

Review

Layered Double Hydroxide-Based Materials for Wastewater Treatment: Recent Progress in Multifunctional Environmental Applications

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Abstract

Layered double hydroxides (LDHs) have gained increasing attention as versatile materials in environmental remediation, particularly for wastewater treatment. Their unique structural properties, such as tunable metal cation composition, interlayer anion exchange, and structural memory effects, make them suitable materials for a broad range of applications. In addition to these intrinsic properties, thermally treated LDH-derived mixed metal oxides have emerged as a key focus, exhibiting enhanced activity through tailored structural, electronic, and textural properties. This review presents an up-to-date and systematic overview of recent advancements in the design and application of LDH-based materials, with a focus on photocatalytic degradation of organic dyes, adsorption of contaminants, and light-activated antimicrobial activity. The review also explores emerging photocatalytic applications in correlation with surface engineering, heterojunction formation, and thermal activation to enhance the overall efficiency. In addition, the synergy between antimicrobial activity and photocatalysis is discussed in the context of achieving multifunctional microbial control in water treatment. Finally, current challenges and future perspectives are addressed, including recyclability, scale-up potential, and the development of LDH composites as sustainable alternatives to conventional photocatalysts. This review aims to support researchers in advancing LDH-based technologies toward more efficient and versatile environmental remediation solutions.

Keywords: mixed oxides; synthesis; photocatalysis; adsorption; wastewater remediation



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1. Introduction

Rapid population expansion and intensified human activities have resulted in increased wastewater generation (production), predominately from industrial sources. Simultaneously, the depletion of freshwater is intensifying, triggering a growing imbalance between water availability and global demand. This poses significant challenges for sustainable development, public health, and ecosystem preservation. Consequently, there is a growing need for innovative and cost-effective wastewater treatment strategies that not only reduce environmental risks but also enable the recovery and reuse of valuable water resources. Among the most persistent contaminants are dyes, antibiotics, pesticides and heavy metals, all posing a great threat to human health because of their toxicity [1–5]. Therefore, ongoing research focuses on identifying adequate methods and materials that can be used for wastewater purification. A wide range of physical, chemical, and biological methods that include adsorption, membrane filtration, electrolysis,

sedimentation, coagulation–flocculation, chemical oxidation, heterogeneous photocatalysis and other treatment processes have been used for the removal of pollutants from wastewater [1,2,6–8]. Among these methods, adsorption and heterogenous photocatalysis have attracted particular interest as efficient, straightforward and eco-friendly processes.

LDHs and LDH-based materials have emerged as promising compounds attributed to their unique structural, physicochemical, and surface properties, which make them suitable for a wide range of applications. Extensive research on application of LDH-based materials in various fields, such as catalysis and photocatalysis, environmental protection, ion-exchange, medicine, biochemistry, adsorption, and electrochemistry, was conducted over the last few decades [2,7,9–13]. Versatile tunable properties of these materials unlocked a great potential for their application in the field of wastewater purification treatment, especially as adsorbents and photocatalysts for removal of various inorganic and organic pollutants (Figure 1) such as heavy metals, organic dyes, pharmaceuticals, per- and polyfluoroalkyl substances (PFAS), and micro- and nanoplastic. These systems have also demonstrated remarkable potential in photo-induced antibacterial wastewater purification [4,14,15]. Alongside this, the ability to engineer their electronic structure and interlayer chemistry paves the way for novel mechanisms of action, broadening their future use in environmental protection.

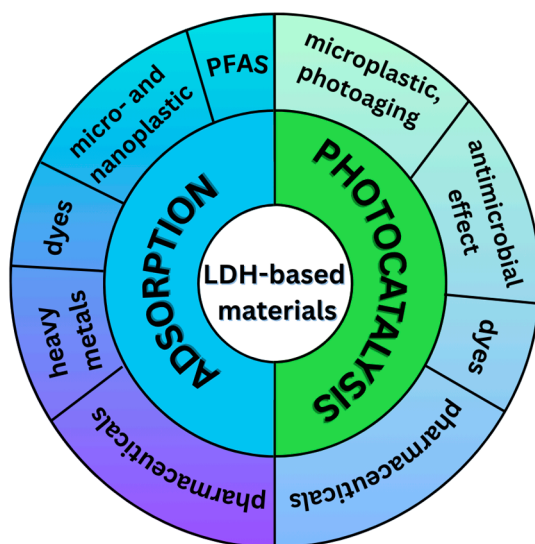


Figure 1. Application of LDH-based materials for wastewater treatment.

This review aims to provide a comprehensive overview of the recent progress in the application of LDH-based materials for wastewater purification, with particular emphasis on adsorption and photocatalytic processes. LDH-based materials are increasingly recognized as highly adaptable systems for sustainable wastewater treatment because they combine compositional tunability, structural versatility and multifunctional reactivity. While many earlier reviews have focused mainly on the dye degradation or general photocatalytic mechanisms, the present work provides an integrated discussion of both adsorption and photocatalytic pathways, supported by unified perspective that links material design, mechanism, and performance. Furthermore, this review extends the discussion to emerging contaminants such as pharmaceuticals, PFAS, and microplastics, broadening the scope of wastewater treatment research. This work aims to highlight the unique potential of LDH-based systems as sustainable and efficient materials for next-generation water treatment technologies.

2. General Properties of Layered Double Hydroxides and Derived Mixed Oxides

LDHs are classified as 2D anionic clays with lamellar structure that consists of positively charged brucite like layers with metal hydroxide octahedral units and anions and water molecules in the interlayer, as presented in Figure 2. The octahedral hydroxyl groups are connected in an infinite layer [5,16,17].

