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## **Magnetic Biosorbents for Wastewater Treatment**

Mazaher Ahmadi\*, Samira Aghajani, Mohsen Majidi and Hesam Al-Din Hojjat Shamami

*Faculty of Chemistry, Bu-Ali Sina University, Hamedan, Iran*

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Wastewater treatment is a global concern nowadays. Although many methods are available for wastewater treatment, the adsorption method has received much more attention from researchers. Significant problems for extending the adsorption method to the industry for wastewater treatment are the high cost of cosmetically available adsorbents, their recovery from the medium, and their regeneration and reuse. Magnetic biosorbents provide easy separation from the treated water at lower prices compared to other commercially available adsorbents. In terms of characteristics and efficiency in the adsorption of organic and inorganic pollutants, magnetic biosorbents have shown growing promise. If biosorbents and the removed contaminants are not handled and disposed properly, they can become causes of environmental pollution. There are numerous review papers on wastewater treatment with magnetic adsorbents. However, this paper provides comprehensive information on the synthesis of magnetic nanoparticles, the magnetization of biosorbents, and the application of magnetic biosorbents for wastewater treatment to pave the avenue for new magnetic biosorbents development that can be scale up from laboratories to the industry.

**Keywords:** Iron oxide nanoparticles, Biomaterials, Waste materials, Magnetic biosorbents, Wastewater treatment

### **INTRODUCTION**

Water pollution is the world's leading ecological problem currently that has to be handled on a sustainable development priority basis. A significant problem is the provision of non-contaminant water for household and industrial usage. Furthermore, wastewater with pollutants, including pharmaceuticals, heavy metals, pesticides, organic waste, bacteria, *etc.*, has been discharged from different sources, untreated or insufficiently treated, leading to fast degradation of freshwater resources. The treatment of wastewater is mainly based on traditional procedures such as membrane separation, oxidation, coagulation-flocculation, floatation, ion exchange, sedimentation, adsorption, reverse osmosis, and filtration. These conventional methods cannot conceivably provide the necessary decontamination range to achieve both precise and affordable requirements. New

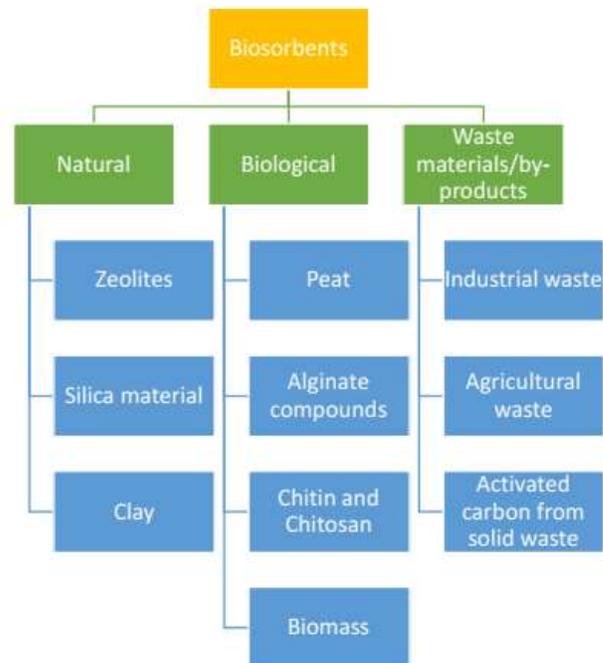
solutions for wastewater treatment are UV photocatalysis or photolysis, adsorption with activated carbon, ozonation, oxidative degradation, and electrochemical methods. However, they are still not cost-effective [1-5]. In the last decade, these methods have been superseded by low-cost biomass adsorbents, in particular, biomass from agricultural wastes. Cellulose is the principal constituent of biomass. It contains hemicellulose, lignin, hemicellulose, minerals, starches, hydrocarbons, proteins, lipids, *etc.* In addition, it contains some surface functional groups that favor contaminant adsorption. Waste biomasses such as rice husk, bagasse, banana shell, walnut shell, sawdust, eggshell, *etc.*, are commonly utilized both in their most basic state and modified for wastewater pollution treatment. Ideally, the water treatment biosorbents should meet the following standards: target pollutant specificity/selectively, high adsorption efficiency, fast adsorption, cost-effectual, ecologically non-toxic, reusability, and easy separation from the treated water [2-5].

\*Corresponding author. E-mail: m.ahmadi@basu.ac.ir

In recent years, several investigations have shown that heavy metals, dyes, pesticides, antibiotics, and other new pollutants have been removed by synthesized magnetic biosorbents (MBSs) [6-10]. The adsorption capacity depends greatly on the type of the contaminant, input materials for the preparation of the adsorbent, type of modifications of the adsorbent, and resultant functional groups. In addition, it is possible to modify and adapt MBSs to remove specific pollutants. Several review papers on wastewater treatment with magnetic adsorbents are available [1-5]. However, this paper provides comprehensive information on the synthesis of magnetic nanoparticles, the magnetization of biosorbents, and the application of MBSs for wastewater treatment to pave the avenue for new MBSs development that can be scale up from laboratories to the industry.

