incineration as a final management technique not only decreases mass and volume of the spent sorbent but also, simultaneously, facilitates the recovery of energy and HMs (Huang et al., 2020a). Ding et al. (2014) demonstrated recovery of caesium ions from hexacyanoferrate-functionalized walnut-shell spent biochar using incineration as a final disposal technique. In their investigation, substantial deceases in volume and mass of the spent sorbent were detected, but volatilization of caesium ions was not observed from the spent sorbent. Martín-Lara et al. (2016) conducted a study on pine-cone shell biomass for removal of Cu^{2+} and Pb^{2+} from aqueous media, and then they used pyrolysis under a controlled N atmosphere as a final disposal technique. A reduced quantity of oxygen, sulphur, and nitrogen, and high quantity of carbon, was observed in the end product, which could be used as a source of thermal energy, instead of coal, with reduced corrosiveness and toxic gas emission.

Dumping of spent adsorbents in landfills is another method of management (Dhillon et al., 2017, Carvalho et al., 2013). This process is similar to domestic landfilling and is cost-effective (Pandey and Shukla, 2019). Nevertheless, before dumping the spent sorbent in landfills, it is imperative to determine the concentration of the adsorbate in the spent sorbent. For example, PFAS with a concentration below 50 mg/kg in a spent sorbent is permitted to be disposed of in landfills (NEMP, 2020). It is crucial that the pollutant remains adsorbed by the spent sorbent for a long period of time, so the chance of its release into the surrounding environment is small. Kasiuliene et al. (2019) demonstrated an innovative technique by applying incineration and co-incineration (mixed with lime) together to manage a spent sorbent loaded with As, Cr, Cu and Zn. The release of As, Cu, and Zn was reduced in the thermally treated spent sorbent compared to the untreated sorbent. Nevertheless, Cr leaching was increased, and that hampered landfilling of the residual ashes. But co-incineration of the spent sorbet mixed with 10 wt% lime remarkably reduced the Cr leaching due to the formation Ca-Cr, a water insoluble compound (Kasiuliene et al., 2019). The spent sorbent, which had HMs, required prior treatment before landfilling, to curb the chance of secondary pollution. After appropriate treatment, the residue could be dumped in a landfill or could be buried, and natural activities would finish the final disposal.

5.4 Other management approaches

Apart from the above methods for spent-sorbent management, there are a few other available methods. Vilar et al. (2007) sterilized a spent sorbent via microwave irradiation and then sealed the sterilized fraction in an inert-material container to avoid leaching of the contaminant. The recovered magnetic spent sorbent could be further applied in construction to manufacture blocks, adhesives, and cement; however, its high proportion may affect the mechanical strength of the mixed, final products (Gupta et al., 2020). Phytocapping and phytoremediation, using selected plant species, are other options for management of spent sorbents loaded with contaminants. Later these plants can be used for biochar synthesis (Fuke et al., 2021, Hassan et al., 2020b, Kumar et al., 2021b). Ramrakhiani et al. (2017) and McCloy and Goel (2017) described the application of inert phosphate glass in the immobilization of spent sorbents loaded with pollutants. Ramrakhiani et al. (2017) applied a spent sorbent loaded with 25 wt% HMs to an inert phosphate glass and observed that there was no leaching of HMs ions from the construct after 35 days of incubation. Additional research is required related to recovery, regeneration, and final disposal of spent adsorbents, so that they can deliver better outcomes and provide a new path of management.

6. Conclusions and future research directions

Wastewater contains various inorganic and organic contaminants that need to be removed before releasing it into the environment. Adsorption using organic and inorganic adsorbents (e.g., biochar, activated carbon, clay minerals, zeolite) is widely used in wastewater treatment to remove undesirable compounds. The spent adsorbents are often recycled for the circular economy through a number of approaches, such as filtration, chemical and thermal desorption, and advanced oxidation processes. The current recovery and regeneration techniques are found to be highly dependent on the type of the contaminant and the adsorbent. The regenerated samples perform remarkably well as adsorbents in wastewater adsorption. Recently, there has been increasing interest in developing advanced, engineered adsorbents with high adsorption capacity to remove and recover contaminants from wastewater streams. At the same time, researchers have focussed on the safety and cost of the adsorbent process as well. Until now, the disposal of end-of-life adsorbents and the subsequent recovery of sorbed contaminants are major practical challenges. The spent adsorbents are subsequently either regenerated for reuse as soil amendments, capacitors, and catalysts or safely disposed through incineration and landfilling. The reuse of the spent adsorbents can not only benefit the environment but also reduce the overall cost of the application. Chitosan-based materials are used in a wide range of applications such as wound healing, drug delivery, bioimaging and tissue engineering (Ahmad et al., 2017, 2019a, 2021). Usage of the spent chitosan-based adsorbents and modifying them for reuse in such applications would be interesting.

Life Cycle Analysis (LCA) is a tool to evaluate the complete environmental effects including the ecological and economic feasibility of the system. This process can be applied to wastewater treatment for the analysis of both positive and negative impacts and thus finding its feasibility before applying it to the practical/real system. Waste management, cost, energy consumption and safety are to be considered important for this analysis. For instance, Vukelic et al., (2018) prepared activated carbon from waste cherry and sour cherry kernels and analysed with LCA. A number of factors such as transportation, various processing, chemical treatment, use of acids and water, electricity, washing, disposing of in landfill, use of waste paper and wastewater were involved in this process. The results show that this process using waste cherry and sour cherry kernels is eco-friendly and economically viable and can be potentially used on an industrial scale with minimal negative impacts on the environment. Researchers need to put more focus on LCA especially with the emerging adsorbents.

