

Review on recent advancements in layered double hydroxides application for effective removal of heavy metals and organic micro-pollutants: Challenges and future perspectives

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Abstract

The rapid expansion of agricultural and industrial activities has led to the release of various pollutants, including heavy metals (HMs), organic pollutants (OPs), pharmaceuticals, and emerging contaminants, posing substantial risks to environmental and human health. Traditional remediation methods struggle with questions of efficiency, specificity, and sustainability. This review offers a comprehensive and critical analysis of recent advancements in adsorbent technologies, with a specific focus on the application of layered double hydroxides (LDHs) for the removal of HMs and OPs. LDHs are considered promising materials for pollutant removal because of their notable characteristics, including stable mineralization, low solubility product constants, anionic intercalation capacity, high surface area, and tunable surface chemistry. However, the review also addresses the challenges associated with large-scale LDH applications. While LDHs demonstrate significant potential as alternatives to conventional adsorbents, further research is required to enhance their surface modification, structural design, and post-treatment recovery processes for more effective applications. Finally, this review develops a foundation for future studies on the mechanical features and practical implementation of LDH-based remediation technologies, consolidating current knowledge and identifying critical research gaps.

Keywords: Heavy Metals; LDHs; Organic Pollutants; Wastewater Treatment; Removal of Pollutants

1. Introduction

The ongoing transformation of global society and the economy has resulted in an intensified focus on critical ecological concerns (Farhan et al. 2024; Pelalak et al. 2023). Human activities, such as agriculture, industrial production, and urbanization, release substantial amounts of pollutants into the environment, contributing to considerable ecological and public health risks (Li ChunMei et al. 2019; Bhatt et al. 2021). Among these contaminants, HMs such as cadmium (Cd), zinc (Zn), chromium (Cr), and nickel (Ni), as well as OPs such as pesticides, antibiotics, and dyes, are particularly concerning. Despite their presence at low concentrations, these substances can lead to severe health effects, emphasizing the necessity for effective removal strategies.

Over the past decade, nanotechnology has emerged as a primary strategy for water purification. Among various adsorbents, LDHs have garnered significant attention due to their exceptional physicochemical features, including a high specific surface area, excellent hydrophobicity, efficient ion exchange ability, cost-effectiveness, and a high interlayer negative charge density (Zubair et al. 2021; Lu et al. 2024). Structurally, LDHs are anionic clays characterized

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by a general molecular formula of $\left\{A_x\right\}_n - mH_2O$. They comprise brucite-like layers in which octahedral metal oxides form the primary lattice, with metal cations occupying octahedral sites (Lu et al. 2016). The molar ratio M^{3+} to $M^{2+} + M^{3+}$ of metal cations typically ranges from 0.2 to 0.4 (Zhang et al. 2016; Zhang, He, et al. 2022). The partial substitution of divalent cations for trivalent metals induces positively charged layers, balanced by intercalated anions. These layers alternate with M^{2+} and M^{3+} ions along the a- and b-axes, while water molecules and neutral anions occupy the interlayer space along the c-axis, forming a well-defined three-dimensional structure (Bai et al. 2024; Zheng et al. 2019).

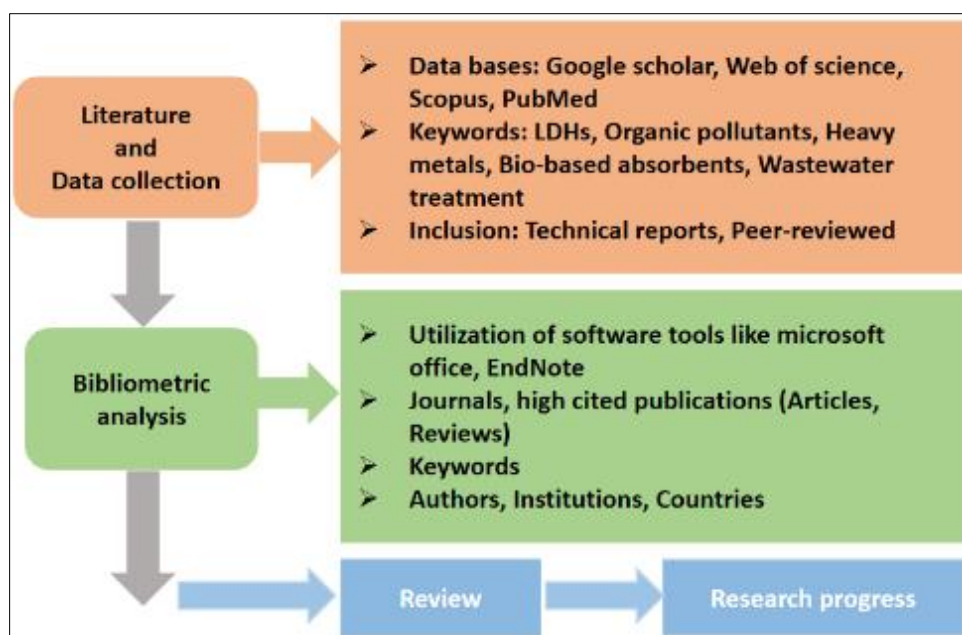


Figure 1 Approach for analyzing LDHs as bio-based absorbents

Table 1 Overview of recent studies on HMs and OPs removal via LDHs

References	Main Subject	Gaps and Contribution
(Sheraz et al. 2024)	Comprehensive assessment of carbon, bio-material, and inorganic-based adsorbents for the removal of the most hazardous heavy metal ions from wastewater.	This paper is the comparative effectiveness of different types of adsorbents in targeting specific HM ions in wastewater.
(Zahara et al. 2023)	LDH Zn/ M^{3+} ($M^{3+} = \text{Al and Cr}$) as highly efficient adsorbent of HMs Pb (II).	The paper focuses on Zn-Al and Zn-Cr LDHs as efficient adsorbents for Pb(II) removal.
(Hamimed et al. 2022)	Nanocelluloses for removal of HMs from wastewater.	This paper presents a promising and innovative solution for the removal of HMs from wastewater.
(Mohiuddin et al. 2021)	Starch-Mg/Al LDH composites as an efficient solid phase extraction sorbent for non-steroidal anti-inflammatory drugs as environmental pollutants.	This paper highlights the potential for biocompatible materials to enhance the removal of environmental pollutants.

