Review

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From Structure to Function: Structure-Property Correlation of Layered Double Hydroxide for Persulfate Activation

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Abstract: The widespread contamination of water by persistent organic pollutants poses a serious threat to both the environment and public health. Sulfate radical-based advanced oxidation processes (SR-AOPs) have attracted considerable interest due to their strong oxidative power and operational adaptability. Central to these technologies is the catalytic activation of persulfate, which drives the development of efficient and stable catalysts. Among various materials, layered double hydroxides (LDHs) stand out as highly promising candidates, owing to their tunable layered structure, high surface area, and compositional flexibility. This review systematically summarizes the use of LDH-based catalysts for persulfate activation in degrading diverse organic contaminants. It further clarifies the fundamental structure–activity relationships that determine catalytic performance, covering key strategies such as electronic structure modulation via doping and defect engineering, interlayer confinement effects, and synergistic interactions among multiple metallic elements. Future research should prioritize the rational design of novel LDH composites with improved stability and selectivity, a deeper mechanistic understanding under environmentally relevant conditions, and the expansion of practical applications toward integrated treatment systems that combine effective pollutant removal with resource recovery. Progress in these areas will be vital for advancing sustainable and efficient SR-AOPs.

Key words: Layered double hydroxides; Persulfate; SR-AOPs; Structure-activity relationship

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1. Introduction

Technological progress, while driving societal development, has unfortunately led to the contamination of vital water resources [1]. Given the significant risks these pollutants pose to human health [2], the development of advanced wastewater treatment technologies is essential to safeguard clean water and sanitation for current and future generations [3]. Traditional advanced oxidation processes (AOPs), despite being highly effective in water treatment, face constraints in widespread application due to challenges such as limited activity and pH dependency.

The persulfate-based advanced oxidation process (SR-AOPs) is widely recognized as a highly efficient method for water treatment. It partially mitigates the reliance on acidic pH conditions, yet still faces challenges of reactive oxygen species (ROS) generation enhancement by catalysts and low oxidant utilization efficiency [4]. Current methods for activating persulfate, including peroxymonosulfate (PMS) and peroxydisulfate (PDS), in wastewater treatment mainly fall into two categories: (I) Homogeneous catalysis, which relies on dissolved metal ions to react directly with persulfate, offering rapid reaction rates and low cost, but often leaving residual metal ions that cause secondary pollution. It has been reported that combining this approach with separation techniques such as nanofiltration could help address this issue [5]. (II) Heterogeneous catalysis, which employs solid catalysts like metal oxides [6], metal hydroxides [7], carbon-based materials [8], single atom-doped materials [9] dispersed in wastewater. This method supports catalyst recovery and reuse, reduces metal leaching, and is more environmentally friendly, making it the current mainstream research direction. Based on a comprehensive review of the literature, the degradation mechanisms involved can be broadly categorized into radical-based and non-radical pathways. However, divergent findings across studies have sparked considerable debate regarding the occurrence and underlying mechanisms of non-radical reactions[10]. This area therefore warrants further in-depth investigation.

Currently, the widely studied persulfate activation methods in the research community encompass a variety of approaches, including dielectric barrier discharge (DBD) plasma [11-13], activated carbon (HGAC) [14], Fe_3O_4 nanoparticles [15], natural titanomagnetite [16], and layered double hydroxides (LDHs) [17], among others. Current research trends

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are mainly oriented toward tuning the band gap, retarding charge recombination, increasing specific surface area, and enhancing the photoelectrochemical effect.

LDHs have captured the attention of researchers due to their unique physicochemical properties[18]. They are increasingly recognized as promising candidates for low-dimensional photocatalytic applications[19], offering advantages such as straightforward synthesis, low cost, and excellent compositional tunability [20]. They are composed of brucite-like layers consisting of positively charged polymetallic cations and interlayer compensating anions[21]. This two-dimensional structure features have high versatility, allowing tunable chemical compositions [22]. Metal ions can be incorporated to modify the two-dimensional structure without damaging the main framework. In addition, LDHs can also be exfoliated into a monolayer structure, and the novel nanosheets with two-dimensional anisotropy exhibit significantly different properties from their stacked counterparts [23]. Through rational design strategies, such as modulating transition metal ions, intercalating metals or metal oxides, and incorporating polymer materials, LDH-based catalysts can be tailored to achieve enhanced magnetic properties, efficient charge separation, and improved catalytic activity in AOPs. They have been successfully applied as visible-light photocatalysts, Fenton-like catalysts, persulfate (PS) activators, and sonocatalysts. Nevertheless, despite the demonstrated high performance in various catalytic applications, certain fundamental and practical aspects remain to be further explored. As illustrated in **Fig. 1a**, a close relationship between LDHs and persulfate activation has been increasingly recognized, with growing research interest in LDH-based persulfate systems (**Fig. 1b**). However, LDHs produced by this method tend to agglomerate and have a limited specific surface area. Therefore, targeted structural design of catalysts is essential to address current limitations in the utilization efficiency of their active sites [22]. As shown in **Fig. 1c**, the structure-activity relationship and application are complementary to each other. Further in-depth research is still required for the currently exfoliated and modified LDHs. This review focuses on modifying LDHs through functional design to develop high-performance catalysts suitable for diverse application scenarios.

