INVITED REVIEW
doi: 10.2306/scienceasia1513-1874.2025.s012

ScienceAsia 51S (1): 2025: ID 2025s012: **1**–**8**

Surface modification of magnetic nanoparticles for selective adsorption of organic dyes: A mini-review

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Received 14 Mar 2025, Accepted 30 May 2025
Available online 25 Jun 2025

ABSTRACT: This review explores the development of magnetic nanoparticles as effective adsorbents for the selective removal of anionic and cationic dyes from wastewater. The inherent magnetic separation feature of these adsorbents allows for easy recovery and reuse. By employing surface modification techniques, the selectivity of these magnetic nanoparticles can be enhanced through the incorporation of various surface modifiers. This review also highlights other factors influencing the selectivity of magnetic adsorbents. Despite promising results in the literature, further research is needed to address the limited studies on other pollutant types and to improve the selectivity of magnetic adsorbents for specific dyes. This review also aims to provide a foundational framework for the future design of advanced magnetic adsorbents in wastewater treatment applications.

KEYWORDS: wastewater treatment, dye selective adsorption, surface-modified magnetic nanoparticles

INTRODUCTION

According to a universal call to action to protect the planet known as the Sustainable Development Goals (SDGs), water pollution has been considered as an ongoing societal concern, largely driven by the growth of industrial activities [1, 2]. Among the various pollutants, organic dyes are significant contaminants often detected in wastewater [3–5]. These compounds pose serious risks to both human health and the environment due to their high toxicity, resistance to biodegradation, and potential carcinogenic effects [3, 4]. Consequently, effective methods for removing and/or recovering these pollutants from contaminated water are essential.

Among various wastewater treatment methods, adsorption stands out as an effective approach for removing pollutants, owing to its high performance, high reliability, straightforward operation and regeneration processes, and reasonable operating costs [6–8]. In order to obtain successful removal, this method basically requires key materials known as adsorbents. An effective adsorbent typically possesses high adsorption capacity, simplicity in its separation, good recyclability, and high selectivity for pollutant removal as shown in Fig. 1 [9, 10]. Notably, adsorbents with high selectivity can be applied for recovering the targeted molecules from a mixture of contaminants and/or serving as a sensor for the detection and identification of certain types of dyes [11, 12]. Furthermore, the design and development of effective adsorbents is one of the most challenging research topics for the future of wastewater treatment.

Recent advancements in the development of efficient adsorbents include the incorporation of magnetic materials as primary or secondary components of the designed materials [13]. Some of the superparamagnetic ferrite materials, such as iron oxide (Fe_3O_4) and magnesium ferrite ($MgFe_2O_4$) nanoparticles, have gained prominence as adsorbents due to their simple and rapid separation from solutions using an external magnet after the adsorption process [14]. This feature contributes to a more reusable and sustainable remediation process. However, adsorbents composed solely of a magnetic component often lack high selectivity, exhibiting preferential adsorption of various types of dyes, including cationic and anionic dyes, by these adsorbents. Therefore, it is crucial to explore additional components, such as organic and inorganic constituents, to provide active sites for specific adsorption. Furthermore, the surfaces of ferrite nanoparticles can be readily modified, making surface modification technique a high potential strategy for enhancing their removal performance [15–18].

Surface modification directly influences the selectivity of adsorbents by introducing specific functional groups (i.e., binding sites for targeted molecules) on their surfaces [19, 20]. In particular, functional groups with either positively or negatively charged functional groups facilitate selective adsorption of charge-bearing organic dyes. For instance, functional groups with negative charges effectively adsorb cationic dyes through electrostatic interactions. Therefore, selection of the appropriate surface modifiers is a critical step for fabricating adsorbents optimized for selective adsorption of dyes. Thus, this review focuses on exploring advancements in the surface modification of magnetic

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[†] Editorial Board of *ScienceAsia* 2023–present.