## BIOSORBENTS

The biosorbents have been classified as natural, biological, and waste materials/by-products (Fig. 1) [2]. Several natural raw materials have adsorbent properties and are readily available in the environment. A wide variety of natural adsorbents such as zeolites, siliceous material, and clay have been used to remove a range of contaminants, including heavy metals, dyes, and pharmaceutical compounds from wastewater or water. Clay is the most plentiful biosorbent that occurs naturally that makes it an integral adsorbent at low cost, strong adsorbing properties, high porosity, wide surface area, and easy chemical functionalization. Clay is made of a layered structure classified as kaolinite, mica, and smectites based on their structural differences [11]. Dolomite, glasses, silica perlite, and alunite are natural silica minerals. Silica beads have silanol functional groups, which have irreversible and non-specific adsorption, resulting from their chemical reactivity. Silica beads offer suitable adsorbents thanks to their availability, high abundance, low price, mechanical stability, and high adsorption capacity. However, their applicability is limited to neutral to acidic mediums since they are dissolvable in basic mediums. Zeolites have high porosity 3D structure with a negatively charged lattice. They are crystalline hydrated aluminosilicate. The structure of zeolites frameworks includes pores occupied by alkaline and alkaline earth cations and water. High surface area, high ion exchange



**Fig. 1.** Various types of frequently used biosorbents in wastewater treatment.

capacity, and low price, make zeolites attractive adsorbents [12].

Biological adsorbents come mostly from microorganisms such as fungus, bacteria, algae, yeasts, and animal shells. Chitin is the second most frequently used cellulose polymer, a significant natural biopolymer. Chitin polysaccharide is rigid, white, non-elastic, nitrogenous, and constitutes material that supports crustaceans, insects, and so on. Chitin is a copolymer of N-acetyl-glucosamine N-glucosamine. Chitosan is a chitin that includes 50 percent or lower units of N-acetyl-glucosamine in the biopolymer [13]. Chitin and chitosan are mainly isolated from crustaceans. Crustacean's exoskeleton is accessible in massive quantities as a food processing metabolite. Many research studies have shown the utilization of biosorbents from chitin as effective adsorption materials. As chitosan is soluble in acidic conditions, it cannot be utilized in acidic mediums. Chemical modification of chitosan can solve the problem of its solubility in acidic mediums and also regulate diffusional characteristics, increases removal performance, and reduces the sorption sensitivity to the environment. Alginate compounds are found primarily on brown seaweeds, consist of linear

polysaccharides, and are highly water-soluble [14]. Peat is a biosorbent that is plentiful, relatively cheap, and widely available. Peat is made of lignin, humic and fulvic acids, and cellulose in its natural state. Polar functional groups, alcohols, carboxylic acids, aldehydes, ketones, ether, and phenolic hydroxides are found in lignin and humic acid. Peat is divided into four categories based on the type of the source material: woody, moss herbaceous, and sedimentary peats [15]. Many research studies have proven the dye adsorption efficacy of peat. Raw peat, on the other hand, has some drawbacks, including poor chemical stability, low mechanical strength and swell, a strong affinity for water, a propensity to shrink, and the capacity to absorb humic compounds. Chemical modifications of peat produce consistent outcomes by increasing adsorption capacity and selectivity. Biosorbents both live or dead biomasses, such as white-rot fungus and different microbial cultures, can also be used in the bioadsorption process. When microbial biomass is created during fermentation operations for antibiotic and enzyme production, a considerable amount of by-products is produced. These by-products can be utilized for pollutant bioadsorption. A wide range of fungal organisms can decolorize a wide range of dyes. Different functional groups in the fungus' cell wall, such as thiol, amino, phosphate, and carboxyl groups, help the dye attach to the fungus. Although dyes biosorption onto the cell surface appears to be a quick process, it has been proven that the complete process takes a long time. Even under constrained growth circumstances, fungal biomass may be utilized for dye adsorption, which is useful when the effluent is cytotoxic. The inactive biomass can be regenerated and reused without any extra nutritional input. Toxic chemicals have been observed to adsorb and biodegrade in living cells. The wide distribution of photosynthetic organisms like cyanobacteria and algae is gaining popularity owing to their biodegrading and biosorption properties. Dye adsorption is possible in algal biomass. When the solution or effluent is hazardous and does not promote the growth of biosorbent, the use of biomass is highly intriguing. The use of dead biomass in such a situation solves the problem. Using a low-cost growing medium, producing fungal biomass as a biosorbent is a simple and cost-effective approach [2,16].

Agricultural wastes, fruit, and vegetable peels are examples of abandoned waste materials that might be useful

