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Agricultural waste materials for adsorptive removal of phenols, chromium (VI) and cadmium (II) from wastewater: A review

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ABSTRACT

Management of basic natural resources and the spent industrial and domestic streams to provide a sustainable safe environment for healthy living is a magnum challenge to scientists and environmentalists. The present remedial approach to the wastewater focuses on recovering pure water for reuse and converting the contaminants into a solid matrix for permanent land disposal. However, the ground water aquifers, over a long period slowly leach the contaminants consequently polluting the ground water. Synthetic adsorbents, mainly consisting of polymeric resins, chelating agents, etc. are efficient and have high specificity, but ultimate disposal is a challenge as most of these materials are non-biodegradable. In this context, it is felt appropriate to review the utility of adsorbents based on natural green materials such as agricultural waste and restricted to few model contaminants: phenols, and heavy metals chromium(VI), and cadmium(II) in view of the vast amount of literature available. The article discusses the features of the agricultural waste material-based adsorbents including the mechanism. It is inferred that agricultural waste materials are some of the common renewable sources available across the globe and can be used as sustainable adsorbents. A discussion on challenges for industrial scale implementation and integration with advanced technologies like magnetic-based approaches and nanotechnology to improve the removal efficiency is included for future prospects.

1. Introduction

Environmental pollution is a challenge particularly when industrial effluents are present in water bodies. The majority of waste from industries is easily transported into wastewaters, and they contain organic and inorganic contaminants. Organic species including phenol and metal ions like chromium(VI), cadmium(II), etc. are toxic to humans either being carcinogenic or affecting the physical-motor functioning of the body. Phenol is discharged by many industries including refineries and organic chemicals manufacturing as it is used as a precursor and an intermediate (Mohammadi et al., 2015). Chromium(VI) is contained in the industrial discharges of dyes and tanneries and is carcinogenic (Rosales et al., 2016). Cadmium(II), along with its compounds, can be found in the industrial discharges related to pigments, mining, plastics, corrosion resistance steel, metal plating, phosphate fertilizer, alloy industries, and the battery industry. Cadmium(II) is highly toxic and suspected to be carcinogenic (Jayakumar et al., 2021; Rahimzadeh et al., 2017).

Word health organization (WHO) sets global norms for potable water quality that provide guidance to various regulatory bodies for establishing standards. The guidelines primarily aim at public health protection in relation to drinking water safety. The permissible phenolic,

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chromium and cadmium(II) concentrations in potable water according to World Health Organization (WHO) guidelines are 0.001 mg/L, 0.05 mg/L, and 0.003 mg/L respectively (Aghav et al., 2011; Faghih Nasiri et al., 2018; Rao et al., 2010; Rosales et al., 2016; WHO, 2017). The WHO guideline value for chromium was initially proposed in 1958 for hexavalent chromium from health perspectives but was later modified to a guideline for total chromium (WHO, 2017). Due to their high toxicity and non-biodegradability characteristics, even minimal discharge of such hazardous contaminants can cause harm to soil, surface waters, aquatic life, humans and environment (John et al., 2018). Over the last two decades, several techniques for the removal of organic and inorganic species have been studied including chemical oxidation (O'Connor et al., 2018; Othmani et al., 2019a); electrocoagulation (Doggaz et al., 2019; Othmani et al., 2017), membrane separation (Poonguzhali et al., 2021), anodic oxidation (Garcia-Segura et al., 2015), sonophotocatalytic process (Theerthagiri et al., 2021), sonoelectrochemical process (Theerthagiri et al., 2020) and coupling anodic oxidation with biosorption (Othmani et al., 2020a, 2020b, 2020b) apart from established techniques such as chemical precipitation, ion exchange, adsorption, etc. All the techniques have certain challenges either in terms of economics or ultimate safe disposal. The metal ions cannot be transmuted to harmless species making it imperative to reduce it to a minuscule volume either for ultimate disposal or for recovery and reuse. However, it is necessary to remove the toxic contaminants to ensure environmental safety (Abdelwahab and Amin, 2013). Several methods have been proposed to resolve environmental issues caused by untreated industrial streams. However, challenges like design, treatment time, fouling, energy consumption and cost still need to be considered for potential scale up (Bhatnagar et al., 2015; De Gisi et al., 2016). Adsorption processes are efficient in the removal of many pollutants from wastewater (Karimi et al., 2019; Rathi and Kumar, 2021a). Commercially, activated carbon is the main adsorbent used in water treatment due to its higher porosity and textural properties. It has also been well documented for the removal of various organic and inorganic species including; phenols, toxic metal ions, pesticides, detergents, besides color, undesirable taste, and odor (Mu'azu et al., 2017). Generally, the adsorption capacity of adsorbents depends on several characteristics, like their textural properties including porosity, surface area, pore size, and surface chemistry that play a crucial role in adsorption performance (Crini et al., 2018). The adsorption capacity is influenced by various parameters including the pH of the solution, the ionic strength, the temperature, the initial concentration of the adsorbates used, and the contact time (El-Aila et al., 2016). Although activated carbon offers higher efficiency, economic viability still needs to be considered. Apart from the high costs of activated carbon, reactivation is cost-intensive and cumbersome. Further, the disposal of contaminants released during deactivation without entering the geo-hydrologic cycle is difficult and challenging.

Since adsorption is technically the best for the removal of trace contaminants, it would be advantageous to look for suitable adsorbents that would be effective, sustainable, cost-effective, and allow easy isolation of the species. Natural products are known to exhibit adsorption of organic and inorganic species (Singh et al., 2018). One of the ways to overcome the existing challenges is to use green materials like an agricultural waste for contaminant removal. Recent studies reveal the focus on the exploitation of agricultural waste materials in wastewater decontamination (Ali Redha, 2020). These materials are natural, inexpensive, widely available, and present promising adsorption efficiencies comparable to commercial activated carbon. Biodegradability, non-toxic nature, and eco-friendliness of these adsorbents are the additional advantages.

A systematic methodology is adopted to collect, comprehend and present the recent literature on the application of green materials originating from agricultural waste for the adsorptive removal of identified contaminants. Research databases including Scopus and google scholar are searched using keywords such as adsorption, decontamination, removal, remediation, wastewater treatment, water purification, adsorbents, green, natural, agricultural waste, phenols, heavy metals, chromium(VI), and cadmium(II) (GracePavithra et al., 2019; Jeevanantham et al., 2019). The literature is included without any bias of researcher, journal, country, etc. to maintain neutrality. The information from the literature has been critically reviewed and discussed under various sections. The structure of the article is as follows. The article initially reviews the harmful effects of the priority contaminants, phenols, hexavalent chromium(VI), and cadmium(II) concerning biota and the environment. Detailed discussions on the characteristics of various agricultural waste materials (AWMs) as adsorbents for sustainable removal of phenol, cadmium(II), and hexavalent chromium in the last few years are presented to indicate that the biosorption to be an effective alternative to conventional pollutant removal techniques from wastewaters. A critical analysis of the challenges to overcome before translating the technology to the field is also provided.

2. Effects of contaminants on biota and environment

Environmental problems generated from water contamination have become a major concern for society, industry, and public authorities (Rasheed et al., 2019; Rathi and Kumar, 2021b). Domestic, agricultural and industrial activities produce wastewaters containing undesirable toxic contaminants including heavy metals, hazardous compounds, and carcinogenic products (Ponnuchamy et al., 2021). In this context, a strong effort must be made to protect valuable water resources. Phenol, hexavalent chromium, and cadmium(II) are placed on the priority list of hazardous pollutants due to their higher toxicity and harmful effects on the flora and fauna (Ahmed Basha et al., 2008; Anyanwu et al., 2018). The environmental and health effects of phenols, chromium(VI) and cadmium(II) along with the major sources of these contaminants are presented here.

2.1. Effect of phenols

Phenols and their derivatives are extensively used in various chemical and allied industries dealing with production and processing of antioxidants, synthesis intermediates, disinfectants, tanning agents, photographic developers, personal care products and additives for lubricants. The wastewater streams emanating from petrorefineries, coking operations, and coal gasification units (H. C. Li et al., 2017) are some of the common sources of phenolic pollution. The discharge from other industries such as textiles, fertilizers, paints, pharmaceuticals, and wood processing also contribute to the contamination of water sources with phenols (Muthu Kumara Pandian et al., 2021; Ponnuchamy et al., 2020; Villegas et al., 2016). Based on the type of pollutant source, the phenolic concentration in the effluent streams can reach up to hundreds or thousands parts per million (Alshabib and Onaizi, 2019). The domestic sewage and surface water runoffs from agro-based activities also allow phenols to find pathways to enter into the ecosystem (Muthamilselvi et al., 2018; Wang et al., 2020). Furthermore, phenols are naturally found in tyrosine, one of the standard amino acids found in epinephrine (adrenaline), proteins and can be also obtained from essential oils of plants (Ribas-Agustí et al., 2018). Even though phenols and their derivatives are extensively used, they are known to be toxic, since they threaten human health and cause many side effects leading to poisoning and death. Generally, they are easily absorbed through the respiratory tract, by inhalation, and through the digestive skin causing liver and kidney damage and central nervous system disruption (Gutiérrez-Grijalva et al., 2016). In the case of eye contact, serious injuries can occur and lead to blindness and when heated, the thermal decomposition of phenols releases toxic vapors, a major source of the fire. Therefore, phenols should be stored in a cooled and well-ventilated area away from light and far from any source of ignition and any strong oxidants (Shahidi and Ambigaipalan, 2015). Compared to humans, phenols are very dangerous for animals and are responsible for many clinical signs (e.g. slow breathing, change in body temperature, shortness of breath, tremors, seizures, lethargy, and coma). After inhalation, animals have shown irritation of the nose and into eyes and a slight loss of coordination with muscle spasms and tremors has been observed. In the case of a high oral dose, the animals die within 5–150 min. Because of their good water solubility and mobility, phenols are likely to reach downstream drinking water and can cause enormous risk for populations even at low concentrations. Moreover, phenols are reported to have negative effects on various biological processes. In the presence of chlorine, phenol forms chlorophenols which are easily absorbed from the gastrointestinal tract causing acute toxicity. The toxicity increases with the degree of chlorination which could generate compounds of chlorophenols, mutagens, and carcinogens (Oluwasanu, 2018).

