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Magnetic Biocomposites for Wastewater Remediation

*Feihu Li** and *Mengyu Ma*

Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, School of Environmental Science and Engineering, Nanjing University of Information Science and Technology, Nanjing 210044, China

10.1 Introduction

Meeting the growing demand for clean water is a critical challenge for the world in the context of global climate change [1]. As the global population continues to grow, so does the volume of wastewater generated through a variety of pathways, with a predicted annual production of 574 billion m^3 by 2050 [2]. These wastewaters, if not treated effectively, no doubt pose a significant threat to human health and the environment since they can spread diseases, pollute waterways, and harmfully affect aquatic life [3]. The growing burden of wastewater treatment is further exacerbating this issue, as conventional treatment methods often rely on energy-intensive processes and chemical usage, and generate large quantities of sludge as by-products [4, 5]. These limitations underscore the urgent need for sustainable and efficient wastewater treatment technologies to meet the United Nations' Sustainable Development Goal (SGD) 2030 of universal access to clean water and sanitation.

In the past two decades, magnetic biocomposites (MBCs) have emerged as a promising and sustainable candidate for wastewater remediation through adsorption processes [6–8]. These materials are typically composed of a mixture of natural biological components, such as chitosan (CS), cellulose, or other biopolymers, along with magnetic nanoparticles, integrating the advantages of magnetic nanoparticles and biological materials thereof [9, 10], and enabling potential applications in biomedical devices, tissue engineering, drug delivery, and

*Author as the corresponding author with Email address at fhli@nuist.edu.cn, and ORCID link at https://orcid.org/0000-0002-2969-8276.

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soft robotics [11–14]. Magnetic nanoparticles (MNPs, e.g., Fe_3O_4) provide high surface area and easy separation using external magnetic fields, while biological components offer high selectivity and specificity for target pollutants. Such composites tend to have high stability, surface area, and many surface functionalities, which prove to be very beneficial for the removal of pollutants from wastewater [15]. Benefiting from their unique properties, such as biocompatibility, tailorability of functional groups, reusability, simplicity of operation, cost-effectiveness, and environmental friendliness, MBCs are effective and attractive in removing a wide range of contaminants from wastewater, including heavy metals, dyes, pharmaceuticals, and pesticides [16–20], offering a sustainable and environmentally friendly approach for improving our water quality.

This chapter aims to provide a comprehensive overview of magnetic biocomposites for wastewater remediation. We first introduce the synthesis strategies and characterization techniques of magnetic biocomposites, with consideration of particular properties required for wastewater remediation applications. In this context, we then discuss the various applications of magnetic biocomposites for adsorptive removal of pollutants from wastewater and evaluate the potential benefits and limitations of using them in wastewater treatment. The most recent advances in wastewater remediation using magnetic biocomposites are presented, with emphasis on the adsorptive removal of inorganic and organic pollutants, and recovery of valuable substances from wastewater. Finally, a perspective on future research directions of MBCs is provided to advance the goal of clean water and sustainability.

10.2 Magnetic Biocomposite: Synthesis Strategy and Characterization Methodology

In this section, some fundamental concepts of MBCs are briefly introduced, with emphasis on the general synthesis strategies of MBCs and relevant characterization methodologies.

10.2.1 Selection of Magnetic Nanoparticles and Biological Matrixes

The successful design of MBCs relies on the judicious selection of MNPs and biological matrixes [21], both of which can offer unique properties that govern the overall functionality and performance of the resulting composites when practicalized for wastewater remediation purposes (Figure 10.1). A crucial aspect of MNP selection is their magnetic behavior. Ideally, superparamagnetic nanoparticles are preferred. These materials exhibit zero remanence magnetization, implying they lose magnetization upon removal of the external magnetic field [22]. Additionally, the size and shape of MNPs play a critical role. Smaller nanoparticles generally

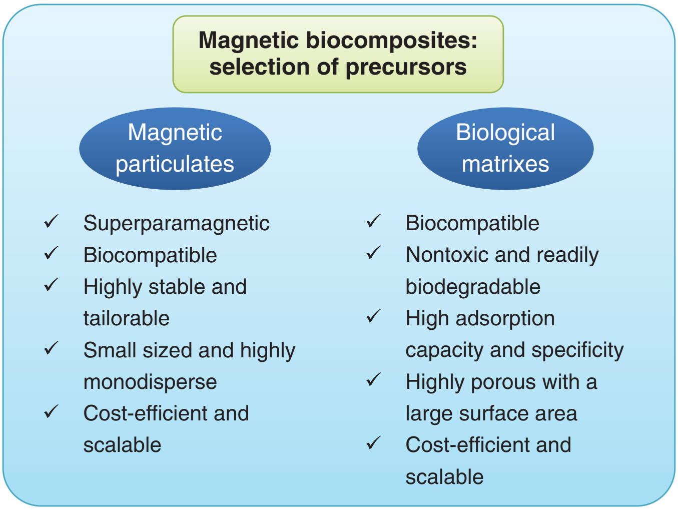

Figure 10.1 The ideal properties of magnetic particulates and biological matrices required for MBCs for wastewater remediation applications.

exhibit enhanced responsiveness to external magnetic fields but may suffer from increased aggregation tendencies [23]. Conversely, larger particles offer better stability but potentially lower magnetic responsiveness. Besides, the surface chemistry of MNPs is another critical consideration. Surface functionalization with biocompatible polymers like polyethylene glycol (PEG) can enhance biocompatibility and dispersibility, and prevent unwanted aggregation [21]. Furthermore, surface modifications can be tailored to capture specific contaminants or elements from wastewater, enabling targeted removal or recovery from the waste stream [19, 24]. Iron oxide nanoparticles, particularly magnetite (Fe_3O_4) and maghemite (γ-Fe_2O_3), are widely employed due to their biocompatibility, tunable magnetic properties, and facile synthesis methods [13]. Other compositions, such as cobalt, nickel, and their alloys, offer distinct magnetic characteristics but might raise concerns regarding cytotoxicity and biocompatibility [23].

Likewise, an ideal biological matrix should not only offer a high surface area and specific functional groups for pollutant binding but also facilitate the incorporation of MNPs. Common biological materials used in MBCs include chitosan, alginate, collagen, fungi, and biomass-based materials, each offering distinct advantages [25–27]. Fungi may possess specific binding sites for targeted contaminants, while algae offer high surface area and potential for in situ remediation [28]. When selecting a biological matrix, factors like the target contaminant type and desired removal efficiency must be considered (Figure 10.1). For example, MBCs with groups of metallophilicity have shown promise in heavy metal removal [7, 29, 30], while algae-based composites may be better suited for organic pollutant adsorption [31, 32]. Additionally, the biocompatibility of the matrix and its potential for regeneration after use are important aspects to consider. Abd Malek et al. [9] demonstrated the effectiveness of a chitosan-based magnetic biocomposite for removing organic dyes from wastewater. Ultimately, selecting the right biological matrix is essential for designing efficient and sustainable MBCs for wastewater remediation.