This review offers a comprehensive overview of LDH applications for the removal of HMs and OPs, providing insight into recent advancements, research trends, and challenges in the research area. The purpose is to implement LDH-based technologies for environmental remediation (Meseguer-Sánchez et al. 2021; Wang, Bai, et al. 2023). To ensure a systematic and effective perspective, this review emphasizes three major objectives: tracing the origin and progression of pollutants, evaluating the efficacy of LDHs in pollutant removal, and identifying current limitations and addressing future research directions. A systematic literature review was conducted using prominent scholarly databases, including Scopus, Web of Science, PubMed, and Google Scholar. Keywords such as "bio-based adsorbents," "LDHs," "OPs," "HMs," and "wastewater treatment" were employed to retrieve peer-reviewed publications, technical reports,

and demonstration projects from the past decade. This approach ensured the inclusion of current and relevant scientific research (Fig. 1). The quality and relevance of the selected literature were evaluated, and duplicates were excluded. The reviewed studies were categorized into key thematic areas, including LDH synthesis, adsorption mechanisms, pollutant removal efficiency, scalability, and environmental impact. Table 1 provides a summary of current research on LDH applications for HM and OP removal.

2. LDHs application benefits

LDHs have gathered significant attention in the recent scientific literature as innovative functional materials for the removal of HMs and OPs (Tian et al. 2024). These materials possess tunable physicochemical characteristics, which include precisely controllable crystal morphology, modifiable interlayer anion configuration, and adjustable molar ratios of host layer cations (Zubair, Syamaladevi, and Ullah 2024; Zubair et al. 2021). LDHs are considered highly promising adsorbents for wastewater treatment, due to their large specific surface area and interlayer anion exchangeability (Guo et al. 2011; Lu et al. 2016; Li et al. 2025). Furthermore, their chemical stability and adaptability make them potential candidates for soil remediation applications (Guan et al. 2022; Zhao et al. 2020).

The Al-Li/ThLDH@CNT nanocomposite, developed by Manea et al. (2022), demonstrated a 98% degradation of malachite green (MG) dye within 45 minutes under visible light irradiation (Manea et al. 2022). Similarly, palladium and tin sulfide (Pd-SnS) nanoparticles containing LDHs exhibited enhanced stability and photocatalytic activity against rhodamine B and hexavalent chromium (Sun, Lee, and Park 2023). Zhang et al. (2022) synthesized a chitosan-LDH composite with high adsorption capacities for various HM ions and organic dyes, operating through diverse adsorption mechanisms (Zhang, Ma, et al. 2022). Additionally, a copper-based oxide/LDH compound was shown to effectively remove tetracycline (TC) and Cr(VI) with a synergistic efficiency of approximately 95%. The copper-based oxide/LDHs-1/15 composite displayed impressive durability and stability during successive photocatalytic cycles in practical applications.

3. Preparation methods of LDHs

LDHs can be efficiently synthesized in laboratory settings due to their favorable thermodynamic properties and the regular interactions of appropriate anions and cations. LDHs are non-toxic and environmentally friendly, as their synthesis involves a series of low-cost, eco-friendly approaches that employ water as a solvent, such as coprecipitation, hydrothermal treatment, and sol-gel methods (Fig. 2). Although LDHs are relatively uncommon, they can also be produced from inexpensive materials (Oh, Hwang, and Choy 2002). The synthesis process is influenced by several factors, including temperature, pH, and reaction time (Zhang et al. 2012). LDH synthesis methods are generally categorized into direct approaches, such as hydrothermal and co-precipitation techniques, and indirect approaches, such as anion exchange (He et al. 2006; Zubair et al. 2017). This review also discusses procedures for synthesizing LDH composites through conventional methods.

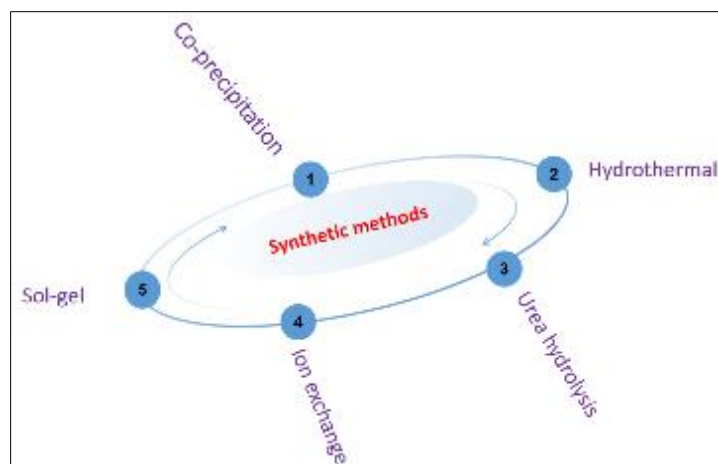


Figure 2 Preparation methods of LDHs

3.1. Co-precipitation

Co-precipitation, also referred to as the salt route or salt-based route, is the most widely applied method for the synthesis of LDHs due to its versatility across a broad pH range (Mittal 2021; dos Santos et al. 2021; Tang et al. 2024). This method involves two types of precipitation: low and high super-saturation, with specific processes employed to adjust the pH (Ng'etich and Martincigh 2021; Evans and Slade 2006). The properties of the resulting LDHs, whether amorphous or highly crystalline hydrotalcite, are influenced by several factors, including aging time, base solution, temperature, metal molar ratios, pH, and cation concentration (Bukhtiyarova 2019; Wang and O'Hare 2012). The process entails the simultaneous precipitation of metal hydroxides from metal salts in deionized water, followed by the insertion of an alkaline solution to enhance the pH (Bukhtiyarova 2019; Ng'etich and Martincigh 2021; Evans and Slade 2006). After sufficient aging, the precipitates are rinsed, filtered, and dried to achieve the final LDH product.