Given the current emphasis on zero waste generation, improved resource efficiency and circular economy in the context of environmental sustainability, , the following research areas could be pursued to make further advances on the adsorption-based removal of contaminants in wastewater streams:

- Development of innovative adsorbents derived from natural resources that are effective in the removal of both inorganic and organic contaminants from wastewater streams.
- Development of low-cost technologies for the recovery of contaminants from spent adsorbents.
- Long-term leaching studies examining groundwater contamination through the movement of contaminants from spent adsorbents disposed in landfill sites.
- Long-term kinetic studies on the value of recycling of spent adsorbents as a nutrient source (in the case of removal of nutrients such as N and P).
- Long-term studies on the stability of recycling and reuse of spent adsorbents.
- Conversion of spent adsorbents into value-added products for recycling and reuse.
- Toxicity studies of adsorbents before their reuse in other applications.
- Energy-efficient regeneration techniques for spent adsorbents.
- Identifying new reuse applications for the management of spent adsorbents.
- Life-cycle analysis and risk assessment of recycling and reuse of spent adsorbents.

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Figures

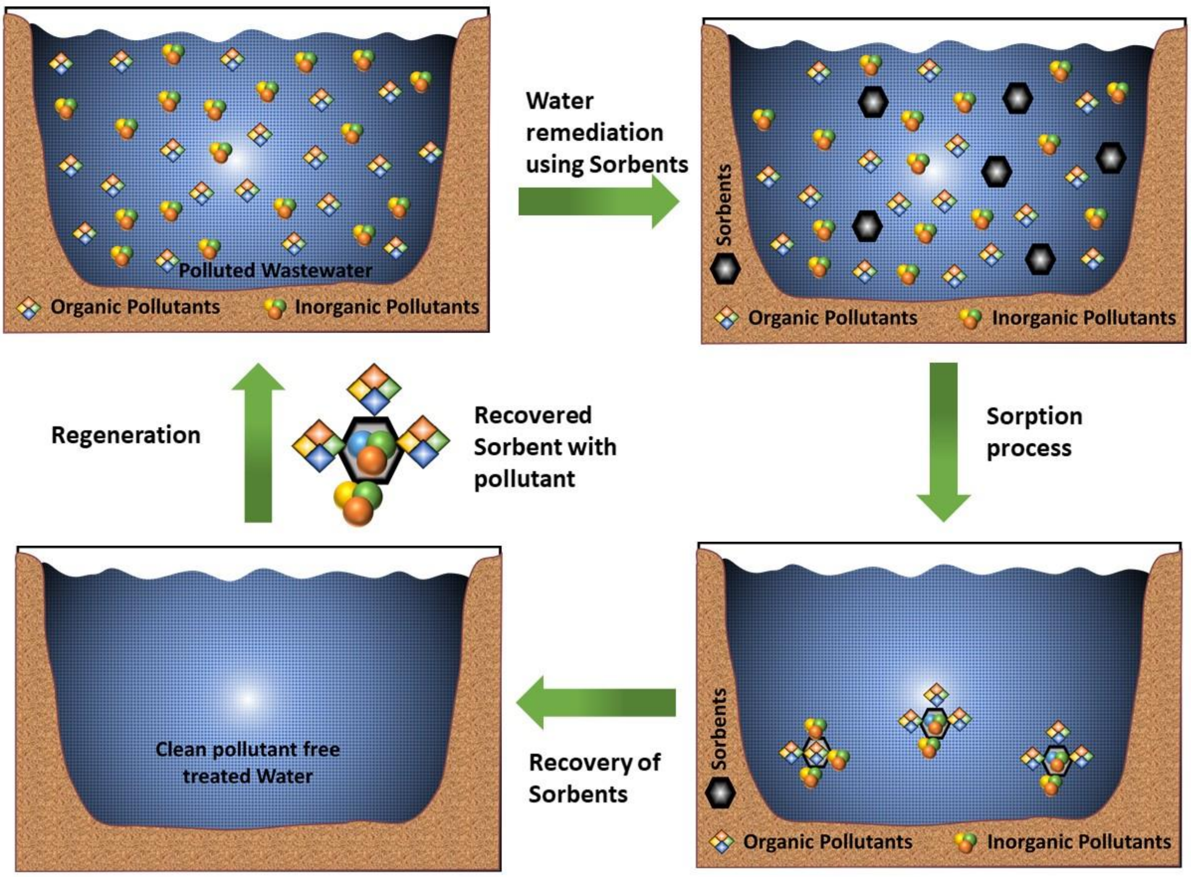

Figure 1. Schematic illustration of the removal of wastewater contaminants using sorbent