10.2.2 Synthetic Approaches of Magnetic Biocomposites

Once the magnetic component and the biological matrix are defined separately, the next step is to prepare the MBCs with a proper approach. Depending on whether the magnetic component is pre-prepared, the synthesis of MBCs can be classified as *in situ* (or one-pot, one-step) synthesis and *ex situ* (or two-step) synthesis (Figure 10.2). In the former method, the MNPs are synthesized within the biological matrix, which offers advantages such as better dispersion of nanoparticles within the matrix and potentially enhanced interaction between the magnetic and biological components [25, 33, 34]. However, it may require additional steps to ensure the uniformity and stability of the nanoparticles within the matrix [35]. In the latter, the magnetic component (such as Fe_3O_4 nanoparticles) is pre-prepared separately by coprecipitation, sol-gel, hydrothermal,

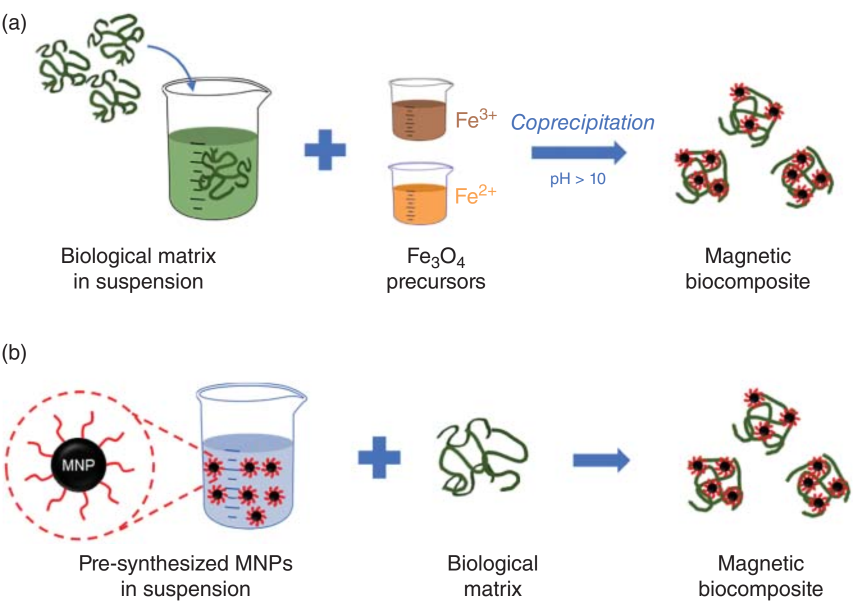

Figure 10.2 Synthesis of MBCs via (a) *in situ* and (b) *ex situ* approaches.

microwave-assisted, and microemulsion methods [23] and then combined with the biological matrix. Based on the bonding nature, the magnetic and biological components can be combined through electrostatic interactions, covalent bonding, or physical encapsulating [36–38]. These approaches allow for more precise control over the magnetic properties and size distribution of the nanoparticles before incorporation into the biocomposite [22, 37]. Besides, layer-by-layer (LbL) assembly, biomimetic synthesis using biomolecules (e.g., peptides, proteins) as templates or scaffolds, and hybrid methods that involve the combination of two or more synthetic methods are also widely applied to create MBCs with enhanced properties [39, 40].

The selection of the appropriate synthetic method for magnetic biocomposite depends on the specific requirements of the application, such as the type of target pollutant, the desired magnetic properties, the stability of the biocomposite, and the cost-effectiveness of the preparation process.

10.2.3 Characterization Techniques

Generally, the physicochemical properties of the synthetic MBCs are fully evaluated before the performance assessment for wastewater remediation in order (i) to establish a convincing relationship between the structural parameters and their activity, and (ii) to understand the underlying mechanisms of pollutant abatement over the surface of the MBCs. To this end, a variety of characterization techniques have been widely used for collecting and analyzing the morphological, crystallographic, structural, chemical, magnetometric, and molecular spectroscopic information of MBCs. Specific techniques include scanning electron microscopy (SEM), transmission electron microscopy (TEM), atomic force microscopy (AFM), X-ray diffraction spectroscopy (XRD), X-ray photoelectron spectroscopy (XPS), X-ray fluorescence spectroscopy (XRF), energy dispersive X-ray spectroscopy (EDX), Fourier transform infrared spectroscopy (FTIR), Raman spectroscopy, nitrogen absorption-desorption isotherm porosimetry, and vibrating sample magnetometry (VSM).

10.2.3.1 Morphological Analysis by SEM, TEM, and AFM

Due to their unique advantages of providing high-resolution images for detailed morphological analysis and inspection, in particular the size and distribution of magnetic particles within the biocomposite matrix, both SEM and TEM are widely used in the morphological characterization of magnetic biocomposite materials [41, 42]. Although both microscopy techniques are very powerful, there are limitations to consider. The sample preparation for SEM analysis would be very time-intensive due to magnetic properties, and a conductive coating on nonconductive samples may also in part affect the morphology. Moreover, SEM provides only surface imaging, and it may not fully capture the internal structure of the composite. This issue can be complemented by TEM, which allows for detailed visualization of the internal structure of the biocomposite at the nanoscale, especially for thin samples [42]. In the case of thick and soft biocomposites, the TEM sample preparation can be challenging. Additionally, TEM is typically limited to two-dimensional (2D) imaging, which may not fully depict the three-dimensional (3D) structure of the biocomposite [43].

Likewise, AFM offers high-resolution imaging and surface profiling capabilities, allowing for the visualization of the topography and mechanical properties of the biocomposite at the nanoscale [44]. AFM can also be used to map magnetic properties, such as magnetic force and magnetic domain distribution, providing valuable insights into the behavior of the magnetic particles within the composite [45]. While versatile and powerful, AFM may struggle with imaging soft or nonconductive samples and can be limited by the scan size and speed [45]. In short, all the above microscopy offers powerful capabilities for characterizing MBCs, providing detailed insights into their structure and properties at the nanoscale. However, researchers should be mindful of the sample preparation requirements and limitations of each technique when choosing a proper morphological characterization approach.

10.2.3.2 Crystallographic and Compositional Analysis by XRD, XPS, XRF, and EDX

XRD analysis identifies long-range ordering (i.e., crystallographic structure) in crystalline materials and short-range ordering in amorphous materials. XRD technique is often used to verify the successful combination of MNPs with biological matrix [17] and to identify the crystallographic or amorphous nature of the biological matrix and other components [46]. It's sensitive to crystalline phases but may be challenging in sample preparation, especially for soft biocomposites.

XRF is a rapid and nondestructive elemental analysis technique that is of high sensitivity for trace elements. Therefore, it's often employed for chemical composition analysis of biocomposite components [47], and biocomposite before and after applying to wastewater remediation [48]. Likewise, XPS analysis can provide surface-sensitive elemental and chemical information (e.g., high-resolution information on chemical states, and quantitative data of elemental composition). It is widely used for an *ex situ* mechanism investigation based on the surface compositional analysis of MBCs, in particular before and after application to contaminant removal [28, 49]. Similarly, EDX analysis offers elemental composition at high spatial resolution and is often integrated with SEM for qualitative mechanism studies according to the surface elemental data of MBCs [10, 49]. However, EDX is less sensitive to light elements, and a combination with other sensitive techniques may be necessary for a more comprehensive characterization of MBCs targeting pollutants with light elements.