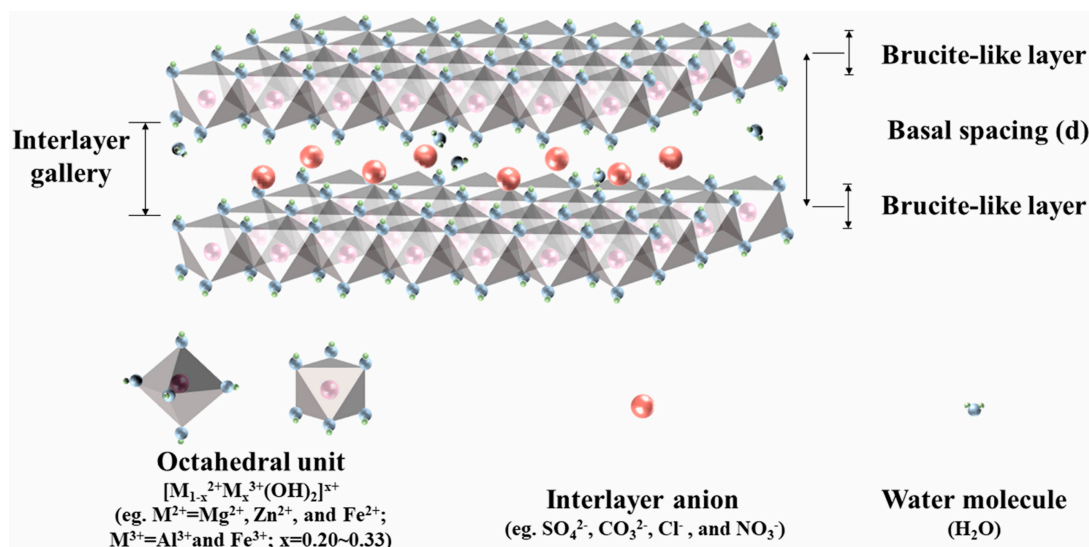


Figure 2. Structure of layered double hydroxides (the figure is reproduced from [10] with permission from Elsevier via Copyright Clearance Centre).

The general structural formula of LDH is $[M_{1-x}^{2+}M_x^{3+}(\text{OH})_2]^{x+}(A^{n-}) \cdot y\text{H}_2\text{O}$, where M^{2+} and M^{3+} are divalent and trivalent metal cations, A^{n-} is an interlayer anion with negative charge, n , x is the molar ratio of M^{3+} to $M^{2+} + M^{3+}$, or charge density, and y is the number of water molecules in the interlayer [2,5,18].

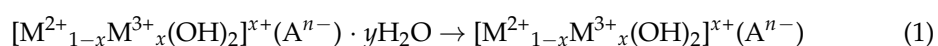
The range of the x value is between 0.2 and 0.33 has been generally known as suitable for the synthesis of single-phase LDHs. Molar ratio variation of metal cations can influence and tailor LDH properties, such as charge density and ion-exchange capacity [10,18,19]. In addition, interlayer anions have an essential role in obtaining electroneutrality and maintaining the layered structure through electrostatic interactions, hydrogen bonding, and ionic bonding [11,19]. Therefore, the type and nature of metal cations as well as anions determine the LDH properties, such as interlayer spacing, which is crucial for understanding their potential use in various processes. Large basal spacing due to weak interactions between metal cations and anions allows higher mobility of anions and possible higher reactivity [19]. Various metal cations have been successfully used for synthesis of LDHs such as Mg^{2+} , Cu^{2+} , Ni^{2+} , Zn^{2+} , Fe^{3+} , Al^{3+} , Cr^{3+} , Mn^{3+} , etc., and the differences in their electronegativity and geometry affect the nature of interactions between layers and anions [3,20–25]. At values of $x > 0.33$, the increased number of neighboring M^{3+} -containing octahedra promotes the formation of $\text{M}(\text{OH})_3$, while for $x < 0.2$, the high density of M^{2+} species within the brucite-like layers results in the precipitation of $\text{M}(\text{OH})_2$. However, these limits should be regarded as approximate, as the actual compositional range may be narrower depending on the specific LDH composition [9].

LDHs consist of positively charged brucite-type octahedral sheets alternating with interlayers that contain carbonate anions in natural minerals, or other exchangeable anions in the synthetic hydrotalcite-like analogs, along with interlayer water molecules. The size-to-charge ratio of the anions plays a crucial role, since large, low-charge anions cannot be evenly distributed between the positively charged layers. A host–guest relationship

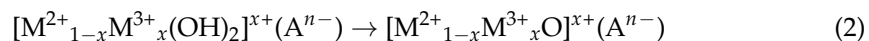
must therefore exist between the inorganic host layers and the anions accommodated in the interlayer space. Depending on anion molecule geometry, several possible arrangements can occur within the interlayer region, including a single layer parallel to the hydroxide sheets, parallel bilayers, or tilted mono- or bilayers. The orientation of interlayer anions is governed by electrostatic forces, tending toward configurations that maximize their attraction to the positively charged host layers. A large number of anions can be present in the interlayer and some of them are inorganic anions (CO_3^{2-} , NO_3^- , ClO_4^- , SO_4^{2-} , F^- , Cl^- , Br^- , I^-), organic anions (carboxylates, benzoates) and biomolecules (DNA, peptides, amino-acids). Among these anions, CO_3^{2-} anions have the highest affinity for incorporation into the LDH interlayer, making them the commonly present LDH interlayer anion. For the preparation of LDH materials with interlayer anions that are different from CO_3^{2-} anions, the method of choice should be conducted in an inert atmosphere without CO_2 presence. The compositional versatility of both the metal hydroxide layers and the interlayer anions gives LDHs a remarkable functional diversity, facilitating their use for different applications [9].