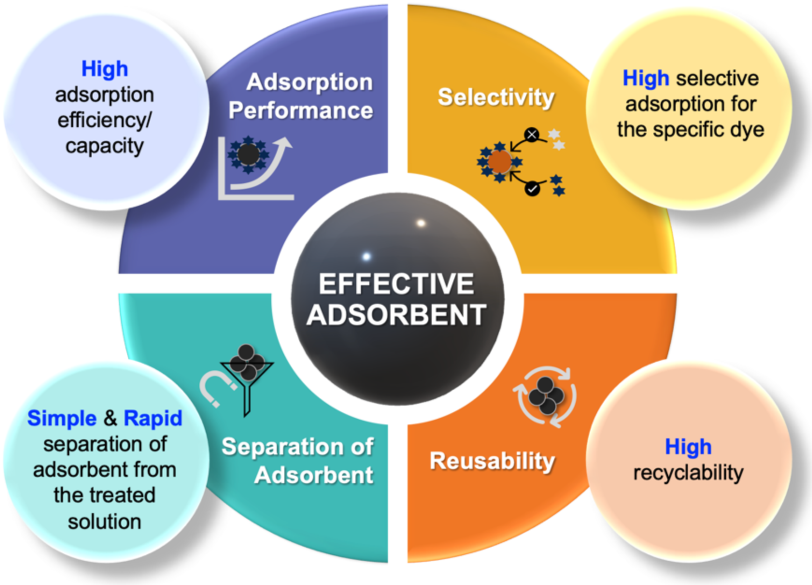

Fig. 1 Typical key characteristics of effective adsorbent.

nanoparticles to enhance the selective adsorption of anionic and cationic dyes.

SELECTIVE ADSORPTION OF DYES BY MAGNETIC ADSORBENTS

To enhance the efficiency of adsorbents, various materials have been integrated into magnetic nanoparticles. This section explores how the selectivity of magnetic adsorbents can be categorized based on the charge-charge interaction between the adsorbent and the dyes. Based on the main charge-charge (electrostatic) interactions, the selectivity of magnetic adsorbents is divided into three groups: (1) selectivity for removing anionic dyes, (2) selectivity for removing cationic dyes, and (3) tunable selectivity for removing either anionic or cationic dyes by adjusting the solution pH.

(1) Selective adsorption of anionic dyes

Metal-organic frameworks (MOFs) have been employed to modify the surfaces of magnetic nanoparticles, enhancing their selectivity for anionic dye removal. For instance, the surfaces of poly(styrenesulfonate)-functionaized Fe_3O_4 nanoparticles were modified with ZIF-67 to create Fe_3O_4-PSS@ZIF-67 adsorbent for selective adsorption of methyl orange (MO) [21]. This adsorbent preferentially removed MO from a binary dye solution containing MO and a cationic dye, methylene blue (MB). The high selective adsorption of MO was attributed to strong electrostatic interaction between the Co(II) center of ZIF-67 and the sulfonate ($-SO_3^-$) group of MO. Similarly, La-MOF-NH_2 was functionalized onto Fe_3O_4 nanoparticles (La-MOF-NH_2@Fe_3O_4) to enhance selective adsorption of Congo Red (CR) [22]. The adsorption study indicated that La-MOF-NH_2@Fe_3O_4 adsorbent selectively adsorbed CR from mixtures of CR with other anionic and cationic dyes, facilitated by electrostatic interactions between the protonated amine ($-NH_3^+$) groups of La-MOF-NH_2 and the $-SO_3^-$ groups of CR. Additional interactions, such as host-guest interaction also contributed to the selective adsorption of CR.