10.2.3.3 Molecular Spectroscopic Analysis by FTIR and Raman Spectroscopy

Both FTIR and Raman spectroscopy can probe the vibrational modes of molecules in biocomposite materials, thereby providing valuable information about the functional groups, bonding configurations, and chemical composition of the magnetic biomaterials based on their interactions with infrared (FTIR) or laser light (Raman) [18, 50]. Nevertheless, FTIR spectroscopy has limited sensitivity to inorganic components [9], and may suffer from spectral interferences, while Raman may also encounter fluorescence interference [51].

Collectively, FTIR and Raman spectroscopy are valuable tools for characterizing MBCs. By utilizing both techniques in conjunction, researchers can obtain comprehensive insights into the surface functional groups and properties of these complex materials, thereby facilitating the mechanism understanding of organic pollutant (e.g., dyes) sequestration [52].

10.2.3.4 Porous Structure Analysis by N_2 Adsorption–Desorption Isotherms

N_2 adsorption–desorption isotherm analysis is a powerful technique usually employed to delve into the intricate porosity of biocomposite materials [53, 54]. By measuring the amount of nitrogen gas adsorbed and desorbed at various pressures, this method provides insights into the surface area, pore size distribution, and pore volume of MBCs [9, 41, 55]. Brunauer-Emmett-Teller (BET) and Barret-Joyner-Halenda (BJH) methods are generally used for the determination of surface area and pore size distribution within the MBCs [49, 56].

However, this technique is only applicable for biocomposites with micropore (<2 nm) and mesopore ($2 \sim 50$ nm), and limited for biocomposites of macropore (>50 nm). To gain a more in-depth understanding of the porosity of biocomposites, it would be necessary to complement N_2 adsorption–desorption analysis with techniques like SEM or TEM with careful interpretation of the data.

10.2.3.5 Magnetometric Analysis by VSM

VSM analysis is a basic technique for investigating the magnetometric properties of magnetic biocomposite materials [46, 55]. This method measures the magnetization of a sample as it is exposed to a varying magnetic field, providing valuable insights into the material's magnetic behavior. VSM is of high sensitivity and versatility, and offers precise quantification of magnetic parameters, providing a comprehensive understanding of the material's magnetic response. But the sample preparation may be challenging, particularly for soft biocomposites as it often requires grinding and packing to ensure accurate measurement. To obtain a more comprehensive understanding of the magnetic properties and their correlation with the biocomposites' microstructure, integrating VSM with microscopic techniques like TEM or SEM is a routine operation [37, 57].

10.3 Approaches to Evaluate the Performance of Magnetic Biocomposites

In this section, some essential approaches for the performance evaluation of MBCs are presented, with a special focus on the adsorption performance and efficiency of magnetic biocomposite materials in wastewater remediation.

10.3.1 Batch Adsorption Studies

Batch adsorption studies are fundamental for understanding and optimizing the adsorption process, providing a cost-effective and versatile method for evaluating the performance of a magnetic biocomposite adsorbent in removing and/or recovering target substances from wastewater [29, 55]. This technique involves immersing or dispersing a given weight of biocomposite adsorbent in a solution containing the target contaminant or substance of interest under controlled conditions. By measuring the change in concentration of contaminants before and after adsorption, the adsorption capacity and efficiency of the biocomposite can be determined [58], allowing for the investigation of various parameters that influence adsorption performance, such as pH, background ionic strength, adsorbent dosage, contact time, temperature, and competitive ions, etc. [59].

Specifically, batch adsorption studies are often conducted for the following purposes:

(I) pH optimization: The pH of the solution can significantly impact the adsorption process. Both the biocomposites' surface charge and the contaminant's speciation are influenced by pH, affecting their interactions. By conducting experiments at different pH values, researchers can determine the optimal pH range for maximum contaminant uptake, leading to efficient removal.

(II) Dosage optimization: The adsorbent dosage plays a critical role in the adsorption process. Increasing the dosage generally leads to higher contaminant removal, but beyond a certain point, further additions may have minimal impact. Finding the ideal adsorbent dosage ensures efficient contaminant removal without unnecessary material expenditure.

(III) Contact time optimization: This parameter explores the time required for the biocomposite adsorbent to reach equilibrium with the target contaminant. The initial adsorption rate is typically high, followed by a gradual decrease until equilibrium is reached. Determining the optimal contact time ensures maximum contaminant removal within a reasonable timeframe. Besides, fitting experimental data to various kinetic models such as pseudo-first-order or pseudo-second-order kinetics can offer mechanistic insights into the adsorption mechanism and the rate-controlling steps, which is essential for optimizing the adsorption process.

(IV) Understanding the effects of background ionic strength and competitive ions: The presence of other ions, especially those with similar affinity for the magnetic biocomposite adsorbent, in the solution can influence the adsorption process. Understanding the effects of background ionic strength and competitive ions is crucial for assessing the selectivity of the biocomposite and predicting its adsorption behavior in real-world scenarios.

(V) Isotherm and thermodynamic analysis: Isotherm studies involve plotting the equilibrium adsorption data obtained from experiments onto various adsorption isotherm models such as Langmuir or Freundlich equations. These models provide insights into the adsorption capacity and affinity of the magnetic biocomposite for specific contaminants in wastewater. Thermodynamic studies involve analyzing the effect of temperature on adsorption to determine the spontaneity, feasibility, and nature of the adsorption process. Parameters like Gibbs free energy ($\Delta \boldsymbol{G}$), enthalpy ($\Delta \boldsymbol{H}$), and entropy ($\Delta \boldsymbol{S}$) changes can be calculated to understand the thermodynamic feasibility of the adsorption process.

(VI) Understanding adsorption mechanisms: Through analysis of batch experimental data, researchers can gain insights into the interactions between the adsorbent and the contaminant, providing a better understanding of the underlying adsorption mechanism.

(VII) Developing predictive models: The experimental data obtained from batch adsorption studies can be used to develop mathematical models that predict adsorption behavior under various conditions (e.g., surface complexation models), allowing for informed decision-making and optimized process design for specific applications.

By harnessing the power of magnetism (i.e., a magnet), magnetic biocomposite adsorbents provide a sustainable and efficient solution for separating spent adsorbents from adsorption suspensions [59], revolutionizing the field of batch adsorption studies and paving the way for faster, more efficient, and environmentally friendly separation processes.

10.3.2 Fixed-Bed Column Studies

To evaluate the performance of MBCs under dynamic, continuous flow conditions, fixed-bed column studies are often performed to simulate real-world applications, providing crucial insights into their practical applicability [60, 61]. In a typical fixed-bed column, the biocomposite material is packed into a vertical column, and a solution containing the target contaminant is passed through the fixed-bed where the pollutants (e.g., Cu^{2+} and Pb^{2+}) are captured continuously [62]. This setup allows for the investigation of various factors influencing adsorption performance, including the adsorbent's properties (e.g., particle size, rigidity, and porosity), and operational parameters (e.g., flow rate, inlet concentration, fixed-bed height, and volume to area ratio of the bed).