Considering the varied operational environments of SR-AOPs, adjustments and optimizations are approached from multiple dimensions to extend their applicability and across a broader pH range. This review mainly provides a detailed analysis of the catalysis of layered double hydroxides. Furthermore, the mechanisms of persulfate activation by LDHs with different structures also has been systematically reviewed, providing valuable insights for the design of future material systems. Finally, a reasoned perspective on the future development of catalysts is presented.

2. LDHs Engineering: Structural Determinants

The contemporary design of LDH-based catalysts has evolved into a systematic engineering discipline. It integrates multi-level control, ranging from atomic-scale arrangements to macroscopic shapes, and from intrinsic activity to surface/interface properties. Its main goal is to achieve precise control over electronic structures, active-site accessibility, and interfacial charge dynamics through synergistic strategies. In this way, the design meets the demands of various applications such as advanced oxidation, photocatalysis, and electrocatalysis.

Regarding electron transfer enhancement, heteroatom doping and single-atom anchoring are effective methods for precise modulation of active centers. For instance, Wang et al. introduced Fe^{2+} to partially replace Ni^{2+} in NiFe-LDH, forming localized Fe–O–Fe configurations (Fig. 2a). This atomic-level substitution directly changes the electron density and coordination environment around the metal centers, significantly boosting PDS activation [24]. Similarly, Li et al. anchored high-valence Ir single atoms onto the surface of NiFe-LDH (Fig. 2b). The strong metal–support interactions there optimize interfacial charge

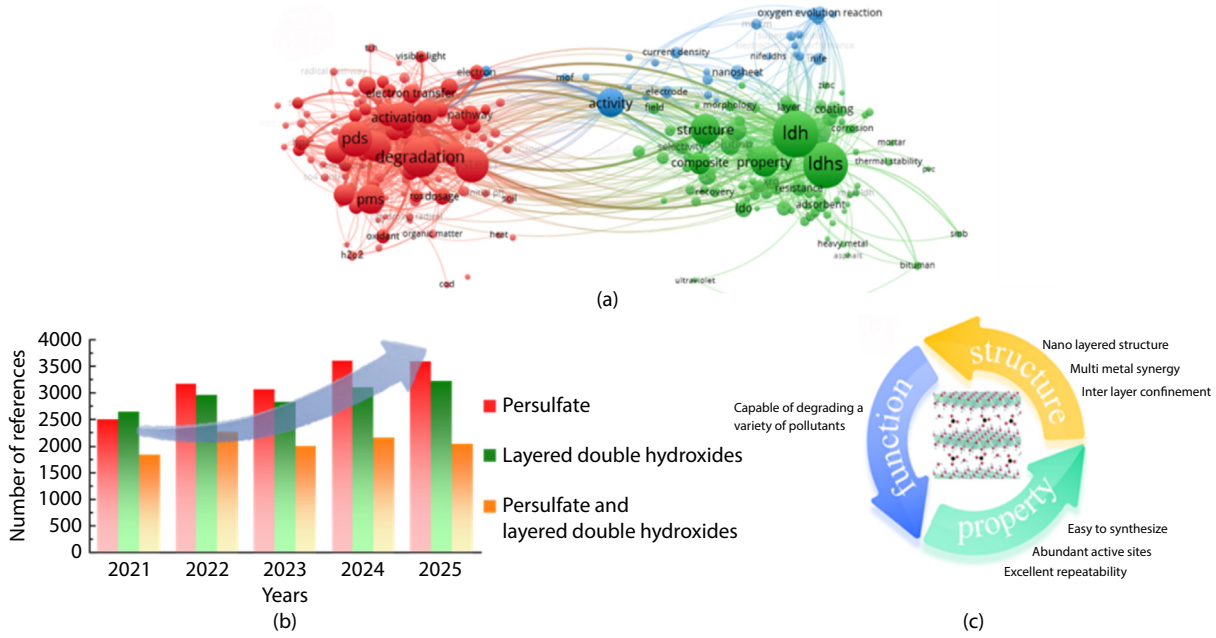

Fig. 1. (a) Data visualization of relevant research on layered double hydroxides catalyzed persulfate in the past five years. (b) Comparison of relevant references in the past five years. Source: Topics "Persulfate", "Layered double hydroxides" and cross-indexing them, respectively in indexed journals from 2021 to 2025; search time: January 2026. (c) The relationship between structure, property, and function.

transfer and markedly improve electrocatalytic water oxidation [25]. In related research, Khan et al. Looked at various metal-doped carbon nitride materials, focusing on recent advancements in doped g-C_3N_4 and its applications in water splitting, photocatalytic pollutant breakdown, bacterial disinfection, and CO_2 reduction [26]. These findings clearly demonstrate that atomic-scale engineering is crucial for controlling rate-determining steps and optimizing the adsorption energetics of reactive intermediates.