Numerous combinations of constituent divalent and trivalent metal ions, an extensive x range for the synthesis of single- and multi-phase LDHs, and variation in divalent and trivalent metal ion ratios enable the design of preferable properties, such as control over the LDH charge density as well as advantageous textural, structural, redox and acid–base properties. These features, alongside with the ability to transform into mixed oxides upon thermal treatment, facilitate tailored physicochemical properties and high surface reactivity suitable for use in wastewater purification.

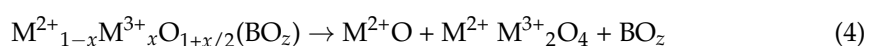
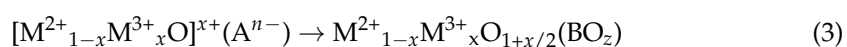
The thermal stability of LDHs arises from the strong interactions between metal cations and hydroxyl groups within the brucite-like layers. Nevertheless, dehydration typically occurs in the temperature range of 100–250 °C, involving the removal of physisorbed and interlayer water without structural collapse. As an example, in Figure 3 the most common thermal decomposition pathway of $[\text{Mg}_{1-x}\text{Al}_x(\text{OH})_2]^{x+}(\text{CO}_3^{2-}) \cdot y\text{H}_2\text{O}$ is given. This process leads to lattice contraction and a consequent decrease in basal spacing (d) compared to the pristine material and it can be presented as [26]



During dehydration, notable rearrangement of the brucite-like octahedral layers may occur, accompanied by partial migration of M^{3+} ions into the interlayer region. The extent of water loss is closely related to the strength of hydrogen bonding between the water molecules and hydroxyl layers, as well as to the structural order, both of which depend on the nature and ratio of $\text{M}^{2+}/\text{M}^{3+}$ cations. Dehydroxylation, which corresponds to the removal of hydroxyl groups from the brucite-like layers, takes place between 350 and 450 °C:



Decomposition of anions (most commonly decarbonation) may overlap with dehydroxylation or occur at slightly higher temperatures (420–470 °C), ultimately causing the collapse of the layered structure and the formation of mixed metal oxides. These processes can be presented as



where BO_z corresponds to decomposed interlayer anion species.

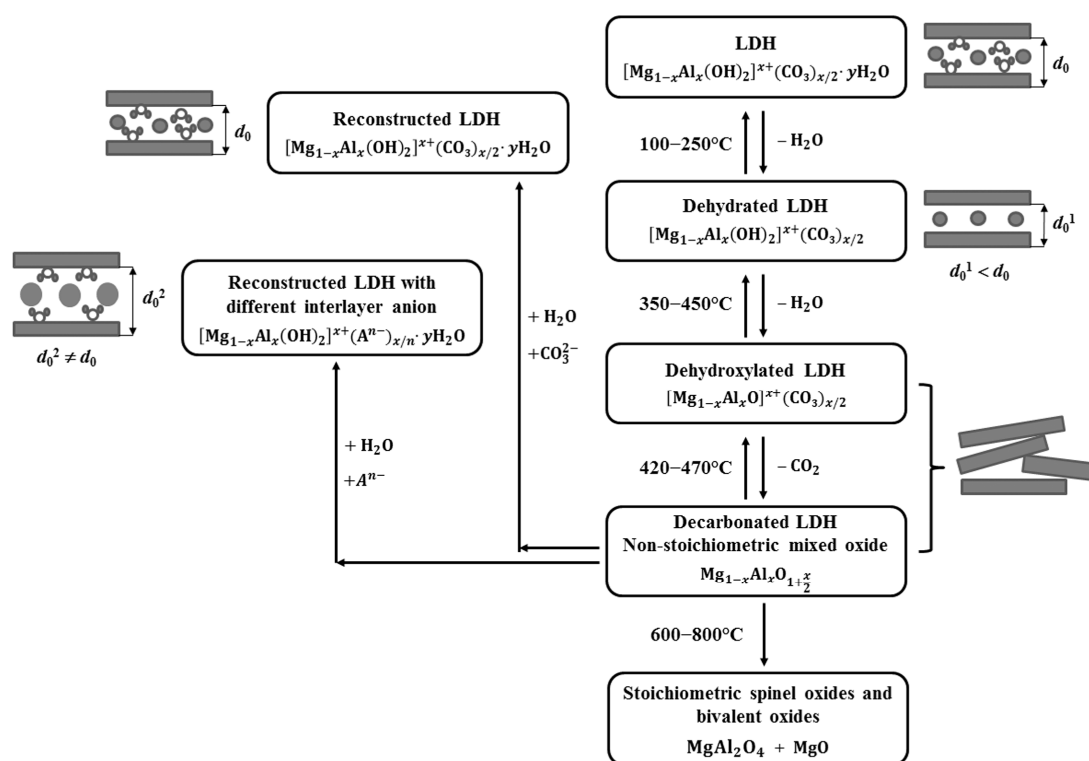


Figure 3. $[\text{Mg}_{1-x}\text{Al}_x(\text{OH})_2]^{x+}(\text{CO}_3^{2-}) \cdot y\text{H}_2\text{O}$ thermal decomposition pathways (reproduced from our previous published work [18]).

Temperatures higher than 600 °C trigger formation of spinel phases [4,18,24,27].