for various purposes. After some processing, these materials might be used as low-cost adsorbents [2]. The primary goal of using these agricultural by-products is to reduce their buildup and repurpose them for profit. As a result, they are being used as biosorbents to remove contaminants from the environment. In 1900-1901, bone char was replaced with activated carbon in the sugar refining process [17]. Activated carbon is the crude form of graphite with a random or amorphous structure. Activated carbon is highly porous, with pore diameters ranging from slits to apparent fissures and crevices with molecular dimensions. Any carbon-based substance such as carbon black, wood char, petroleum coke, fish, bone char, and fertilizer waste can be used as a starting point for making activated carbon. Activated carbon is the most frequently used sorbent for the adsorption of contaminants from wastewater among all sorbent materials. There have been several reports of coal-based adsorbents being used. Activated carbon is efficient in removing both organic and inorganic pollutants in wastewater effluents. Activated carbon remediation is one of the best-controlled methods in the world, according to the US Environmental Protection Agency [18]. Coal is primarily utilized in the production of activated carbon. However, it is frequently non-selective and useless; moreover, the better the quality, the higher the expense [2]. Agricultural solid waste biosorbents are inexpensive and widely accessible. Tree fern, an agricultural byproduct, is also utilized to absorb contaminants. Lignin and cellulose are found in tree ferns. Bark, a polyphenol-rich by-product of the lumber industry, is highly efficient in dye and other chemicals removal from aqueous solutions. Tannin in the bark is primarily responsible for the bark's adsorption capabilities. The sole investment expense associated with such forest trash is the cost of transportation to and from storage buildings and other places where the waste will be valorized and utilized. Industrial wastes like sawdust from the wood sector are commonly utilized as adsorbents. Sawdust includes polyphenolic organic substances that aid in the dye molecules binding onto sawdust through several processes. If the incubation pH is higher than its neutral pH, sawdust becomes either positively or negatively charged. One of the primary issues with sawdust materials is that pH levels are pretty crucial in sorption processes. Blast furnace, Red mud, metal hydroxide, fly ash, and sludge slag are among the other industrial wastes

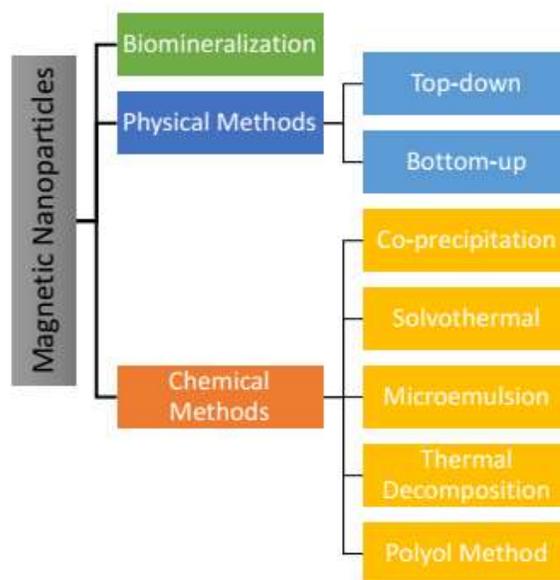
utilized as adsorbents [2].

## METHODS FOR THE SYNTHESIS OF MAGNETIC NANOPARTICLES

Methods for the production of magnetic nanoparticles (MNPs) may be divided into three categories: physical techniques, biomineralization, and chemical approaches (Fig. 2) [19]. The next part provides a brief overview of the main approaches for the synthesis of MNPs.

### Biomineralization

This method is based on MNPs production using live microorganisms [20]. Magnetotactic bacteria, which have recently been identified as excellent model organisms for magnetite biomineralization, are among those living microorganisms [21]. Magnetotactic bacterium magnetosomes generated by this method are MNPs surrounded by biofilms and arranged in a chain-like form. The resulting MNPs have a regular form, ordered particle size, and excellent crystallinity. The phospholipids and fatty acids found in the magnetosome's membrane operate as a nanoreactor, regulating the precise production of MNPs. Furthermore, proteins in the magnetosome membrane play a significant role in the biomineralization process by regulating iron transport, nucleation, redox reaction, and MNPs synthesis. The magnetosome, which is protein-coated with iron oxide, is an example of MNPs produced by bacteria. Considering that bacteria naturally use MNPs as a compass to find their preferred home in the anaerobic environment of depths [22]. MNPs were effectively produced in the laboratory in the 20-50 nm range under anaerobic circumstances, comparable to their original habitat [23-25]. Furthermore, magnetosomes have been discovered to have good magnetic behavior, which could be employed in biomedical applications [26,27]. Current magnetosome research efforts are focused on the clarification and improvement of the biomineralization method to enhance wet chemical synthesis procedures that replicate the biologic environment, providing MNPs with the same magnetic characteristics [28,29]. However, The magnetosome biomineralization process is yet unknown, and large-scale synthesis of magnetosomes requires additional research [30].



**Fig. 2.** Frequently used methods for synthesis of magnetic nanoparticles.

### Physical Methods

The two primary methods for the physical synthesis of MNPs are top-down and bottom-up methods. MNPs are produced at the nanoscale using the top-down method, which involves milling bulk magnetic materials into tiny particles. The primary drawback of this method is that it is difficult to regulate the ideal particle shape and size [31]. Milling processes produce lattice flaws, which cause magnetic properties to deviate from typical particles of the same size [32]. In bottom-up methods, on the other hand, MNPs are created by condensation in either the gaseous or the liquid phases. Surprisingly, laser evaporation is regarded as one of the most promising bottom-up techniques for MNP preparation. In this process, a laser beam evaporates a powder of rough metal oxides of a few micrometers [33,34]. The particle size produced by evaporation techniques ranges between 20-50 nm [35].

### Chemical Methods

The chemical techniques are similar to the bottom-up synthesis strategies of MNPs. We will address the most prevalent chemical methods briefly.