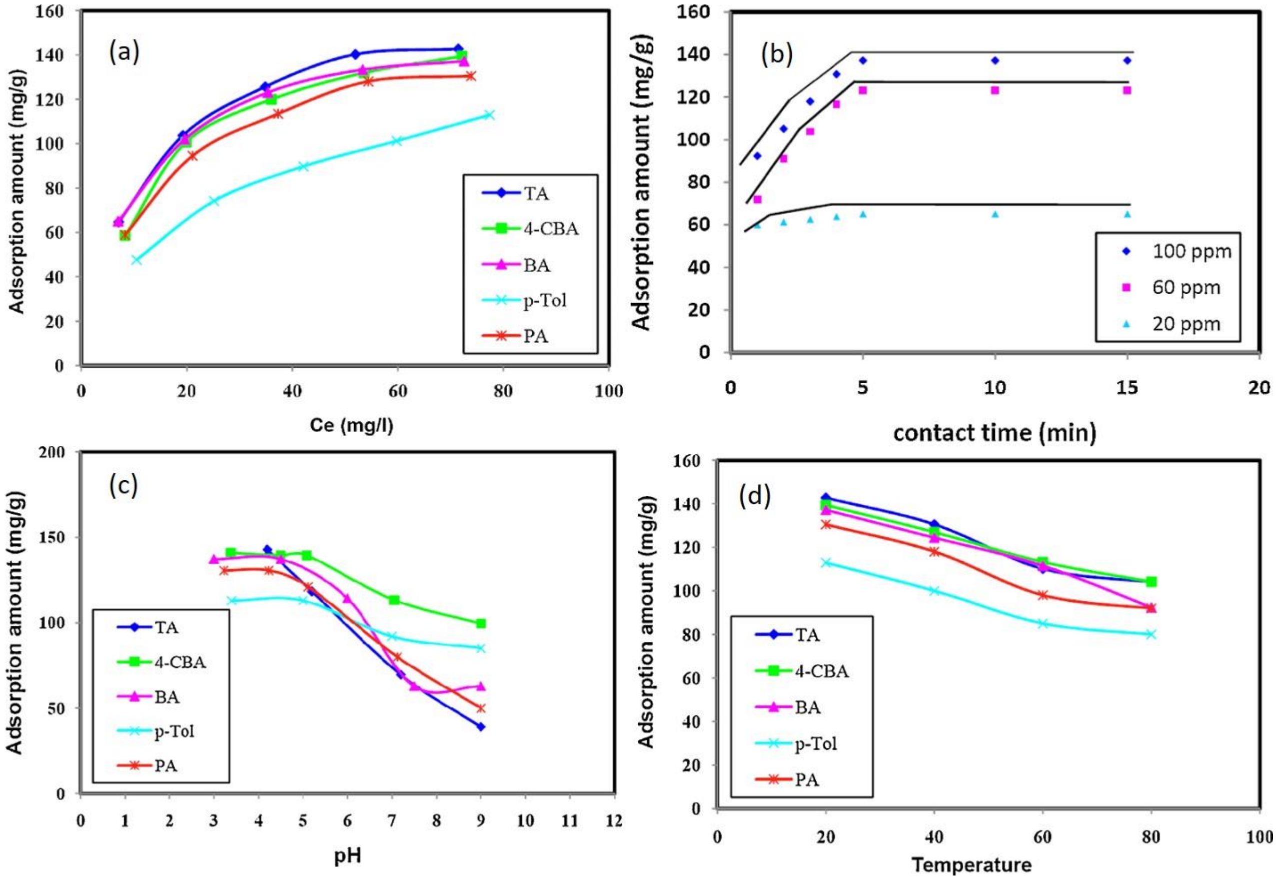

Figure 2. Sorption capacity performance of acidic compounds TA, 4-CBA, BA, p-Tol and PA using CMK-1/PDDA as sorbent (a). Comparison of adsorption of acidic compounds with respect to pollutant concentration (b). Effect of contact time and concentration of BA on adsorption (c). Effect of pH on adsorption (d). Effect of temperature on adsorption

Tables

Table 1. Contaminants in wastewater sources

Wastewater source	Location	Contaminants	Concentrations	Reference
River	Barmah-Millewa	Dissolved organic carbon	2–6 mg/L	Rees et al.
	Forrest, Murray	(DOC)	< Limit of Detection (LOD)–4 μg/L	(2020)
	Darling Basin,	Oxides of nitrogen (NOx)	12–15 μg/L	
	Australia	Ammonium (NH_4^+)	< LOD μg/L	
		Filterable reactive phosphorus		
	Mid-Murray,	DOC	3.5–6 mg/L	
	Murray Darling Basin,	Particulate organic carbon	15–30 mg/L	
	Australia	(POC)	1–2 mg/L	
		Particulate organic nitrogen	2 μg/L	
		(PON)	2–6 μg/L	
		NOx	12–15 μg/L	
		Filterable reactive phosphorus		
		NH_4^+		
	Lower Murray,	DOC	3 mg/L	
	Murray Darling Basin,	Total N	< LOD–800 μg/L	
	Australia	Total P	50–90 μg/L	
		NOx	5–10 μg/L	
		NH_4^+	12–15 μg/L	

Conventional drinking-water-treatment plant	USA	Stimulant	Caffeine: 0.119 μg/L	Stackelberg et al. (2004)
		Anticonvulsant	Carbamazepine: 0.258 μg/L	
		Nicotine metabolite	Cotinine: 0.025 μg/L	
		Nifedipine metabolite	Dehydronifedipine: 0.004 μg/L	
		Fragrance manufacturing	7-acetyl-1,1,3,4,4,6-hexamethyl tetrahydronaphthalene: 0.49 μg/L 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethyl Cyclopenta-γ-2-benzopyran (HHCB): 0.082 μg/L	
		Fixative	Anthraquinone: 0.072 μg/L	
		Plasticizer	Benzophenone: 0.13 μg/L	
		Trihalomethane	Bisphenol A: 0.42 μg/L; Bromoform: 21 μg/L	
		Insecticide	N,N-diethyl-meta-toluamide (DEET): 0.066 μg/L	
		Herbicide	Prometon: 0.096 μg/L	
		Solvent	Tetrachloroethylene: 0.1 μg/L	
		Plasticizer	Tri(2-butoxyethyl) phosphate: 0.35 μg/L	
		Flame retardant	Tri(2-chloroethyl) phosphate: 0.099 μg/L	
		Flame retardant	Tri(dichlorisopropyl) phosphate: 0.25 μg/L	
		Flame retardant	Tributyl phosphate: 0.1 μg/L	
		Cosmetics	Triethyl citrate (ethyl citrate): 0.062 μg/L	
Wastewater treatment plant effluents	Boulder Creek, Colorado, USA	Acidic organic compounds	Ethylenediaminetetraacetic acid: 140 μg/L Nitrilotriacetic acid: 1.0 μg/L	Barber et al. (2013)
		Neutral organic compounds	Bisphenol A: 0.084 μg/L Caffeine: 0.18 μg/L	
		Antibiotic compounds	anhydro-Erythromycin: 0.33 μg/L Ofloxacin: 0.13 μg/L	