The collapse of the layered structure and formation of mixed oxide phases can be confirmed with structural XRD analysis. In our previous published work, the XRD analysis of LDH samples with Mg^{2+} and Zn^{2+} as divalent anions was conducted before and after calcination at 500 °C. The LDH samples showed characteristic reflections for LDH phase, that corresponded to (003), (006), (012), (015), (018), (110) and (113) reflection planes, with hexagonal unit cells on the basis of rhombohedral $R\text{-}3m$ symmetry (Figure 4a). However, after calcination and the collapse of layered structure, new phases were detected that corresponded to mixed oxides (Figure 4b). The structure of calcined samples also changed to close-packed wurtzite structure for Zn samples and cubic structure for Mg samples, concluding that the type of metal cation greatly influences the structural properties of calcined samples [3].

Mixed oxides exhibit desirable properties, such as high surface area, improved thermal and structural stability, and adjustable composition and porosity. One of the most distinctive properties of mixed metal oxides is the so-called “memory effect”. This very specific property of mixed oxides enables the reconstruction of the initial layered LDH structure in mild conditions (ambient temperature, atmospheric pressure) upon prolonged exposure to aqueous solutions or air (Figure 5). This effect offers a novel approach to modifying LDH materials, enabling the preservation of their original active sites while simultaneously increasing the number of available sites, thereby expanding their potential for applications in adsorption and catalytic processes [10,19,28]. However, it is very important to emphasize that the reconstruction of layered structure is possible only for certain calcination temperatures. If the calcination temperature is too high (>600 °C) and the formation of the spinel phase occurs, the memory effect will not take place or it will be insignificant [18,19,29,30].

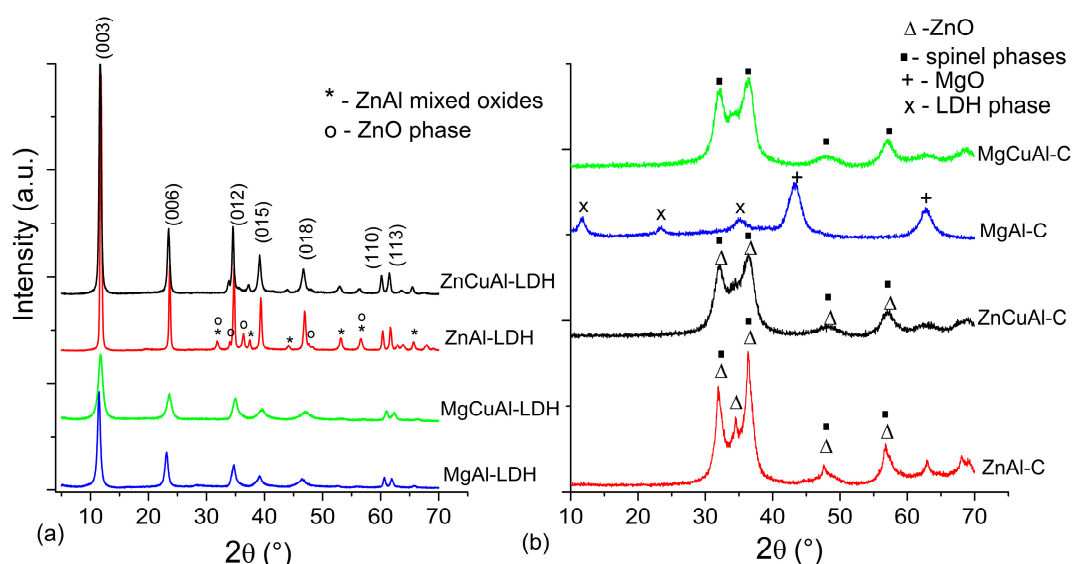


Figure 4. XRD analysis of different LDHs (a) before and (b) after calcination (reproduced from our previous publication [3] (CC) 2022 SCS).

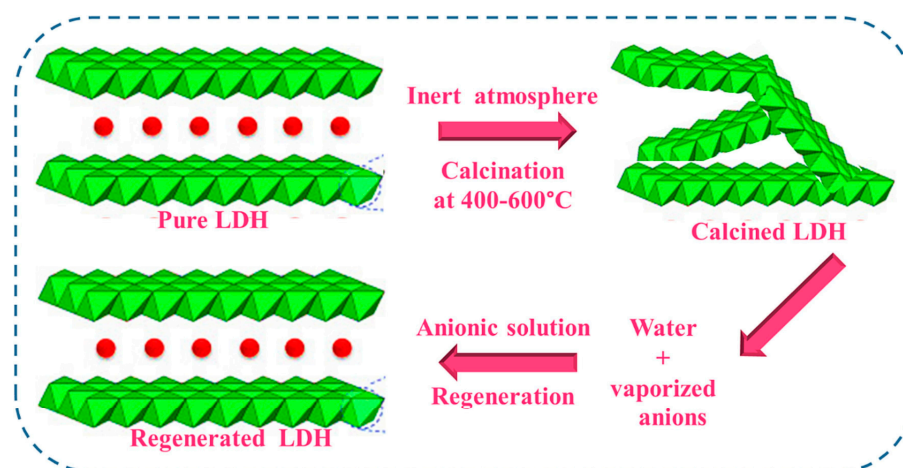


Figure 5. Thermal activation of LDHs and memory effect (reproduced from [5] with permission from Elsevier via Copyright Clearance Centre).