**Co-Precipitation.** The co-precipitation method is a straightforward approach for producing MNPs. In this method, an aqueous medium is the most often employed. Magnetic iron oxides are usually synthesized by co-precipitation of a mixture of ferrous and ferric iron salts, followed by adding of the base in water. The crystal phase and particle size of the nanoparticles may be altered by varying the pH, iron salts ratio, the kind of base employed, and the temperature [36]. By adjusting the reaction conditions, the particle size may be raised to 40 nm. Many changes have been made to this technique in recent years. Single crystals of superparamagnetic with sizes between 10 to 15 nm may be produced by applying high pressure during the precipitation process [37] or by slowing the reaction conditions [38], exhibiting high promising properties for medical applications [39,40].

**Hydrothermal.** Hydrothermal synthesis occurs in an aqueous phase at temperatures above 200 °C in autoclaves under high pressures exceeding 2000 psi. This method demonstrates the capacity of water to be hydrolyzed and dehydrate metal salts at elevated temperatures. Because of the low solubility of the discovered metal oxide particles in certain solvents at such temperatures [41,42], precipitation occurs. The particle size and morphology may be tailored by varying temperature, concentration, and autoclaving duration [43,44]. Increasing the autoclaving duration increases particle size but also widens the size distribution. The diameters obtained ranged between 10 and 50 nm, and by shortening the autoclaving durations, monodisperse particles were produced [44].

**Microemulsion.** This method is based on a two-phase system that produces tightly monodisperse MNPs. A water-in-oil microemulsion system is created by dropping nanosized water droplets over the oil phase, with surfactants acting as a stabilizers at the water/oil interface [45]. Because of the limitations in nanoparticle nucleation and growth, the MNPs produced from this system have a relatively uniform shape [46]. Because of their narrow size distribution, MNPs synthesized using the microemulsion methods are a potential tool for various medicinal applications [47].

**Thermal decomposition.** Thermal decomposition includes boiling organometallic compounds in an organic solvent to produce MNPs from nonmagnetic precursors. The particles obtained have a limited size distribution. Iron

carbonyls and acetylacetonates are frequently utilized as nonmagnetic precursors, with fatty acids acting as surfactant molecules. The shape and size of the synthesized MNPs can be modified by altering the ratio of precursors, surfactants, and solvents. Thermal breakdown of nonmagnetic precursors yields highly pure iron metal. The resulting metal particle can then be oxidized to iron oxide in oxidative conditions at high temperatures. The thermal decomposition of reagents containing cationic iron centers like tris(acetylacetonate) iron introduces a one-step method for synthesizing MNPs [48]. Park *et al.* [49] produced excellent results in the synthesis of MNPs by thermal decomposition, on which various modifications have been made by some research groups, resulting in nearly monodisperse MNPs smaller than 30 nm.

**Polyol method.** The polyol method involves the preparation of MNPs in high-boiling multivalent alcohols. The oxidative base of iron salts is hydrolyzed in a polyol combination of N-methyl diethanolamine and polyethylene glycol in this technique. The size and structure of the produced MNPs can be altered by changing the reaction conditions or the solvents used. However, the resulting particles have a low monodispersity. This technique may produce MNPs with a floral form under particular reaction conditions [50], which have an excellent heating effect for hyperthermia [51]. These particles having a multicore structure are composed of single cores of 8 to 10 nm sizes, which may form a cluster with a size of up to 30 nm. As previously stated, these particles have intriguing properties for hyperthermia therapy [39]. Other synthesis methods for MNPs, such as microwave-assisted synthesis and laser pyrolysis [52], the sol-gel technique [53], and glass crystallization [54], are not discussed in this paper since the resultant particles are rarely employed in analytical applications.

## SYNTHESIS OF MAGNETIC BIOSORBENTS

For the synthesis of MBSs, the MNPs can be synthesized in the presence of pre-treated biosorbents. Alternatively, many previously reported techniques could be used to magnetize initially diamagnetic biological materials. The presence of magnetic labels (particularly MNPs) within the treated biomaterials' pores, on the biomaterials' surface, or

within the biopolymer gels causes magnetic modification in most situations. The classic chemical co-precipitation technique for magnetization is used to create magnetic iron oxide particles by incubating a stoichiometric combination of ferric and ferrous ions in aqueous alkaline media in the presence of modified biological components, followed by heating. It is possible to produce magnetic biocomposites with various amounts of iron oxides on their surface. Procedures that have been slightly changed, such as the use of inert gas in the synthesis, have been detailed elsewhere. Ferrites can be utilized as a magnetic label. They can be synthesized by substitution of ferrous ions with other divalent cations. Water-based magnetic fluids are an effective post-magnetization technique; in many cases, the modification was done in methanol or simply by combining ferrofluid with the modified biomaterial and drying it. Recent research studies have reported the rapid and easy production of magnetic iron oxide particles using the microwave irradiation of ferrous sulfate at high pH. Recently, one-pot direct magnetization techniques using  $\text{Fe}^{2+}$  salt at high pH in the presence of modified materials have been proposed. In this method, the suspension is microwaved for the necessary time in a standard home microwave oven. Individual particles and aggregates of microscale MNPs produced during microwave irradiation deposit on the surface of the treated material. To magnetize sensitive materials, an indirect microwave-assisted modification was developed. Firstly, magnetic iron oxide nano- and microparticles were synthesized in a microwave oven from  $\text{FeSO}_4$  at a high pH. Materials to be magnetized were then extensively mixed with MNPs suspension after the particles were washed and dried entirely at a slightly higher temperature. Microwave-generated MNPs contained in a water-soluble organic solvent are used to transform agglomerate-forming diamagnetic biomaterials into their magnetic. The same process could be used to modify materials that are sensitive to high temperatures. However, this method employs subzero temperatures to dry the treated material. Magnetic derivatives of diamagnetic biosorbents could also be performed via mechanochemical methods. At room temperature, hydrated ferric and ferrous chlorides are crushed in a mortar with an excess of NaCl; after adding the target diamagnetic powdered material, the combination is ground for the necessary duration. The mixture is then homogeneously added to powdered alkaline

hydroxide, and the mechanochemical treatment continues.