Polymer-based materials have also been utilized as surface modifiers. The surfaces of Fe_3O_4 nanoparticles were coated with catechol and polyethylenimine (PEI) via a co-polymerization reaction (Fe_3O_4@catechol/PEI) [23]. Adsorption results showed that this adsorbent selectively removed several anionic dyes (i.e., Methyl Blue, Orange G, and Amaranth) from mixtures of these dyes with cationic dyes. The selective adsorption was attributed to electrostatic interaction between the protonated amine and imine groups of the catechol/PEI component and the negative charges of the anionic dyes. Interestingly, the adsorption isotherm indicated a preferable adsorption for Methyl Blue over other anionic dyes. Recently, alkaline lignin (AL), chitosan (CTS), and PEI were cross-linked onto Fe_3O_4 nanoparticles in the presence of glutaraldehyde, yielding AL-CTS@MNPs adsorbent [24]. This adsorbent effectively removed MO from binary dye solutions containing MO and cationic dyes. The selective adsorption for MO was mainly driven by electrostatic attraction between the protonated amines of CTS/PEI and negative charge of MO. Additionally, the π-π^* electron-donor acceptor interaction partially facilitated the selective MO removal.

Other types of modifiers have also been reported for selective adsorption. For example, the surfaces of $MgFe_2O_4$ nanoparticles were functionalized with ethanolamine ($MgFe_2O_4-NH_2$ NPs) [25, 26]. Adsorption studies demonstrated that $MgFe_2O_4-NH_2$ adsorbent effectively removed anionic dyes, particularly CR and Indigo Carmine (IC). The proposed adsorption mechanism involved electrostatic attraction between the $-NH_3^+$ groups of the adsorbents and the $-SO_3^-$ groups of the dyes. However, this adsorbent removed both CR and IC from the CR/IC binary dye solution. Subsequently, the $MgFe_2O_4-NH_2$ adsorbent was further modified through silica encapsulation and amino functionalization using 3-aminopropyltriethoxy silane (APTES) [26]. This modified adsorbent exhibited high selectivity for CR removal from the CR/IC solution. The improvement of selectivity was rationalized by the presence of amine and silanol groups, derived from APTES and silica, respectively. In addition to electrostatic attraction, hydrogen bonding interaction also contributed to the enhanced selectivity for CR adsorption.

(2) Selective adsorption of cationic dyes

In addition to anionic dyes, magnetic adsorbents can be designed for the selective removal of cationic dyes. MXenes, a new class of two-dimensional (2D) materials, have been used to modify magnetic nanoparticles for the selective adsorption of dyes. For instance, titanium carbide ($Ti_3C_2T_x$) MXene was modified onto the surfaces of Fe_3O_4 nanoparticles via an *in situ* growth method (MXene@Fe_3O_4) [27]. In alkaline conditions, the MXene@Fe_3O_4 adsorbent demonstrated selective adsorption of MB from the mixtures containing MB and anionic (MO) and cationic (Rhodamine B, RhB) dyes. The selectivity for MB removal was attributed to electrostatic attraction between the deprotonated hydroxyl ($-O^-$) groups of MXene and the cationic MB. A later synthesis of the $Ti_3C_2T_x/Fe_3O_4$ adsorbent also revealed preferential adsorption of both MB and RhB from ternary dye solutions comprising these dyes and the cationic Chrysoidine G (CG) dye [28]. The selective removals of MB and RhB were rationalized by the presence of surface functional groups (e.g., $-OH$, $[Ti-O]^-$, and $[Ti-F]^-$) on $Ti_3C_2T_x$, which facilitated electrostatic attraction with the cationic dyes. Notably, even though the chemical compositions of these adsorbents were similar, the selectivity for dye removal varied across systems.