On account of their advanced properties, mixed oxides derived from LDHs have found a wide range of practical applications as efficient catalysts and photocatalysts for the removal of organic pollutants, such as dyes and phenolic compounds, and in the purification of industrial wastewater. Additionally, because of their high surface reactivity and chemical versatility they are frequently used in organic synthesis, electrochemical energy storage and CO₂ capture processes. Also, their structural stability and adjustable composition can contribute as electrode materials to improve performance of sensors and solar cells. These diverse applications highlight their growing potential in environmental remediation [9,11,31,32]. Improvement of LDH-derived mixed oxide properties, essential for their use as adsorbents and photocatalysts, can be achieved through understanding of the relationship between phase and chemical composition as a function of their performance efficiency. This can be achieved by careful LDH synthesis that initiates the formation of defects in the layered structure, developing high potential surface energy sites and semiconductor properties, enhancing the movement of photo-generated electrons toward the surface.

3. Synthesis of LDHs

When compared to other materials, one of the advantages of LDHs is the possibility of synthesis via different methods that enable tailoring of targeted properties. Nevertheless, every method has certain advantages and disadvantages and the best way to choose the method for LDH synthesis is to take into the consideration the necessary properties needed for the future use of LDHs and their derived mixed oxides. The following methods have been used for the successful synthesis of LDHs: co-precipitation, urea, hydrothermal, functionalization, sol–gel, ion exchange method, reconstruction (delamination) method based on “memory effect”, salt oxide (or hydroxide) method, non-equilibrium and non-conventional aging method, surface synthesis, electro synthesis, template synthesis, etc. [18–20,22,33–35]. The most commonly used synthesis method, co-precipitation, as well as the methods that enable the formation of required LHD physicochemical properties (hydrothermal, urea method and functionalization), will be explained in more detail.

3.1. Co-Precipitation Method

Most frequently used method for LDH synthesis with various ranges of interlayer species is the co-precipitation method, owing to its cost-effectiveness, time-saving, ease of operation, mild conditions and higher yield when compared to other methods. However, it does not offer a high level of morphology control [5,7,10,19,36,37]. Co-precipitation of metal salts (nitrates, chlorides, sulfates) and bases (NaOH, KOH, $\text{NH}_3 \cdot \text{H}_2\text{O}$) leads to the simultaneous precipitation of two or more metal cations under supersaturation conditions. The synthesis of single-phase LDHs was studied in detail and several key factors that influence this process regarding successful synthesis were identified, such as molar ratio of metals, pH and temperature [38]. Metal molar ratio and its influence on charge density, charge surface and formation of certain phases was already discussed above, and some of the recent studies further emphasized it [4,37,39,40]. Bencherif et al. reported that a Zn/Cr molar ratio of 3 was chosen in order to influence the formation of certain phases (ZnO and spinel phase ZnCr_2O_4) after calcination of LDHs, thus influencing its future application [39]. Lee et al. came to the conclusion that the variation of Mg/Al ratio during precipitation affects surface charge, as well as charge density. High Mg/Al ratio reduces interactions between the layers and interlayer anions, thus decreasing positive charge density of LDHs [40]. Temperature set during co-precipitation also has great influence on properties of precipitates, especially crystallinity. Lee et al. showed that temperature-controlled co-precipitation leads to higher crystallinity of Mg/Al LDH [40]. To ensure successful co-precipitation, the pH value should be adjusted to greater than or equal to the precipitation pH of the more soluble hydroxide [10,19,38,41]. The pH was also noted as a very important factor in a research conducted by Wei et al. which showed that pH strongly affects nucleation and crystal growth during synthesis of LDH [42].

The co-precipitation method can be conducted under variable pH (high supersaturation method—HS; Figure 6b) and constant pH (low supersaturation method—LS; Figure 6a). HS method is very challenging regarding particle size control, owing to fast co-precipitation, a high number of nuclei sites, simultaneous nucleation and crystallization and the presence of impurities in obtained LDHs. On the contrary, LS method is very common technique for the synthesis of high-purity, high-crystallinity and smaller particles of LDHs, attributable to uniform nucleation rate, but it is often more time-consuming and labor-intensive when compared to HS method [12,19,38,43].

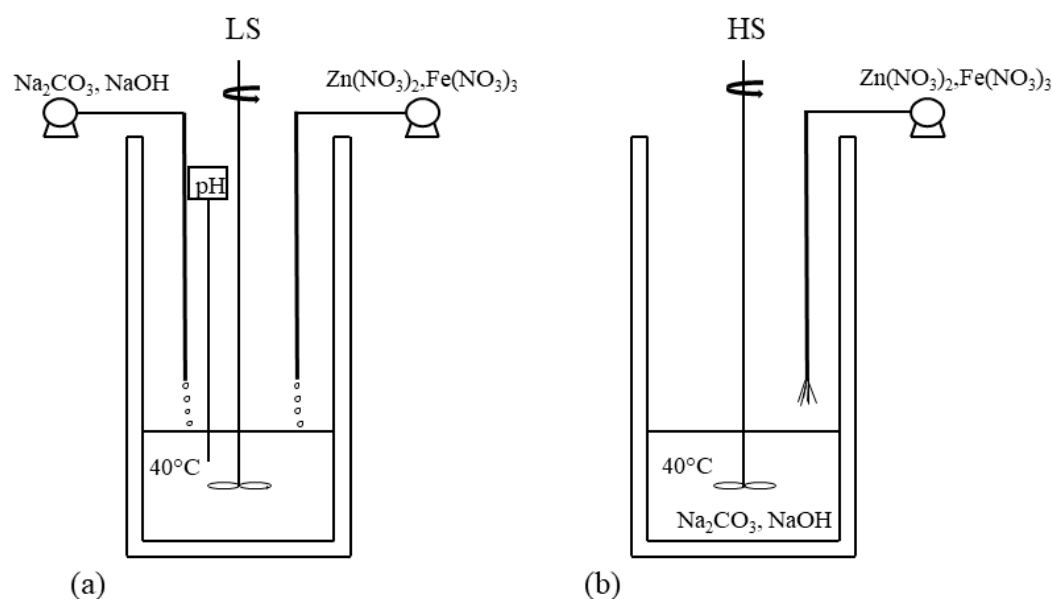


Figure 6. Co-precipitation method under (a) low-supersaturation and (b) high-supersaturation conditions (reproduced from our previous publication [44] (CC) 2024 Serbian Chemical Society).