## APPLICATION OF MAGNETIC BIOSORBENTS FOR WASTEWATER TREATMENT

### Removal of Inorganic Cations

MBSs remove a broad spectrum of cations from wastewater, including  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Cr}^{6+}$  [3]. Han *et al.*, for example, produced magnetic activated carbon MBSs to remove lead cation, with adsorption capabilities ranging from 11.6  $\text{mg g}^{-1}$  to 35.7  $\text{mg g}^{-1}$  [55]. Novel multifunctional MBSs can remove more than 97 percent cobalt from wastewater through the formation of bis and tris complexes with surface carboxylic acid functional groups of the adsorbent [56].  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ , and  $\text{Cr}^{6+}$  removal efficiencies using biosorbents modified with zerovalent iron were 99.4, 99.1, and 66.7 percent, respectively, by complexation with -OH/-COOH functional groups or delocalized p electrons. Heavy metals can be absorbed by iron-based functional groups by moving into  $\text{Fe-O-M}^{n+}$  complexation [57]. MBSs have a greater removal efficiency than their raw counterparts. Yan *et al.*, for example, synthesized MBSs containing ZnS nanocrystals that had 10 times higher adsorption capacity than the unmodified biosorbents [58]. Through a coordination bond between the lattice oxygen and C=O group (electron acceptor), iron chloride modified MBSs enhanced mercury adsorption capability [59]. Compared to unmodified biochar, zinc borate pre-treatment increased biochar removal capability and enhanced the availability of the surface oxygen group, which increased the adsorbent capacity for nickel ions removal by 3-10 times [60]. Similarly, compared to pristine biochar, manganese-modified wood-derived biosorbents increased adsorption capacity by 2.8 times for copper, 2.1 times for lead, and 5.9 times for cadmium due to the induction of more oxygen-containing functional groups and a greater surface area [61]. Due to increased oxygen-containing functional groups, the adsorption capability of lead by manganese-induced biosorbents was enhanced up to 2-20 times in other investigations [62,63]. Furthermore, nitrogen-induced magnetic biosorbents enhanced lead adsorption capacity (893  $\text{mg g}^{-1}$ ) within a short equilibrium time of less than 10 min [64] due to surface coordination of  $\text{Pb}^{2+}$  and O=C-O or C=O, as well as the presence of aliphatic

N [64]. The primary separation processes were Cd-O/Pb-O or hydroxyl binding and ion exchange reactions, which revealed an adsorption capacity of 45.8 mg g<sup>-1</sup> of Cd and 248.0 mg g<sup>-1</sup> of lead for amorphous MnO<sub>2</sub> modified biosorbents [65]. This suggests that by designing functional groups of biosorbents, greater adsorption capacity for cation removal from wastewater can be achieved [64].

The efficiency of Cd removal by MBSs was compared to that of different sorbents, revealing that MBSs outperform most conventional adsorbents [3]. MBSs provide better adsorption efficiencies than polymers, activated carbon, plant wastes, minerals, nanocomposites, and nanomaterials. Due to their complex structures and various elemental compositions, microbial wastes have a high adsorption capacity, which helps to create more active sites to adsorb ionic pollutants. MBSs are excellent for removing Cd from wastewater because of their large surface area and ease of separation. Due to competition with cadmium ions in the adsorption medium, a moderate positive connection was discovered between the weight percent of hydrogen and adsorption capacity for Cd removal. In contrast, a limited negative correlation was established between cadmium adsorption and weight percent of metal nanoparticles. As a result, metal nanoparticle impregnation in MBSs should be limited to minimize competition for the active sites of the adsorbent [3]. Co-precipitation, surface complexation, and chemisorption are all methods for adsorbing cations [66]. Chemisorption, chelation, ion exchange, complexation, and hydrogen bonding are all methods for attracting and bonding cations [67]. The sorption processes of cations can be influenced by metal-induced multifunctional groups in MBSs. Apart from raw biomass-derived biochars, the presence of iron and other metals and salts in industrial waste and magnetic biochar generated functional groups that are more varied, carboxyl, ester, metal, and amine groups are the major functional groups that contribute to cation elimination [3]. Ion exchange reactions are caused mainly by the hydroxyl group, carboxyl group, amino group, carboxylic acid, and M-O/OH, whereas electrostatic attraction is caused by carboxylic acid and carboxyl groups. Carboxyl, hydroxyl, amino, carboxylic acid, and amino groups can all help in cation adsorption by forming complexes on the surface of adsorbents. Chelation is caused by carboxyl, hydroxyl, and carbonyl groups [68]. The inclusion of  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> nanoparticles enhanced the adsorption