2.2. *Effect of chromium(VI)*

Chromium is a hazardous contaminant found in water originating from natural as well as anthropogenic sources. Chromium compounds are generally oxidized into the corresponding chromic form and chromium is found in two forms: Cr(III), the trivalent form in chromites, and Cr(VI), the hexavalent form, in chromates and dichromates. Chromium salts have varying degree of solubility in water, except hydroxide and carbonate for trivalent chromium. In solution, the salts are completely dissociated to form ionic compounds mainly Cr^{3+}, CrO_4^{2-}, and $Cr_2O_7^{2-}$ (Senthil Kumar et al., 2018). Compared to the hexavalent form, the trivalent form is recognized to encounter less danger for the water bodies, aquatic fauna, and flora. In contrast, the hexavalent form of chromium has been considered very toxic and legislations have generally imposed severe limit values for waste rejection. The hexavalent form of chromium is extremely toxic due to its high oxidation potential and high solubility. The natural occurrence of chromium(VI) is typically in the form of chromite in ultramafic rocks or in the complexed form along with other metals (Oliveira, 2012). The microbial interactions with rocks accompanied by geogenic processes like leaching allow chromium(VI) to be released into the environment (Suganya and Senthil Kumar, 2018; Tumolo et al., 2020). Chromium(VI) is a very common pollutant in the effluents of several types of industries, such as the electroplating, textiles, cement, pulp and paper processing units generating a large volume of discharge streams containing toxic heavy metals (GracePavithra et al., 2019; Liknaw et al., 2017; Mitra et al., 2017). The tanning process is one of the biggest sources of chromium(VI) pollution globally. The hexa chrome laden mines wastewater also poses a serious threat to the environment (Sinha et al., 2017).

Due to the wide range of applications in different industries, large quantities of Cr(VI) are released into the environment posing a greater risk for the environment and human health (Sun et al., 2015). Chromium (VI) has been identified as a carcinogen through the route of inhalation, and stomach cancers have been linked to ingesting water contaminated with chromium(VI). Its presence in the aqueous environment poses toxic effects to human beings and aquatic organisms. It can also cause dermatitis, liver, nerve tissue, and circulatory damage (Mishra and Bharagava, 2016).

2.3. *Effect of cadmium(II)*

Cadmium(II) is one of the abundant natural metals, which is massively used in the production of colorants, neutron absorbers, alloys, solders, as well as nickel-cadmium batteries, fluorescent paints, and fireworks (Martelli et al., 2006). Cd(II) is present in ores of species such as lead, copper, and zinc and is involved in processing and extraction procedures. Mineral processing is known to emit an enormous amount of cadmium(II) into the hydrosphere, atmosphere as well as soil leading to severe environmental contamination. The sources of cadmium(II) pollution also include wastewater discharges from industries dealing with corrosive reagents, stabilizers and fertilizers (Ayub et al., 2019). The presence of cadmium(II) as an environmental contaminant is noted in non-ferrous metal smelting and electronic waste recycling (Awasthi et al., 2016; Genchi et al., 2020).

Cadmium(II) possesses a relatively higher degree of mobility in the environment in comparison to other heavy metallic species (Kubier et al., 2019; Rocco et al., 2018). Consequently, the activity of cadmium (II) in the water-soil-plant system is enhanced. The high rate of translocation of cadmium(II) from soil to plant makes it very likely to enter the food chain (Satarug, 2018). Staple foods such as rice and wheat along with vegetables form prominent sources of dietary intake of cadmium(II). The exposure of cadmium(II) to the biota is subsequently followed by retention and accumulation considering its extensive long life ranging up to 30 years (Buha et al., 2019; Kiruba et al., 2014). The detrimental health effects of toxic cadmium(II) have been observed on endocrine, pancreatic, cardiovascular and reproductive systems (Andjelkovic et al., 2019; de Angelis et al., 2017). Cadmium(II) has been linked to the induction of oxidative stress and immunosuppression in kidneys (Zhang et al., 2017). Cadmium(II) and compounds containing cadmium(II) are designated to be cancer-causing in nature. An accumulation of cadmium(II) content in tissues has been reported to be associated with carcinogenicity in pancreas (Djordjevic et al., 2019). Further, cadmium(II) exposure also adversely impacts the skeletal system. Environmental cadmium(II) exposure is likely to play a substantial role in posing health risks related to osteoporosis and low bone mineral density (Ma et al., 2021). Effective environmental remediation techniques are necessary to prevent acute and chronic health effects caused by cadmium(II).

Table 1 presents some effects of these hazardous pollutants on biota and the environment. These contaminants have very dangerous effects on human health due to their high toxicity even at low concentrations and the improper disposal of chemicals, animal wastes, human wastes and industrial byproducts can contaminate the drinking water sources.

3. Current technologies for the removal of contaminants

A wide variety of removal approaches are reported in the literature for pollutant removal from the environment. Electrochemical treatments (Doggaz et al., 2019), ion exchange (Gaikwad et al., 2010), adsorption (Ortiz-Martínez et al., 2018), membrane filtration (Kim et al., 2018), ultrafiltration (Yaqub and Lee, 2019), reverse osmosis (Egea-Corbacho Lopera et al., 2019), chemical and electrochemical precipitation (Grace Pavithra et al., 2017; Oncel et al., 2013; Rasalingam et al., 2014), coagulation-flocculation (Tang et al., 2016), advanced oxidation processes (Garrido-Cardenas et al., 2020; Ghime and Ghosh, 2020), solvent extraction (Hu et al., 2005), photocatalytic and sonolytic oxidation (Dehghan et al., 2019; Farhadi et al., 2020; Theerthagiri et al., 2018) are considered among the most useful methods for the removal of hazardous pollutants. However, many of these technologies present

Table 1
Effects of phenols, Cr(VI), and Cd(II) on biota and environment.

Contaminants	Effects on human health	Reference
Phenols	It causes many chemical burns on contact. Long-term exposure to phenol may definitively cause cancer and many harmful effects like the damage to the nervous system, protoplasmic poisoning, and diarrhoea.	(Aghav et al., 2011; Gohiya and Dwivedi, 2016; Parikh, 2015)
Cr(VI)	Cr(VI) form is found to be 500 times more toxic than Cr(III) responsible for severe diseases in humans including lung cancer, kidney and liver damage.	Tumolo et al. (2020)
Cd(II)	Cadmium(II) has many hazardous effects such as the loss of appetite, the lung fibrosis. It causes also cancer, kidney damage, bronchitis, fibrosis, lumbago and dyspnea.	Cobbina et al. (2015)

some drawbacks including the formation of metal oxides, generation of sludge, high cost, partial removal of some ions, and a large amount of sludge production. These technologies pose certain limitations in meeting industrial requirements since they can generate by-products more toxic than the parent compound which made them critical for cost-effective treatment methods (Kehrein et al., 2020).

Several techniques have been developed for the removal of contaminants from wastewater particularly, phenol, chromium(VI) and cadmium(II). They can be broadly classified as physical, physico-chemical, and chemical processes. Conventionally, processes such as adsorption, coagulation-flocculation and ion exchange processes are predominantly deployed. These processes are primarily equilibrium controlled requiring large foot print. Chemical precipitation and coagulation-flocculation produce large quantities of sludge, which may lead to secondary pollution when disposed as a landfill. Ion exchange process requires regeneration very often and the regenerated stream would contain the offending contaminants. Adsorption with conventional adsorbents also faces challenges with reference to safe disposal as the adsorbents are not incinerable or biodegradable. Bioprocesses involve sorption due to physical adsorption or ion exchange or chelation depending on the functional group present in the biomaterial. Developing processes are mostly physico-chemical in nature and are energy intensive and include electrochemical precipitation, chemical oxidation, Fenton and Fenton-like treatment and membrane processes. Membrane processes such as reverse osmosis (RO) and nano-filtration (NF) are essentially pressure driven and are not amenable to complete separation. Size enhanced ultrafiltration (SEUF) and membrane based solvent extraction processes are under development. Table 2 collects the main methods used for the removal of contaminants from environmental wastewaters and discusses their relative merits and demerits. According to the literature, biosorption remains a low-cost alternative compared to other technologies including activated carbon-based adsorption technology. Generally, the use of natural materials offers high performance and represents a promising approach to pollutant removal. Natural materials are abundant and can be obtained from several sources; both from forestry and agricultural resources, and can effectively lower the cost of the removal of hazardous pollutants from environment. In the current review, a special interest is given to the agricultural waste materials used for the removal of phenol, hexavalent chromium and cadmium(II) with biosorption as a promising technology for contaminant removal.

Table 2
Merits and demerits of techniques used for removal of contaminants.

Techniques	Merits	Demerits	References
Chemical precipitation	Simple and efficient	Production of sludge	Oncel et al. (2013)
Coagulation-flocculation	Removes colloids	Sludge volume is high, large foot print, slow	Tang et al. (2016)
Ion exchange	Energy efficient, low maintenance, easy operation	Regeneration is required	Ghime and Ghosh (2020)
Electrochemical Precipitation	Easy operation, high selectivity, low cost	Production of sludge Production of dendrite, loose or spongy deposit	Oncel et al. (2013)
Adsorption on activated carbon	Large specific surface area High capacity and high rate of adsorption Fast kinetics A high quality-treated effluent is obtained	Performance is dependent on the type of carbon used.Expensive	Karimi et al. (2019)
Biosorption	Cheap, possibility of pollutant recovery and regeneration, Environmentally friendly	Sensitive to operating parameters. The problem of early saturation and limited potential for biological process improvement	Othmani et al. (2020a)
Ultrafiltration	High selectivity, high diffusion rate, low energy demand	High capital cost; requires high operation and maintenance	Yaqub and Lee (2019)
Chemical oxidation	Low consumption of reagents and energy costs, operating under mild conditions (temperature and pH)	Potential need for large amounts of chemicals Resistance of some contaminants to oxidation Limited ability to penetrate low permeability soil and groundwater zones.	(Dhangar and Kumar, 2020; Othmani et al., 2020a)
Reverse osmosis	Concentration of organic and inorganic constituents is possible, no regeneration required, no sludge formation	No absolute separation, the necessity of a pretreatment before the reverse osmosis	Egea-Corbacho Lopera et al. (2019)
Fenton and Fenton-like treatment	Controlling degradation kinetics Higher degradation rates of organic pollution The on-site production of H_2O_2 Feasibility of overall mineralization at relatively low cost Simplicity	Cost of UV lamps Fouling of the surface of the UV tubes Operation pH between 2 and 4, pH adjustment likely to increase operating costs Generation of sludge through the removal of iron ions	Mirzaei et al. (2017)
Sonication	The design of the generator is very simple, low cost Frequencies ranging from 100 Hz to 3000 kHz	Necessity to add oxidant to improve the efficiency of the treatment, thereby increasing cost Generation of heat Inability to lyse all cells equally.	Dehghan et al. (2019)
Membrane filtration	Low consumption of chemicals and	Low flow rates High running cost	Kim et al. (2018)

(*continued on next page*)

4. Adsorption using agricultural waste materials

Agricultural waste material (AWM) is an abundant renewable resource almost found on the entire globe (Padam et al., 2014; Saravanan et al., 2021). Their biological and chemical properties, as well as their low operating cost, encourage their use to effectively and durably solve the persistent environmental problems, and thanks to these properties, they have gained interest in production of adsorbents as alternatives to commercial activated carbon (Ferronato and Torretta, 2019; Isikgor and Becer, 2015).