In addition to MXenes, polydopamine (PDA) and lignin have also been reported as effective surface modifiers for enhancing the selective removal of dyes. Dopamine, as a monomer, was polymerized onto the surfaces of Fe_3O_4 nanoparticles in the presence of Fe(III) ions as a stabilizer (Fe_3O_4@PDA-Fe(III)) [19]. This adsorbent exhibited selectivity for MB adsorption from mixtures containing MB and MO and RhB, with selectivity arising from electrostatic attraction between the $-O^-$ groups of PDA matrix and cationic MB. Additionally, several other interactions, such as π-π stacking, hydrogen bonding, and complexing interactions involved the selective adsorption of MB due to the presence of amines, Fe(III), and aromatic rings within the PDA-Fe(III) matrix. In another study, the Fe_3O_4@PDA was further polymerized with various deep eutectic solvents (DESs) [29]. The as-obtained adsorbents selectively removed various cationic dyes, particularly Crystal Violet (CV), from the mixture comprising CV and anionic Tartrazine dye. The synergistic effect of electrostatic interaction, hydrophobic force, hydrogen bonding, and π-π stacking was noted, emphasizing that the chemical structure of the dyes influenced selectivity even when the adsorbent favored cationic dye adsorption.

Silica (SiO_2) is another promising modifier for producing adsorbents with selectivity for cationic dye adsorption due to its abundant silanol (Si—OH) groups. In one study, $MgFe_2O_4$ nanoparticles were encapsulated within a silica shell ($MgFe_2O_4$@SiO_2) [30]. The adsorption behavior revealed that the $MgFe_2O_4$@SiO_2 adsorbent selectively adsorbed MB from mixtures containing MB and anionic, cationic, and neutral dyes. The high selectivity for MB removal was primarily attributed to electrostatic attraction between the deprotonated silanol ($Si-O^-$) groups on the silica surface and the positive charge of MB. Furthermore, it was emphasized that the chemical nature of the dyes (molecular charge and size), the surface charge of the adsorbent, adsorption conditions, and interaction among dyes and between dyes and adsorbents should all be taken into account when assessing selectivity. Other materials, such as MOFs-based materials (e.g., ZIF-8 [31] and polyDES-functionalized HKUST-1-NH_2 [32]), lignin-based materials (e.g., lignin regenerated from ionic liquid solutions [33]), hyperbranched polymers (e.g., hyperbranched polyglycerol [34]), have also been reported as efficient surface modifiers for the selective adsorption of cationic dyes.

(3) pH-tunable selective adsorption of anionic and cationic dyes

Moreover, the selectivity of dye removal can be fine-tuned by tailoring the pH of the dye solution. Basically, adjusting the solution pH directly influences the overall surface charge of adsorbents, enabling selective adsorption of either cationic or anionic dyes. Zwitterionic polymers are promising materials for achieving pH-tunable selective adsorption, as these polymers contain at least two functional groups capable of exhibiting either positive or negative charges depending on the pH of the solution [35]. By switching the surface charges of the adsorbents, it becomes possible to selectively remove dye molecules with the opposite charge.

For instance, poly(cysteine methacrylate)