Catalyst defect engineering, particularly when applied to LDH structures, significantly enhances persulfate activation for AOPs. By introducing atomic vacancies and lattice distortions, defect engineering allows precise regulation of the material's electronic performance and surface properties. These defect sites can serve as electron-rich centers, promoting the decomposition of persulfate to generate radical and non-radical reactive species. Moreover, defects can optimize the band structure and electron transport, lower the reaction energy barrier, and improve the adsorption and degradation efficiency of target pollutants. Flower-like nickel-iron layered double hydroxide (NiFe-LDH) rich in oxygen vacancies (**Fig. 2c**) exhibits excellent performance in PMS activation [27]. Yang et al. constructed a heterojunction by combining nitrogen-defective graphitic carbon nitride (g-C_3N_4) with LDHs (**Fig. 2d**), forming high-density active sites and multiple vacancies, further enhancing the generation and utilization efficiency of reactive oxygen species [28]. LDH/g-C_3N_4 composites offer an efficient, solar-driven advanced oxidation process for pollutant degradation, leveraging their strong visible-light absorption and generation of highly reactive radical species[29]. Mutahir et al. prepared a highly efficient oxygen-doped graphitic carbon nitride (O-CN) and layered double hydroxide (LDH) nanocomposite photocatalyst and utilized it for the degradation of organic dyes in wastewater [30]. In addition to improving catalytic activity, defect engineering also helps enhance the stability and reusability of catalysts, providing a reliable strategy for the treatment of refractory organic pollutants in complex aqueous environments.

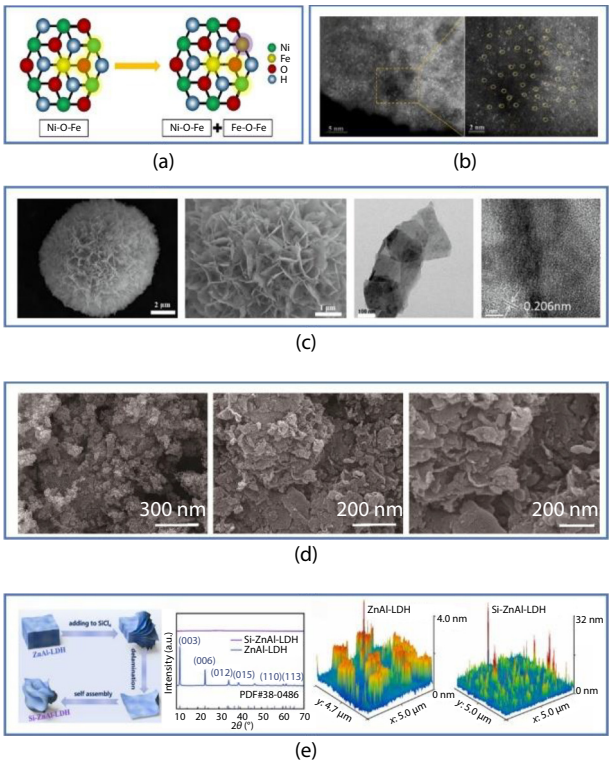

Fig. 2. (a) Atomic models of NiFe-LDH and Fe-substituted NiFe-LDH at Ni sites [24]. Copyright 2023, Elsevier. (b) AC-STEM image of Ir/NiFe LDH [25]. Copyright 2025, Elsevier. (c) SEM and TEM image of synthesized Ni-Fe-LDH [27]. Copyright 2023, Elsevier. (d) Two-dimensional heterostructure of V n-CN/Co and LDH [28]. Copyright 2023, Elsevier. (e) Schematic diagram of the $SiCl_4$ exfoliation process for ZnAl-LDH nanosheets, along with XRD patterns of nAl-LDH, Si-ZnAl-LDH nanosheets and the standard card, as well as AFM results of ZnAl-LDH and Si-ZnAl-LDH nanosheets [31]. Copyright 2024, Elsevier.

Combining layer exfoliation and chemical doping offers a synergistic strategy that enhances catalyst performance on two fronts: physical structure and electronic structural. When bulk LDHs are exfoliated down to monolayer or few-layer nanosheets, the specific surface area increases sharply and unsaturated coordination sites become exposed. Simultaneously, introducing heteroatoms during this process creates additional defects and modifies the local electronic states of the active centers. For instance, Bao et al. introduced silicon into ultrathin ZnAl-LDH nanosheets (**Fig. 2e**). This incorporation not only promoted the formation of oxygen vacancies but also effectively suppressed electron–hole recombination. This leads to a clear improvement in photoelectrocatalytic water splitting [31]. In a related study, Chi et al. synthesized monolayer NiFe-LDH nanosheets without using organic solvents. These nanosheets contained abundant oxygen and metal defects, which resulted in enhanced oxygen evolution reaction (OER) activity [32]. These studies illustrate the combined benefits of structural nano-engineering and chemical modification in improving mass transfer, charge separation, and intrinsic catalytic activity.

Although considerable progress has been made in engineering LDH-based catalysts, there are still several critical limitations. Precise control over atomic-scale modifications, such as heteroatom doping, single-atom anchoring, or defect engineering often demands complex synthesis conditions and suffers from poor reproducibility. These issues hinder large-scale practical application. Another challenge lies in the incomplete understanding of how multiple modulation factors (e.g., doping, vacancies, and morphology) interact with one another. As a result, isolating their respective contributions to the overall catalytic performance proves difficult. This complexity frequently leads to catalyst design that is empirical rather than predictive. Furthermore, most performance evaluations are carries out under idealized laboratory conditions. Such conditions may not accurately reflect catalyst behavior in realistic, dynamic environments including variable pH, competing ions, or long-term stability requirements. Future design strategies should therefore focus on more than just refining the modulation approaches. Equal priority needs to be given to developing operando characterization techniques and theoretical models that can elucidate synergistic effects. Combining high-throughput computations with well-controlled experimental synthesis may pave the way for more rational design frameworks. Such frameworks would enable targeted optimization of LDH-based catalysts for specific applications, while also bridging the gap between fundamental research and industrial viability.