AWM is mainly composed of cellulose, hemicellulose, lignin, and extractable materials. The capacity uptake of AWM based adsorbents and their performance are highly dependent on their chemical composition, the influencing parameters on the biosorption process, the surface chemistry, the chemical and physical properties, and the affinity between available sites onto the biosorbent surface and the pollutant (Bilal et al., 2018; Gupta et al., 2015).

Table 3 presents the chemical composition of the selected waste materials. As shown in the table, the chemical composition of the selected AWMs differs and this can directly influence the adsorption capacity for pollutants. The aforementioned species are among the wastes widely used for environmental remediation. *P. australis* is reported to have a good adsorption capacity for organic pollutants such as basic dyes as well as heavy metals (Bourgeau-Chavez et al., 2013;

Table 2 (*continued*)

Techniques	Merits	Demerits	References
	minimal generation of sludge Limited production of solid waste High efficiency >95% for single metal	Decrease in performance in the presence of many metals	
Electrokinetic coagulation	Cheap and affordable Rapid and efficient process. Production of high quality of the treated water	Large sludge generation Chemicals required High cost Accumulation of sludge causing disposal problems	Tang et al. (2016)
Nanotechnology	Cheap, effective, and environmentally friendly in most cases	Decomposition/ degradation of biosorbents	Jassby et al. (2018)
Electrodialysis	Separation of ionic species, no chemicals required, works under wide pH range	High power consumption	(Banasiak and Schäfer, 2009; Onorato et al., 2017)
Ozonation	Applied in gaseous state, alteration of volume	Short half life	Ikehata et al. (2008)

Kankiliç et al., 2016). This higher capacity for metal removal is due to its chemical composition and its nature since this latter is known to be invasive with a fibrous structure (Shi et al., 2018) and a higher percentage of lignin and cellulose that account for around 13% and 21%, respectively. Generally, lignocellulosic compounds have been reported to be capable of heavy metals uptake since they contain some functional groups that contribute to high sorption efficacy (Tran et al., 2017). As shown in Table 3, cellulose, hemicellulose, and lignin content in L. cylindrica is around 54%, 23%, and 11%, respectively(Khiari et al., 2012), and the Cd(II) removal in this climbing plant was reported to reach up to 99% of removal at pH 6 compared to other waste (i.e sugarcane bagasse and Jatropha oil cake) (Garg et al., 2008). This removal capacity in L. cylindrica is linked to the presence of a highly developed fibrous vascular system that allows the elimination of pollutants from water (Boynard and D'Almeida, 2000). Besides, these fibers are biodegradable, natural, and renewable (Ncibi et al., 2009; Segun Esan et al., 2014), with a wide large application in the packaging industry and insulation. Moreover, P. oceanica known as 'the olive of the sea' presents a quite heterogeneous and compact internal structure (Alvarez et al., 2013) that allows to uptake until 30.22 mg/g of Cd(II) in 80 min and acid pH (Boulaiche et al., 2019) as well as methylene blue (Douissa et al., 2013). Similar observations were found in C. stigmas or Zea mays *that is* composed of different percentages of wax, grease, pectin; lignin, hemicellulose, and cellulose (Mbarki et al., 2018) with a lamellated and non-porous structure and considerable numbers of heterogeneous layers and channels (C. H. Li et al., 2017) and offers a potential alternative application for the treatment of wastewater. A. Americana can be hydrothermally carbonized to be applied as an effective adsorbent for the tetracycline removal but also other heavy metals; this removal potential is related to the fibrous structure (Krishnadev et al., 2020; Madhu et al., 2020).

The species discussed are among numerous AWMs widely used for biosorption such as almond shell (Omri and Benzina, 2014); pulp and paper waste (Sthiannopkao and Sreesai, 2009), rice husk (Jittin et al., 2020; Shamsollahi and Partovinia, 2019), red mud (Wang et al., 2019), fly ash (Adegoke et al., 2017), waste peels (De Goes Sampaio et al., 2019) that have been also explored for phenol, Cr(VI), and Cd(II) removal.

4.1. *Production of agro-based adsorbents*

Several methods are adopted for the production of adsorbents from agricultural waste materials. The simplest approach involves physical modification of agricultural waste materials using size-reducing techniques such as crushing and grinding (Jeyaseelan and Gupta, 2016). In such cases, AWMs such as leaves and peels are typically subjected to cleaning and drying followed by size reduction to obtain the sorbent in the form of powder that offers higher surface area compared to physically unmodified material (Ponnuchamy et al., 2020). However, in order to enhance adsorption performance, advanced techniques are used for the preparation of biosorbents that usually involve carbonization and activation (Reza et al., 2020).

Biomass in its carbonized form, designated as biochar, has been demonstrated to be highly effective adsorbent material for several environmental remediation applications (Inyang et al., 2016; Jacob et al., 2020). The technologies involved in production of biochar can be broadly classified into three categories including pre-treatment, thermal processing and post-treatment (Xiang et al., 2020). Pre-treatment technologies can be further categorized into physical, chemical, and biological methods. Physical pre-treatment methods of biomass feedstock commonly include steps like washing, drying, crushing and sieving. The sequence and duration of methods may also vary depending on the type of raw material used. Chemical pre-treatment methods aim at modifying the composition and characteristics of the biomass by chemical reactions. Biological pre-treatment techniques employ bioprocesses for engineering biochar production. The biological methods such as bacterial treatment and bioaccumulation have shown promise in production of biochar with improved characteristics (Wang et al., 2017).

The production of biochar from pre-treated biomass can be accomplished by several thermal carbonization techniques such as thermochemical pyrolysis, microwave-assisted pyrolysis, hydrothermal carbonization and gasification (Prasannamedha et al., 2021; Zbair et al., 2018). Thermochemical pyrolysis enables biomass decomposition under anoxic or hypoxic conditions (Cha et al., 2016). A number of factors such as type of feedstock, process temperature, rate of heating and residence time influence the characteristics of biochar product (Yaashikaa et al., 2019). Microwave-assisted pyrolytic approach offers distinct merits such as rapid, homogeneous, controlled, selective and efficient heating, with reduced processing times and lesser energy requirements. Thus, microwave based technology is emerging as a sustainable alternative for biochar production (Ethaib et al., 2020). Hydrothermal carbonization process involves transformation of wet feedstock biomass into biochar in the absence of pre-drying step, typically in temperature range of 120–260 °C, which is relatively lower compared to pyrolysis (Xiang et al., 2020). In gasification technique, the biomass gets converted to gaseous fuel by employing gasification agents at temperatures typically higher than 800 °C (You et al., 2017). The biochar produced is generally

Table 3
Overview of chemical composition of some agricultural waste materials.

AWM	Cellulose	Hemicellulose	Lignin	Pectin	Wax and grease	References
P. australis	21%	29%	13%	13%	24%	Dallel et al. (2018)
P. oceanica	40%	21%	28%	9%	2%	Khiari et al. (2012)
L. cylindrica	54%	23%	11%	5%	7%	Othmani et al. (2019b)
A. americana	38%	21%	27%	10%	4%	El Oudiani et al. (2009)
C. stigmas	17.1%	28.91%	28.97%	14.1%	10.87%	Mbarki et al. (2018)

high in alkali content that can facilitate removal of contaminants by precipitation.

Post-treatment technologies aim at modification of biochar to improve its properties such as specific surface area, pore volume, etc. as well as to introduce specific functional groups for achieving enhanced overall adsorption capacity. Various physical and chemical modification strategies have been investigated for post-engineering of biochar (Dai et al., 2020; Tan et al., 2016). The pre and post modification technologies can significantly impact the remediation performance of adsorbent. However, the aspect is relatively less explored and needs greater attention to translate the developments to field applications.

4.2. *Adsorbent characterization techniques*

The characterization of adsorbent before and after adsorption is essential to examine the properties of adsorbent material and gain insights into the mechanism of the phenomena involved. X-ray diffraction (XRD) is used for analysis of the amorphous and crystalline nature of the adsorbents being investigated. Well-defined sharp peaks are observed in crystalline materials whereas broad peaks denote non-crystallinity of the material (Kaur et al., 2013). The change in XRD patterns after adsorption indicates the change in nature of substrate due to sorption of adsorbate on the surface. Scanning electron microscopy (SEM) examines the morphological characteristics of adsorbents (Ighalo and Adeniyi, 2020). In a study on a biosorbent derived from guava leaves, SEM images revealed that chemical modification of biomass made the surface more ridged (Abdelwahab et al., 2015). Such morphological changes can play a role in determining the surface availability and thereby impact the adsorption performance of biosorbents. The morphological characteristics can also be studied using transmission electron microscopy (TEM) that provides a greater resolution and more detailed imaging information including that of the inner structure of the sample. However, its usage for bisorbent characterization has been relatively less explored in comparison to SEM, possibly due to higher cost of TEM (Ighalo and Adeniyi, 2020). The individual elements present in the sample and their quantitative compositional information is obtained by energy dispersive X-ray analysis (EDX). Another surface analysis technique that has been relatively less explored is Time-of-flight secondary ion mass spectrometry (TOF-SIMS) that can be used to know in detail about elemental and molecular species present on a surface with high mass resolution and high spatial resolution (Kempson et al., 2010).

The specific surface area of the adsorbent is estimated by Brunauer, Emmett and Teller (BET) method of nitrogen adsorption and desorption. The surface analysis measurements including surface area, porosity, and pore size distribution and pore volume play a critical role in determining available sites for adsorption. The chemical characterization with respect to presence of the functional groups present in the adsorbent can be acquired from Fourier transform infrared spectroscopy (FTIR) by measurements of their absorption of infrared radiation typically in wavelength range of 2.5–25 μm (Khaled et al., 2020). Further insights into the chemical structure of the material can be obtained using Raman spectroscopy as each Raman spectrum acts like a chemical fingerprint for specific molecule (Rápó et al., 2020). X-ray photoelectron spectroscopy (XPS) also designated as electron spectroscopy for chemical analysis (ESCA) is useful to acquire information about chemical bonding states of the adsorbent before and after adsorption (Ren et al., 2018). Nuclear magnetic resonance (NMR) spectroscopy allows determination of active sites present on the adsorbent material (Şen et al., 2015). Further, thermal stability of the adsorbent can be evaluated by thermogravimetric analysis (TGA) (Ghanbari and Niu, 2019).