		Pharmaceutical compounds	Codeine: 0.056 μg/L
			Erythromycin: 0.18 μg/L
		Steroid and steroidal hormone compounds	Coprostanol: 14 μg/L
			Estrone: 0.11 μg/L
		Pesticide compounds	Atrazine: < 0.007 μg/L
			Deethylatrazine: < 0.006 μg/L
			Fipronil: < 0.016 μg/L
			Metolachlor: < 0.006 μg/L
			Prometon: < 0.01 μg/L
	Fourmile Creek, Iowa, USA	Acidic organic compounds	Ethylenediaminetetraacetic acid: 170 μg/L
			Nitrilotriacetic acid: 0.9 μg/L
		Neutral organic compounds	Bisphenol A: 0.010 μg/L
			Caffeine: 0.020 μg/L
		Antibiotic compounds	anhydro-Erythromycin: 0.53 μg/L
			Ofloxacin: 2.2 μg/L
		Pharmaceutical compounds	Codeine: 0.25 μg/L
			Erythromycin: < 0.05 μg/L
		Pesticide compounds	Atrazine: 0.026 μg/L
			Deethylatrazine: 0.006 μg/L
			Fipronil: 0.037 μg/L
			Metolachlor: 0.075 μg/L
			Prometon: < 0.01 μg/L
River		PFCAs	PFHxA: 0.4–4.7 ng/L

	Ganges River Basin, India	PFSAs	PFBS: < Method Quantitation Limit (MQL)–10.2 ng/L	Sharma et al. (2016)
Groundwater		PFCAs	PFBA: < MQL–9.2 ng/L	
		PFSAs	PFBS: < MQL–4.9 ng/L	
River	Northern France	PFBS	$^{13}C_2$ – PFHxA: 95 ng/L	Boiteux et al. (2017)
		PFHxS	$^{13}C_4$ – PFOA: 83 ng/L	
		PFHpS	$^{13}C_4$ – PFOA: 83 ng/L	
		PFOS	$^{13}C_4$ – PFOS: 82 ng/L	
		PFDS	$^{13}C_2$ – PFDoDA: 81 ng/L	
		PFBA	$^{13}C_4$ – PFBA: 85 ng/L	
		PFPeA	$^{13}C_2$ – PFHxA: 84 ng/L	
		PFHxA	$^{13}C_2$ – PFHxA: 74 ng/L	
		PFHpA	$^{13}C_2$ – PFHxA: 94 ng/L	
		PFOA	$^{13}C_4$ – PFOA: 81 ng/L	
		PFNA	$^{13}C_4$ – PFOA: 84 ng/L	
		PFDA	$^{13}C_2$ – PFDA: 100 ng/L	
		PFUnDA	$^{13}C_2$ – PFUnDA: 103 ng/L	
		PFDoDA	$^{13}C_2$ – PFDoDA: 93 ng/L	
		PFTrDA	$^{13}C_2$ – PFDoDA: 70 ng/L	
		PFTeDA	$^{13}C_2$ – PFDoDA: 15 ng/L	
Wastewater Treatment Plants	Italy	PFAA (effluents)	PFOA: 8-260 ng/L PFSA: < Limit Of Quantitation (LOQ)–$17_{\text{(only PFOS)}}$ ng/L short-chain PFCA: < LOQ–134 ng/L long-chain PFCA: < LOQ–207 ng/L	Castiglioni et al. (2015)

Drinking water	North of Milan (industrialized area), Italy	PFAA	PFOA: 10–47 ng/L PFSA: 1–32 ng/L short-chain PFCA: 2–44 ng/L long-chain PFCA: 2–14 ng/L	
Drinking water	Metropolitan area of Milan (urban area), Italy	PFAA	PFOA: 2–17 ng/L PFSA: 2–29 ng/L PFCA short-chain: 3–18 ng/L long-chain PFCA: < LOQ ng/L	
Drinking water	South of Milan (agricultural area), Italy	PFAA	PFOA: < LOQ ng/L PFSA: < LOQ ng/L short-chain PFCA: < LOQ ng/L long-chain PFCA: < LOQ ng/L	
Bottled Water	Brazil France Spain	PFASs	15.0 ng/L 14.9 ng/L 11.3 ng/L	Schwanz et al. (2016)
Tap water	Brazil France Spain	PFOS	15.83 ng/L 7.73 ng/L 15.33 ng/L	

Sewage effluent	Five drainage systems in Lithuanian Institute of Agriculture	Heavy metals	Cr: 0.035 mg/L Cd: 0.002 mg/L Pb: 0.003 mg/L Ni: 0.011 mg/L Cu: 0.002 mg/L Zn: 0.059 mg/L	Antanaitis and Antanaitis (2004)
Farm effluent	North Island, New Zealand	Heavy metals	Cu: 0.5–10.5 mg/L	Bolan et al. (2003)
Farm effluent	Not Available	Heavy metals	Cu: 0.26 mg/L Zn: 0.58 mg/L	Lowe (1993)
Storm water	Austin, Texas, USA	Heavy metals	Cr: 0.04 mg/L Cd: 0.04 mg/L Fe: 2.429–10.3 mg/L Pb: 0.073–1.78 mg/L Ni: 0.053 mg/L Cu: 0.022–7.033 mg/L Zn: 0.056–0.929 mg/L As: 0.058 mg/L Hg: 3.22 mg/L	Barrett et al. (1995)
Industrial effluent	Bytom, Silesian Voivodeship, Southern Poland	Heavy metals	Cd: 1.8–4.1 mg/kg Cr: 34.9–68.3 mg/kg Cu: 104.1–194.0 mg/kg	Tytła (2019)