Specifically, fixed-bed column studies enable the study of the following key parameters:

(I) Particle size: The size and uniformity of particles impact flow dynamics and mass transfer within the bed, influencing the adsorption process.
(II) Rigidity: The mechanical strength and structural integrity of the adsorbent influence its long-term stability and performance under continuous flow conditions.
(III) Porosity: The pore structure and surface area of the adsorbent govern its adsorption capacity and the diffusion of contaminants within the material.
(IV) Flow rate: The rate at which the feed solution passes through the column affects the residence time of the contaminant, influencing the extent of adsorption.
(V) Inlet concentration: The concentration of the contaminant in the feed solution directly impacts the adsorption capacity and breakthrough behavior of the bed.
(VI) Fixed-bed height: The height of the adsorbent bed impacts the overall adsorption capacity and the time required for a breakthrough.
(VII) Volume to area ratio of the bed: This ratio, also known as the bed aspect ratio, significantly influences the flow distribution and mass transfer within the column.

To further understand and predict adsorption behavior in fixed-bed column studies, various mathematical models are employed [61], including Thomas Model (THM, to describe the breakthrough curve and predict the breakthrough time based on the adsorption rate constant and other parameters), Bed Depth Service Time Model (BDST, to relate the bed height to the service time—time until breakthrough, and predict the performance of adsorbents under different conditions), and Yoon-Nelson Model (YNM, to provide a simplified representation of the breakthrough curve and estimate of adsorption capacity and breakthrough time). By integrating the fixed-bed column data with appropriate mathematical models, we can effectively evaluate the performance of biocomposite adsorbents under dynamic conditions [63], gain insights into the impact of various factors, and optimize the design and operation of adsorption processes for removal of specific pollutants from wastewater.

10.3.3 Regeneration and Reusability Assessment

Whether in batch adsorption or fixed-bed column adsorption, the magnetic biocomposite adsorbent will inevitably reach saturation after a period of adsorption. Therefore, how to effectively restore its adsorption capacity and extend its reusability is a crucial step forward. Besides, assessing the regeneration and reusability of the spent MBCs is essential to optimize its long-term performance, reduce costs, and promote sustainability [25, 59].

Regeneration aims to restore the adsorption capacity of spent adsorbents, making them reusable for multiple cycles. Based on the underlying mechanism of action, regeneration can be generally categorized into chemical regeneration, thermal regeneration, biological regeneration, and physical regeneration. Chemical regeneration is usually achieved by employing various chemical agents to desorb adsorbed contaminants. Agents like sodium hydroxide (NaOH), hydrogen chloride (HCl), and ethylenediamine tetraacetic acid (EDTA) are often used to desorb heavy metals from MBCs [7, 25, 29]. Sodium chloride (NaCl), dilute nitric acid (HNO_3), and organic solvents like acetone, methanol, and ethanol are effective in desorbing metal species and organic pollutants, respectively [29, 64]. Thermal regeneration involves heating the spent adsorbent to release adsorbed contaminants, which is effective for removing organic impurities from magnetic biochar adsorbents, facilitating the restoration of its adsorption capacity [59, 61].

As mentioned earlier, one of the inherent merits of MBCs is the easy and rapid separation from suspension by external magnets, which facilitates the following regeneration units. Biological techniques using specific microbes to break down adsorbed contaminants over MBCs are time-consuming and, thus, do not match so well to the preceding magnetic separation on the same time scale. Likewise, methods like ultrasonication, agitation, and centrifugation can be employed to remove adsorbed contaminants and restore the adsorbent's surface [65, 66]. However, such physical techniques are seldom used for restoring magnetic biocomposite adsorbents. For a specific MBC adsorbent, cycle life testing via repeated regeneration and adsorption cycling with various regeneration techniques is crucial to evaluate the long-term stability and reusability of the magnetic biocomposite adsorbent and identify the most effective and sustainable methods, paving the way for their widespread application in wastewater remediation [29, 59].

10.3.4 Environmental Risk and Safety Considerations

MBCs, while offering a promising solution for wastewater remediation, also present potential environmental risks that cannot be ignored, particularly for biocomposites involving pyrolysis and those after long-term operation in the removal of toxic compounds [28, 67]. For instance, the preparation of magnetic biochar is likely to release toxic compounds into the environment, particularly if improper methods are used [68]. Additionally, the application of biocomposites can lead to unintended consequences, such as the leaching of toxic compounds from the biocomposite material itself, especially after long-term adsorbing pollutants with weak affinity [67]. If not properly managed, the release of accumulated contaminants (e.g., toxic heavy metals and metalloids) from the biocomposite material into the environment during disposal or breakdown could cause significant harm, effectively shifting the pollution burden rather than eliminating them [59]. Therefore, potential hazards and environmental risks should be considered in the preparation and application of magnetic biocomposite materials to minimize harm to the environment. Short-term simulation studies cannot guarantee that long-term application to water treatment is risk-free. Therefore, long-term water tests and risk assessments are essential for ensuring the sustainable and safe utilization of MBCs in water treatment.

10.4 Applications in Wastewater Remediation

As mentioned earlier, combining MNPs and biological matrices to provide high stability, surface area, and many surface functionalities makes the MBCs particularly interesting in the adsorption of pollutants, recovery of valuable elements, and other state-of-the-art applications for wastewater remediation. The most recent advances will be covered in this section, categorized in the removal of inorganics, organics, recovery of valuable elements, and other applications.

10.4.1 Adsorptive Removal of Inorganic Pollutants

Inorganic pollutants including heavy metals, metalloids, and nutrients in wastewater pose a significant environmental and health risk, yet their removal remains a challenging task in wastewater treatment. Conventional methods, such as chemical precipitation and ion exchange, often struggle to effectively remove such contaminants due to limited adsorption capacities, high operational costs, and the generation of secondary pollutants [24]. Additionally, traditional adsorbents, in particular those on the nanosized scale, can be difficult to separate from the treated water, leading to further environmental contamination. By integrating the high adsorption capacities of biomaterials with the ease of magnetic separation, MBC adsorbents offer a promising solution to these challenges [59, 69].

Over the past two decades, a variety of MBC adsorbents with varying biological components have been developed for the adsorptive removal of heavy metals and other inorganics of concern from wastewater [70, 71]. A part of these novel MBC adsorbents invented over the last 5 years are summarized in Table 10.1. A fuller and more comprehensive list beyond this time scale can be found in the earlier review papers. Heavy metals like Cd(II), Cr(VI), Cu(II), Pb(II), Hg(II), Ni(II), etc., have shown high affinity to these MBC adsorbents under environmentally relevant conditions. For instance, Staron et al. [73] reported an MBC consisting of hydrochar obtained by hydrothermal carbonization of raffia fibers, *Lindnera jadinii* yeast, and MNPs, labeled as Fe_3O_4@hydrochar/*Lindnera jadinii*, exhibits a maximum adsorption capacity of 16.3 $mg\,g^{-1}$ for Cd(II) at pH 6. Under similar conditions, another fungal-based MBC adsorbent, Fe_3O_4@*P. chrysosporium*, demonstrated a maximum capacity of 18.2 $mg\,g^{-1}$ toward Cd(II) [72]. Touihri et al. [10] synthesized a novel biocomposite adsorbent—magnetic pine cone gel beads (MPCB) for the removal of Cu(II) and Cr(VI) from aqueous solution—and found that the maximum adsorption capacity for Cu(II) and Cr(VI) were 68.6 and 212.2 $mg\,g^{-1}$, respectively. Omer et al. [74] integrated ZIF-67, Fe_3O_4 nanoparticles, and aminated chitosan matrix, yielding core-dual shell Fe_3O_4/ZIF-67@aminated chitosan MBC beads with a maximum capacity of 119.1 $mg\,g^{-1}$ for Cr(VI). Such hierarchical core-dual shell structure seems to be beneficial for improving their adsorption capacity toward Cr(VI) (Table 10.1), with maximum Cr(VI) capacities of 282.0 and 294.1 $mg\,g^{-1}$ for Fe_3O_4/CS@aminothiazole and Fe_3O_4/CS@*Azolla pinnata*, respectively [75, 76]. Chitosan (CS) has a large contribution to these large affinities for Cr(VI) since it is an alkaline polysaccharide containing a large number of amino and hydroxyl groups that can chelate with various metal ions [97].