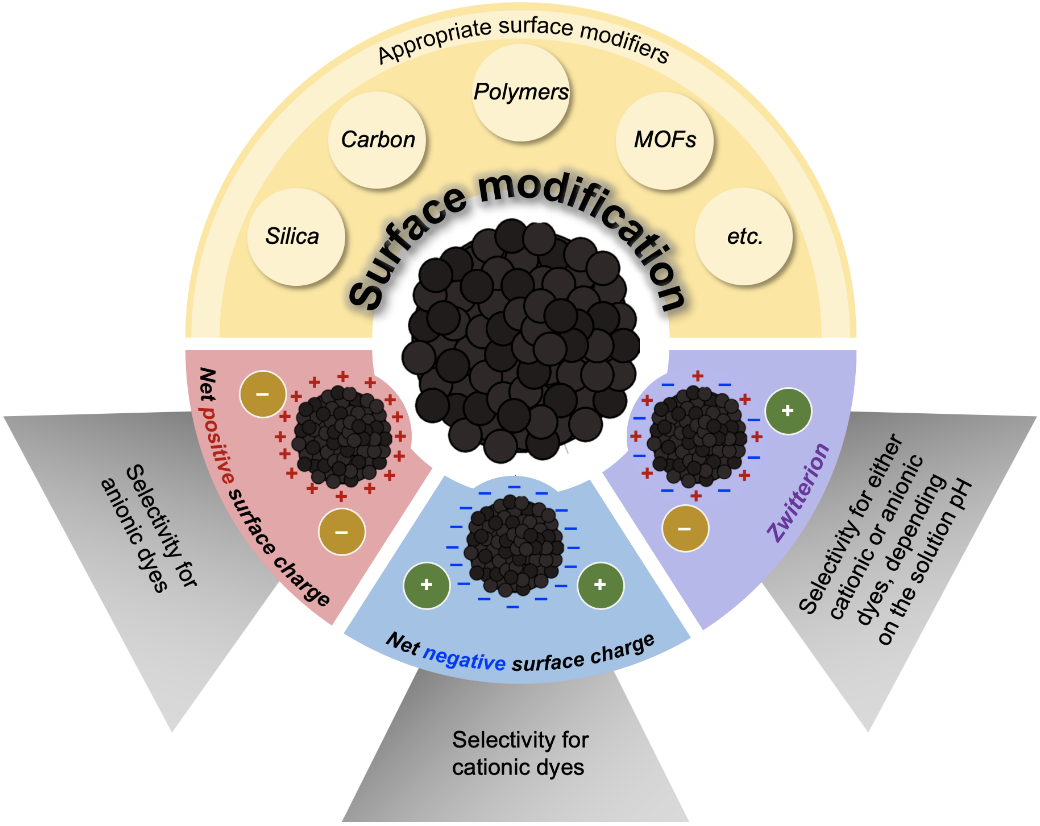

Fig. 2 Surface modification of magnetic nanoparticles for selective adsorption of organic dyes.

(PCysMA) was modified onto the surfaces of $MgFe_2O_4$ and $MgFe_2O_4@SiO_2$ nanoparticles [36]. The as-obtained $MgFe_2O_4$/PCysMA adsorbent demonstrated selective adsorption of anionic IC and cationic MB dyes by tuning the solution pH to ∼2 and ∼10, respectively. The selectivity for the removal of these dyes was attributed to electrostatic attraction between the $-NH_3^+$ (at pH ∼2) or $-COO^-$ (at pH ∼10) groups of PCysMA and the respective dyes. Additionally, the $MgFe_2O_4@SiO_2$/PCysMA adsorbent exhibited higher selectivity and adsorption capacity for the removal of MB and IC. The presence of silica influenced the chemical composition of the adsorbent, contributing to the changes in selectivity and adsorption capacity of the adsorbent.

Similarly, lysine and poly(glycidal methacrylate) (Lys-PGMA) were coated onto the surfaces of Fe_3O_4 nanoparticles to produce Fe_3O_4@Lys-PGMA adsorbent [37]. This adsorbent exhibited the selective removal of anionic Lemon Yellow (LY) and cationic MB dyes at appropriate pH ∼4 and ∼10, respectively. The presence of amine and carboxylic groups in the Lys-PGMA component contributed to the pH-tunable selectivity of the adsorbent. Furthermore, protonated amine-modified hydrochar (PAMH) was combined with the Fe_3O_4 nanoparticles to achieve the pH-tunable selective adsorption of dyes [38]. According to the literature, it appears that the surface modification of magnetic nanoparticles with suitable components to create pH-tunable selective adsorbents has been infrequently reported to date.

Overall, the surface modification of magnetic nanoparticles directly affects the chemical composition of the adsorbent surface. The presence of appropriate surface functional groups is crucial for the selectivity of the adsorbent. Generally, functional groups that can display positive charges (e.g., primary, secondary, tertiary, and quaternary amines) are effective surface modifiers for the selective adsorption of anionic dyes. Conversely, functional groups that can exhibit negative charges (e.g., hydroxyl and carboxylic groups) are suitable for producing adsorbents with selectivity for cationic dye removal. Therefore, the choice of surface modifier significantly influences the selectivity of the design adsorbents, as illustrated in Fig. 2.