			Ni: 55.0–98.1 mg/kg Pb: 97.6–189.2 mg/kg Zn: 1092.2–1851.6 mg/kg Hg: 0.3–1.1 mg/kg	
Industrial effluent	Nairobi, Kenya	Heavy metals	Hg: <0.1 mg/L Pb: 15.31 mg/L Cd: 8.12 mg/L Cr: 0.09 mg/L Ni: 0.05 mg/L Tl: 4.96 mg/L	Kinuthia et al. (2020)
Municipal wastewater treatment	Kentucky, USA	Perfluoroalkyl sulfonates Perfluoroalkyl carboxylates	PFOS: 7.0–149 ng/L PFOA: 22–334 ng/L	Loganathan et al. (2007)
	Georgia, USA	Perfluoroalkyl sulfonates Perfluoroalkyl carboxylates	PFOA: 1–227 ng/L PFOS: 1.8–22 ng/L	
Wastewater treatment plants	Australia	PFAS (influent)	0.98–444ng/L	Gallen et al. (2018)

Table 2. Adsorption performance of various aqueous contaminants by organic, inorganic and industrial by-products sorbents

Primary Sorbent Type	Pollutant	Adsorbent/ Sample Name	Surface Area (m^2/g)	Operating conditions (pH/ Temperature, ∘C)	Sorbent concentration (g/L)/ Initial pollutant concentration	Sorption capacity (mg/g)	Reference
				Inorganic Sorbents			
Metal Oxide	Cr^{3+}	Phosphated TiO_2	278	4/–	0.1/ 0.5 mmol/L	92	Wang et al. (2020e)
Metal Oxide	Bisphenol A Methylene Blue	α-MnO_2	110	2.7 /– 6.3 /–	0.5/100 mg/L 1/100 mg/L	86 98	Mathew and Saravanakumar (2021)
Metal Oxide	Methyl orange Tetracycline hydrochloride	Co/Cr-co-doped ZnO	75	7/25	0.3/800 mg/L	1058 874	Li et al. (2018)
Magnetic Metal Oxide	Catechol Tetracycline hydrochloride	Iron (III) oxide	13	8/–	10/0.3 M	361 86	Abugazleh et al. (2020)
Magnetic Metal Oxide	Naphthalene	Kaolin/Fe_3O_4	157	6.5/–	4.8/10 mg/L	97%	Arizavi et al. (2020)
LDH	Pb^{2+}	MgAl-LDH/biochar	405	7/25	0.2/500 ppm	591	

	CrO^{4-}			2/25	0.2/300 ppm	331	Wang et al. (2020c)
LDH	Perfluorooctanoic acid (PFOA)	Al-Mg-Cl	37	–/25	2.5/20 mg/L	90%	Ahmed et al. (2020)
Silica	Methylene blue dye	Spherical mesoporous silica	348	0.5/RT	1/83 mg/L	61	de Paula et al. (2021)
Silica	Ru	TRPO/SiO_2–P	58	–/25	20/1059.8 mg/L	55	Zhang et al. (2020g)
Silica	Acetaminophen	Silica microspheres	105	5/30	0.1%/100 ml of 20 ppm	89	Natarajan et al. (2021)
Clays	Malachite green dye	Chitosan–halloysite nanotubes	–	–/30	2.5/750 mg/L	238	Peng et al. (2015)
Clays	Cu^{2+} Ni^{2+}	Bentonite/Graphene Oxide	63	6/25	0.5/100 mg/L	98% 82%	Chang et al. (2020)
Zeolite	Ammonium	Synthetic zeolite (ZFA)	13	7/25	10/100 mg/L	32	Tang et al. (2020)
Zeolite	Phenol 2-chlorophenol 2-nitrophenol	Fe-nano zeolite	981	–/RT	2.5/500 mg/L	139 159 171	Tri et al. (2020)

MOF	Amoxicillin	MIL-53(Al)	1288	7.5/30	0.1/150 mg/L	~505	Imanipoor et al. (2021)
MOF	Hg^{2+} Pb^{2+}	MOF-MA	296	4/25	0.5/800 mg/L	1080 510	Wang et al. (2020a)
Mxene	Ba^{2+} Sr^{2+}	$Ti_3C_2T_x$	10	–	1/2000 mg/L	180 225	Jun et al. (2020)
Mxene	Phosphate Nitrate	$Ti_3C_2T_x$	11	6/30	2/100 mg/L	89 71	Karthikeyan et al. (2021)
			Organic Sorbents				
Activated Carbon	PFOA	HMB900	1322	–/25	0.2/100 mg/L	1269	Zhou et al. (2021)
Activated carbon	Chromium Zinc Lead Phenol	SAS	796	4/25	1/50 mg/L 1/50 mg/L 1/50 mg/L 1/110 mg/L	9 4 0.4 10	Elboughdiri et al. (2021)
Activated Carbon	4-chlorophenol	Activated carbon from pomegranate husk (MPHAC)	1168	6/–	2/50 mg/L	98.4%	Hadi et al. (2021)
Activated Carbon	Congo red Tetracycline	Ni-Co-S-Activated carbon (AC-NCS)	74	7/RT 7/RT	0.33/20 mg/L 0.33/10 mg/L	57 25	Chowdhury et al. (2021)