The synthesis route and the molar ratio of M^{3+} to $M^{2+} + M^{3+}$ (charge density) have a pronounced influence on the structural ordering of LDHs (Figure 7). Samples prepared by both LS and HS methods have sharp and symmetric reflections from (003), (006), (110) and (113) planes, along with broad, non-symmetric reflections from (102), (105) and (108) planes. The increase in M^{3+} content lowers the intensity of characteristic XRD reflections and induces the formation of an additional M^{3+} hydroxide phase for both LS and HS methods. In general, samples prepared by the HS method exhibit broader and less symmetric XRD reflections, indicating a partially disordered stacking of layers (a randomly oriented set of layers lacking orderly stacking), whereas LS-derived samples show a more regular layer arrangement and improved crystallinity. HS method can cause incomplete incorporation of the trivalent metal cations into the LDH lattice and formation of additional single hydroxide phases at lower x values. In contrast, the LS method favors a more homogeneous incorporation of trivalent cations, leading to better-defined layered structures. Overall, these differences point to a stronger structural order and phase purity in LS-prepared LDHs, while HS synthesis tends to produce materials with higher defect density and partial layer misalignment [44,45].

3.2. Urea Method

The urea method is also widely employed for the synthesis of LDHs. It is based on the precipitation of metal ion solutions accompanied by the hydrolysis of urea [46]. Compared to conventional co-precipitation, this method is considered more advanced, as it yields products with high crystallinity and uniform particle size distribution, but its disadvantages include longer synthesis duration and lower yield. In this approach, urea is used instead of a basic solution, since it is a weak Brønsted base, highly soluble in water, and remains neutral at lower temperatures, thereby enabling the formation of a homogeneous mixture with the metal ion solution. These methods yield well-defined hexagonal microcrystals with the desired stoichiometry [19,36]. Di Michelle et al. [36] highlighted the influence of different process parameters on morphology. The reaction time, urea/(Al + Zn) molar ratio, and metal ion concentration were carefully optimized to synthesize ZnAl LDH in a single step, incorporating the desired anions into the interlayer region. It was revealed that urea/metal ratio determines morphologies of obtained LDHs.

Urea method can also be successfully used for the synthesis of trimetal LDHs (Fe-, Co-, Ni-, Cu- and Zn-substituted MgAl-LDH) as shown in the study by Naseem et al. [34]. Chagas et al. reported the successful synthesis of MgCoAl and NiCoAl LDHs by the hydrothermal urea hydrolysis method, obtaining pure, crystalline, and Al-rich materials. The study showed that the Mg-containing samples exhibited smaller basal spacing and higher crystallinity than the Ni analogs, attributed to stronger electrostatic interactions between hydroxyl groups and interlayer carbonate anions. Upon calcination, mixed oxides with rock-salt-type structures and high surface areas were formed, making them suitable as catalyst supports for hydrodesulfurization reactions [47].

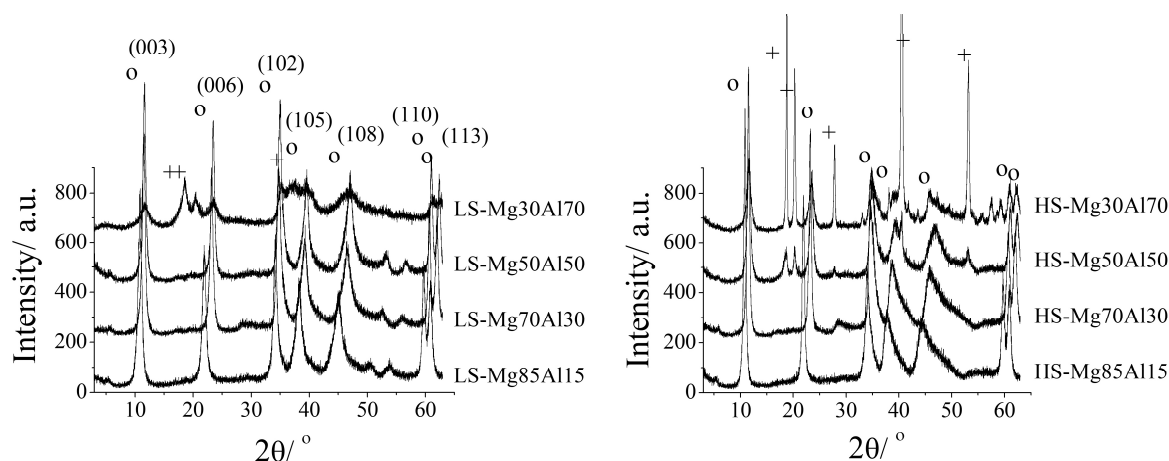


Figure 7. XRD patterns of samples with different Mg-Al content synthesized by LS and HS co-precipitation method (o LDH; + Bayerite- $\text{Al}(\text{OH})_3$) (reproduced from our previous publication [45] with permission from Springer Nature via Copyright Clearance Center).