4.3. *Adsorptive removal of contaminants*

The adsorptive removal of hazardous contaminants including, phenol, Cr(VI), and Cd(II), using different AWMs presents cheap, effective and environmentally friendly technology. The performance of this technology is usually optimized based on the adsorbate concentration and process conditions. Table 4 summaries some important findings reported in previous works for the removal of phenol, Cr(VI), and Cd(II) for various green materials.

4.3.1. *Phenol removal*

Various research studies have been done in recent years on phenol removal using different types of green adsorbents obtained from agricultural waste materials. For instance, corn husk waste was used to derive magnetized activated carbon for the removal of phenol from aqueous environments (Mishra et al., 2019). A comparative study was performed for two thermal treatment temperatures 250 °C and 500 °C. The corn husk waste-derived adsorbent treated at 500 °C showed relatively better adsorption performance with 96% phenol removal efficiency and 7.8 mg/g maximum uptake capacity compared to 81% removal efficiency and 5.8 mg/g maximum uptake capacity for 250 °C treated sorbent. Similar studies were reported for biochar derived from pine fruit shell biomass to understand its ability to sorb phenol (Mohammed et al., 2018). The sorption behavior for three biochars prepared at 350 °C, 450 °C, and 550 °C pyrolytic temperatures was compared. The high pyrolytic temperatures favored the formation of more effective adsorbents. Biochar prepared at 550 °C exhibited 26.738 mg/g maximum adsorption capacity compared to 15.974 mg/g and 10.373 mg/g adsorption capacities shown by biochars prepared at 450 °C and 350 °C at pH of 6.5 and temperature of 25 °C respectively. In another study, black wattle bark waste produced from tannin extraction industries was used for phenolic sorption (Lütke et al., 2019). The activated carbon derived from agro-waste exhibited a specific surface area of 414.97 m^2/g with 0.064 cm^3/g total pore volume on a unit mass basis. The maximum phenol adsorption capacity of 98.6 mg/g was recorded at 6.5 pH and 55 °C process conditions. The adsorbent was shown to be reusable twice with less than a 5% reduction in removal capacity. An efficiency of 95.89% for phenol removal from simulated wastewater was achieved.

4.3.2 Chromium(VI) removal

Chromium(VI) removal using adsorbents has equally received attention majorly in the research arena due to the negative effects it has caused in the environment. Various fruit and vegetable waste adsorbents have been investigated to provide a cheaper and sustainable approach towards removal. A recent study was conducted using data seed-derived activated carbon was studied in batch experiments and the maximum uptake capacity of 42.57 mg/g was recorded at pH 2, 30 °C temperature, and 1 h contact duration. The adsorbent showed successful regeneration and could be used up to 3 times with less than 10% reduction in removal capacity (Rambabu et al., 2019). In another study, the maximum chromium(VI) adsorption capacity of 36.01 mg/g was obtained using activated carbon derived from apple peel waste. The optimal experimental conditions were highly acidic with pH 2, 28 °C temperature, 4 h contact time, activated carbon dosage of 0.05 g for 50 mL aqueous solution for an initial chromium concentration of 50 ppm (Enniya et al., 2018). A facile and eco-friendly approach was also used to prepare chromium(VI) removing adsorbent from waste corn stalks. Potassium hydroxide solution at various concentrations was used for activation of corn stalk powder and adsorbent with micro and mesoscale pores was obtained. Unlike conventionally used KOH activation procedures, the method involved the recycling of KOH before carbonization. The pores were formed due to alkali treatment and calcination The highest chromium (VI) adsorption capacity of 89.5 mg/g was achieved for 4% potassium hydroxide activated corn stalks-based adsorbent (Zhao et al., 2020). In recent years, a multivariate optimization approach has also been evaluated for determining the most suitable process conditions for the removal of chromium(VI) from contaminated water using sugar beet bagasse derived activated carbon (Ghorbani et al., 2020). The removal of 50.45% was obtained at a pH of 4.5, the adsorbent dosage of 1.49 g/L, and the initial chromium(VI) concentration of 10.13 mg/L. Further, the

Table 4
Performance of adsorbent materials based on agricultural waste for the removal of phenols, Cr(VI) and Cd(II).

Contaminant	Adsorbent	Operating conditions			Adsorption capacity (mg/g)	References
		pH	Time (min)	Temperature (°C)		
Phenol	*L. cylindrica*	7	120	20	10.37	Abdelwahab and Amin (2013)
	P. australis	–	120	30	29.6	Shi et al. (2018)
	Dall mill residue waste	8	24	40	6.18	Girish and Ramachandramurty (2013)
	Garlic peel	2	420	30	14.49	Muthamilselvi et al. (2016)
	Corn husk activated carbon	7	120	25	7.8	Mishra et al. (2019)
	H_3PO_4 activated spent tea waste	8	120	32	154.39	Pathak et al. (2020)
	H_2SO_4 activated spent tea waste	8	120	32	185	Pathak et al. (2020)
Cr(VI))	Red mud	2	1440	–	75	Gupta et al. (2001)
	Tea factory waste	2	–	60	54.65	Malkoc and Nuhoglu (2007)
	L. cylindrica	8	60	–	188.5	Nwosu-Obieogu and Okolo (2020)
	Groundnut shell	8	120	–	2.96	Bayuo et al. (2019)
	Banana peel	2	60	25	10.42	Parlayici and Pehlivan (2019)
	Polyethylene functionalized corn bract	2	–	50	438	Luo et al. (2017)
	Grafted macadamia nutshell powder	2	180	50	39.21	Ntuli and Pakade (2020)
	Cranberry kernel shell	2	90	25	6.81	Parlayici and Pehlivan (2019)
	P. oceanica	2	–	–	14.48	Krika et al. (2012)
	Rich husk ash –	2–3	180	–	25.64	Bhattacharya et al. (2008)
Cd(II)	*P. oceanica*	6	80	25	30.22	Khiari et al. (2012)
	Areca waste	5.6	120	–	1.32	Zheng et al. (2008)
	Almond shell	–	120	–	3.18	Bulut and Tez (2007)
	L. cylindrica	7	60	22	6.71	Shahidi et al. (2015)
	Maize corncob	2–7	60	–	105.6	Garg et al. (2008)
	P. oceanica	7	60	20	117.65	Kaouah et al. (2014)
	P. australis	<10	60.4	–	11	Bello et al. (2018)
	A.americana	5	30–60	20	12.5	Hamissa et al. (2010)
	Lentil husk	5	60	30	107.31	Basu et al. (2017)
	Sodium hydroxide treated aloe vera waste	–	360	25	104.2	Noli et al. (2019)
	De-oiled palm kernel waste	6	120	–	1.09	Hebbani et al. (2020)
	A. rubescens	5	30	20	27.3	Sari and Tuzen (2009)

cost analysis revealed the economic efficiency of the process with an estimated cost of adsorbent being 1.5 US dollars per kg based on 34% yield.

In another study, a comparative analysis of three agricultural waste material-based adsorbents for chromium(VI)-contaminated water remediation was performed (Mokkapati et al., 2018). The powdered form of banana bunch stem, sorghum stem, and casuarinas fruit were modified by acid treatment for use in batch adsorption studies. The studies using the Dunwald-Wagner model based on intraparticle diffusion revealed that diffusion coefficients were 1.03×10^{-4} mm^2/min for banana bunch stem powder, 0.65×10^{-4} mm^2/min sorghum stem powder, and 0.56×10^{-4} mm^2/min for casuarinas fruit powder suggesting the diffusive mass transfer in this order. Both monolayer and multilayer adsorption mechanisms were involved in the binding of chromium(VI) on binding sites. Focus has also been given in conducting continuous column studies for the removal of hexavalent chromium using H_3PO_4 activated Trapa natans, commonly called water caltrop shell waste (Kumar et al., 2020). However, continuous studies are relatively few compared to batch studies and more attention is required in this direction to design unit operations for field applications.

4.3.3 Cadmium(II) removal

Cadmium(II) has been shortlisted as a potential metallic toxicant harming the entire biological community due to its hazardous nature. The metal has been shown to accumulate in living organisms and damage vital organs like kidneys, liver, and lungs, and play a role in causing cancer and hypertension (Basu et al., 2017). Thus, agricultural waste materials have been investigated with an aim of effective adsorptive decontamination of cadmium(II) from the environment. In a recent study, waste corncob was pyrolyzed at 350 °C to prepare biochar grafted with acrylonitrile for adsorptive removal of cadmium(II). The adsorption process was found to be pH-dependent and stabilized at pH values in the range of 5–7. The maximum uptake of 85.65 mg/g was reported and the mechanism was governed by ion exchange and sorption-complexation (Luo et al., 2018). Similarly, aloe vera waste was investigated regarding its ability to remove cadmium(II) from aqueous solutions (Noli et al., 2019). The adsorption capacities of 70.4 mg/g, 104.2 mg/g, and 66.2 mg/g were observed for unmodified waste biomass, NaOH treated and H_3PO_4 treated waste respectively. In another study, the adsorbent derived from Sorghum x drummondii, commonly known as sudangrass was used for the removal of cadmium (II) (Saraeian et al., 2018). A series of surface modification trials were conducted and 0.05 M NaOH surface-modified adsorbent exhibited the maximum adsorption capacity of 7.76 mg/g. A five-fold improvement in adsorption capacity compared to unmodified adsorbent was achieved. The uptake was observed to increase as agitation speed was increased from 50 rpm to 200 rpm and decreased thereafter. The behavior was attributed to the lack of sufficient time for cadmium(II) ions to reach the available sites on the sudangrass-based adsorbent surface at higher speeds. The activated carbon based on Leucaena leucocephala biomass also showed capability in adsorbing cadmium(II) with 70.42 mg/g maximum uptake capacity at optimal pH 7 and temperature 30 °C. The adsorbent was produced by the alkali activation process with a 3:1 alkali to carbonized biomass ratio at 800 C and exhibited 776 m^2/g Brunauer–Emmett–Teller (BET) surface area (Wan Ibrahim et al., 2019). A recent investigative study for cadmium(II) adsorption was conducted on biochar prepared from sodium silicate-modified oil tea camellia shell agricultural waste. The silicate modification process was found to be highly effective in improving available surface area by 45–112%. The porosity was increased by 5–12% and consequently, enhanced internal diffusion of cadmium(II) ions resulted in improved adsorption (Cai et al., 2021). These studies add another perspective in understanding the potential of waste valorization and its removal characteristics.