3.2. Hydrothermal

The hydrothermal synthesis reaction is conducted by combining M(II) and M(III) metal salt solutions dissolved in an alkaline solution, exhibiting characteristics akin to those of the coprecipitation process. This reaction occurs in a heated autoclave at temperatures of 100–180°C and a pH range of 8–10 for 10–48 hours (Daud et al. 2016; Mohapi et al. 2020; Bai et al. 2024). The synthesized solids are extracted via centrifugation and subsequently washed with ethanol and deionized water. Hydrothermal synthesis has demonstrated superior efficacy compared to the coprecipitation method in producing LDHs with high crystallinity and distinct morphologies. This method underscores its significance in the preparation of advanced inorganic nanomaterials for contemporary green technologies, facilitating enhanced chemical reactions and temperature conditions (Rao et al. 2007; Wang et al. 2021).

3.3. Urea hydrolysis

Urea hydrolysis typically entails rapid supersaturation with the precipitant (OH^-), leading to the nucleation of mixed hydroxides, independent of Ostwald particle growth, which includes a range of particle sizes (Pang et al. 2019; Karim et al. 2022). Alkaline retarders, such as urea, are used to decouple nucleation from particle formation, thereby enabling aging retardation (Adachi-Pagano, Forano, and Besse 2003). The process of urea hydrolysis occurs in two stages: the initial formation of intermediates that govern the rapid hydrolysis to ammonium carbonate. The rate of hydrolysis is temperature-dependent, and LDHs can be synthesized through various heat treatments. The urea-to-metal salt ratio significantly influences particle size and can produce large sheets and well-crystallized hexagonal LDHs (Ogawa and Kaiho 2002; Benali et al. ; Okamoto, Iyi, and Sasaki 2007).

3.4. Ion exchange

The anion exchange process in LDHs involves dispersing the precursor LDH in an aqueous solution containing the target anion, followed by continuous stirring at room temperature for several hours (Mohapi et al. 2020; Omwoma et al. 2014). The solid sediments are filtered via filtration, repeatedly cleaned with deionized water, and subsequently dried. Additionally, water molecules in the interlayer play a crucial role, with hydrogen bonding being important for their arrangement across the inorganic layer. The high anion exchange capacity of LDHs facilitates interlayer ion exchange, simplifying the synthesis of LDH precursors (Guan et al. 2022; Cui et al. 2023).

3.5. Sol-gel

The sol-gel technique, employing condensation and hydrolysis methods, is considered a straightforward, cost-effective, and efficient moist-chemistry approach for synthesizing highly pure metal oxides from LDH precursors (Prince et al. 2009; Danks, Hall, and Schnepf). In comparison to coprecipitation, sol-gel-derived LDHs exhibit a higher specific surface area and superior thermal stability but are characterized by lower crystallization levels (Aramendía et al. 2002; Elhalil et al. 2019).

4. Modification techniques of LDHs

The electron state of functional groups significantly influences the zeta potential of LDHs, enabling effective material customization through component combinations and synthesis methods. Their composites serve as efficient adsorbents for OPs HMs due to their exchangeable interlayer anions (Dai et al. 2024). Nanosheets exhibit dynamic tunability, with surface bonds that are more reactive than bulk formulations. Minor adjustments in small-scale synthesis can substantially alter adsorption properties (Fig. 3). Consequently, physicochemical modifications are crucial for optimizing performance in adsorbing HMs and OPs (Miyah et al. 2024). Table 2 provides a summary of LDH-based material synthesis and modification strategies, while additional preparation techniques are detailed in Table S1.

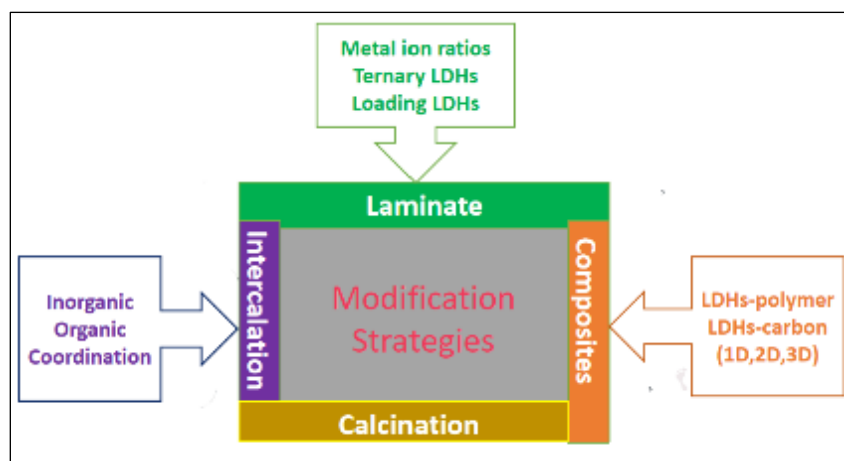


Figure 3 Modification techniques of LDHs

Table 2 Recent utilization of LDH-based components

Material	Modification method	Pollutants	Adsorptive capacity (mg/kg)	Reference
CuFeAl-LDO	Laminate	Tetrabromobisphenol A	99.91%	(Hou et al. 2022)
ZnCr-LDH	Calcination	Pyrophosphate	79.00	(Fu et al. 2021)
ZnIn-LDH	Calcination	Methylene bleu	98.00%	(Liu et al. 2020)
MgFe-LDHs-AC	Composites	Cu^{2+}	2.03	(Song et al. 2024)
CoAl-LDH	Composites	Ranitidine	97.00%	(Asif, Kang, and Zhang 2022)

4.1. Laminate

4.1.1. Metal ion ratios

The ratio of metal ions significantly influences the efficacy of pollutant adsorption into LDHs. Recent studies have demonstrated that a 5:1 MgAl ratio achieves superior phosphorus removal efficiency (Lee et al. 2019; Deng et al. 2021; Jung et al. 2021). Additionally, research has shown that varying metal ion proportions (2:1, 3:1, 4:1) during LDH preparation impact their performance, with a 4:1 ratio maintaining optimal performance (Li et al. 2016; Wan et al. 2012). While pore capacity decreases with higher metal ion ratios, the pore surface area increases, enhancing pollutant adsorption. The current approach involves dispersing LDHs into biochar (BC) ponds (Vallet-Regi et al. 2004; Montiel-Centeno et al. 2025; Pandey et al. 2021). Furthermore, as Mg/Al ratios increase, the lower charge density facilitates the entry of pollutant ions into larger interlayer spaces, thereby improving adsorption capacities (Li et al. 2016; Wan et al. 2012).