3.3. Hydrothermal Method

In hydrothermal synthesis, freshly precipitated metal oxides obtained through mechanical stirring are subjected to treatment under controlled pressure and temperature. The process can be conducted under high temperatures (50–200 °C) and pressures (50–200 bar) to enhance the rate of crystallization while reducing reactant loss as a result of evaporation. This method has superior control over the crystallinity, morphology, and particle size of the resulting products, but when compared to the co-precipitation method, the yield is usually lower and it requires expensive equipment and high energy consumption [19,32,48]. Wu et al. studied the effect of molar ratio of ZnAl LDHs synthesized by hydrothermal method on the structure, morphology and photocatalytic ability. The results indicated that the crystallinity, lamellar structure, and photocatalytic efficiency increased with the increase in Zn/Al molar ratio [49]. Bao et al. reported that 3D flower-like, photocatalytic active CoFe LDHs can be successfully synthesized by the hydrothermal method for efficient azo dye removal [50]. Similarly, a CaAl-LDH was successfully synthesized in a polytetrafluoroethylene reactor placed in a sealed stainless-steel autoclave at 100 °C for 36 h, using a simple hydrothermal method, and demonstrated high efficiency for Cu(II) and Cd(II) removal from aqueous solutions. The material exhibited typical structural features of LDHs, including well-defined basal reflections, good crystallinity, and a hexagonal lamellar morphology, confirming the successful formation of the layered structure after hydrothermal synthesis [51].

3.4. Functionalisation/Composites

Pristine LDHs often exhibit limited activity and effectiveness in environmental applications because of the absence of sufficient functional groups or structural components. To overcome these limitations, functionalized LDHs have been developed through the

incorporation of additional functional groups or structural units. Such modifications enable the design and synthesis of advanced LDH-based nanocomposites with enhanced functional performance [2,5,19]. Additionally, the dispersion of active substances on the layers of LDHs can cause defects in the structure and influence the band gap, enhancing the absorption of light, therefore promoting the photocatalytic performance of LDHs [36,52]. The unique combination of chemical stability, high conductivity, texture, and mechanical robustness makes carbon-based materials particularly attractive for hybridization with inorganic nanomaterials, where synergistic effects can be exploited to yield nanocomposites with tailored structures and enhanced catalytic performance [53,54]. For instance, highly functional graphene oxide-based ZnCr LDH nanocomposites were synthesized via in situ crystallization of ZnCr LDH on graphene oxide. When compared to pristine ZnCr LDH, the nanocomposite exhibited higher antibacterial and photocatalytic activity [8]. Also, a nanocomposite with 3D structure that facilitates easier mass and electron transfer in photodegradation and adsorption, composed of graphitic carbon nitride (g-C₃N₄), NiAl LDH and CeO₂, was synthesized in situ in order to trigger synergistic effects [55].

Metal–organic frameworks (MOFs) are crystalline materials consisting of metal ions or clusters connected by organic linkers, whose highly ordered and tunable porous structures provide large surface areas [56]. Coupling MOFs with LDHs in order to initiate heterojunctions was investigated for ZnTi LDH. It was reported that a ZnTi LDH/MOF composite with a 30% ZnTi LDH mass ratio had the best photocatalytic and adsorption properties when compared to MOF, pure ZnTi LDH and composites with other molar ratios [57]. Soltani et al. also studied functionalization of LDHs with metal–organic frameworks and revealed that LDH/MOF nanocomposites exhibited high adsorptive capacity for uptake of heavy metals from aqueous solution [58].

Functionalization of LDHs can also be achieved with agents like glycerol, as demonstrated in a study by Abo El-Reesh, where NiFe LDH synthesized with glycerol by a simple co-precipitation method showed higher adsorptive capacity than pristine NiFe LDH [59].

Although functionalized and composite LDH materials offer enhanced performance and broader applicability, several aspects still require further attention. The synthesis procedures can be relatively complex and costly, and achieving uniform distribution of functional components remains a challenge for future research.

A comparative summary of the discussed synthesis methods, including their main advantages and disadvantages, is presented in Table 1.

Table 1. Summary of the discussed synthesis methods with comparison of their advantages and disadvantages.

Synthesis Method	Advantages	Disadvantages
Co-precipitation	• Simple and low-cost process • High yield and mild synthesis conditions • Suitable for large-scale production	• Limited control over particle size and morphology • Possible impurity incorporation • Lower structural uniformity in HS variant
Urea method	• Produces uniform and highly crystalline particles • Enables precise stoichiometric control • Facilitates homogeneous anion incorporation	• Long synthesis time • Lower yield compared to co-precipitation • Sensitive to urea/metal ratio

Table 1. *Cont.*

Synthesis Method	Advantages	Disadvantages
Hydrothermal method	• Excellent control of crystallinity and morphology • Produces well-defined lamellar structures • Enhances purity and structural order	• Requires expensive equipment and high energy input • Lower yield • Limited scalability
Functionalization/Composites	• Improved surface reactivity and adsorption capacity • Tunable band gap and enhanced photocatalytic activity • Enables design of multifunctional materials	• Complex and costly synthesis steps • Difficult to achieve uniform functionalization • Reproducibility issues

4. Application of LDHs in Wastewater Treatments

The contamination of aquatic systems with toxic pollutants, including organic dyes, pesticides, pharmaceuticals, and heavy metals, has emerged as a critical global environmental issue, posing severe risks to ecosystems and human health [1,4,7,10,28,60]. Consequently, the development and optimization of wastewater treatment technologies have attracted growing attention, with continuous efforts directed toward achieving more efficient and sustainable removal of these hazardous substances. The design of LDH-based materials with favorable properties represents a promising strategy to substantially enhance their functional efficiency.