3. LDHs applications in SR-AOPs Contaminant Degradation

Owing to their distinctive layered structure, LDHs are widely recognized as highly promising catalysts for activating persulfate in the degradation of organic pollutants. Their appeal is attributed to their straightforward synthesis, high catalytic activity, and broad applicability to various contaminants, including pharmaceuticals, dyes, and pesticides (**Fig. 3**) [33]. Guided by insights into the structure–activity relationships of LDHs, researchers have employed diverse design strategies to develop and optimize LDH-based composite materials for the removal of organic pollutants from aqueous environments via persulfate activation.

Based on the comprehensive research conducted above, **Table 1** summarizes metal leaching levels reported for various LDH-based catalytic systems, with values typically ranging from <0.1 to 5 mg/L depending on catalyst composition, reaction conditions, and operational duration. These findings underscore the importance of assessing metal stability, as leaching not only reduces catalytic activity but also poses potential environmental risks. Strategies to minimize leaching include optimizing metal ratios to form more stable phases, introducing protective coatings such as carbon layers or polymers, and operating under mild pH conditions to suppress dissolution. Despite these mitigation efforts, most studies have been conducted under controlled laboratory settings, leaving a critical gap in understanding catalyst behavior in complex, real-world wastewater matrices.

In the area of simple compositing and structural modification, Ma et al. synthesized a Mg-Fe-LDH@biochar composite capable of effectively activating PMS for doxycycline degradation in aqueous solutions [39]. Mutahir et al. used cinnamic acid-modified mulberry wood waste biochar (CA-MWB) to remove methylene blue dye (MB) from aqueous solutions[40]. confirming the efficacy of biochar as a support for enhancing catalytic activity. Similarly, Li et al. dispersed Mg/Al-LDH onto activated carbon and further decorated it with Co_3O_4 nanoparticles, constructing a composite catalyst with confined spatial architecture that significantly improved the degradation efficiency of levofloxacin [41]. This approach highlights the benefits of hierarchical structuring in optimizing active site distribution and utilization.

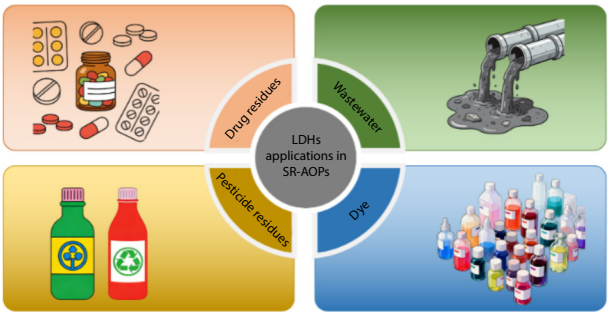

Fig. 3. LDHs applications in SR-AOPs.

Beyond composite construction, defect engineering offers another effective route to enhance LDHs performance. For example, Xu et al. synthesized NiCo-LDH enriched with single-oxygen vacancies, which exhibited excellent PMS activation and efficient ciprofloxacin degradation over a wide pH range, highlighting the role of defects in improving both activity and environmental adaptability [42]. In a related approach, Bian et al. fabricated both copper-containing and copper-free CoAl-LDH nanosheets via urea co-precipitation for PMS-activated degradation of rhodamine B, showing that the controlled introduction of metal components provides a direct method for tuning catalytic behavior [43]. Mutahir et al. synthesized a highly efficient ZnO/g-C_3N_4 nanocomposite via a facile one-step calcination of zinc glutamate MOF and melamine, and utilized it for the photocatalytic degradation of the organic pollutant rhodamine B [44]. The introduction of varying metal components offers a straightforward strategy for tuning the catalytic performance of LDH-based systems.

In the pursuit of practical application and functional integration, research has expanded the forms and applications of LDH-based materials. Liang et al. prepared a polyacrylonitrile/cellulose nanofiber@CoFe-LDH nanofiber membrane through electrospinning and in situ growth. This membrane combines good mechanical strength, superhydrophilicity, and self-cleaning catalytic capability, showing promising potential in integrated water purification systems [45]. The MIL-101(Fe)/NiFe-LDH composite membrane developed by Shehriyar et al. via interfacial polymerization exhibits exceptional performance, achieving 91% methyl orange removal and 88% methyl green removal at a high water flux (>100 $L{\cdot}m^{-2}{\cdot}h^{-1}{\cdot}bar^{-1}$). Such work advances LDH-based catalysts from powdered forms toward practical, multifunctional membrane materials that combine separation with catalysis. Qian et al. synthesized a self-supporting and recyclable catalyst by growing CoFe-LDH on nickel foam via a one-step solvothermal method, which effectively degraded monomethylhydrazine in propellant wastewater and offered a solution for catalyst recovery [46]. Furthermore, Guo et al. used NiFe-LDH to activate PMS for the degradation of organophosphorus pesticides while simultaneously enabling phosphate recovery, demonstrating an integrated treatment strategy that transitions from pollutant removal to resource reclamation [47]. Collectively, these studies illustrate the significant potential of LDH-based systems in synergistic environmental remediation and resource recovery.