Likewise, as shown in Table 10.1, MBC adsorbents with a hierarchical core/shell/shell structure also demonstrated significantly high affinity toward Cu(II) [18, 77], Pb(II) [79, 81], and Hg(II) [82], probably due to more binding sites are provided from these varying components during the preparation process [77, 82]. The abundance of hydroxyl groups on the surface of nanocellulose (NC) provides an excellent platform for considerable surface modifications, permitting the grafting of various functionalities onto the cellulosic portion, thus allowing easy immobilization of pollutants. Therefore, the MBC adsorbent $CaCO_3$/Fe_3O_4@cellulose nanofiber demonstrated a maximum adsorption capacity of 1010.0 $mg\,g^{-1}$ toward Pb(II) [81]. Compared to the simple core/shell structured MBC adsorbent, viz. Fe_3O_4/CS [83], the core/shell/shell structured MBC—Fe_3O_4@CS/PSSMA [78], showed higher maximum capacity for Ni(II) under similar conditions (113.1 versus 30.0 $mg\,g^{-1}$).

In addition to heavy metals, MBC adsorbents were also widely used for the removal of metalloids and nutrients from wastewater. For example, our group has developed a magnetic biochar composite (i.e., Fe_3O_4@biochar/Mg/Al oxides) with a maximum capacity of 34.5 for As(V) at pH 5 [58]. Luo et al. [57]

Table 10.1 Magnetic biocomposites for the adsorptive removal of pollutants from wastewater reported in the literature over the last 5 years.

MBCs		Target pollutants	Adsorption conditions				Q_{max} (mg g^{-1})	Reference
			pH	T (°C)	$t_{eq.}$ (min)	C_0 (mg L^{-1})		
Fe_3O_4@*P. chrysosporium*	Heavy metals	Cd(II)	6.0	25	60	100	18.2	[72]
Fe_3O_4@hydrochar/*Lindnera jadinii*	Heavy metals	Cd(II)	6.0	25	1440	150	16.3	[73]
Fe_3O_4/ZIF-67@aminated chitosan	Heavy metals	Cr(VI)	2.0	25	120	50	119.1	[74]
Fe_3O_4/CS@aminothiazole	Heavy metals	Cr(VI)	2.0	22	2880	10.4 ~ 499.2	282.0	[75]
Fe_3O_4/CS@*Azolla pinnata*	Heavy metals	Cr(VI)	4.0	25	15	50	294.1	[76]
Fe_3O_4/citric acid@chitosan	Heavy metals	Cu(II)	6.5	25	66	50	232.6	[77]
Fe_3O_4@CS/PSSMA	Heavy metals	Cu(II)	5.0	25	720	5 ~ 200	141.7	[78]
Fe_3O_4@starch-g-PAA/cellulose nanofibers	Heavy metals	Cu(II)	5.0	25	540	600	555.6	[18]
Fe_3O_4@TCA@chitosan	Heavy metals	Pb(II)	5.5	25	180	20 ~ 250	204.9	[79]
Fe_3O_4/CS@polymer-grafted silica	Heavy metals	Pb(II)	6.0	25	480	50 ~ 400	111.1	[80]
$CaCO_3$/Fe_3O_4@cellulose nanofiber	Heavy metals	Pb(II)	n/a	25	1440	50 ~ 2000	1010.0	[81]
Fe_3O_4@CS@polypeptide	Heavy metals	Hg(II)	5.0	45	120	25 ~ 600	218.8	[82]
Fe_3O_4@CS/PSSMA	Heavy metals	Ni(II)	5.0	25	720	5 ~ 200	113.1	[78]
Fe_3O_4/CS	Heavy metals	Ni(II)	6.0	25	120	20 ~ 300	30.0	[83]
Fe_3O_4/CS	Metalloids and nutrients	As(V)	3.0	25	1440	5 ~ 500	15.2	[84]
Fe_3O_4@biochar/Mg/Al oxides	Metalloids and nutrients	As(V)	5.0	10	1440	2 ~ 30	34.5	[58]
Fe_3O_4@camphor seeds husk	Metalloids and nutrients	As(III)	7.2	n/a	480	10 ~ 100	39.7	[57]
Fe_3O_4@Mg/herbal medicine residues	Metalloids and nutrients	PO_4^{3-}	8.0	25	180	10 ~ 500	635.4	[85]

Fe_3O_4@peanut husks@NH_2-MIL-101(Fe)		PO_4^{3-}	5.6	25	240	10 ~ 70	20.4	[86]
Fe_3O_4@polyethyleneimine lignin		PO_4^{3-}	6.5	25	1080	10 ~ 100	43.0	[87]
Fe_3O_4@Mg/herbal medicine residues		NH_4^+	8.0	25	180	10 ~ 500	615.6	[85]
Fe_3O_4/AC@*Yarrowia lipolytica* cell		MB	>5.8	25	1440	50 ~ 250	32.1	[19]
Fe_3O_4/CS@salicylaldehyde/nanoclay		AR88	4.0	25	550	20 ~ 200	173.5	[88]
Fe_3O_4/CS-cross-linked-PAM		SY	5.0	25	120	5 ~ 1500	360.2	[89]
Fe_3O_4/corn cob	Dyes	MB	7.7	55	300	75	13.2	[90]
Fe_3O_4/peanut husk		MB	5.7	30	240	30 ~ 240	245.0	[91]
		RhB	6.8			30 ~ 240	57.3	
Fe_3O_4/papaya seed powder		CR	5.0	30	180	100 ~ 1000	216.9	[56]
Fe_3O_4/CS/GO		CR	≤8.0	35	1440	200	395.8	[92]
Fe_3O_4@SiO_2/SA-PEI		X-3B	7.0	25	1440	50 ~ 500	370.9	[93]
Fe_3O_4/alginate/bentonite@CS		CPX	6.0	23	120	100 ~ 600	264.8	[16]
Fe_3O_4-eggshell@ZIF-8		NOR	5.0	25	240	10	53.5	[46]
Fe_3O_4@SiO_2/SA-PEI		DS	5.0	25	1440	25 ~ 200	241.5	[93]
$CuFe_2O_4$/AG-g-PAO	Others	OPP	6.0	25	15	50 ~ 300	769.2	[94]
Fe_3O_4@CS/*chlorella vulgaris*		TC	6~8	25	240	10 ~ 200	834.0	[95]
		CIP					394.9	
		AMX					150.8	
Fe_3O_4/$HCP_{Bn\text{-}glu}$		nitrophenol	6.0	25	30	2 ~ 150	83.3	[96]