However, achieving the right balance of surface charge (i.e., net positive or net negative charges) on magnetic adsorbents for the selective adsorption of both anionic and cationic dyes presents a challenge. In addition to selecting potential candidates

Table 1 Selectivity of magnetic adsorbents for removal of cationic and anionic dyes.

Adsorbent	Selectivity of adsorbent[1]				Additional advantages	Ref.
	Surface modifier	Specific dye	Binary dye systems	Adsorbent-dye interaction[2]		
Selectivity for anionic dye removal						
Fe_3O_4@NH_2/PEI	PEI	ARS, MO	ARS/MB MO/MB	Electrostatic	Amine-rich groups High adsorption capacity	[39]
Fe_3O_4@PDA/PEI	PDA PEI	OG, RB	OG/CV OG/MB RB/MB	Electrostatic	Rapid adsorption Alkaline resistance	[40]
MWCNTs/Fe_3O_4@(PDA/PEI)	PDA PEI MWCNTs	CR, MO	CR/MB MO/MB	Electrostatic, H-bonding	High specific surface area High adsorption capacity	[41]
γ-Fe_2O_3@C@UiO-66-NH_2	Carbon UiO-66-NH_2	AO, AF	AO/MB AF/MB	Electrostatic	Good structural stability	[42]
Fe_3O_4-MgAl	MgAl	PR	PR/MB	Electrostatic, H-bonding	High adsorption capacity	[43]
Selectivity for cationic dye removal						
$MnFe_2O_4$@SiO_2@ZIF-8	SiO_2 ZIF-8	MG	MG/MO	π-π stacking	High specific surface area High adsorption capacity	[31]
Fe_3O_4@HKUST-1@PDES	HKUST-1 PDES	MG, CV	MG/LY CV/LY	Electrostatic, H-bonding, Lewis acid-base reaction	High adsorption capacity Used as MSPE	[32]
Fe_3O_4@Lignin	Lignin	MB, MV	MB/MO MV/MO	Electrostatic, H-bonding, π-π stacking	High adsorption capacity	[33]
HPG-based MMG	HPG Carboxylic acid	MB, MV	MB/MO MV/TTZ	Electrostatic	Rapid adsorption High adsorption capacity	[34]
Fe_3O_4-MgAl/SDS	MgAl SDS	MB	PR/MB	Electrostatic, Hydrophobic	High adsorption capacity	[43]
HPIM/Fe_3O_4	HPIM	MB	MB/MO	Electrostatic	High adsorption capacity Adsorption stability under strong ionic environment	[44]
BC/AA/MN@Fe_3O_4	BC AA MN	MB, MG, RhB	MB/MG CV/MG RhB/MG MG/MO	Electrostatic, H-bonding, Ion-exchange	Highly porous material High adsorption capacity	[45]
Fe_3O_4@Ti-SiO_2	Ti-SiO_2	MB, MG	MG/MO MB/MO	Electrostatic, H-bonding	High adsorption capacity	[46]

[1] Selectivity revealed under optimal adsorption conditions.

[2] These interactions mainly involve selective adsorption; MWCNTs: Multi-walled carbon nanotubes; HPG-based MMG: Hyperbranched polyglycerol-based multi-carboxylic magnetic gel; MgAl: magnesium-aluminium layered double hydroxide; SDS: sodium dodecyl sulfate; PDES: polymeric deep eutectic solvents; MSPE: magnetic solid-phase extraction; HPIM: hydrolyzed polymers of intrinsic microporosity; BC/AA/MN: Bamboo nanocellulose/acrylic acid/montmorillonite nanosheets; ARS: Alizarin Red S; OG: Orange G; RB: Rose Bengal; AO: Acid Orange; AF: Acid Fuchsin; CM: Carmine; SY: Sunset Yellow; PR: Ponceau Red; MG: Malachite Green; LY: Lemon Yellow; MV: Methyl Violet; TTZ: Tartrazine.