5. insights into the adsorptive behavior

5.1 mechanism

The mechanism of adsorptive remediation using agro-waste is a complex phenomenon governed by several factors related to characteristics of adsorbates and adsorbents as well as process conditions. These factors include physicochemical features of adsorbent prepared from agricultural wastes such as porosity, types of binding sites and their availability, chemical and stereochemical characteristics of adsorbate species, and process parameters like pH, the concentration of species, temperature, and presence of competing species.

Various types of mechanisms involved in the adsorption of organic contaminants such as phenol include pore filling, partitioning, hydrogen bonding, π-π stacking, and electrostatic interaction (Ahmad et al., 2014; Ambaye et al., 2020). Often more than one of these mechanisms may be involved in the adsorptive removal process. The electrostatic interactions between phenols and adsorbate play an important role in determining the removal efficiency of phenol. The underlying mechanism of phenol removal using corn husk waste-based adsorbent was primarily governed by the electron donor mechanism. The presence of functional groups on corn husk waste-derived activated carbon facilitated the donation of electrons to an aromatic phenolic structure leading to favorable adsorption (Mishra et al., 2019). Studies on activated carbon obtained from other sources such as sludge revealed that experimental conditions that favored electrostatic force interactions enhanced phenolic removal (Raza et al., 2019). However, pH values beyond 8 resulted in a decrease in adsorptive removal of phenol that could be attributed to electrostatic repulsive forces becoming stronger due to negative surface charge on the adsorbent. Further, the interactions between functional groups present on the adsorbent surface such as –COOH and phenol were postulated to govern the adsorption mechanism. The surface of the adsorbent can also be modified by incorporating charged functional groups such as hydroxyl, amine, etc. to facilitate the adsorption of charged species. Studies on the adsorption of phenol onto activated biochar obtained from spent tea waste suggested that the removal was primarily governed by the interactions between the phenolic group and functional groups on the adsorbent. The interactions of the phenolic group with alkenes, alkynes, amino, and carbonyl groups were involved in the adsorptive mechanism (Pathak et al., 2020).

Some of the commonly observed mechanisms for removal of heavy metal species using adsorption include surface sorption, ion exchange, electrostatic interaction, surface precipitation, and complexation (Ahmad et al., 2014; Ambaye et al., 2020). Surface sorption is a physical phenomenon that involves interaction between metal ions and the surface of the adsorbent. The physical characteristics and affinity between surface and metallic species play a crucial role in governing this mechanism. When the surface of the adsorbent possesses a charged configuration, electrostatic interactions come into play. The electrostatic forces between the charged sorbent surface and metal ions immobilize the toxic ions and facilitate their removal from the solution. The anionic metal attraction takes place when the pH of the solution is less than a point of zero charge and cationic metal attraction takes place when pH is more than the point of zero charges.

Hexavalent chromium is present in a variety of ionic forms in aqueous solutions, including H_2CrO_4, $HCrO_4^-$, CrO_4^{2-} and $Cr_2O_7^{2-}$. Under highly acidic conditions, chromium(VI) is reported to exist mainly as $HCrO_4^-$ (De Goes Sampaio et al., 2019). Since chromium(VI) is existing in a negatively charged form under these conditions, the surface charge characteristics of the adsorbent can play an important role in determining the removal of chromium(VI). The mango peels and cultivars of seeds were therefore found to be effective in the removal of chromium(VI) as they possessed a positive charge in acidic conditions when pH was below their point of zero charges. The complexity of the adsorption phenomena of hexavalent chromium was also demonstrated in an investigative study on adsorbents obtained from tea waste and date pits (Albadarin et al., 2013). The mechanism was found to comprise three steps. The first step consisted of anionic sorption of hexavalent chromium onto the adsorbent favored by the presence of positively charged amino and carboxyl groups. Thereby, hexavalent chromium was reduced to trivalent chromium under acidic conditions. This was followed by ion exchange or surface complexation.

In a study on lentil husk for cadmium(II) removal, characterization studies such as scanning electron microscopy with energy dispersive X-ray analysis, FTIR spectroscopy was conducted for husk-derived adsorbent before and after exposure to cadmium(II) solutions to elucidate the mechanism (Basu et al., 2017). The results showed that the ion exchange mechanism was involved in the adsorption with calcium replacing cadmium(II) on the sorbent. Further, the interactions of functional groups present on lentil husks such as carboxyl, hydroxyl, amide, and amino groups also participated in the adsorptive phenomenon. The pH of an aqueous solution containing cadmium(II) ion was found to directly influence the adsorption behavior of areca waste adsorbent as it impacted the surface charge and interaction between the adsorbate and adsorbent (Zheng et al., 2008). The adsorption of cadmium(II) from aqueous solutions was attributed to the exchange of cationic species with H^+ ions in functional groups on areca waste adsorbent, namely $–C_6H_5OH$ and –COOH. The mechanism was in agreement with pH-dependent adsorption behavior. The maximum removal was observed at a pH of 5.6. The solution at lower pH consisted of a high concentration of H^+ ions that offered competition to metal ions for binding to active sites on areca waste resulting in lower adsorption. As the pH of the aqueous solution was increased beyond the optimal value, the increase in hydroxyl complex formation caused a decrease in adsorptive removal. Other mechanisms also include the exchange of cadmium(II) with H^+ ions from positively charged surfaces originating from the electromeric effect in acidic conditions, followed by coordination of cadmium(II) ions. In a study involving biomasses derived from palm bagasse, corn crop, orange crop, and palm crop, the incorporation of alumina nanoparticles was found to enhance the adsorption efficiencies (Herrera et al., 2020). The proposed mechanism was governed primarily by the binding of cadmium(II) ions onto the nanoparticle surface. The presence of an aqueous medium resulted in hydrolysis of the surface of alumina nanoparticles and the formation of AlO^- occurred due to the removal of protons from Al–OH functional groups on the surface. The presence of a negative charge made the surface more amenable to the cationic binding. The hydroxyl functional groups on the biomass surface also provided favorable sites for binding by cadmium (II) ions. Thus, adsorption technology works based on various mechanisms that can be suitably tailored to achieve high performance. This makes adsorption using green materials one of the promising alternatives compared to conventional technologies in terms of cost, ease of adsorbent availability, low energy consumption, and efficacy.

5.2. Influencing factors

Several factors influence the adsorption of contaminants on agricultural waste material based adsorbents such as contact time, adsorbent dosage, solution pH, temperature, initial concentration of contaminant, ionic strength, adsorbent characteristics, etc. A discussion on the effect of some of the key factors on the adsorptive remediation process using AWM based adsorbents is presented here.

5.2.1. Contact time

The contact time affects the adsorption kinetics and plays an important role in governing the adsorption phenomenon. The initial adsorption rates are faster due to abundant availability of active sites on the adsorbent surface, large number of adsorbate species in the bulk solution and intermolecular attractive forces between the adsorbent and the contaminant adsorbate species. The rates gradually decrease with the course of time. Once the equilibrium is attained, the extent of adsorption remains constant with time (Anantha and Kota, 2016). The influence of contact time was studied for the adsorption of phenol onto waste corn husk derived sorbent. The phenol removal percentage exhibited a rapid increase from 0 to 60 min followed by a gradual increase from 60 min to 120 min. Subsequently the adsorption percentage remained unchanged with time suggesting that the state of equilibrium was reached (Mishra et al., 2019). A similar trend was observed for removal of hexavalent chromium using polyethylenimine functionalized corn bract. The sorption capacity exhibited a rapid increase till 6.6 h. The rate of increase was then slowed down and ultimately equilibrium was reached after 24 h (Luo et al., 2017).

5.2.2. *Adsorbent dosage*

The adsorbent dosage affects the availability of adsorbent surface area and the number of active exchangeable sites on the surface. Generally, an increase in the dosage results in an increase in the extent of adsorption. However, it must be noted that beyond a certain dosage limit, the extent of adsorption may remain unaffected or even decrease if there is an interference due to the interaction of active sites (Iftekhar et al., 2018). Such behavior was observed during phenol adsorption studies on corn husk based activated carbon. The percentage removal of phenol increased as the adsorbent dosage increased from 0.1 g/L to 2 g/L. The maximum phenol removal observed at 2 g/L was 81.6% and 96.56% for adsorbents prepared at 250 C and 500 C respectively. Further increase in the dosage to 3 g/L either did not affect the percentage removal or decreased it slightly. The observed behavior at higher dosage levels was attributed to agglomeration of adsorbent particles at higher concentrations resulting in enhancement of particle size and reduction in specific surface area and available binding sites (Mishra et al., 2019). The adsorptive removal using cashew nut shell resin functionalized nanoparticles revealed that cadmium(II) removal increased as the dosage increased from 0.5 g to 1 g for 100 mL aqueous solution. Further increase in dosage had no effect on the removal (Devi et al., 2017). Further, economic aspects associated with the use of higher adsorbent dosage must also be considered.

5.2.3. *Solution pH*

One of the important factors that determine the uptake of contaminant species by the adsorbent is pH. The solution pH influences the extent of adsorbate ionization and plays a key role in adsorptive processes where electrostatic interactions are primarily involved in governing adsorption. The surface characteristics of an adsorbent determine the pH at which net surface charge is zero, known as point of zero charge. When pH is less than the point of zero charge, the adsorbent is positively charged and favors sorption of negatively charged contaminant species and vice versa. In a study using cashew nut shell resin (CNSR) covalently bonded with magnetic Fe_3O_4 nanoparticles, the adsorption of cadmium(II) increased with increase in pH with the highest removal observed at pH 10. The phenomenon was attributed to deprotonation of functional groups at high values of pH thus becoming negatively charged and consequently enhancing the interactions with positively charged cadmium(II) species (Devi et al., 2017).

Activated carbon derived from sugar beet bagasse agro-waste was demonstrated as an effective adsorbent for removal of hexavalent chromium(VI) from aqueous solutions (Ghorbani et al., 2020). The highest chromium(VI) removal was observed at pH of 2.98. The remediation process was observed to be dependent on pH of solution due to the existence of various ionic forms of chromium and type of functional groups present on the agro-waste derived activated carbon. Chromium manifested different ionic forms due to variation in equilibrium states at different pH values. The predominant form at lower pH values was $HCrO_4^{-}$. Consequently, electrostatic attraction occurred between protonated sorbent surface and negatively charged metallic species in aqueous solution. The prominent ionic forms at higher pH included CrO_4^{2-} and $Cr_2O_7^{2-}$. The presence of more OH^{-} ions at higher pH resulted in lesser protonation of the sorbent surface, which in turn was responsible for electrostatic repulsion between negatively charged chromium species and sorbent surface having negative charges.