AC – activated carbon; AG – Arabic gum; AMX – amoxicillin; AR – Acid Red; C_0 – initial concentration; CIP – ciprofloxacin; CPX – ciprofloxacin; CR – Congo Red; CS – chitosan; DS – diclofenac sodium; GO – graphene oxide; HCP – hyper-cross-linked polymer; MB – Methylene Blue; NOR – norfloxacin; OPP – organophosphorus pesticide; PAA – poly(acrylic acid); PAM – polyacrylamide; PAO – polyamidoxime; PEI – poly (ethylene imine); PSSMA – poly(4-styrenesulfonic acid-*co*-maleic acid); Q_{max} – maximum adsorption capacity; RhB – Rhodamine B; SA – sodium alginate; SY – Sunset Yellow; T – temperature; TC – tetracycline; TCA – tetracarboxylic acid; $t_{eq.}$ – equilibrium time; X-3B – reactive red X-3B.

reported recently a magnetically modified biomass camphor seeds husk (i.e., Fe_3O_4@camphor seeds husk) delivering a maximum adsorption capacity of 39.7 mg g^{-1} based on the Langmuir model at pH 7.2, which is much higher than its counterpart without magnetic property. Moreover, nutrient species like phosphate (PO_4^{3-}) and ammonium (NH_4^+) can also be effectively captured by MBC adsorbents, which are likely to be further reused as slow-released fertilizers if handled properly. For instance, Wang et al. [85] developed an MBC adsorbent based on loading magnesium (Mg) to Chinese herbal medicine residues (i.e., Fe_3O_4@Mg/herbal medicine residues). It exhibited remarkable performance for simultaneously removing and recovering phosphate and ammonium from

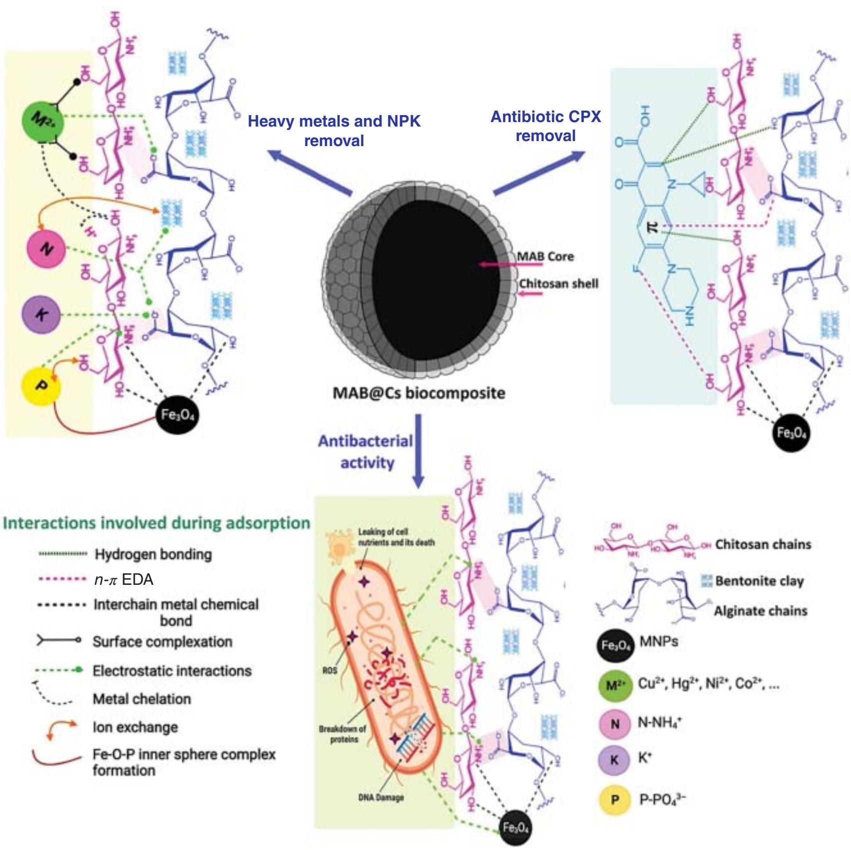

Figure 10.3 Proposed adsorption mechanism of organic and inorganic pollutants by one typical magnetic biocomposite (i.e., Fe_3O_4/alginate/bentonite@CS, labeled as mAB@Cs) and its antibacterial activity. *Source:* Reprinted with permission from [16]. Copyright 2024 Elsevier B. V.

wastewater, with maximum adsorption capacities of 635.35 and 615.57 mg g^{-1}, respectively. Pot tests have verified the feasibility of these nutrient-loaded MBCs as a fertilizer substitute for the growth of a typical forage grass, alfalfa (*Medicago sativa* L.). As depicted in Figure 10.3, the binding mechanisms for the above inorganic pollutants over MBCs usually involved (i) electrostatic interaction, (ii) ion-exchange, (iii) surface complexation, (iv) interchain metal chemical bond, and (v) metal chelation [16].

10.4.2 Adsorptive Removal of Organic Contaminants

The removal of organic pollutants from wastewater is often hindered by their complex properties, such as low biodegradability and high solubility, which can render traditional treatment methods ineffective [19, 98]. Moreover, the high energy intensity and costs associated with advanced treatment processes, such as advanced oxidation and membrane bioreactors, can limit their accessibility, particularly in resource-constrained communities [99, 100]. As a crucial class of organic contaminants, organic dyes are persistent and inherently toxic, which has raised a major environmental challenge and health concern, especially in low-income and middle-income countries [99]. However, recent advancements in MBC adsorbents have demonstrated great potential in overcoming the challenges of synthetic dyes [47, 71].

For instance, research has shown that Fe_3O_4/corn cob can effectively capture methylene blue (MB), with a maximum capacity of 13.2 mg g^{-1} at 55 °C [90], while the yeast-based MBC, viz. Fe_3O_4/AC@*Yarrowia lipolytica* cell has been demonstrated to possess a maximum adsorption capacity of 32.1 mg g^{-1} of MB at 25 °C [19]. Recently, Aryee et al. [91] prepared an MBC based on peanut shells using the coprecipitation method for efficient sequestration of alkaline dyes using MB and rhodamine B (RhB) as model dyes, which showed maximum adsorption capacities of 245.0 and 57.3 mg g^{-1}, respectively, at near-neutral pH (Table 10.1). Nascimento et al. [101] reported a nanosized MBC using yeast waste and MNPs as parent materials by coprecipitation method to remove three vinyl sulfonic acid reactive dyes (i.e., blue 19 (B-19), red 198 (R-198), and yellow 15 (Y-15)) from effluents of industrial-like dyeing processes. However, the adsorption capacities for all dyes were not as high as expected since the saturation of such MBC adsorbents was not reached in these suspensions. Similarly, by incorporating CS inside the MBC adsorbents, for example, three hierarchical core/shell/shell structured MBCs, viz. Fe_3O_4/CS@salicylaldehyde/nanoclay, Fe_3O_4/CS-cross-linked-PAM, and Fe_3O_4/CS/GO have been shown greatly improved capacities toward acid red (AR) 88, sunset yellow (SY), and Congo red (CR), respectively [88, 89, 92]. Munagapati et al. [56] observed that MBC adsorbent based on loading papaya (*papaya L.*) seed powder over MNPs (i.e., Fe_3O_4/papaya seed powder) could sequester up to 216.9 mg g^{-1} of CR at pH 5. A recent study by Wu et al. [93] has unveiled a novel core/shell/shell structured MBC based on loading poly (Benzyldimethyl[2-[(2-methyl-1-oxoally)oxy]ethyl] ammonium chloride) over the magnetic sodium alginate (SA) composite, showing a maximum adsorption capacity as high as 370.9 mg g^{-1} toward reactive red X-3B.