4.1.2. Loading LDHs

ZnO, TiO₂, Cu₂O, and similar semiconductors are recognized for their low cost and excellent photocatalytic performance, though they exhibit poor transport properties (Gao, Yu, and Xu 2018; Hu et al. 2025; Mortaheb et al. 2008). To mitigate these limitations, various metal materials, such as Cu, Ag, and Ni, as well as carbon materials like carbon nanotubes, are employed due to their capacity for electron storage and transport (Li, Liu, et al. 2023). Combining these materials with LDHs enhances visible light absorption and hot carrier performance. For instance, the cadmium sulfide/CoAl-LDHs nanocomposite demonstrated improved light absorption and achieved a bandgap of 2.15 eV (Cai et al. 2017). Additionally, the extended activity of hot carriers in cerium, with its 3.2 eV bandgap, offers a significant advantage. Research in this area has further improved the photodegradation efficiency of phenolic pollutants (Valente, Tzompantzi, and Prince 2011).

4.2. Intercalated LDHs

4.2.1. Inorganic intercalation

LDHs contain a common nanostructure, rendering them highly valuable for environmental remediation (He et al. 2018; Khandaker, Fujibayashi, and Kuba 2025). However, their low selectivity and poor affinity for HMs and OPs stem from the absence of surface functional groups (Chaudhuri and Yun 2022; Ampong et al. 2023; Rojas 2014; Velusamy et al. 2021). Recent studies have focused on enhancing LDHs by introducing sulfonic acid, thiol, and sulfide groups. These molecules significantly improve their selectivity, particularly for soft metal ions, and strengthen their adsorption performance. For example, Rahman et al. (2018) demonstrated that high-retention adsorption can be achieved by intercalating nitrate Mg-Al LDHs without organic anion exchange (Rahman et al. 2018). Similarly, Poudel et al. (2021) synthesized LDH@ through a hydrothermal method, achieving effective removal of chromium (400.40 mg/g) and divalent lead ions (Pb(II), 426.76 mg/g) via mechanisms such as electron migration, isomeric replacement, and surface complexation (**Fig. S1**) (Poudel, Awasthi, and Kim 2021).

4.2.2. Organic intercalation

LDHs are widely selected for modification, particularly for grafting functional groups. Organic molecules, such as ethylenediaminetetraacetic acid (EDTA) (Chen et al. 2024; Shen et al. 2024; Lin et al. 2024; Jasem-Feisal, Amiripour, and Ghasemi 2024), amino acids (Koilaraj, Kalusulingam, and Sasaki 2019), thiol carboxylic acids (Jiang et al. 2023), and citric acid (Wang, Zhang, et al. 2023), have been employed for this purpose. For instance, Zhang et al. (2020) utilized MgAl-Cys-LDH for the adsorption of Cd(II), Cu(II), and Pb(II), leveraging the ability of the -SH group to immobilize metal ions into metal sulfide structures (Zhang et al. 2020). Similarly, Chen et al. (2014) synthesized ion-exchanged platinum/ZnTi-LDHs with enhanced surface sites and pore volume (Chen et al. 2014). Further studies are necessary to fully explore the potential of modified LDHs for the adsorption of HMs and organophosphates in environmental applications.

4.3. Composite materials of LDHs

4.3.1. LDHs polymer

The interactivity of LDHs and polymers involves creating nanocomposites with unique structural characteristics, such as gas barrier properties and photostability (Alexandre et al. 2002; Mallakpour and Tabebordbar 2020; Pathak and Singh 2024). LDH production in a mixed polymer solution enhances the interfacial exchange between LDHs and the host polymer. This is achieved by coprecipitating two basic metal salts into a suitable polymer solution (del Valle Ponce et al. 2022; Mallakpour and Tabebordbar 2020). For instance, polyvinylpyrrolidone (PVP) nanocomposites were produced by inserting LDHs into a mixed polymer solution through a polyester substance (Stimpfling et al. 2016). ZnAl-LDH nanocomposites were synthesized via a coprecipitation process in water at a constant pH of 9 (Gaume et al. 2013). Additionally, the proton conductivity of sulfonated polyether ether ketone (SPEEK) was improved by incorporating MgAl-LDH (Kim et al. 2015). Du et al. (2007) produced nylon-MgAl-LDH by melting nylon 6 with organically modified MgAl-LDH intermediate layers (Du, Qu, and Zhang 2007). Similarly, Suresh et al. (2017) employed an internal mixing mechanism to create polystyrene/NiAl-LDH nanocomposites, where the interaction of LDH nanoparticles with polystyrene enhanced material resistance and encouraged thermal degradation (Suresh et al. 2017).

4.3.2. LDHs carbon

Several studies have demonstrated that LDHs can effectively be applied to various carbon-based materials, including graphene (Cao, Li, and Li 2016; Nayak and Parida 2019; Wang et al. 2018; Zhang et al. 2017), graphite carbon (Abazari et al. 2019; Sahoo et al. 2020; Tonda and Jo 2018; Zou et al. 2017), carbon nanotubes (CNTs) (Bhuvaneswari et al. 2021; Fan et al. 2020; Jia et al. 2016; Zhang, Sun, et al. 2019), carbon quantum dots (Koilaraj, Kamura, and Sasaki 2017; Rahmanian, Dinari, and Abdolmaleki 2018; Wei et al. 2016), BC (Gholami et al. 2020; Vithanage et al. 2020), and carbon fibers (Fang et al. 2019; Peng et al. 2018; Wang and Guo 2019). These investigations highlight the exceptional features and high conductivity of these materials, which enhance their applicability (Bi et al. 2011; Pourfarzad et al. 2019; Zhu et al. 2015). The combination of LDHs with carbon-based materials has been proven to improve the adsorption and removal efficiency of HMs and OPs, as well as catalytic degradation processes. These materials prevent the aggregation of LDH laminar structures and reduce the recombination of photogenerated carriers (Liang et al. 2019; Daud et al. 2016). For instance, Liang et al. (2019) reported an increased capacity for photocatalytic ciprofloxacin degradation in wastewater under visible light by the integration of graphene oxide (rGO) with LDHs (Liang et al. 2019). Further, Wang et al. (2024) synthesized E-Mg/Al-LDHBC using ethylenediaminetetraacetic acid as the primary inserted substance and evaluated its adsorption capabilities for Cd and Pb (Wang et al. 2024). New prominent patterns associated with Cd and Pb were observed at the E-Mg/Al-LDHBC interface following the adsorption of Pb(II) and Cd(II) (**Fig. 4**). Additional examples of catalytic activity are summarized in **Fig. S2**.