To evaluate the potential of LDHs for practical applications, several studies have demonstrated the performance of LDH-based catalysts in continuous flow reactors. For instance,

Table 1. Application List of Selected Layered Double

Materials	Oxidant	Pollutants	Removal rate	Cycle number	Ref.
S/CoFeAl-LDH	PMS	Norfloxacin	>95% (10 min)	4 cycles (84.3% retention)	[34]
CuFe-LDHs	PS	Ofloxacin	>90% (60 min)	5 cycles (66.8% retention)	[35]
NiFe-LDHs	PMS	Doxycycline	>90% (60 min)	3 cycles (82% retention)	[36]
CuFe-LDH/NF	PMS	Sulfamethoxazole	98.07%	10 cycles (90.82% retention)	[37]
BC@FeMg-LDH	PDS	2,4-dichlorophenol	>95% (60 min)	5 cycles (88% retention)	[38]

Pei et al. developed a non-contact electron transfer process (NCETP) for antibiotic degradation in an LDH/PMS system, which effectively avoids side reactions and maintains high selectivity, achieving stable levofloxacin (LVX) removal during 24-hour continuous operation [47]. In another approach, Seyed Ali Hosseini et al. synthesized a novel LDH catalyst using metal–organic frameworks (MILs) as precursors, enabling synergistic adsorption and photocatalytic degradation of meropenem (MER) in pharmaceutical wastewater [48]. Additionally, R.S. Nascimento et al. first proposed the use of layered double oxides (LDOs) derived from mining waste for the rapid and efficient removal of 4R dye from aqueous solutions [49]. These cases collectively illustrate the broad applicability of LDH-based materials across diverse fields, highlighting their significant potential and the value of continued research.

Beyond catalytic performance, using LDH-based materials in water treatment is beneficial to the environment, fully in line with green and sustainable remediation principles. The high mineralization efficiency of these catalysts often turn organic pollutants completely into harmless by-products like CO_2 and H_2O [50]. As a result, the ecological toxicity of treated wastewater drops significantly. Furthermore, LDHs are inherently stable and reusable. That helps minimize secondary pollution from metal leaching, solving a key problem in practical applications. Recent advances have further shown that LDHs play a useful role in resource recovery. For example, they can reclaim phosphate from wastewater or capture valuable metals from waste streams. In this way, a former disposal problem becomes an opportunity for a circular economy. LDH-based catalytic systems combine high degradation performance, low environmental footprint, and resource recovery capability. These features collectively enable such systems to contribute to the United Nations Sustainable Development Goals (SDGs), particularly those related to clean water and responsible consumption.

4. LDHs Structure-Activity Relationship in Catalysis

The functional properties of LDHs stem from their unique structural characteristics. As illustrated in **Fig. 4**, the combination of polymetallic valence interconversion, layered architecture, and structural tunability enables their broad application in catalysis.

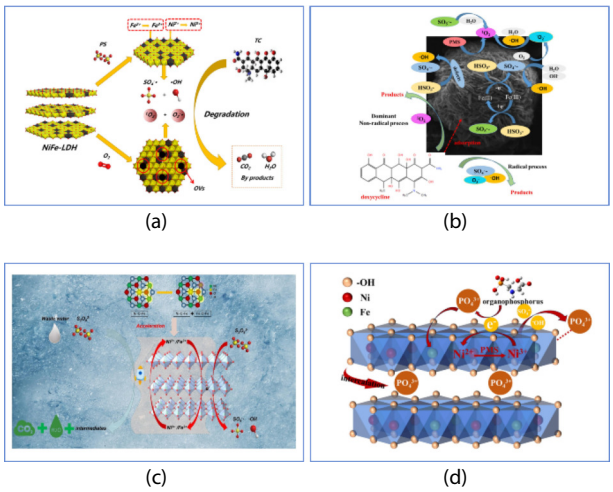

Fig. 4. (a)Possible reaction mechanism for the degradation of TC by defective NiFe-LDH[51]. Copyright 2022, Elsevier. (b) The possible mechanism for the DOX degradation in the Mg-Fe-LDH@biochar/PMS system[39]. Copyright 2021, Elsevier. (c) Possible reaction mechanism for the degradation of Orange II by Fe^{2+}-NiFe-LDH/PDS system[24]. Copyright 2022, Elsevier. (d) Mechanism diagram of NiFe-LDH/PMS degradation of glyphosate and simultaneous recovery of phosphate[47]. Copyright 2025, Elsevier.

4.1. Nano layered structure

LDHs are generally represented by the formula $[M^{2+}_{1-x}M^{3+}_{x}(OH)_2]^{x+}(A^{n-})_{x/n}{\cdot}mH_2O$, where a portion of the divalent metal cation sites in the brucite-like octahedral layers is isomorphously substituted by trivalent cations, resulting in positively charged sheets. The parameter x, typically between 0.20 and 0.33, denotes the molar ratio of M^{3+} to total metal ions, while A^{n-} represents charge-balancing anions located in the interlayer region [52]. The general structural arrangement is illustrated in **Fig. 5**.