It is well-defined that the solution pH determines the charge on the surface of the adsorbent and also controls the degree of ionization of dissolved substances (e.g., dye molecules), thereby controlling the overall adsorption performance [102]. Studies have shown that the removal of anionic dyes increases at lower pH values, while the opposite is true for cationic dyes [103]. Regarding the binding mechanism of synthetic dyes, Jawad et al. [104] claimed that the reactive groups of MBC adsorbent CTS-GL/MgO/Fe_3O_4 play a crucial role in the adsorption process of Reactive Blue 19 (RB-19). Concretely, the positively charged groups including protonated amino groups ($—NH_3^+$), ($—OH_2^+$), and $Mg(OH)^+$ can effectively bind to the negatively charged ($—SO_3^-$) groups of RB-19 dye by electrostatic attraction. Besides, hydrogen bonding and interactions between the electron donors of the MBC and the acceptors inside the RB-19 dye may probably have contributed to the efficient binding of the dye. In addition, a study by Abd Malek et al. [105] has indicated that the amino and hydroxyl-rich CS component within the MBC, viz. m-CS-PVA/FA, is likely to protonate in acidic media and thus displays a strong affinity toward the anionic dye, Reactive Orange 16 (RO16), by strong electrostatic attraction. Dipole-dipole hydrogen bonding and n–π stacking interactions may also play an important role in the adsorption process.

Apart from organic dyes, organic contaminants like antibiotics, anti-inflammatories, and pesticides, also known as emerging pollutants, have shown remarkable affinity over the surface of many MBCs (Table 10.1). For instance, Mirizadeh et al. [95] investigated the adsorption performance of a novel magnetic chitosan/microalgae biocomposite, Fe_3O_4@CS/*chlorella vulgaris* for capturing three antibiotics, i.e., tetracycline (TC), ciprofloxacin (CIP), and amoxicillin (AMX) from wastewater, and unveiled that the maximal adsorption capacities were as high as 834.0, 394.9, and 150.8 mg g^{-1} for TC, CIP, and AMX, respectively. Hassanzadeh-Afruzi et al. [94] reported a novel MBC adsorbent based on Arabic Gum-grafted-poly amidoxime and $CuFe_2O_4$ magnetic nanoparticles ($CuFe_2O_4$/AG-g-PAO) recently, which displayed a remarkable maximum adsorption capacity as high as 769.2 mg g^{-1} for organophosphorus pesticide (OPP) under ambient conditions (pH 6, 25 °C). This core/shell/shell structured MBC also demonstrated fast adsorption kinetics and excellent recyclability. A recent work by Chkirida et al. [16] reported a new hybrid MBC based on an alginate-bentonite core and a chitosan shell layer (Fe_3O_4/alginate/bentonite@CS), where the MNPs were in situ generated within the alginate-bentonite core via a simple nonthermal coprecipitation approach. The biocomposite has a high ability to magnetically separate and remove organic (ciprofloxacin (CPX), $\boldsymbol{Q}_{max} = 264.8\,mg\,g^{-1}$ at pH 6 and 23 °C) and seven heavy metals from wastewater. The interaction between MBC and these organic and inorganic contaminants is complex, and the high antibacterial activity of the biocomposite is believed to be attributed to the chitosan-MNPs antibacterial coating (Figure 10.3). Despite significant advances of the MBC adsorbents in adsorptive removal of inorganic and organic pollutants from wastewater, the implementation of this technique from laboratory scale to pilot scale and even industrial scale applications is still to be accomplished.

10.4.3 Recovery of Valuable Elements from Wastewater

The recovery of valuable elements, such as precious metals (e.g., gold, palladium, rhodium, silver), radiative metals (e.g., uranium, thorium), rare earth elements (e.g., lanthanum), and energy-critical metals (e.g., lithium) from waste streams is crucial for sustainable resource management and reducing the environmental impact of mining and processing these elements. However, the efficient and cost-effective recovery of these elements from complex waste matrices poses significant challenges. MBCs offer a promising solution due to their ability to selectively adsorb and concentrate target elements through tailored surface modifications, enabling efficient magnetic separation. This approach minimizes energy consumption and chemical reagent use compared to traditional methods. For instance, a MBC based on magnetic chitosan adsorbent (CDF-CS) has been successfully employed to recover gold from electronic waste [106], where a maximum adsorption capacity of $478.47\,mg\,g^{-1}$ and a >95% recovery rate were achieved at pH 7 and 25 °C. A study by Mohamady [107] has shown that magnetic-chitosan nanocomposite was effective in capturing and recovering Th(IV) from aqueous solutions, with a maximum adsorption capacity of $142.3\,mg\,g^{-1}$ at pH 3.5. In another study, a mesoporous MBC based on chitosan nanocomposite functionalized with cysteine (Cys) was used to adsorb uranium from contaminated water with a high efficiency of $136.9\,mg\,g^{-1}$ at pH 3.5 ∼ 5 and 25 °C [108].

Additionally, researchers have demonstrated the effective recovery of rare earth elements (REEs) from mining residues or waste streams using MBCs [109–111]. By using ion-imprinting technology, Liu and coworkers developed a Fe_3O_4/carboxymethyl cellulose (Fe_3O_4/CMC) MBC and found that Fe_3O_4/CMC exhibited high adsorption affinity and selectivity toward La(III) in aqueous solution, with $\boldsymbol{Q}_{max}$ up to $61.5\,mg\,g^{-1}$ at 25 °C [109]. Another study by Mohammadi et al. [110] indicated that the naturally magnetic biomass from the magnetotactic bacterium *Magnetospirillum magneticum* AMB-1 can capture $37.2\,mg\,g^{-1}$ of La(III) on a dry basis of biomass at pH 6, overperforming that reported for some other bacterial species. More recently, Ye et al. [111] developed a new