capacity of catalyst-doped magnetic carbon composites compared to simply magnetic biochar [66]. Similarly, employing  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> instead of bare iron oxide nanoparticles increased Cd adsorption capacity from 31.80 to 167.22 mg g<sup>-1</sup> [66]. In general, nanoparticles containing biochar have a lower adsorption capacity than MBSs that are further modified for cation removal at many steps. The adsorption capacity of Fe<sub>3</sub>O<sub>4</sub>@layered double hydroxide@biocomposite was improved to 258.00 mg g<sup>-1</sup> [69], whereas the adsorption capacity of magnetic oak wood biochar was 66.22 mg g<sup>-1</sup> due to functional group variation [69]. Similarly, magnetic EDTA/Chitosan/TiO<sub>2</sub> exhibited an adsorption capacity of 209.21 mg g<sup>-1</sup>, but Fe<sub>3</sub>O<sub>4</sub> coated sawdust carbon had an adsorption capacity of just 51.00 mg g<sup>-1</sup> [70]. When EDTA, TiO<sub>2</sub>, or other chemicals were added to magnetic biochar, more functional groups were provided, which promoted adsorption. Similarly, MnFe<sub>2</sub>O<sub>4</sub> biochar had an adsorption capacity of 181.49 mg g<sup>-1</sup> due to Mn-doped iron oxide that induced the Mn-O functional group. In contrast, ferromanganese ferromanganese binary magnetic biochar had an adsorption capacity of 101 mg g<sup>-1</sup> [71,72].

The pH of the solution might affect the sorption of cations by MBSs. The deprotonation/protonation of surface functional groups influences cation adsorption performance, demonstrating that electrostatic interaction is one of the most critical processes for cation adsorption. Naidu *et al.* investigated the interrelationships between pH, point of zero charges (pzc), background electrolytes, and cadmium adsorption capacity [73]. However, due to the magnetization of biosorbents, the pzc of MBSs may drop [55]. The surface of MBSs is protonated at pH < pH<sub>pzc</sub>, making it electropositive and causing electrostatic attraction for anions but electrostatic repulsion for cations. The hydroxyl group on the MBSs surface covers the adsorbents at pH > pH<sub>pzc</sub>, resulting in electrostatic attraction for cations but electrostatic repulsion for anions. The electrostatic attraction and repulsion, as well as the competitive impact of similarly charged ions, may be controlled by co-existing cations and anions. The electrostatic attraction or repulsion may be regulated by creating a monolayer of coexisting cations and anions on the MBSs. Although the effects of pH and pH<sub>pzc</sub> on the removal of cations and anions are firmly linked, the influence of solution pH on the adsorption of non-ionic

organic pollutants is negligible [3].

### Removal of Inorganic Anions

This section summarizes the removal of main anions from water and wastewater by MBSs, which revealed that MBSs had a greater adsorption capacity than raw biomass or pure biochars. Because of the enhanced functional groups and nano-flakes on the biosorbent's matrix after modification with Al/Mg/Fe metals, magnetic biochar has a considerably greater phosphate adsorption capacity ( $290\text{-}887\text{ mg g}^{-1}$ ) than pristine biochars ( $290\text{-}887\text{ mg g}^{-1}$ ) [74-77]. MBSs have a high nitrate adsorption capacity, recorded as  $95\text{ mg g}^{-1}$  on  $\text{MgCl}_2$ -treated biosorbents [78]. Similarly, biosorbents modified with zinc nitrate/iron/calcium agents increased Cr(III) and Cr(VI) sorption capacity to nearly twice that of virgin biochars [79-81]. The magnetic biochar has a greater adsorption capacity for As(V) and Cr(VI) than the non-magnetic biochar [80]. Wen *et al.* created magnetic porous carbonaceous materials with enhanced adsorption capabilities of  $38.03\text{ mg g}^{-1}$  for As(V) and  $21.23\text{ mg g}^{-1}$  for Cr(VI). To remediate Cr(VI) from water, Zhong *et al.* produced magnetic biochar composite by single-step microwave pyrolysis of iron sulfate and rice husk (feedstock materials). Due to increased surface area, porous graphitic structure, pore volume, and reactive magnetite to sorption or reduction of Cr(VI), magnetic biochar has 11.7 and 3.2 times greater reduction and sorption capacity of Cr(VI) [82]. Furthermore, anionic dye and humic acid may be separated using positively charged magnetic adsorbents [83].

The sorption of As ( $\text{H}_2\text{AsO}_4^-$  and  $\text{HAsO}_4^{2-}$ ) by magnetic adsorbents is utilized as a case study to better understand the link between sorption capacity and other variables. With several kinds of metal oxide integrated into biosorbents, MBSs exhibited excellent adsorption ability for arsenic ranging from 5 to  $500\text{ mg g}^{-1}$  [84,85]. The adsorption capacity of a zero-valent iron (nZVI) modified biochar is  $124.50\text{ mg g}^{-1}$ , which is greater than that of other metal oxide impregnated biosorbents [86,87]. However, nZVI is quickly oxidized and forms aggregation following physical contact with a variety of pollutants (mainly organic), whereas metal oxide nanoparticles pose no such risk. When it comes to biosorbents, metal oxide is more useful than zero-valent iron [88]. Another research found that sawdust modified with  $\text{MnFe}_2\text{O}_4$  had an adsorption capability of  $507\text{ mg g}^{-1}$  for As

[89]. For the production of MBSs, metal doping iron oxide is more efficient than raw metal oxide nanoparticles [89,90].