Nanolayered structure exhibits high structural tunability, a large specific surface area, and abundant active sites. For example, Li et al. synthesized NiFe-LDH modified biochar (NiFe-LDH/BC) and applied it to tetracycline degradation via PMS activation. The enhanced catalytic performance was attributed to

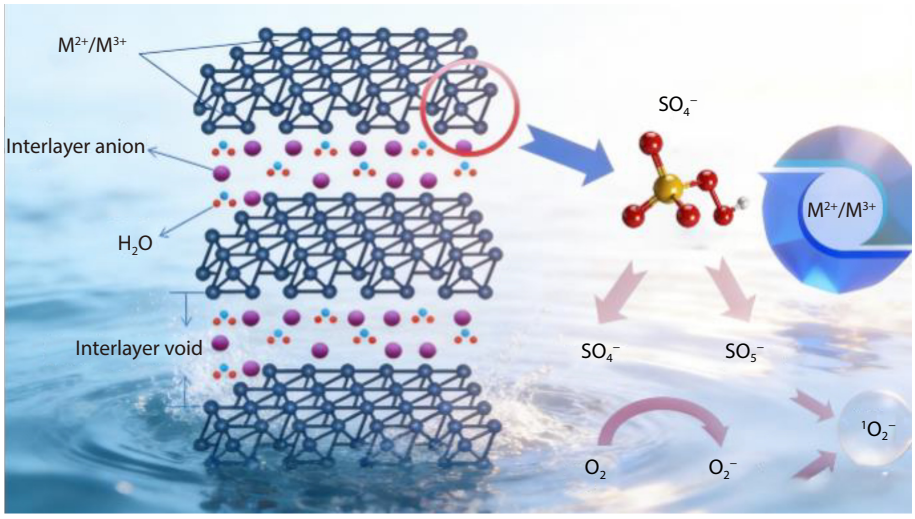

Fig. 5. Mechanism of LDH-Catalyzed PMS.

the increased oxygen vacancies introduced by biochar, which facilitated the redox cycling of Ni(III)/Ni(II) and Fe(III)/Fe(II), altered the singlet oxygen generation pathway, and improved catalyst stability [53]. Mutahir et al. prepared boron-doped g-C_3N_4 (BCN), MXene, and CuO via soft chemistry techniques to fabricate a novel dual Z-scheme CuO/MXene/BCN heterojunction for the photocatalytic degradation of malachite green pollutant. This dual Z-scheme mechanism promotes improved charge separation, minimized electron-hole recombination, and enhanced light absorption and redox capacity [54]. Similarly, Wu et al. applied this principle to prepare CoNiFe-LDH materials for efficient photodegradation of microplastics (MPs). The improved removal efficiency of MPs using optimized CoNiFe(VZn-Al)-LDHs can be attributed to several factors: (Ⅰ) extended solar absorption and effective electron-hole separation due to reduced d-orbital occupation on metal sites; (Ⅱ) high-valence metal sites acting as electron-trapping centers that promote electron transfer during PMS activation; and (Ⅲ) a reduced energy barrier for O–O bond cleavage in PMS [55]. In summary, the nanolayered architecture provides a highly adaptable platform for catalytic design and mechanistic study.

4.2. Multi metal synergy

The synergistic interaction among multiple metals in LDHs enhances persulfate activation through coordinated mechanisms. Variations in metal electronegativity generate distinct local charge densities at active sites, which promote the adsorption and polarization of the O–O bond in PMS [56]. Additionally, the flexible and interconvertible valence states of different metal ions facilitate multi-path electron transfer and sustain continuous redox cycles, thereby driving PMS activation [57]. Furthermore, multi-metal configurations effectively mitigate the pH sensitivity typically associated with single-metal catalysts, broadening the operational pH range while reducing undesirable metal ion leaching.

For instance, Ma et al. synthesized transition-metal-doped Li/Al-LDHs via a one-pot method. Transition metal doping enhances the stability of Al–O octahedra, reduces the lattice constant, suppresses Al dissolution, and strengthens lithium adsorption capacity [58]. Similarly, Shuai's group developed a $BiVO_4$/Ag/CoMnZn-LDH composite photoanode. Here, Ag nanoparticles improve light absorption via localized surface plasmon resonance and promote charge separation through hot-electron injection and hole transport, while the CoMnZn-LDH overlayer forms a p–n heterojunction to enhance charge separation and provides multivalent redox sites to accelerate oxygen evolution kinetics [59]. Overall, polymetallic synergy significantly enhances catalytic performance. Future studies should further emphasize the development of green, cost-effective catalyst systems.