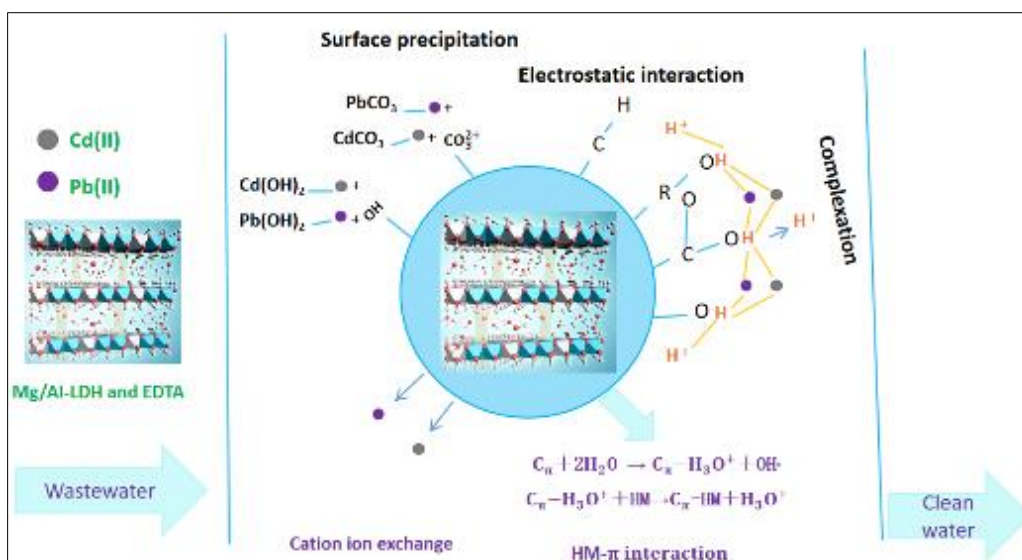


Figure 4 Diagram of pollutants removal mechanism (Wang et al. 2024). Copyright 2024 Wang et al

4.3.3. LDHs calcination

To enhance the efficiency of LDH preparation, recent studies have employed calcined LDH to produce layered double metal oxides (LDOs) (Xu et al. 2023; Shen et al. 2020). These investigations demonstrated that calcination significantly enhanced the surface sites and adsorption characteristics of metal oxides, thus improving the adsorption capacity of HMs. For instance, Yuan et al. (2013) utilized a nanostructured filter to create graphene/LDH for Cr removal from wastewater (Yuan et al. 2019). A calcination temperature of approximately 500°C was applied to produce graphene-LDO with high efficiency for Cr removal. Moreover, LDOs have been found to be promoters that enhance the catalytic degradation of OPs. With band gaps ranging between 2.0 eV and 3.0 eV, LDOs exhibit excellent photocatalytic activity (Zhang, Yan, et al. 2019; Benito et al. 2008). Additionally, catalytic nanocrystals are facilitated by effective electron transfer from high-valence metals (Chen, Zhang, et al. 2019; Chen, Zeng, et al. 2019). For example, Ju et al. (2018) developed a nano-adsorbent capable of degrading bisphenol A at low temperatures by employing calcined ZnAlTi-LDHs at approximately 500°C (Ju et al. 2018). To improve toluene adsorption, calcined MgAl-LDH was integrated into MgAl-LDO (Laipan et al. 2015). Furthermore, the memory effect of LDOs during rehydration allows them to revert to their original structures, enabling reuse in multiple cycles.

5. Utilization of LDHs for the removal of pollutants

The mechanisms underlying the elimination of HMs and OPs are depicted in **Figs. 5** and **6**, respectively, based on prior analyses. The formation of anion-metal complexes and the precipitation of hydroxides on the surface of LDHs occurs due to the presence of hydroxyl groups in these materials. Surface complexation and precipitation are identified as the major mechanisms by which LDHs adsorb HMs (González et al. 2014; Laipan et al. 2020). LDHs demonstrate exceptional ion exchange and adsorption properties, attributed to their exchangeable anions and positively charged layers (Li et al. 2016). Furthermore, chelator-synthesized LDHs have been employed as effective adsorbents for HMs in aqueous solvents, owing to the strong chelating interactions between ligands and HMs (Koilaraj, Kalusulingam, and Sasaki 2019; Chen et al. 2021). Functional groups such as aldehyde, carboxyl, and sulfhydryl can form complexes with HMs, thereby enhancing the contaminant removal capacity of LDHs. The interlayer anions can be reduced to less hazardous valence states or equivalent metal forms, while isomeric substitution has been proposed as a pathway for HM stabilization. The removal of OPs is commonly accomplished through three principal advanced oxidation processes (AOPs), including photocatalysis, Fenton-like reactions, and peroxydisulfate catalysis. The main mechanisms through which LDHs facilitate OP degradation through AOPs include electron transfer, single oxygen generation, and radical formation. Additionally, LDHs have been applied for removing common contaminants, such as malachite green, Cd(II), Cr(VI), and tetracycline, as noted in the bibliometric analysis. Recent studies further highlight the application of LDHs to address these contaminants.