The relationship between magnetic biosorbent characteristics and arsenic sorption capabilities was investigated using a software. A positive association was discovered between the weight percent of Fe, the weight percent of H, and the pore size and arsenic sorption capacity. Due to the opposing charges of metal nanoparticles and arsenic, the correlation values show that the weight percent of iron content favorably influences arsenic adsorption without competing with arsenic on the active sites of the adsorbents [3].

Metal oxides, nanocomposites, laterite soil, minerals, residual materials, biochar, red mud, and other low-cost adsorbents are all examined for removing arsenic from wastewater [3]. Although activated carbon, polymer, industrial waste materials, and thermally and chemically (base and acid) modified biochar were frequently utilized, their removal efficiency was lower. In contrast to other sorbents, metal oxide nanoparticles, nanocomposites, modified iron oxide nanoparticles, and magnetic biochar showed greater arsenic removal efficiency. The highest adsorption capacity for arsenic removal was found in metal nanomaterial and nanocomposites-derived adsorbents. Carbon nanotubes modified with various metal oxides have a sorption capacity of less than  $1\text{ mg g}^{-1}$  [91-93]. Minerals such as gibbsite, kaolinite, goethite, hematite, and zeolite, with adsorption capacities of less than  $7\text{ mg g}^{-1}$ , can also adsorb arsenic [94-96]. The adsorption capacity of metal oxide nanoparticles was about  $5\text{-}20\text{ mg g}^{-1}$ , but modified iron oxide had a significantly greater adsorption capacity for arsenic removal between 50 and  $250\text{ mg g}^{-1}$  [97-100]. This results might be owing to the formation of additional functional groups during the modification with other nanomaterials, photocatalysts, acids, bases, and graphene [3].

The major functional groups for adsorbing arsenic species from water include the amino group, carboxyl group, carboxylic acid, and thiol groups. At low pH, electrostatic interactions are responsible for the removal of anionic pollutants. Still, at high pH, electrostatic interactions are effective for the removal of cationic contaminants such as copper, lead, cadmium, and zinc ions [101-103]. Adsorption capability for anions, for example, was poor at high pH but high at low pH [85,104]. Zhou *et al.* noted the influence of

background electrolytes at the pzc. In the presence of co-existing anions, the adsorption of anions is lower than that of cations [105]. Electrostatic repulsion between the adsorbents and the anions causes these occurrences. As a result, the phenomenon can only be described at  $\text{pH}_{\text{pzc}}$ , higher, or lower. When the pH of the biosorbents is less than  $\text{pH}_{\text{pzc}}$ , the surface of the biosorbents is positively charged, favoring arsenic adsorption. The presence of anions may reduce adsorption capacity owing to anions' competition. Still, the presence of cations may enhance adsorption capacity due to cation monolayer development on the negatively charged adsorbents' surface. When  $\text{pH} > \text{pH}_{\text{pzc}}$ , the presence of cations improved arsenic adsorption capacity by creating a monolayer of positively charged cations on the adsorbents that may bind anions via electrostatic interactions, but the presence of anions may reduce anions adsorption.

### Removal of Organic Pollutants

Many organic chemical substances, such as endocrine-disrupting chemicals, organic water-soluble dyes, crude oil derivatives, pharmaceuticals, and personal care products, so on, have been recognized as ecologically harmful [1]. Organic dyes are a major category of pollutants that the textile dyeing and finishing industry produces in large quantities. Environmental contamination is also caused by the leather, paper, paint, cosmetics, plastics, ink, and pharmaceutical sectors. Simultaneously, these contaminants, together with heavy metal ions, are one of the most studied model adsorbates [106]. Despite the availability of various removal methods, biosorption is still thought to be the most effective and cost-effective. Various inorganic and organic compounds has been thoroughly investigated as potential organic dye adsorbents. In general, excellent adsorbents should have high adsorption effectiveness, be inexpensive, and be accessible in large quantities. These needs could be met using biomass from the food and agriculture industries [106]. Waste materials that have been magnetically modified can be easily manipulated using an external magnetic field.

Diverse magnetized by-products or waste biomaterials have been utilized for dye adsorption, such as waste tea leaves [7], spent coffee grounds [107], corn straw [108], peanut husks [109], sugarcane bagasse [110], *Platanus orientalis* leaves [6], and sawdust [111]. Preparation of their magnetic derivatives was performed by treatment with

perchloric acid [112], or tetramethylammonium hydroxide [113] stabilized magnetic fluids,  $\text{Fe}_3\text{O}_4$  particles prepared from different precursors, namely  $\text{FeSO}_4$  under microwave irradiation and NaOH alkalization [114],  $\text{FeCl}_3$  and  $\text{FeCl}_2$  alkalized with ammonia [115], and  $\text{FeCl}_3$  and  $\text{FeSO}_4$  alkalized with ammonia [116] or  $\text{CuFe}_2\text{O}_4$  particles synthesized from  $\text{FeCl}_3$  and  $\text{CuCl}_2$  precursors alkalized with NaOH [111]. Compared to native materials, the maximum adsorption capacity of magnetically modified biomaterials may somewhat decrease. The partial blocking of adsorption sites by trapped magnetic iron oxide particles is most likely the source of this condition. However, the presence of magnetic particles in the magnetized biomaterial might improve the adsorption capacity of certain compounds in some situations [117].