The experimental studies of phenol adsorption onto waste corn husk derived activated carbon prepared at 250 C and 500 °C demonstrated the effect of pH on phenol adsorption. The removal of phenol increased from 55% to 81% with an increase in pH from 4 to 6 for sorbent prepared at 250 °C. Whereas an increase in removal from 78% to 98% was observed for other sorbent prepared at 500 °C when pH was raised from 4 to 7. Further increment in pH of solution resulted in slight decrease in adsorption. The influence of pH was attributed to variation in degree of ionization in solution with change in pH (Mishra et al., 2019).

5.2.4. *Temperature*

The temperature of solution can influence the mobility of ionic species, pore size, and also alter the exchangeable sites on the adsorbent surface. Any variation in these aspects can affect mass transport and thereby adsorption efficiency. The adsorption of phenol onto cornhusk derived activated carbon was investigated at three different temperatures of 25 °C, 35 °C, and 45 °C while keeping other experimental conditions constant. The studies demonstrated a decrease in adsorption percentage with an increase in temperature. The observations suggested the exothermic nature of the phenolic adsorption (Mishra et al., 2019). On the other hand, adsorption of hexavalent chromium onto polyethylenimine functionalized corn bract showed a different trend. The adsorption capacity increased as the solution temperature was raised from 20 °C to 50 °C pointing to the endothermic nature of the adsorption phenomenon. The observed behavior was possibly due to enhanced rate of diffusion of chromium(VI) across the boundary layer at higher temperatures (Luo et al., 2017).

5.2.5. *Initial contaminant concentration*

The initial concentration of contaminant in aqueous solution determines the number of contaminant adsorbate molecules in bulk solution available for binding to active surface sites on the adsorbent. The effect of initial phenol concentration onto corn husk based activated carbon was studied by varying feed concentration from 5 mg/L to 30 mg/L. The percentage removal of phenol showed a decreasing trend with increase in initial concentration. The highest adsorption percentage was observed at the lowest initial phenol concentration of 5 mg/L. The observed behavior could be possibly explained on the basis of the relative number of adsorbate molecules and adsorbent binding sites. At lower phenol concentration, the number of adsorbate species were lesser in comparison to the number of available adsorbent surface binding sites. Thus, all phenolic species could easily interact with the adsorbent and be removed from the bulk solution. An increase in initial phenol concentration allowed the number of adsorbate molecules to exceed the number of adsorbent surface binding sites, thereby resulting in a decrease in adsorption percentage (Mishra et al., 2019). A similar observation was made for removal of hexavalent chromium using sugar beet bagasse based activated carbon. The adsorption percentage decreased from 40.11% to 30.96% as chromium(VI) concentration was increased from 5 mg/L to 130.68 mg/L (Ghorbani et al., 2020).

6. Regeneration of adsorbents

Regeneration refers to the recycling of spent adsorbents for possible reuse by employing techno-economic approaches (Momina et al., 2018). Various regeneration techniques have been investigated to examine their efficacies in reclaiming adsorptive capacities of the adsorbents.

6.1. *Chemical regeneration*

In chemical regeneration approach, the spent adsorbent is treated with specific chemical reagents to desorb the adsorbed species or decompose them. The regeneration performance is dependent on factors including pH of solution, nature of regenerating solvent and solubility of adsorbed species in the solvent. The solution pH must be carefully controlled as it affects the charged state of adsorbed and adsorbing species, which in turn influences the regenerative performance. The regeneration of guava tree bark as phenol adsorbent was studied using distilled water and ethanol. The regenerated adsorbent exhibited 40.15% and 57.25% phenol removal after the first cycle with distilled water and ethanol as regenerating agents, respectively. However, the adsorption performance reduced to 20.23% and 33.30% phenol removal after the second cycle when distilled water and ethanol were used as regenerating agents, respectively (Mandal et al., 2020). The chemical regeneration of adsorbents derived from groundnut, walnut and almond shells was studied using sodium hydroxide solution. The removal of hexavalent chromium was 87.8% for fresh groundnut shell adsorbent and was diminished to 69.8% and 41.5% after first and second regeneration cycles. The chromium(VI) removal using walnut shell was 92.2% for fresh adsorbent and was reduced to 82.7% and 58.7% after first and second regeneration respectively. Similarly, the chromium(VI) removal using almond shell was 90.2% for fresh adsorbent and was reduced to 75.1% and 50.4% after first and second regeneration respectively (Das et al., 2019).

6.2. *Thermal regeneration*

In thermal regeneration technique, the spent adsorbent is subjected to heat treatment that facilitates decoupling of adsorbed species from the adsorbent surface. This may involve weakening of intermolecular interactions and/or cleavage of bonds between adsorbent and adsorbate. The regeneration capability is generally a function of temperature as well as time of heat treatment. In most cases, thermally induced degradation would require a minimum threshold temperature. In a recent study, the performance of adsorbent prepared from fallen leaves of Magnoliaceae was investigated after thermal regeneration (Guo et al., 2020). The contaminant uptake capacity of 139.4 mg/g was observed even after three consecutive thermal treatment cycles suggesting viability of the regenerated adsorbent for reuse. The requirements of steam or inert gas supplies for maintaining high temperature makes this technique relatively costly. Further, repeated exposure to high temperature may affect structural integrity of the adsorbent and also cause reduction in adsorbent mass in successive regeneration cycles.

6.3. *Photoassisted regeneration*

Photoassisted treatment aims at the degradation of pollutants in presence of reactive free radicals generated by photocatalytic and photosensitized oxidation. The technique is highly effective for organic contaminants and allows their rapid degradation. The approach involves incorporation of photocatalysts or photosensitizers with the biomass. Metallic oxides such as CuO, TiO_2, ZnO, etc have been successfully studied for photocatalytic degradation of organic pollutants (Benabbas et al., 2020). A self-regenerative biosorbent was prepared by incorporating copper oxide into the phosphoric acid-pretreated biomass originating from *Callitriche obtusangula*. The hybrid material exhibited good regeneration performance and sustained adsorption efficiency higher than 90% for azo dye over four biosorption-regeneration cycles (Benabbas et al., 2020).

6.4. *Biological regeneration*

Biological regeneration relies on biodegradation of adsorbed contaminants, typically organic species by microbial action (Momina et al., 2018). In this method, the contaminant-laden adsorbent matrix is mixed with microbes along with nutrients and dissolved oxygen (Omorogie et al., 2016). The ensuing degradation and desorption of adsorbents allow renewal of the adsorbent for subsequent usage. The performance of bioregeneration technique depends on various factors including type of microorganism used, optimal conditions for its activity, structural properties of the adsorbent, nature of the adsorbate and relative concentration of microorganism and adsorbent. The biological regeneration of modified wheat straw adsorbent was explored using heterotrophic bacteria (Tan et al., 2012). A comparative analysis of the adsorption capacities for fresh and regenerated adsorbents revealed the efficacy of bioregeneration. The perchlorate adsorption uptake of virgin adsorbent was 23.94 mg/g and was slightly reduced to 21.97 mg/g after the first regeneration cycle and 20.85 mg/g after the second regeneration cycle. The high reduction efficiencies of 91% and 87% for the two cycles suggest the potentiality of bioregenerative approach.

6.5. *Ultrasound regeneration*

Ultrasound regenerative technique subjects spent adsorbent to ultrasonic treatment for desorbing pollutants from the adsorbent matrix. The technique is being explored as an economical alternative to thermal regeneration process (Omorogie et al., 2016). Bench scale and pilot scale studies were carried out to investigate the regeneration performance of biological activated carbon using ultrasound treatment (Liu et al., 2017). The experimental parameters including ultrasound frequency, sonication intensity, and water temperature had a discernible effect on the regeneration performance. Whereas, the application time of the biological activated carbon did not show any significant effect on the regeneration. The ultrasound treatment was able to regenerate the spent adsorbent as indicated by iodine and methylene blue values. Iodine value showed an increase from 480 mg/g to 680 mg/g whereas methylene blue value increased from 100 mg/g to 133 mg/g.

6.6. *Microwave regeneration*

In microwave regeneration technique, the adsorbent is exposed to microwave irradiation. The electromagnetic energy is converted to heat energy that allows the release of adsorbed contaminants from the adsorbent matrix. Unlike conventional heating, microwave technology enables volumetric heating and has potential for the regeneration of carbonaceous substances. The efficacy of microwave regeneration for activated carbon derived from durian shell and jackfruit peel was examined using a modified domestic microwave oven operated at a frequency of 2.45 GHz with 600 W power for 3–4 min irradiation time. The adsorption capacities of the regenerated sorbents could be maintained at 181.43–207.57 mg/g after five regeneration cycles suggesting good adsorption potential of the renewed adsorbent (Foo and Hameed, 2012). A summary of advantages and limitations of various adsorption regeneration methods is presented in Table 5. The literature findings revealed that regeneration of agricultural waste material based adsorbents in the context of removal of phenol, chromium(VI) and cadmium (II) have mainly focused on chemical regenerative approach. Future studies should aim at investigation of other approaches as well and comparing their efficacies with chemical treatment. Apart from the regenerative techniques discussed here, several promising developments have been taking place in adsorbent regeneration using supercritical fluid regeneration, electro-assisted regeneration and plasma regeneration (Sanchez-Montero et al., 2018; Xing et al., 2018; Zhou et al., 2016). A techno-economic analysis is recommended to select the most suitable regeneration method for a particular combination of adsorbate and adsorbent.

7. Biosorption: A promising approach for environmental remediation

Due to the complexity of the biosorption process, the choice of the biosorbent is primordial for better performance at the industrial scale. A promising biosorbent should be cheap and easily available in large

Table 5
Advantages and limitations of adsorbent regeneration methods.

Regeneration method	Advantages	Limitations
Chemical regeneration	• Rapid regeneration • Ease of integration with advanced techniques	• Toxicity issues • Need for further treatment of waste liquid
Thermal regeneration	• Suitability for adsorbent matrices laden with heterogeneous adsorbate mixture	• High energy consumption • Effect on structural integrity • Loss of carbon surface area
Photoassisted regeneration	• Rapid regeneration • Environmental friendliness	• Formation of byproducts, that may be harmful in some cases
Biological regeneration	• Safety • Cost effectiveness • Environmental friendliness	• Low efficiency • Slow • Applicability for biodegradable pollutants
Ultrasound regeneration	• Low energy requirements • Simplicity of equipment • Low carbon loss • High recovery of valuable materials	• Less effectiveness for substances that are difficult to desorb or degrade
Microwave regeneration	• Volumetric heating • Rapid and precise temperature control • Enhancement of adsorption capacity of adsorbent • Small space requirements	• Limited applicability for continuous regeneration • Possibility of volatilization of some substances without mineralization

amounts in nature. The physicochemical characteristics and biological factors are reported to largely impact the biosorption process, thus, a better understanding of the involved mechanisms during the pollutant removal is a must to monitor the process well. The adsorption capacity, selectivity, kinetics, and the possibility of regeneration or reuse of biosorbent are of paramount importance for the enhancement of the process (Fig. 1).