	Ciprofloxacin			4.5/RT	0.33/10 mg/L	24	
Biochar	Zn^{2+} Pb^{2+}	Jujube seeds biochar	48	5/30	2.2/50–500 mg/L 3.4/50–500 mg/L	221 119	Gayathri et al. (2021)
Biochar	Tetracycline	Zn-loaded biochar	11	6/25	0.2/50 mg/L	159	Wang et al. (2021b)
Hydrochar	Methylene blue Cd^{2+}	CFHC (bamboo powder as source)	28	–/30	0.8/1000 mg/L 0.8/90 mg/L	1155 91	Li et al. (2019)
Cellulose	Fluoride	Cellulose−CeO_2	–	3/25	35/100 mg/L	48	Yao et al. (2021)
Chitosan	Au^{3+}	CS-GTU	–	5/30	0.66/1000 mg/L	696	Zhao et al. (2021)
Ordered carbon	Uranium	P− Fe−CMK-3	187	4/25	0.2/20 mg/L	150	Husnain et al. (2017)
Ordered carbon	p-toluic acid Benzoic acid Terephthalic	CMK-1/PDDA	658	–/25	0.2/100 mg/L	141 166 164	Anbia and Salehi (2012)

Graphene	As^{5+}	Alginate coated-Fe-Al-LDH/reduced graphene oxide	151	7/RT	0.07/100 mg/L	191	Priya et al. (2021)
Graphene	In (III)	Silica gel/graphene oxide	177	2.5/25	0.5/100 mg/L	147	Li et al. (2021a)
CNT	Phenol p-cresol	Diatomite−Carbon Nanotube	50	7/25	2/50 mg/L	8 17	Wang et al. (2019a)
CNT	Ni^{2+} Zn^{2+} As^{3+} Co^{2+}	PAMAM/CNT	–	7/25	0.03/30 mg/L	3900 3650 3500 3800	Hayati et al. (2016)
Polymers	Imidacloprid insecticide	Polypyrrole/peanut husk Polyaniline/peanut husk	–	3/–	0.2/25 mg/L	~7 ~9	Ishtiaq et al. (2020)
Polymers	Congo red Crystal violet Rhodamine B dyes	Polyaniline@TiO_2	–	6.8/28	0.5/1000 mg/L	93 80 94	Maruthapandi et al. (2020)

Ion exchange resin	$Au3^{+}$	Crosslinked polyethyleneimine resin	–	2/–	0.2/300 mg/L	944	Liu et al. (2020a)
Ion exchange resin	Acid fuchsin Acid orange G Acid blue 80	StAM–Arg (guanidine-containing starch-based resin)	–	3/–	5/ 0.25 mmol/L	~28 ~25 ~33	Zhang et al. (2020b)
			Industrial by-products				
Red mud	PFOS	Red mud modified sawdust	121	3.1/25	0.57/248.48 mg/L	195	Hassan et al. (2020a)
Red mud	Diclofenac Phosphorus	Redmud/polypyrrole	102	5/25	0.1/10 mg/L	195 31	Li et al. (2020b)
Fly ash	Cl−	Alkali-combined roasting-modified fly ash hydrotalcite	20	8/60	10/10,000 mg/L	68.1%	Qi et al. (2020)
Fly ash	Phosphate	BTP-FA CTP-FA	–	6/–	8/10–1,000 mg/L	62 4	Park et al. (2021)
Sludge	Acetamiprid Thiamethoxam Imidacloprid (pesticides)	AC-$ZnCl_2$ (Activated carbon with $ZnCl_2$ activation)	558	–/25	1.5/50 mg/L	129 127 166	Sanz-Santos et al. (2021)

Sludge	Orthophosphorus Condensed phosphorus	Alum sludge	39	4/25	12/25 mg/L 12/15 mg/L	5 4	Maqbool et al. (2016)

LDH - Layered Double Hydroxide, **MOF** - Metal organic framework, **CNT** - Carbon nanotube, **PFOA** - Perfluorooctanoic acid, **PFOS** - Perfluorooctanesulfonic acid, **TRPO/SiO$_2$-P** - Silica-polymer based adsorbent, **ZFA** - Zeolite from fly ash, **MIL-53(Al)** - (MIL, Materials of Institute Lavoisier) or aluminum 1,4-benzenedicarboxylate or $\{Al(OH)[O_2C{-}C_6H_4{-}CO_2]\}$, **MOF-MA** - mercaptosuccinic anchored metal organic framework, **HMB900** - hierarchically microporous biochar, **SAS** – Steam activated sawdust, MPHAC - magnetized activated carbon pomegranate husk, **AC-NCS** - activated carbon loaded with **Ni-Co-S** nanoparticles, **CFHC** - carboxylate-functionalized hydrochar, **CS-GTU** - chitosan-based adsorbent from guanylthiourea, **P− Fe−CMK-3** – P, Fe doped ordered mesoporous carbon from mesoporous silica **SBA-15, CMK-1/PDDA** – ordered mesoporous carbon from mesoporous silica **MCM-48 -** modified with polydiallyldimethylammonium chloride, **PAMAM/CNT**-Poly(amidoamine)/carbon ananotube, **StAM-Arg** – Corn starch modified with polyacrylamide and arginine, **BTP-FA** – Fly ash from biothermal power plant, **CTP-FA** – Fly ash from coal thermal power plant, **AC-ZnCl$_2$** – Activated carbon with $ZnCl_2$ activation

Table 3. Selected references for recovery, regeneration, and further application of spent sorbents