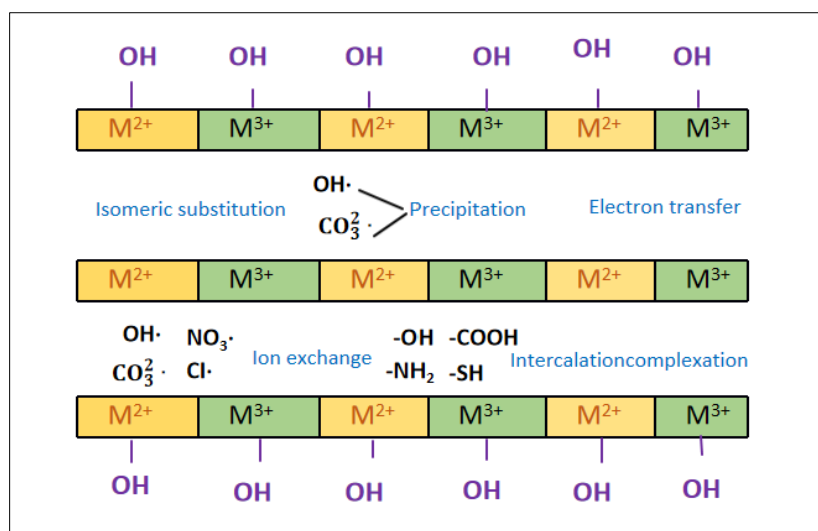


Figure 5 LDHs applications pathways for HMs removal

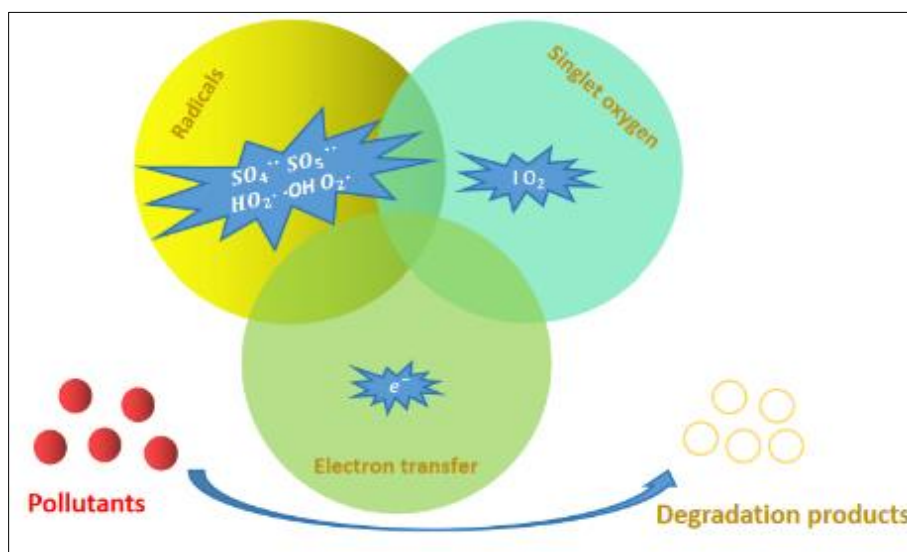


Figure 6 OPs elimination via LDHs

5.1. Chromium (VI)

Hexavalent chromium (Cr (VI)) is identified as one of the most hazardous HMs because of its intense toxicity and tendency to biological aggregation (Bao et al. 2020; Cong et al. 2022; Zhao et al. 2021; Zhou et al. 2022; Li et al. 2024).. Naturally occurring in the environment, Cr exists primarily in two forms: the highly toxic Cr (VI) and the less toxic trivalent Cr (III). Special attention has been devoted to Cr (VI) because of its potential risks. Anionic forms of Cr (VI), such as chromate and dichromate, further complicate its environmental impact. The removal of Cr (VI) usually involves ion exchange, coordination, and redox processes. For example, the MgAl-LDH composite, described by Ma et al. (2017), demonstrates impressive adsorption capabilities, with an efficiency of 130 mg/g and a 93% capture of Cr(VI) (Ma et al. 2017). This reduction of Cr (VI) to Cr (III), a milder Lewis acid, occurs through a heterogeneous redox process. A portion of Cr (III) remains in the interlayer after forming bonds with sulfides. Fourier transform infrared spectroscopy (FTIR) has confirmed the stability of LDHs during these redox processes, indicating that the host layer provides a stable setting for anion exchange. V. Rives (2001) revealed that uncalcined Cl-intercalated LDHs had a more significant positive impact on Cr(VI) removal than their calcined alternatives (Rives 2001). The principal mechanism involves the partial exchange of Cl^- with chromate. Advanced materials, such as a nanofiber film of magnesium aldehyde/polyacrylonitrile (MgAl-LDH/PAN), have also been developed, enabling in situ growth of LDHs coated with boehmite sol (Gore et al. 2016). This approach effectively reduced Cr (VI) to Cr (III), as confirmed by the valence state of adsorbed Cr ions. Additionally, NiCo-

LDH microspheres, synthesized via a single-stage hydrothermal method (Li et al. 2024), demonstrated exceptional Cr(VI) removal efficiency due to their high surface area, positive surface charge, and unique pore structure, achieving an adsorption capacity of 123 mg/g. Similarly, a NiFe-LDH/Polypyrrole pseudocapacitive electrode, created through in situ polymerization, exhibited a Cr(VI) removal rate of 111 mg/g from wastewater (Yang et al. 2024). These advancements underscore the potential of LDH-based materials for addressing Cr (VI) contamination (Fig. 7).

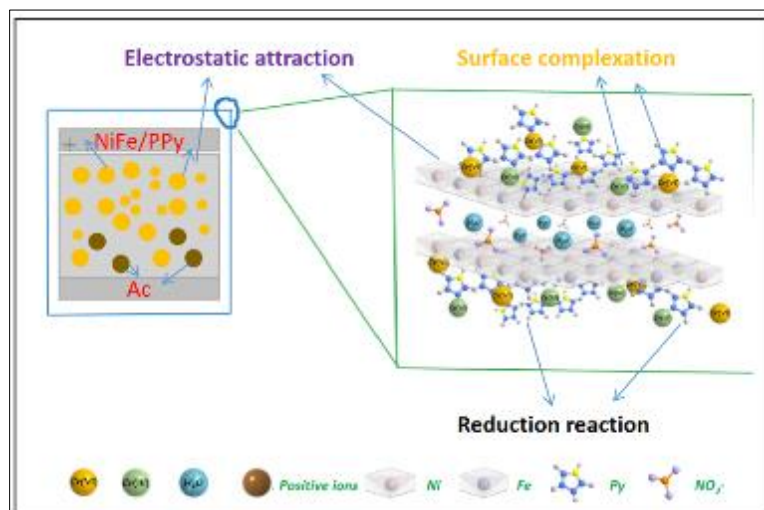


Figure 7 Cr(VI) electrosorption by NiFe/PPy (Yang et al. 2024). Copyright 2024 D Yang et al