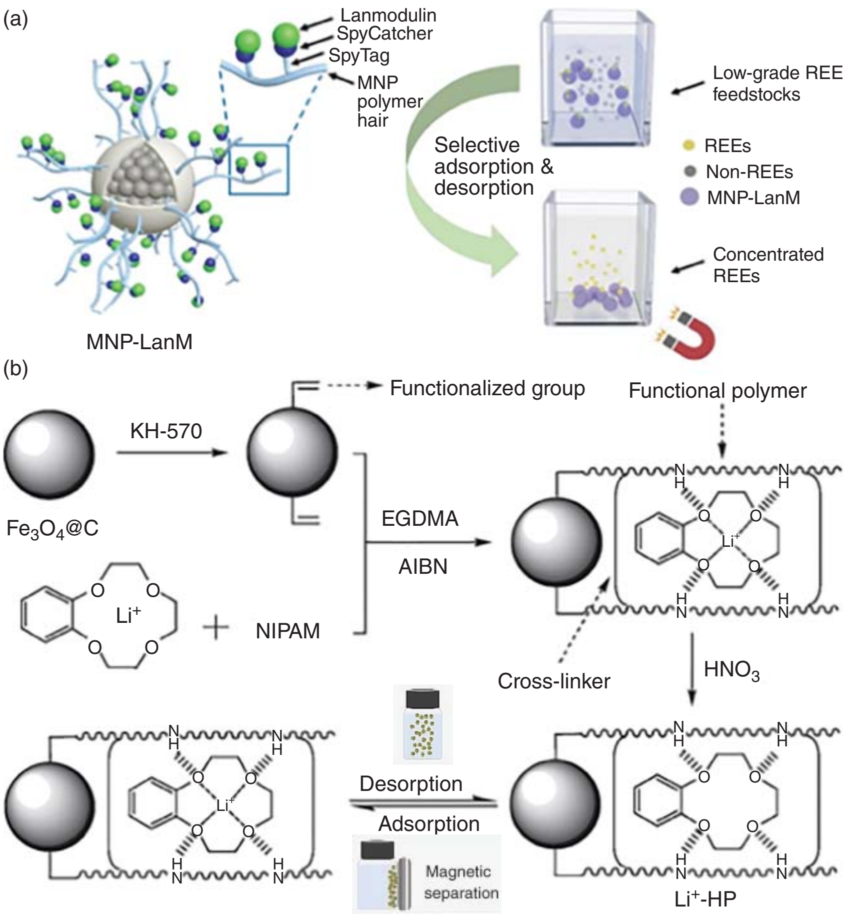

Figure 10.4 Examples of recovery of valuable elements by MBCs from wastewater. (a) Lanmodulin-SpyCatcher-loaded MNPs (MNP-LanM) for selective recovery of rare earth elements (REEs) from waste streams (Reprinted with permission from [111]. Copyright 2023 American Chemical Society). (b) Magnetic ion-imprinted polymers (IIP) for capturing low-concentration lithium ion from wastewater. *Source:* Adapted with permission from [112]. Copyright 2023 Elsevier B. V.

type of MBC (labeled as MNP-LanM) by immobilizing Lanmodulin-SpyCatcher (LanMSpycatcher) on the surface of SpyTag-functionalized MNPs. This MBC had demonstrated an adsorption capacity of $0.95\,mg\,g^{-1}$ for terbium (Tb) with fast adsorption kinetics and high selectivity, providing a promising solution for selective and efficient separation and recovery of REEs from low-grade feedstocks, e.g., the leachate of coal fly ash and geothermal brine (Figure 10.4a). In addition to REE recovery, Liang et al. [112] recently fabricated an ion-imprinted polymer (IIP) over the surface of magnetic carbon nanospheres, which can effectively and selectively capture as much as 23.5 mg g^{-1} of Li(I) from waste streams at 25 °C within 60 minutes. The recognition mechanism of this IIP toward Li(I) is due to the synergism of dehydration effect, electrostatic attraction, and size and imprinting effect (Figure 10.4b), facilitating the adsorption/desorption operations and recovery of Li(I) from aqueous medium. These successes highlight the potential of MBCs for a greener and more sustainable future in resource recovery from waste streams.

10.4.4 Other Applications

Apart from the above applications, MBCs have the potential to effectively break down organic pollutants through catalytic oxidation. Besides, MBCs can be used as adsorbents to capture organic substances from wastewater for the subsequent degradation in advanced oxidation processes. In combination with common advanced oxidation technologies such as photocatalytic oxidation, ozone oxidation, and advanced oxidation processes (e.g., Fenton reaction and photochemical oxidation) [113], MBCs usually play a crucial guiding role in facilitating the oxidant to target the pollutants more accurately in wastewater. For instance, Bornas et al. [114] fabricated a new Mn-Fe_3O_4-SiO_2/amine-glutaraldehyde/chitosan biocomposite (MIOSC-N-et-NH_2@CS-Mn) for the advanced oxidation process. This MBC was combined with an advanced oxidation process for the degradation of methylene orange with a removal efficiency of 98.9%. Ara et al. [115] reported a nanostructured chitosan-modified nickel magnetite biocomposite (CS-NM) recently and found that the MBC can effectively destruct MB and methyl red (MR) under the irradiation of sunlight, with photocatalytic efficiencies of 99% and 96% in 90 minutes for individual MB and MR, respectively. A recent study by Aryee et al. [116] shows that the MBC based on magnetic biomass/MOF composites (MPN@NH_2-MIL-101(Fe)) doped with nitrogen atoms was efficient in activation of peroxydisulfate (PS) for oxidative degradation of tetracycline compounds from wastewater (degradation efficiency ~ 87.6% in 60 minutes). Likewise, it was reported that the MBC based on microwave-assisted method using the methyl cellulose (MC) as a substrate ($CoFe_2O_4$@MC) can act as high-efficient activator of peroxymonosulfate (PMS), yielding a degradation efficiency up to 97% in degradation of reactive blue 19 under optimum conditions [117].

A more recent work by Verma and Moholkar [40] has shown that the Fe_3O_4@Laccase nanocomposite, based on the immobilization of laccase on the surface of amino-functionalized magnetic Fe_3O_4 nanoparticles, was very effective in treating industrial wastewater in the presence of sonication for chemical oxygen demand (COD), total organic carbon, and toxicity reduction. The optimization led to 90.7% COD removal and 70% toxicity reduction due to the synergistic effect of the enzymatic activity and the application of 35 kHz sonication. The Fe_3O_4@Laccase biocatalyst also demonstrated excellent recoverability and 72% retention of enzymatic activity for six consecutive treatment cycles.

10.5 Conclusions

Over the past two decades, magnetic biocomposites (MBCs) have emerged as a promising technology, offering a versatile platform for removing and recovering substances of environmental or valuable concerns from wastewater. Benefiting from their potential biological and ecological compatibility, tunable magnetic behavior, and affinity for various contaminants, these materials offer distinct advantages aimed at developing magnetic separation technologies of interest for water treatment units. These MBC adsorbents offer significant magnetic separation advantages by applying an external magnetic field in a confined space. This aspect is of great importance in waste storage or recycling and the reuse of adsorbents. While this technology holds immense potential, limitations and challenges persist, including the need for greater material scalability, stability, and regeneration methods to optimize their cost-effectiveness and environmental impact. In particular, information on large-scale evaluation and application of MBCs is scarce as most of the reports are laboratory-scale studies. Additionally, further research into tailoring the surface chemistry and structure of MBCs is essential for maximizing their adsorption capacities and selectivity for specific pollutants. To bridge the gap between laboratory results and full-scale application, more research is needed on the optimization of both the MBCs properties and the operation parameters in pilot-scale tests, with consideration of the complexity of real wastewater.

Looking ahead, the future of MBCs in wastewater treatment is bright, particularly with the integration of magnetic biocomposite technology with advanced oxidation processes (AOPs) such as heterogeneous photocatalysis or Fenton reactions for enhanced treatment. This synergistic approach of MBCs-AOPs would harness the unique properties of MBCs for efficient pollutant capture while utilizing AOPs to break down recalcitrant pollutants into less harmful by-products. As research continues to advance, efforts focused on optimizing this integration and developing scalable, cost-effective, and sustainable systems will be critical in transforming wastewater treatment and achieving a cleaner, healthier environment.

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