Spent sorbent	Application	Separation and regeneration technique	Regeneration condition	Highlights of the study	Reference
Fe_3O_4-graphene-biochar composite	Crystal violet (CV)	Recovery via magnetic separation and regeneration via chemical treatment	30 °C for 2 h	The CV absorbability of the recovered composite was 157.31 mg g^{-1}, which was slightly lower than pristine (199 mg g^{-1}). These findings highlighted the recovery and reusability of the spent sorbent.	Du et al. (2020)
Biochar/iron oxide	MB	Chemical treatment	Drying at 80 °C	The biochar/iron oxide composite exhibited a minor reduction in adsorption efficiency after five cycles, but the efficiency remained within an acceptable limit thoroughly.	Zhang et al. (2020e)
Biochar	Removal of SMX and methyl paraben (MP)	Advance oxidation process	H_2O_2:Fe ratio 29:0.29 mM	This investigation highlighted the important of the electro-Fenton process in the elimination of the contaminants its recycling, regeneration, and reuse.	Acevedo-García et al. (2020)
Pit biochar	Removal of Pb^{2+}	Chemical treatment	–	This investigation releveled that the removal efficiency was around 70% of the initial adsorption capacity after the last round therefore, this process can minimize the working cost associated with adsorption process.	Gao et al. (2020)
CaO-based adsorbent	Adsorption of CO_2	Thermo-chemical treatment, water washing and vacuum filtration	105 °C for 24 h	The sorbent regeneration/reactivation via water washing displays an improved capacity of 0.390 g g^{-1} after 40 cycles. Furthermore, NaCl impregnation combined with water washing also enhance CO_2 capture stability. The use of filtration during	Sun et al. (2020)

				acidification reactivation procedure can efficiently improve the initial CO_2 capture potency. .	
Multi walled carbon nano tubes (MWCNTs)	Removal of diquat dibromide (DQ)	Chemical treatment	–	OMWCNT can be recycled at least five times without significantly decreasing the adsorption and desorption efficiency.	Duman et al. (2019)
Activated carbon (ACs)	Removal Toluene	Thermal and KOH activation	750 °C for 1 h, and 850 °C for 3 h	The SSA of spent AC was 680 m^2 g^{-1}, and increased up to 710 m^2 g^{-1} via heating. When the spent AC was activated by the chemical agent KOH, the SSA increased to 1380 m^2 g^{-1}. The toluene adsorption capacity of regenerated ACs (0.154 g g^{-1}) was more than commercial ACs (0.142 g g^{-1}).	Park et al. (2019)
Peat-based adsorbent	Removal of heavy metals	Hydrothermal carbonization (HTC)	230 °C for 3 h	HTC was futile in desorbing an adequate quantity of metaloidss from spent sorbents to synthesize a clean hydrochar that could be applied as a soil amendment without environmental jeopardies. The leaching of As, Cu, and Zn from hydrochars was improved remarkably in comparison to the spent sorbents, therefore the hydrochars would not be appropriate for landfilling without pre-treatment.	Kasiuliene et al. (2019)
Graphite Intercalation Compound (GIC)	Emulsified oil	Electrochemical	–	The adsorptive capacity of the GIC was 100% recoverable by electrochemical regeneration. Energy consumption for the adsorbent regeneration process was found to be 22 kWh kg^{-1} of COD removed for treatment of the synthetic emulsion and 36 kWh kg^{-1} of COD for produced water.	Fallah and Roberts, (2019)

Magnetic AC	Removal of perfluorooctane sulfonate (PFOS)	Recovery via magnetic separation and regeneration via methanol-wash	Shaking for 12 h and drying at 60 °C	The regenerated MAC could be reused for more than 5 time and remain stable adsorption capacity after 3 cycles.	Meng et al. (2019)
MgAl-LDH/Biochar composites	MB	Recovery via filtration and regeneration via chemical treatment	Shaking for 2 h and drying at 60 °C for 2 h	After 6 cycles the capacity of removal of the composite decreased from 65-70 mg g^{-1} to 40-45 mg g^{-1}. The presence of the biochar favored the stability of the adsorptive capacity.	Meili et al. (2019)
Metal azolate framework-6 (MAF-6)	Removal of PPCPs	Solvent washing (ethanol and water)	–	An insignificant reduction in sorption capacity toward ibuprofen (IBP) over five rounds of recycling, except for a slight reduction after the first round. Notably, the sorption capacity of CDM6-k1000 on IBP in the fifth cycle was still about twice that of fresh AC.	An et al. (2018)
MWCNTs	Removal of AAP	Filtration, thermal treatment, ultrasonication and water washing	100 °C for 8 h	Successive reductions in sorption efficiency from 95% (1^{st} cycle of regeneration) to 25% (4^{th} cycle of regeneration).	Yanyan et al. (2018)
Bio-metalorganic framework -derived carbons (BMDCs)	Removal of PPCPs	Solvent washing (washed with deionized water, and soaked in acetone)	25 °C for 12 h	The reusability of spent sorbent for atenolol removal did not reduce noticeably with an increase in the number of cycles up to the fourth run. More importantly, the performance after the fourth run was still around 10 times higher that the fresh AC.	Bhadra et al. (2018)
Bio-adsorbent	Removal of Pb^{2+} and Hg^{2+}	Chemical treatment	–	The adsorbent exhibited good stability, its regeneration being made possible by the use of EDTA-Na_2 solution (0.05M) as a regenerative agent	Ifthikar et al. (2018)

				without significant biochar alteration after five regeneration cycles, keeping a similar adsorption capacity.	
MWCNTs	Removal of pharmaceutical and personal care products (PPCPs)	Thermal treatment	380 °C	The adsorption capacities of the regenerated MWCNTs were 3.59–3.73 mg g^{-1} during the reuse cycles, resulting in 89.8–93.3% removal of triclosan (TCS) from the feedwater, which was greater than 72.0% removal obtained by the pristine MWCNT. Similarly, the adsorption capacities of MWCNT for ibuprofen (IBU) and acetaminophen (AAP) were 2.59–3.09 mg g^{-1} and 3.19–3.83 mg g^{-1} in the reuse cycles, respectively, which corresponded to 87.1–93.2% and 64.8–77.3% removal of the two compounds, respectively.	Wang et al. (2017c)
MWCNTs	Removal of PPCPs	Sonication	15–60 min sonication duration	The adsorption capacity of the MWCNT for AAP reached 1.99 mg g^{-1} after regeneration or 96% of that of the pristine MWCNT sample. The ratios of recovery were 95% for IBU and 95% for TCS, respectively.	Wang et al. (2017b)
Zeolitic material synthesized from coal fly ash (Ze–Na and Ze–K)	Recovery of ammonium	Alkaline regeneration, and water washing	25 °C for 4 h	In the case of Ze–Na, the maximum sorbent capacity was obtained during the first sorption cycle whereas in the case of Ze–K, it was obtained during the last working cycle due to the alkaline regeneration. Moreover, after the last sorption-desorption working cycle, loaded zeolites can be used as fertilizer after a separation process by filtration	You et al. (2017)