4.3. Inter layer confinement

The confined interlayer spaces within LDHs significantly enhance PMS adsorption and interfacial electron transfer. Specifically, this nanoconfinement environment can restructure the hydrogen-bond network of water molecules, thereby influencing proton transfer pathways [60]. Moreover, such confined spaces help suppress PMS self-decomposition and mitigate radical quenching, extending the catalyst's operational lifetime [61]. The layered architecture also "anchors" metal active sites within the crystalline framework, serving as an efficient nanoreactor that reduces metal leaching [62]. Additionally, the layered sheets can "shield" the transient complex formed between PMS and metal sites, lowering the activation energy for PMS decomposition and improving the cyclic stability of the catalyst.

Consistent with this understanding, the rational modulation of the interlayer chemical environment and interfacial electronic structure has emerged as a fundamental strategy for optimizing the catalytic properties of layered double hydroxides. Hu et al. synthesized NiFe-LDH materials intercalated with distinct molecular species, namely tetrakis(4-carboxyphenyl)porphyrin (TCPP), 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (DOTA), and carbonate —yielding hybrids denoted as NiFe-TCPP, NiFe-DOTA, and NiFe-CO_3, respectively [63]. These materials were subsequently evaluated as electrocatalysts for water oxidation under industrially relevant alkaline conditions. Among the three, NiFe-TCPP demonstrated superior catalytic performance, achieving the lowest overpotential and exhibiting the highest reproducibility across repeated electrochemical cycling. Mechanistic investigations integrating both theoretical calculations and experimental characterization revealed that the enhanced performance originates from the strong coordination interactions between the conjugated carboxylic ligand TCPP and the iron species situated within the LDH layers. This specific coordination environment effectively suppresses iron dissolution by substantially increasing the energy barrier for metal leaching while simultaneously promoting the material's intrinsic self-healing capacity. Collectively, these synergistic effects contribute to markedly improved catalytic stability and sustained electrocatalytic activity under demanding operational conditions. Furthermore, the charge redistribution induced by this strong coordination optimized the d-band center to −2.81 eV and reduced the reaction energy barrier to 1.47 eV, ultimately enhancing catalytic activity[63]. In another study, Hu et al. elucidated the mechanism by which single-atom sites (SAs) on nitrogen-doped graphitic carbon-encapsulated nanoparticles enhance PMS activation. They found that constructing a nanoparticle–single-atom catalyst (NP-SAC) by leveraging the electronic modulation effect of nanoparticles on SAs resulted in highly efficient catalytic performance. The Co NPs-SAs@N-C/PMS system showed outstanding activity in degrading various organic pollutants, along with excellent stability and minimal Co^{2+} leaching over 20 cycles. The system also exhibited broad pH adaptability, strong salt tolerance, and effectiveness in long-term real wastewater treatment, demonstrating its practical potential [64]. In summary, the activation of persulfate by LDHs involves a multifaceted and intricate mechanism. The structural design of LDHs is intrinsically linked to their activation pathways, and due to their inherent structural complexity, these mechanisms are seldom singular, often requiring comprehensive multidimensional analysis. **Table 2** examines the catalytic mechanisms of LDH-based materials from three interrelated perspectives: nanostructured layered configuration, polymetallic synergistic effects, polymetallic synergistic effects, and interlayer confinement effects, with illustrative examples demonstrating how distinct structural features lead to varied catalytic outcomes.

Table 2. Structure-Activity Relationship of Layered Double Hydroxides

Materials	Structure	Characteristics	Performance	Ref.
Graphitic carbon nitride (NRCN-2)	Nano layered structure	Tunable structure, High active sites, Large specific surface area	Methyl orange: k =2.968 × 10 $^{-2}$ min^{-1} Trans-resveratrol: k =2.490 × 10 $^{-2}$ min^{-1}	[65]
NiFe-LDH			Tetracycline: ~80% removal (30 mg·L^{-1})	[51]
CoNiFe-LDH			Diverse MPs: Effective removal achieved	[55]
Li/Al-LDHs		High electron transfer activity	90-minute saturated adsorption, stable after 10 cycles	[58]
$BiVO_4$/Ag/CoMnZn-LDH	Multi metal synergy		Improved $BiVO_4$/Ag/CoMnZn photoanode: 3.2 mA/cm^2 photocurrent density, 4.15 times pristine $BiVO_4$	[59]
LaNiAl-LDH		High photocatalytic activity	La doping reduces band gap to 2.57 eV, 84.85% tetracycline degradation in 90 min	[66]
NiFe-LDH	Inter layer confinement	Inhibiting PMS self-decomposition and radical quenching	Improved self-healing to inhibit Fe leaching	[63]
NP-SAC			Enhance long-term stability	[64]

5. Environmental and Energy Footprint Analysis of LDH-Based SR-AOPs

While LDH-based catalysts have shown excellent performance in persulfate activation, a comprehensive assessment of their environmental and energy footprint is essential for evaluating their practical sustainability and alignment with green chemistry principles.

5.1. Synthesis Energy and Resource Consumption

The synthesis of LDH materials usually depends on high-energy-consumption methods, such as co-precipitation, hydrothermal treatment, or calcination. Each of these steps significantly increases the overall environmental footprint. Following the analytic framework proposed by Khan et al. for energy analysis in AOPs [67], recent studies have begun quantifying the energy consumption of different LDH fabrication routes. For example, conventional hydrothermal synthesis usually requires prolonged heating at 120-200°C for 12 to 48 hours. Such process demands a large energy input. In contrast, emerging green synthesis methods, such as mechanochemical methods and room-temperature coprecipitation, offer potential ways to reduce the energy footprint while maintaining catalytic performance. Future research should focus on systematic energy audits that compare different synthesis methods to determine the optimal balance between catalytic activity and energy efficiency.