5.2. Cadmium (II)

Cadmium ions are highly toxic and persistent HMs that pose significant threats to the environment and human health. LDHs can effectively remove these ions through mechanisms such as complexation, precipitation, interface adsorption, and isomorphic replacement (Asiabi et al. 2017; Zhang, Chen, et al. 2022; Zhao et al. 2023). The mineralization process involving LDHs is notably stable, as demonstrated by experiments (Fig. S3). LDHs are positively charged laminates composed of brucite-like layers stacked and bound by interlayered anions. These structures can be modified through isomorphic substitution, where divalent metal cations are replaced with trivalent ones (Fan et al. 2014). This substitution, combined with the abundance of hydroxide (-OH) groups, enables LDHs to interact with a wide range of inorganic and organic compounds (Bing et al. 2018). Strong coordination of these -OH groups facilitates the removal of contaminants. For instance, Liu et al. (2019) achieved a 99.7% removal rate using calcined MgZnFe-LDH (CMZF) as an adsorbent (Liu et al. 2019). Additionally, metal ions in LDH laminates can exchange with metal ions in other metal salts. Kong et al. (2021) demonstrated efficient removal of cadmium ions through the mineralization of CaAl-LDH, achieving a high adsorption capacity of 592 mg/g and a rapid adsorption rate of 5 minutes (Kong et al. 2021). The efficiency of CaAl-LDH immobilization on soil exceeded 96.9%. The isomorphic substitution of CaAl-LDH to CdAl-LDH during adsorption further enhanced the stability of the mineralized product. This consistency was corroborated by density functional theory simulations, confirming the resilience of the process.

5.3. Tetracycline

Tetracycline (TC), a highly chemically stable antibiotic, is released into the environment through urine and feces and cannot be fully absorbed by the body (Yuan et al. 2022; Zhang, Meng, et al. 2022; Zhang, Cheng, et al. 2022). AOPs, including peroxymonosulfate catalysis, Fenton-like effects, and photocatalysis, are employed for antibiotic degradation using LDH-based composites (Xie et al. 2021). Under light irradiation, LDH-based materials typically generate reactive species, initiating redox processes as photoelectrons transfer from the valence band (VB) to the conduction band (CB), resulting in the formation of free radicals such as $\cdot\text{O}_2^-$ and $\cdot\text{OH}$. These radicals efficiently oxidize and degrade antibiotics. LDHs play a critical role in activating peroxymonosulfate (PMS), producing substantial amounts of $\cdot\text{OH}$ during Fenton-like reactions, which rapidly inserted antibiotics into smaller molecules. Furthermore, LDHs facilitate PMS reactions, leading to associated reactive radicals. Research by Li et al. (2022) demonstrated that persulfate activation, combined with LDHs synthesized via Z-scheme heterojunction mechanisms, effectively degraded TC (Li et al. 2022).

A straightforward hydrothermal method was employed to synthesize $\text{MoO}_2/\text{CoFe-LDHs}$ Z-type multilayer junction, which was successfully activated by Co-Fe dual-site mechanisms, facilitating tetracycline degradation under visible light (Li, Meng, et al. 2023). The results indicated that the synergistic destruction effect of this junction on tetracycline was

3.7 times more effective than that of the multilayer junction alone. Additionally, it was 2.7 times higher than the value reported by Xu et al. (2022) (Xu et al. 2022). The latter applied a hydrothermal process to develop a pH-universal electric Fenton (EF) cathode, which exhibited excellent tetracycline degradation (94.2%) across a broad pH range. Additionally, Fig. S4 illustrates the production of $\text{In}_2\text{O}_3@\text{LDHs}$.

5.4. Malachite green

The plastic industry, printing, and textiles frequently use malachite green (MG), a dye made from triphenane molecules. Its effectiveness against fungal and protozoan diseases in aquatic species makes it a popular fungicide in aquaculture. However, due to its high toxicity and solubility, MG can harm the liver, kidneys, and lungs of various animals, even at low doses (Vareda 2023; Yuan et al. 2023). Common adsorption mechanisms include chemical bonding, physical adsorption, and hydroxide precipitation. Tang et al. (2023) synthesized magnetic ZnAl-LDH (MZA) for MG adsorption, utilizing molecular intercalation, hydrogen bonding, and electrostatic attraction as the main mechanisms (Tang et al. 2023). MZA maintained a high adsorption capacity over four adsorption-desorption cycles. Manea et al. (2022) developed flower-like Al-Li/ThLDH@CNT nanocomposites for MG photodegradation by combining exfoliated Al-Li/Th LDH nanosheets (Fig. 8), with F-CNTs via a hydrothermal method (Manea et al. 2022). The Al-Li/ThLDH@CNT exhibited a maximum adsorption capacity of 172.4 mg/g and achieved 98% MG decolorization in 45 minutes. Gao et al. (2021) created an innovative synthetic adsorbent (LDH@PDA@MPNs) for MG removal using metal complexation and mussel-inspired chemistry in a two-step process (Gao et al. 2021).