Because plant-derived materials have reduced adsorption of target chemicals, they are frequently modified or processed before being used for xenobiotic elimination. The bulk of strategies for improving adsorption efficiency are based on treating the material with different hydroxides, acids, or a mix of them. However, other methods, such as carbonization or hydrolysis, have also been described. The highest adsorption capacity of all the dyes studied increased more than four times when barley straw [118] or rye straw [119] were modified with citric acid and then treated with NaOH. An increase in carboxylic group quantity and a rougher adsorbent surface were credited for the substantial efficiency boost. Furthermore, there were only minor variations in adsorption effectiveness between native and magnetic counterparts. Yu *et al.* investigated the adsorption process utilizing sugarcane bagasse magnetic derivatives modified with pyromellitic dianhydride. They found that the adsorption effectiveness for basic magenta and methylene blue removal was more than six times that of the native material. The introduction of new carboxylic groups produced increased adsorption following pyromellitic dianhydride modification. The adsorptive elimination of both dyes was likewise minimal when magnetic particles were used alone [110].

### Removal of Radionuclides

In uranium mining and milling sites, as well as nuclear waste management facilities, removing radionuclides from

nuclear waste solutions is a major environmental problem. Only a few scientific articles have detailed the usage of magnetic derivatives of waste biomaterials, although a broad spectrum of magnetically sensitive organic materials has been identified [1].

Wang *et al.* used microwave-synthesized magnetic iron oxide particles to magnetize wheat bran and then investigated uranium adsorption. Adsorption effectiveness was shown to increase with rising pH value and to improve marginally with increasing temperature. The Langmuir isotherm adequately represented the adsorption process, suggesting monolayer adsorption, with a maximum adsorption capacity of 29 mg g<sup>-1</sup> [120].

Yamamura *et al.* investigated uranium adsorption on sugarcane bagasse that had been thermally treated at 300 °C for 30 min and then magnetized using an in situ co-precipitation method [121]. At pH 5, the best adsorption efficiency was found. The Langmuir equation was used to explain the equilibrium isotherm, and a maximum adsorption capacity of 17 mg g<sup>-1</sup> was achieved. Rahnema *et al.* investigated sugarcane bagasse carbonized at 300 °C and in situ magnetically modified with co-precipitated Fe<sub>3</sub>O<sub>4</sub>. The adsorption process was described using the Langmuir equation, the optimum pH value was found to be pH 3, and the maximum adsorption capacity for uranium was 72 mg g<sup>-1</sup> [122].

Cheng *et al.* used chitosan as a bridging agent to investigate the adsorption of strontium using Fe<sub>3</sub>O<sub>4</sub>/sawdust. Magnetic particles were added to chitosan dissolved in an acidic medium containing sawdust using a co-precipitation method in a nitrogen environment. The final magnetic composite was produced with the addition of sodium hydroxide, followed by washing with water and drying at 80°C. The Langmuir model was used to explain the adsorption process, and the maximum adsorption capacity of approximately 13 mg g<sup>-1</sup> was determined [123].

## CONCLUSION AND FUTURE PERSPECTIVES

In terms of characteristics and efficiency in the adsorption of organic and inorganic contaminants, MBSs have shown growing promise. If biosorbents and the removed

pollutants are not handled and disposed of properly, they can become causes of environmental pollution. More study is needed to ensure the management and safe disposal of MBSs and adsorbed contaminants [124]. Magnetic sedimentation and centrifugal sedimentation can be used to separate and reuse of iron oxide/iron biosorbents. MBSs recovery is a time-consuming and challenging procedure. The recovered MBSs may be utilized to make bricks and cement in the construction sector, but excessive volumes could compromise the mechanical characteristics of the products. Its usage in agriculture for moisture retention and soil aggregation may cause pollutants to accumulate in the soil, posing a significant threat to aquatic and human life. The pollutants can then be disposed of in a controlled setting once they have been isolated. Another viable method is to bury the bioadsorbents in the soil and then employ plants for the phytoremediation of the pollutants. These plants can then be utilized as raw materials for the biochar manufacturing process [124,125].

The cost-effectiveness of adsorbents is often ignored, but it should be thoroughly examined to guarantee that the adsorbents used for large-scale wastewater treatment are feasible. To decrease operational expenses, the main desirable characteristics of adsorbents should be recovered via recycling. Due to the ease with which magnetic fields may separate MBSs, they may be a viable choice. The magnetic adsorbents cost, on the other hand, is determined by the availability of raw biomass, the modification procedures, and the price of MNPs.

MBSs can be tailored or designed to have specific properties that are beneficial for the removal of target pollutants. The availability of several functional groups on the surface of MBSs, as well as their chemistry, favors greater adsorption capacity, which has to be investigated further. Only a few research studies have looked at the field use of MBSs so far, indicating that further work is needed before MBSs may be commercialized. Future research efforts should focus on developing a more efficient operating mechanism for the separation of adsorbents in real wastewater treatment facilities. Environmental scientists, economists, engineers, policymakers, and governments should collaborate in this situation to maximize the technology's design, installation, and implementation.

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