Granular activated carbon (GAC)	Removal Chloramphenicol	Microwave and ultraviolet irradiation	2450 MHz for 10 min	The mineralization percentage of chloramphenicol amplified to 37% from 5% when adds the electrodeless lamp into the regeneration reactor. Besides, 83% of the total chloride in chloramphenicol can transmute into inorganic chloride. Add ultraviolet radiation in microwave regeneration reactor can improve the oxidizability of microwave regeneration process. Moreover, the adsorption capability of GAC can uphold at a high level after five absorption/regeneration cycles.	Sun et al. (2017)
AC	Adsorption of Cu^{2+}	Filtration and acid treatment	60 °C for 1 h	Use of 6 M HCl resulted in only 13.3% loss of adsorption potency after 10 consecutive adsorption desorption cycles. The maximum loss in adsorption potency of happened after the first cycle but material performance was almost steady.	Da'na and Awad, (2017)
Zeolite	Recovery of ammonium	Chemical treatment	–	The ammonia (NH_3) recovery ratio exceeded 98% and the spent NH_3/NaOH streams once NH_3 is eliminated can be re-used for regeneration of the ammonium exhausted zeolites filters.	Sancho et al. (2017)
Granular CNTs	Removal of pharmaceuticals	Thermal treatment	400 °C	The spent granular CNTs were effectively regenerated without reducing the adsorption potency in five regeneration cycles. The adsorbed carbamazepine (CBZ) and diclofenac sodium (DS) were totally mineralized, while the adsorbed tetracycline (TC) was moderately oxidized and the	Shan et al. (2016)

				residual was advantageous for the successive adsorption.	
$CNT/CoFe_2O_4$ composites	Removal of sulfamethoxazole (SMX) and 17β-estradiol	Magnetic separation and thermal treatment	300 °C	Adsorption efficiency slight reduced over five cycles while the weight of the composite reduced remarkably after the first cycle.	Wang et al. (2015a)
$CNTs/Fe_3O_4$ nanocomposites	Removal of bisphenol A (BPA)	Recovery via magnetic separation and regeneration via methanol-wash and chemical oxidation	75 °C for 12 h	The recyclable $CNTs/Fe_3O_4$ nanocomposites can uphold a high recovery extent (~98%) via magnetic separation and hold their adsorption performance after several adsorption–deactivation–regeneration cycles.	Li et al. (2015)
Magnesium hydroxide-coated pyrolytic bio-char	Removal of anionic dye	Microwave irradiation	At 320, 480, 640 W for 5 min	Spent magnesium hydroxide-coated pyrolytic bio-char was treated by microwave irradiation, and yield of regeneration was 98.5%, 89.0%, 85.5% in the case of microwave irradiated time 5 min at 320W, 480W, and 640W respectively.	Zhang et al. (2014)
Durian shell and jackfruit peel ACs	Removal Methylene blue dye (MB)	Microwave treatment	Operated at 2.45 GHz and irradiation time of 3 and 4 min	The adsorption uptake and carbon yield of the regenerated activated carbons could maintain at 181.43–207.57 mg g^{-1} and 80.51–81.63%, even after five adsorption–regeneration cycles. Microwave treatment preserved the porous structure of the spent ACs efficiently to restore the original active sites and adsorption capacity.	Foo and Hameed, (2012)

Table 4. Desorption studies of eluents by different adsorbents

Solvent	Adsorbent	Pollutant	Desorption Efficiency (%), cycles	Reference
0.1M HNO_3	Biochar	Co^{2+}	76%, 3 cycles	Kołodyńska et al. (2017)
0.01M EDTA	Magnetic chitosan-palygorskite	Pb^{2+}	70%, 4 cycles	Rusmin et al. (2022)
H_2SO_4	Silica based hybrid	Ni^{2+}	26%, 1 cycle	Xu et al. (2016)
methanol	Activated carbon-chtosan	methylparaben	96%, 5 cycles	Mashile et al. (2020)
0.1M HCl	polysiloxane-graphene oxide gel	Pb^{2+}	99%, 5 cycles	Zhou et al. (2015)
Ethanol	Nano zeolite	Ortho-nitrophenols	72.8%, 5 cycles	Pham et al. (2016)
0.1M HCl	Gallic acid-conjugated iron oxide nanocomposite	Al^{3+}	85%, 5 cycles	Guan et al. (2017)
0.08M HCl	Activated sewage sludge	Cd^{2+}	100%	Zhai et al. (2004)
$NaNO_3$	$C@MnO_2$	Pb^{2+}	81.47%, 5 cycles	Li et al. (2021b)
0.05M NaOH	Polyaniline coated onto wood sawdust	methyl orange	45%	Ansari et al. (2011)
0.1M HCl	Mg-Fe-LDH	Cu^{2+}	84%, 5 cycles	Awes et al. (2021)
NaOH	Halloysite/biochar	Pb^{2+}	95.61%, 1 cycle	Wang et al. (2021a)
Acetone	$CNT/ZnCo_2O_4$-DES	eosin Y dye	65%, 5 cycles	Lawal et al. (2019)
0.5 mol L^{-1} NaOH	Zeolitic bagasse fly ash	BFA	91.98%	Shah et al. (2021)