for surface modification, optimizing the surface modification conditions of magnetic nanoparticles is also important. Some surface modification processes may require multiple steps and/or complex reactions to fabricate magnetic adsorbents with high selectivity, which can be time-consuming and increase production costs. Furthermore, the surface modification of magnetic nanoparticles directly influences their magnetic properties. A decrease in the magnetization of the adsorbents after modification is often observed, owing to the presence of non-magnetic constituents (i.e., surface modifiers). Thus, the amount of surface modifiers introduced must be optimized to ensure it is sufficient to achieve high selectivity without compromising the magnetic separation capability of the adsorbent.

In addition to charge-based interactions, the presence of other functional groups that do not carry any charge can further enhance selectivity through other adsorbent-adsorbate interactions. Beyond the chemical composition of the adsorbents, the chemical nature of adsorbate (i.e., the dye molecules) also plays a pivotal role in determining selectivity. The literature also indicates that the adsorption behavior of various magnetic adsorbents can vary significantly across different systems (Table 1), highlighting the complexity and system-specific nature of adsorption processes.

CONCLUSION AND FUTURE PERSPECTIVES

The surface modification of magnetic nanoparticles represents a promising approach for fabricating magnetic adsorbents with selectivity for the removal of anionic and cationic dyes. By carefully selecting suitable surface modifiers, it is possible to achieve magnetic adsorbents that exhibit the desired selectivity for dye adsorption. These highly selective magnetic adsorbents could be practically applied in wastewater treatment. In addition to their capability for selective removal of target pollutants, these adsorbents are easily reusable after the treatment process, owing to their magnetic separation feature. However, there is still considerable room for research on the selectivity of magnetic adsorbents for the removal of organic dyes. Addressing these challenges as listed below will pave the way for more effective and versatile adsorbent materials in the fight against water pollution.

- The number of studies focused on designing and fabricating magnetic adsorbents with selectivity for dye adsorption through surface modification is limited. Specifically, the selective removal of other types of pollutants, such as pesticides, has rarely been reported.

- While several magnetic adsorbents have been shown to exhibit high selective adsorption of either anionic or cationic dyes, some adsorbents have been reported to remove multiple types of anionic or cationic dyes, indicating low selectivity. Enhancing selectivity for specific dyes remains a challenge.

- Many studies on selective adsorption using magnetic adsorbents have been conducted predominantly in binary dye systems. However, real wastewater typically contains complex mixtures of multiple dyes and contaminants. Future studies should therefore move beyond binary systems to investigate adsorption behaviors in more realistic, multi-component systems, such as ternary or even more complex combinations. Such investigations are essential for accurately assessing and enhancing the practical selectivity and applicability of adsorbents in real-world wastewater treatment scenarios.

- Although some materials have been reported as pH-tunable adsorbents, there has been little research on achieving pH-tunable selective adsorption through the surface modification of magnetic nanoparticles.

- Future research should prioritize the development of multifunctional magnetic adsorbents with tunable selectivity and efficient recyclability. These materials would support adaptable treatment processes for diverse pollutant mixtures and varying environmental conditions, significantly enhancing the practical applicability and long-term sustainability of adsorption technologies.

- In addition to experimental approaches, computational modeling and machine learning could offer promising strategies for predicting and optimizing adsorbent performance, specifically selectivity. Advanced computational techniques, such as molecular dynamics and density functional theory calculations can clarify molecular interactions, while machine learning enables rapid screening of high-performing materials, significantly streamlining the adsorbent design process.

- Future studies should address the inherent trade-off between enhancing surface functionality to improve selectivity and maintaining sufficient magnetic properties for effective adsorbent recovery. Strategies to balance surface modification with magnetic performance, such as precise control over the degree of functionalization or the design of thinner, multifunctional coatings, represent a critical area for future investigation.

Acknowledgements: The authors acknowledge helpful discussions and suggestions from the anonymous colleagues.

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