5.2. Metal Leaching and Secondary Pollution Risks

Metal leaching from LDH catalysts during operation is another critical environmental concern. Although LDHs generally show high structural stability due to their layered architecture and strong metal-oxygen bonds, the release of metal ions—especially cobalt, copper, nickel, or iron—can cause secondary pollution and pose ecotoxicological risks. As Garner et al. pointed out in their assessment of nanomaterial environmental risks[68], even low concentrations of leached metals may accumulate in aquatic ecosystems and affect sensitive organisms. Table 1 summarizes metal leaching levels reported for various LDH-based systems, with values typically ranging from <0.1 to 5 mg/L depending on catalyst composition, reaction conditions, and operational duration. Several strategies help minimize leaching, including optimizing metal ratios, applying protective coatings, and operating under mild pH conditions. Nevertheless, long-term stability studies under realistic wastewater matrices are urgently needed to fully understand the environmental implications of metal release.

5.3. Lifecycle Assessment Perspectives

A holistic lifecycle assessment (LCA) of LDH-based SR-AOPs remains largely unexplored. Such analyses should systematically evaluate environmental trade-offs across the entire catalyst lifecycle: raw material extraction, synthesis, transportation, operational use, and end-of-life disposal. Some preliminary considerations suggest that while LDH catalysts are advantageous in reusability and high degradation efficiency, the energy and chemical inputs required for their synthesis may offset some environmental benefits. Furthermore, the potential for catalyst regeneration and metal recovery at the end of the catalyst's life could significantly improve overall sustainability. Future studies should adopt standardized LCA methodologies to enable meaningful comparisons between LDH-based systems and alternative treatment technologies. It's essential for guiding the development of greener, more sustainable approaches that balance treatment efficiency with environmental stewardship.

5.4. Toward Sustainable LDH Design

Integrating environmental footprint considerations into catalyst design represents a paradigm shift from performance-only optimization toward holistic sustainability. Luo's team constructed a novel 2D/2D Z-scheme heterojunction photocatalyst CuFe-LDH/BiOBr (4-CFB), which exhibits excellent stability, minimal metal leaching (Cu^{2+} < 1.1 mg/L), and broad-spectrum efficacy against various antibiotics over multiple reuse cycles[69]. Meanwhile, Wang's team also synthesized oxygen-vacancy-rich ZnAl-LDO via co-precipitation, followed by calcination at different temperatures. Finally, tests showed that ZnAl-LDO calcined at 500°C (ZA-500°C) exhibited the highest photocatalytic performance for CLZ degradation through peroxydisulfate (PDS) activation. Quenching experiments confirmed that •OH and $SO_4^{-\bullet}$ were the main reactive oxygen species responsible for CLZ degradation [70]. In recent studies, Ye's team designed a Ni-Zn layered double hydroxide (NiZn-LDH) catalyst that forms a self-buffering neutral microenvironment through the amphipathicity of $Zn(OH)_2$ groups. They demonstrated a closed-loop PS-P-AOPs strategy integrating selective pollutant removal, polymer product valorization, and catalyst reuse, providing a low-emission strategy for sustainable wastewater treatment [71]. Future research should focus on achieving this goal.

Key priorities for future research include: (i) developing energy-efficient, solvent-free synthesis routes; (ii) engineering LDH structures with enhanced stability to minimize metal leach-

ing; (iii) conducting comprehensive LCA studies to identify environmental hotspots; and (iv) exploring circular economy approaches that enable catalyst recovery, regeneration, and valuable resource reclamation from spent materials. By addressing these aspects, LDH-based SR-AOPs can fulfill their potential as truly sustainable technologies for water purification, contributing to the United Nations Sustainable Development Goals while minimizing unintended environmental consequences.

6. Conclusion and outlook

LDHs demonstrate versatile functionality in SR-AOPs, with applications spanning water treatment, electrocatalysis, and other fields, highlighting their potential as highly effective catalysts. However, further research is still required regarding the targeted regulation of the activated persulfate molecular configuration and the precise control over the generation of reactive species. Future research and development should prioritize the following directions:

(Ⅰ) A novel LDH-based catalyst was designed to enhance its persulfate activation performance through strategies including metal doping and intercalation modification, thereby achieving regulated generation of reactive species. Future research endeavors should focus on the synergistic integration of these complementary strategies, as their rational combination holds promise for the development of next-generation LDH-based catalysts with superior performance.

(Ⅱ) Expanding the application scenarios of catalysts should align with the principles of green chemistry, with efforts focused on optimizing solid-liquid separation efficiency to improve process sustainability. Concurrently, enhancing environmental benefits and extending catalyst service life are essential for advancing practical implementation and maximizing long-term ecological value.

(Ⅲ) Further elucidating the underlying mechanisms to establish a robust theoretical foundation, thereby guiding the rational design and optimization of next-generation catalysts. At present, studies on the mechanism of action are still insufficient, and more efforts should be devoted to this direction in the future.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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