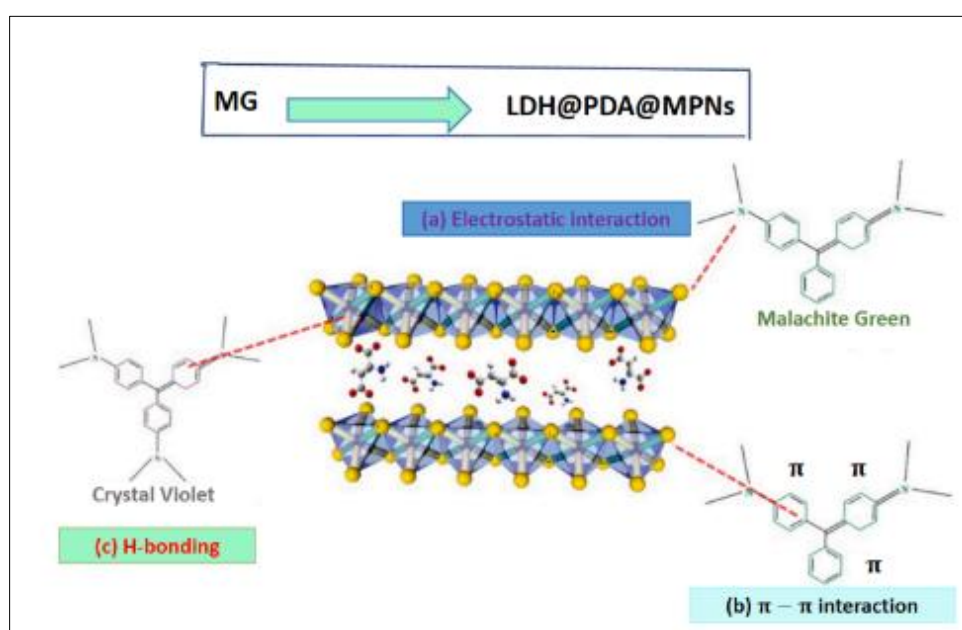


Figure 8 Interaction mechanisms between MG and LDH@PDA@MPNs (Manea et al. 2022)

6. Challenges and futures perspectives

The development of adsorbents for wastewater treatment, particularly for the removal of hazardous or toxic pollutants, remains a critical area of ongoing research. This study reviews recent advancements in the application of LDHs for the adsorption-based removal of various contaminants, including OPs and HMs. The findings indicate that LDHs are highly effective in adsorbing a wide range of organic contaminants.

However, despite significant progress in the application of LDHs for treating HMs and OPs, several challenges continue to hinder their broader implementation:

6.1. Assessment of potential environmental effects

Examining the potential hazardous effects of LDHs and their modified components on the environment is crucial for assessing their applicability in environmental remediation. However, data in this area remain minimal. For example, toxic metals such as Co/Ni frequently occur in LDHs with high catalytic activity, and their production may involve harmful chemicals. Consequently, efforts should prioritize identifying less toxic alternatives and minimizing the

leaching of hazardous compounds. Additionally, studying LDH movement and transformation into natural environments is essential to indirectly assess potential adverse environmental impacts.

6.2. Enhancement of synthesis methods

The molar concentration of metal ions significantly influences the effectiveness of LDH-based components. To enhance pollutant removal through LDHs, researchers should explore modified components incorporating different trivalent cations and divalent ions in various proportions. Additionally, to improve the economic feasibility of LDH composites, more attention should be paid to optimizing preparation techniques. Reaction conditions and preparation methods also play a crucial role in determining the properties of LDHs, with some methods being overly complex, resource-intensive, and unsuitable for large-scale synthesis. Consequently, future research should prioritize the development of innovative, cost-effective, efficient, and environmentally friendly synthesis techniques.

6.3. Emerging pollutants

Emerging pollutants, due to their diverse chemical structures and properties, can be classified into several groups. Each group exhibits distinct characteristics that may influence the adsorption capacity of LDHs. Previous studies introduced LDH-based materials for the removal of emerging contaminants, demonstrating their potential. However, existing research remains insufficient to fully elucidate the underlying mechanisms. Consequently, further research is needed to explore LDH materials capable of selectively adsorbing specific emerging contaminants, alongside efforts to clarify the associated mechanisms.

6.4. Long-term stability analysis

Certain LDHs may release structural components, functional categories, or specific ions into the environment under specific conditions during pollutant removal activities, potentially leading to performance degradation, secondary pollution, and reduced reusability. To ensure the reliability and longevity of LDH-based components in practical applications, researchers must carefully evaluate the materials' structural and chemical stability, as well as their performance under varying conditions. It is possible to achieve long-term stability through isomorphic substitution; however, it is effective for only a limited range of HMs. Moreover, the lack of clear explanations regarding isomorphic substitution in several studies highlights the need for further research to elucidate its precise mechanism.

6.5. Comprehensive mechanistic research

The exact reduction process remains uncertain despite efforts to investigate LDHs' pollution removal mechanisms. This includes studies suggesting that LDHs and their components can convert Cr (VI) into Cr (III). Furthermore, it is crucial to determine whether a specific structure or functional group is essential for Cr (VI) elimination and how LDH compounds influence this process. Consequently, more comprehensive and detailed characterization techniques are necessary. LDH-based catalysts in sulfate radical-AOPs facilitate organic molecules degradation via both reactive and nonradical pathways. Nonradical-dominant systems exhibit superior tolerance to interfering contaminants, whereas the radical pathway often demonstrates better oxidation potential for OPs. While recent studies have focused on identifying reaction pathways, managing the present mechanisms remains a challenge. Therefore, future research should prioritize controlling reaction pathways alongside identifying active chemicals to achieve optimal pollutant degradation efficiency.

6.6. Investigating for soil remediation applications

Several studies have been conducted on the use of HMs and OPs for soil remediation. However, LDH composites exhibit significant removal capacity for both contaminants in sewage treatment. Investigating the potential applications of LDH and its components in soil remediation is crucial, particularly soil amendments that supply elements such as nitrogen and phosphorus. Additionally, researchers aim to recycle LDH composites after adsorption to enhance resource utilization efficiency.

7. Conclusion

This study conducted a comprehensive bibliometric analysis of research advancements, focal points, and trends in the use of LDHs for the removal of HMs and OPs. In recent years, LDH research has expanded rapidly, with significant growth observed in recent academic studies across various countries. This trend highlights the growing relevance of LDHs in diverse scientific applications and the collaborative efforts of researchers worldwide. Keyword analysis, including citation bursts, clustering, and co-occurrence, revealed that initial research primarily focused on using LDH nanoparticles for water purification. Recent studies have identified MG, Cd (II), Cr (VI), and TC as the most prevalent

pollutants, which are expected to remain key areas of focus. To enhance pollutant degradation in AOPs, researchers have increasingly applied LDHs as alternatives to persulfates and photocatalysts. Additionally, the development of LDH-biochar composites has emerged as a prominent topic. These composites, known for their exceptional catalytic degradation capabilities, high anion exchange capacity, superior electrical properties, strong chemical resistance, and versatile interfacial functionalities, are highly effective for pollutant removal.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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