Surface modification is often required to enhance the binding capacity of the material. Despite the huge number of papers dealing with the use of agricultural waste materials as adsorbents and their positive effects on removal and suitability, the desorption process, kinetics, and feasibility studies are still at the embryonic stage with minimal investigation focusing on large scale utilization. According to the literature, one of the main drawbacks of the use of agricultural wastes materials is their low uptake capacity compared to activated carbon (De Gisi et al., 2016). Current attempts done to valorize AWMs for environmental remediation are generally investigating one biosorbent. The combination of two or more biosorbents for the decontamination process may offer new horizons for metal removal and waste management. The combination of different physicochemical properties and characteristics of various waste materials should be well studied. To gain in performance, coupling more than two metals oxides such as ZnO and Al_2O_3 to prepare a performing hybrid material useful for wastewater decontamination is promising. However, the reports regarding modification of the agricultural waste materials by metallic oxides are relatively limited. A few investigations have been done and this approach offers promising opportunities for further work. Table 6 collects some reported works dealing with the modification of natural fibers or AWMs with metal oxides like TiO_2 and ZnO and their use for the removal of contaminants. According to the literature, some investigations have been dedicated to the modification of waste materials by metal oxides for wastewater remediation (Kesraoui et al., 2019; Y. Li et al., 2015). A study examined the possibility to synthesize the zinc oxide (ZnO) nanoparticles and their deposition on the surface of cotton fabrics through ultrasound irradiation (Perelshtein et al., 2009). The results showed that the antibacterial activities of the ZnO-fabrics composite against *Escherichia coli* (gram negative) and *Staphylococcus aureus* (gram positive) cultures were significant. These observations were in agreement with another study that successfully synthesized hybrid materials TiO_2-wood using a hydrothermal method (Sun et al., 2011). The results showed that TiO_2-wood hybrid materials could yield many interesting properties such as hydrophobic and photocatalytic features for pollutant removal. In another study, researchers optimized the performance of the synthesized ZnO nanoparticles using bacterial cellulose (BC) and natural cellulose based on their photolytic degradation of methyl orange (Hu et al., 2010). They

Table 6
Agricultural waste material (AWM) based adsorbents, modification process and their applications in contaminant removal.

AWM	Modification	Applications	Synthesis method	References
Cellulose	TiO_2	Biosorption of Pb^{2+} ions	In situ hydrolysis	(Y. Li et al., 2015)
Bacterial cellulose	ZnO	Photolytic degradation of Methyl orange	Decomposing bacterial cellulose infiltrated with zinc acetate	Hu et al. (2010)
Wood	TiO_2	Photocalytic degradation of pollutants	Hydrothermal method	Sun et al. (2011)
Cotton fiber	ZnO	Breakdown of bacteria	Ultrasound irradiation	Perelshtein et al. (2009)
L. cylindrica	(1%, 2%, and 4% ZnO)	Biosorption of methylene blue, industrial wastewater, and phenol	Precipitation method in presence and absence of alternating current	(Othmani et al, 2019a, 2020b, 2019a)
L. cylindrica	(1%, 2%, and 4% Al_2O_3)	Biosorption of methylene blue and methyl orange	Precipitation method	Kesraoui et al. (2019)

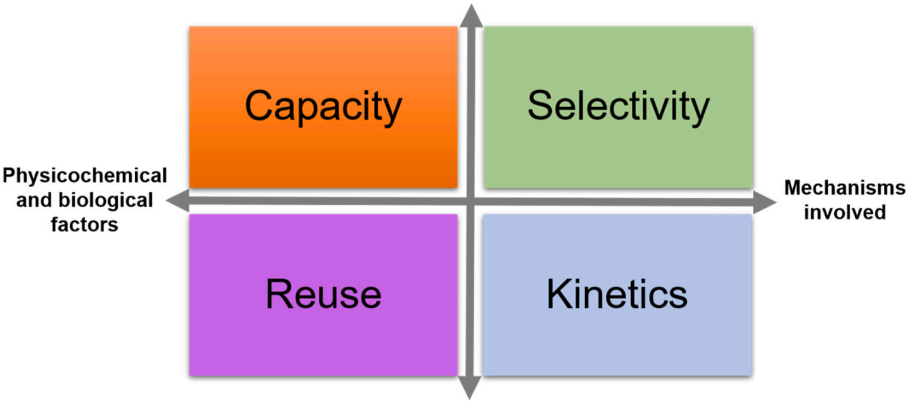

Fig. 1. Main factors influencing the adsorption complexity.

confirmed that the methodology was environmentally friendly simple and efficient for the removal of methyl orange from an aqueous solution. Besides, synthesis of TiO_2/cellulose nanocomposite by in situ generations of titanium dioxide (TiO_2) nanocrystals was carried out on cellulose fibers (CF) via hydrolysis of $TiOSO_4$. The results showed that the synthesized composite possessed a good adsorption capacity for Pb^{2+} ions, four times higher than pure cellulose fibers (Y. Li et al., 2015).

Most of these studies have discussed the method used for the modification of natural or agricultural waste material by metal oxides using several methods such as coating, electrodeposition, irradiation, and hydrothermal reactions. However, a deeper understating of the phenomena that may occur during the modification is still required. Therefore, the quest for the relationship between acid-base properties of natural materials and their adsorbing capacities can simplify the identification of various chemical interactions that may occur during the removal of pollutants from contaminated water. Further often, the determination of the surface functions can help to understand the effect of the modification of raw materials on their adsorbing capacities.

In this regard, several researchers have given attention to acid-base properties of the adsorbent as a tool to identify differences between raw and modified materials. A recent study has proposed a new alternative based on the modification of L. cylindrica by metal oxides (ZnO, Al_2O_3) by precipitation method (Kesraoui et al., 2019). They found that the addition of Zn^{2+} increased the carboxylic and lactonic groups, at the same time, it decreased the phenolic groups. Therefore, an increase in the number of sites has led to an increase in adsorption capacity. On the other side, the addition of Al^{3+} decreased the carboxylic groups resulting in a strong binding to pectin and hemicellulose and an increase of the phenolic groups caused by the high interaction between aluminum and OH groups.

In a recent study, researchers studied the dependence of the acid-basic properties of L. cylindrica with the adsorption capacity of methylene blue and industrial textile wastewater (Othmani et al., 2019b). The raw material was modified later by different percentages of zinc oxide through precipitation method in the presence and absence of alternating current. They found that the alternating current during the modification process of the raw material reduced the number of carboxylic and phenolic groups and increased the number of lactonic groups. The results also showed that the presence of lactonic groups was responsible for the increase of the negative charge of the modified surface material that favorably impacts the adsorption capacity of L. cylindrica. Moreover, the use of fractal mathematical model confirmed the increase of the number of the available sites onto the material surfaces through the decrease of the equilibrium time τ_c and the adsorbed quantities.

Furthermore, the use of L. cylindrica modified by different percentages of zinc oxide for the biosorption of phenol from aqueous solution was demonstrated in another study (Othmani et al., 2020b). The results showed an enhancement of the uptake capacity of the phenol compared to the raw materials as well as a decrease in the time required for the contaminant removal. Based on these studies, the modification of AWMs with metal oxides can be considered among the green performing alternatives used for environmental remediation.

8. Challenges and future prospects

The efficacy of the biosorption process using agricultural waste materials for environmental remediation has been extensively reviewed. Despite the huge number of papers dealing with the use of AWMs, the feasibility of using such materials is still limited to laboratory scale. Table 7 presents some concerns with the use of AWM based adsorbents for the removal of hazardous contaminants and prospects for future studies. AWM derived adsorbents differs in type, characteristics (lignin, hemicellulose, cellulose, wax content) as well as chemical properties (porosity, water retention, rate of crystallinity, ability to mold, tensile strength, surface area, adsorption capacity, specific functional groups, etc.). The biosorbents derived from agricultural species namely, *P. australis*, P. oceanica, L. cylindrical, A. Americana and C. stigmas are identified for their potential use in water decontamination. However, the application of these green materials in particular and agricultural waste, in general, would always depend upon factors such as climatic conditions, socioeconomic status, geography, etc. Despite the abundance, cost-effectiveness, availability, and renewable nature of agricultural waste materials, still, a significant quantity is burned down. Thus, safe management and disposal strategies are essential for these materials. AWMs, if left to degrade and decompose at source, may ultimately create acute and chronic pollution to human life and the surrounding environment. Therefore, their valorization to produce value-added products (e.g., biosorbents) at an industrial scale in an eco-friendly manner may offer potential opportunities for safe management. However, the cost associated with AWM processing and transportation questions the commercial viability of the process and cost-energy-environment input-output balance is of paramount importance before any further reuse. On the other side, environmental pollution due to wastewater is a major global problem especially in heavily populated areas and wastewater treatment is indispensable. Taken together, green materials are considered as potential alternatives to remove organic (phenols, dyes, rhodamine B, crystal violet, etc.) and inorganic compounds with toxic and carcinogenic nature (Cobbina et al., 2015). These adsorbents offer a promising option to deal with the higher operational cost of activated carbon and to provide cost-benefits (Bhattacharya et al., 2008). Numerous AWMs have been explored as low-cost adsorbents such as the shells of fruits, waste resulting from products of cereals coir pith, sawdust, and sugarcane fiber (De Goes Sampaio et al., 2019; Xing et al., 2020). The enhanced removal can also be obtained by mixing different biosorbents from agricultural waste materials. These

Table 7
Overview of main challenges for the use of agricultural waste material derived adsorbents and some prospects for future studies.

Challenges	Prospects for future studies
• Inability to reuse some adsorbents after a certain number of cycles of sorption/desorption. • Stability of agricultural waste materials is unknown. The removal efficiency cannot be considered as a measure of stability. • Decrease of the removal efficiency with real wastewater applications can pose a limit to the use of these materials. • Low adsorption capacities of many natural material based adsorbents. • Suitability of agro-waste derived adsorbents for continuous remediation in industrial applications remains to be established.	• Optimization of the life cycle of agricultural waste materials. • Studies on the regeneration of the adsorbents. Various compatible regeneration techniques can be compared for a particular adsorbent. A combination of regeneration techniques can also be explored. • Investigation of pretreatment processes before the use of adsorbent to overcome the competition of existing anions in the aqueous media • Evaluation of combination of two or more adsorbents for the same decontamination process. Such a solution can offer a high performing ability for wastewater purification due to the combination of different physicochemical properties, characteristics, and performance of the combined materials. This alternative can be better performing in case of coupling more than two metal oxides to prepare hybrid materials useful for wastewater decontamination. • Studies to examine the effect of a mixture of several pollutants in aqueous solution on the efficiency of adsorption. • Studies to identify the suitable treatment and modification strategies for agricultural waste material derived adsorbents to improve their adsorption performance. • Investigation of efficacy of agricultural waste material derived adsorbents in fixed-bed column set up.

combinations tend to improve the adsorption capacity while avoiding possible competition for different catalytic active sites existing in AWM. Besides, the investigation of a binary or multicomponent mixture of adsorbents derived from different agricultural waste materials with different properties in complex aqueous solution contaminated by organic and inorganic pollutants is still at the embryonic stage. More research is needed to improve the reproducibility and sensitivity of the biosorption approach. Therefore, detailed studies are required in the future to study the performance of every waste solely before any possible combination. The adsorptive performance of green materials can be increased by adding functional groups, metal oxides and ligands, which can greatly contribute to higher adsorption capacity and more selectivity. Nevertheless, the preparation of novel biosorbents via surface modification techniques requires extensive attention to make the process a convenient operation with the unique merits of designability and reproducibility. A comprehensive research on the choice of biosorbent and its selectivity is still required. Also, kinetics, thermodynamics, adsorption/desorption behavior, isotherms, and regeneration process such as the choice of an effective regeneration solution, its type, and concentration, the optimum time and volume of regeneration have to be studied carefully (Othmani et al, 2019b, 2020b, 2019b). Moreover, a deep knowledge of the adsorption mechanism and the interaction between the adsorbent and the adsorbate requires extensive investigation since the removal of phenols, Cr(VI), Cd(II) as well as other competitor anions in the same aqueous media is likely to involve a combination of different mechanisms such as physisorption, chemisorption, ion exchange, etc. The interaction between the solution phase and the solid matrix, in turn, depends on the nature of the adsorbent and the adsorbate, reaction time, pH of the solution, temperature, solid\liquid ratio, chemistry of the aqueous phase, etc. Besides, species such as Cd(II) and Cr(VI) do not usually exist alone and they are mainly found with a mixture of ions that compete for the existing sites in the biosorbent. The coexistence of ions makes it imperative to remove or minimize the competitiveness of different ions and suspended solids to achieve success in the biosorption process.

Further, any absorbent should have the capability to be easily regenerated by the desorption process so that it effectively regains the adsorption capacity after the consecutive treatment cycle. Generally, the regeneration of activated carbon and its possible reuse is tedious, expensive, and represents a major concern for researchers who are still looking for commercially applicable ways of regeneration including thermal desorption, acid/alkaline treatment, magnetic separation, electromagnetic desorption, etc. and their potential adaptability to AWM. Following the water treatment process, the recovery of residual adsorbent biomass after the adsorption process, and its environmental stability problems (i.e., leaching activity and metal release) should be studied since these issues may prevent their reuse and their scale-up from the lab to pilot scale.

Considerable efforts have been put over the years in development of novel adsorbents for removal of pollutants and improving their efficiencies. However, the aspects of recovery of useful substances such as heavy metals remain relatively less explored. Heavy metals adsorbed on the adsorbent can be recovered during desorption or regeneration steps. Acids such as sulfuric acid, nitric acid, hydrochloric acid, etc., alkalis such as sodium hydroxide, sodium bicarbonate, potassium hydroxide, etc., salts such as sodium chloride, calcium chloride, potassium nitrate, etc., chelating agents like ethylene diamine tetraacetic acid (EDTA) can be potentially useful as desorbing agents for regeneration of spent adsorbent and recovery of metallic species. A critical evaluation of literature by Lata et al. inferred that acids, alkalis and chelating agent EDTA proved to be efficient for desorption of metals from biosorbents, chemically modified adsorbents and biomass respectively (Lata et al., 2015).

The post-treatment processing of the biosorption should be performed to achieve industrial robustness in the wastewater treatment plant (WWTP) and remediation units. Keeping in view the progress made in biosorption research and environmental remediation, the recovery of biosorbents biomass and their possible reuse into products is promising (Sara et al., 2016). To attempt this, a multi-product approach and detailed studies should be developed to elucidate the technical and economic impact of using residual AWM after the biosorption process for possible applications in the energy, environment, and agriculture sectors under the framework of the circular economy. Therefore, a comprehensive approach, including technological advancement, integrated waste management strategies, policies, and plans are required that can drive forward to find the possibilities of utilizing green materials to fulfill the local energy and biofertilizer demands replacing fossil fuels and synthetic fertilizers respectively and to produce other value-added products through biorefinery approach. Overall, agricultural waste material can be valorized into biogas (e.g., methane from anaerobic digestion) not only offering a valuable resource for renewable energy and decreasing fossil fuel dependence for energy but also contributing to a significant reduction in greenhouse gas emissions. Moreover, AWM is a potential intermediate feedstock to produce a valuable platform chemical for the production of syngas, dimethyl ether, methanol, etc. as well as the production of AMW-based catalysts for the transesterification of oil to produce biodiesel (Odude et al., 2019) which offers new alternatives to the current process used for the transesterification and other related bioprocesses (Magdouli et al., 2018) to meet the global demand of energy. Thus, AWM has potential in bio-based industries and green chemistry to be utilized as a source, support, and feedstock. Furthermore, agricultural waste is rich in phosphorous and nitrogen, and thus, could be utilized as a fertilizer or a soil-stabilizer, however, the sorption of heavy metals in fertilizers needs to be considered and their bioavailability, fractionation, and speciation along with the air, soil, food chain, and the environment must be examined.

Recently, adsorption has been reviewed to be an effective tool not only limited to the environmental sector and WWTP but also in the mining industry where many approaches such as advanced oxidation processes (AOP), physical and chemical technologies can be combined with biosorption to gain performance for inorganic metal removal and to remove anions competitiveness (Etteieb et al., 2020; Madhavan et al., 2019; Zolfaghari et al., 2020). This is generally conducted by changing the physical adsorption-based-zeolite process to a green process based on biosorption and agricultural waste may open new horizons towards the green mine of tomorrow. Further, AWMs can be used as support for microbes used in contaminated soil and water. Nevertheless, these biosorbents are sometimes subject to microbial attack and may lose their performance during the treatment process. Therefore, a performant biosorbent should be chemically and physically stable, resistant to environmental conditions and microbiological degradation upon consecutive adsorption-desorption processes. More attention has to be paid to the management of residual AWMs after desorption processes. Recent alternatives suggest fast and slow pyrolysis for the production of biochar that can be activated and used in turn for the treatment of heavy metals in the different matrices such as soil, contaminated treated wood, water, etc. (Zolfaghari et al., 2020). More studies have to be done on this topic to elucidate a clear idea on the characteristics of green materials after pyrolysis (surface area, porosity, water content, etc.) as well as the effect of pyrolysis conditions (i.e., the temperature of pyrolysis, oxygen, residence time) on the adsorption capacity of AWM based biochar to open further opportunities for the potential use of biochar for other alternatives than heavy metals adsorption. Rather than pyrolysis, carbonization and hydrothermal conversion can be also studied for the production of physically activated carbon with a wide range of applications (Yunus et al., 2020). Besides, thermal or chemical modification of the resultant activated carbon by carbonization or biochar after pyrolysis may lead to the production of highly performant adsorbent that can be impregnated or coated with some metals oxides such as ZnO or TiO2 to treat water contamination (M. Li et al., 2015; Othmani et al., 2019b). Thus, nanoengineered AWM and/or biochar could increase the performance of the remediation process and help to target the contaminant of interest in a short time (Jayaraman et al., 2018). Another area of research could lie in combining natural adsorbents with magnetic properties for the removal of organic pollutants and heavy metal ions. For instance, recyclable magnetic chitosan-based microspheres have been developed for addressing the biocompatibility aspect (Luo et al., 2015). The hybrid heavy metal ion system used a combination of chitosan and cellulose embedded with magnetic gamma-Fe_2O_3 nanoparticles producing adsorption efficiency of 92% (Cu^{2+}), 89% (Cd^{2+}), and 85% (Pb^{2+}) post-regeneration approximately 10 times. Additionally, the kinetics showed regression values > 0.98 giving an excellent fit. Thus, such nanocomposites coupled with magnetic separation technologies could be explored for future work. Further, decontamination approaches should be integrated with water quality monitoring tools such as microfluidic analytical devices to enable onsite assessment of water quality before and after treatment (Balasubramanian et al., 2021; Ghosh et al., 2019). Finally, although the efficiency of biosorption for the removal of contamination at a laboratory scale is promising, more research needs to be done on real contaminated water under operational and climatic variations to generate sufficient data before the industrial implementation of a large-scale wastewater treatment process.

9. Conclusion

Industrial effluents from different sources like tanneries and textile processing units pose a harmful impact on the environment and human health because of the presence of complex organic and inorganic pollutants. Despite the efficiency of conventional treatment processes, the associated high costs and non-environment friendly nature adversely impact their viability. In this study, we have reviewed the adsorptive removal of phenols, chromium(VI) and cadmium(II) using adsorbents derived from agricultural waste. The valorization of agricultural waste-based materials as green adsorbents for the removal of wastewater contaminants has been considered to address the aspects of sustainability, waste utilization, and environmental safety. The literature findings conclude biosorption using agro-based materials to be a promising alternative compared to conventional technologies and carbon-based adsorbents thereby making a review on this topic timely. The adsorbents derived from agricultural waste materials offer unique merits in terms of cost-effectiveness, abundance of raw materials, amenability to surface modification and environmental friendliness. Moreover, after the adsorption process, residual biomass can be either directly converted to energy through thermochemical transformation or can be suitably valorized. The introduction of green materials into wastewater treatment and environmental remediation technology also poses some challenges. Extensive research should be effectively conducted in terms of the cost of processing, regeneration, selectivity, and reproducibility for large-scale applications. The practical application of agriculture waste materials as adsorbents on a commercial scale requires systematic investigation. Recent advances in nano and biotechnology can help to establish a sustainable and scalable route for environmental remediation and recovery of valuable products. Also, commensurate considerations should be given to the design and economical aspects for successful industrial level implementation.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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