4.1. Adsorption Processes

Adsorption has been recognized as superior to many other treatment techniques on account of its design flexibility, simplicity, low initial cost, tolerance to toxic pollutants, and ease of operation. Additionally, adsorption does not generate harmful by-products [6,61]. Adsorption processes have several benefits: the ability to reuse adsorbents, high effectiveness, and relatively quick removal of various pollutants such as heavy metals, pharmaceuticals, micro- and nanoplastic, per- and polyfluoroalkyl substances and dyes from wastewater [2,46,62]. The adsorption of anions from solutions can be explained by surface adsorption, exchange with anions in interlayers attributed to high anion-exchange capacity and the “memory effect” [11,63]. Detailed understanding of the pollutant adsorption mechanisms by LDH-based adsorbents is essential for their successful use in treating actual wastewater. The adsorption mechanism is governed by both the nature of the adsorbate and the physicochemical characteristics of the adsorbent. In particular, parameters such as pore structure, particle size, surface area, and the type and distribution of surface functional groups play a crucial role in determining the strength and specificity of adsorptive interactions. The overall adsorption behavior thus arises from the interplay between the structural features of the adsorbate and the textural and surface properties of the adsorbent [2]. Generally, several mechanisms, such as electrostatic interactions, hydrogen bonding, ion-exchange, surface complexation and π - π interactions, take place simultaneously [3,64,65].

The research group of Farghali et al. investigated the adsorption mechanism of Congo red dye (CR) by MgAl-LDH nanosorbent and suggested two possible adsorption mechanisms: adsorption on the external surface and ion exchange. They concluded that the CR adsorption that occurred on the outer LDH surface was initiated by electrostatic attraction, triggered between the negatively charged sulfonate group of CR dye and the

positively charged LDH surface, and by hydrogen-bonding generated between the hydroxyl groups of the LDH surface and negatively charged groups of CR dye. The replacement of the anions in the interlayer spacing (CO_3^{2-} , HCO_3^- , or OH^-) with CR anions through the insertion of CR molecules was defined by the authors as the ion-exchange process responsible for CR adsorption [66].

The adsorption of levofloxacin onto ZnCoFe/LDH and Cu–Cyanoguanidine–ZnCoFe/LDH in a study by Abdelkawy et al. occurred through several complementary mechanisms, predominantly surface complexation and π – π interactions. Surface complexation plays a central role, where Lewis-acidic metal cations (Zn^{2+} , Co^{2+} , Fe^{3+}) coordinate with the carboxyl and carbonyl functional groups of levofloxacin, forming stable inner-sphere complexes. Equally important, π – π stacking between the aromatic rings of levofloxacin and the electron-rich Cu-modified LDH layers significantly strengthens adsorption, contributing to higher selectivity and stability. Electrostatic attraction and hydrogen bonding also assist in the overall adsorption process, especially near neutral pH, where levofloxacin exists in its zwitterionic form [67]. Apart from these, other possible adsorption mechanisms are outlined in Table 1.

More in-depth understanding of mechanism and adsorption rate-limiting steps can be achieved by applying different kinetic models on experimental data. Kinetic models that usually describe adsorption of heavy metals and organic pollutants with LDH adsorbents are pseudo-first-order (PFO) and pseudo-second-order (PSO) models [2,6]. In the PFO kinetic model, the adsorption rate is assumed to be proportional to the concentration of the adsorbate, with the process mainly governed by diffusion through a boundary layer. This model is typically applied when the rate of site occupation is directly related to the number of available active sites on the adsorbent surface. In contrast, the PSO model emphasizes the role of chemisorption, where the adsorption rate is influenced by the interaction between adsorbate molecules and the surface of the solid adsorbent. The model assumes that the rate of adsorption depends both on the availability of unoccupied sites and on the concentration of heavy metal ions or dye molecules in solution [2,58,68]. However, Soltani et al. discussed the possibility of fitting different kinetic models in the adsorption process [58]. Based on the obtained kinetic parameters of adsorption of heavy metals (Pb^{2+} and Cd^{2+}) on LDH/MOF nanocomposite, both the PFO and PSO models provided a good fit to the experimental data. It can be inferred that pseudo-first-order model better describes the initial stage of the adsorption process. This suggests that adsorption occurs rapidly at first, which is typical of physisorption, while the partial conformity with the PSO kinetic model indicates that chemisorption interactions become more relevant at later stages. Therefore, the overall adsorption mechanism can be described as a combination of both models, with the PFO model being more dominant.

Some LDH-based materials used for adsorption of different pollutants from wastewater, as well as their adsorption kinetics and mechanism, are given in Table 2.

Table 2. Summary of pollutant adsorption by LDH-based materials: adsorption mechanism and kinetic model.

LDH	Pollutant	Adsorption Kinetics	Adsorption Mechanism	Ref.
NiAl LDH—S	Congo red	PSO	H-bonding and electrostatic attraction	[65]
ZnCoCr LDH	Congo red	PSO	Electrostatic attractions	[69]

Table 2. Cont.

LDH	Pollutant	Adsorption Kinetics	Adsorption Mechanism	Ref.
MgAl LDH nanosheet-modified montmorillonite	Methyl orange, Methylene blue	PSO	Intercalation-split mechanism, ion exchange	[70]
MgAl LDH nanosorbent	Congo red	PSO	Electrostatic interactions, surface complexation	[67]
ZnFe LDH	Methyl orange	Fits well into several models	Electrostatic interactions, H-bonding	[71]
MgAl-NO ₃ LDH MgAl-CO ₃ LDH	Light green dye	PSO	Electrostatic interactions, H-bonding	[72]
MgFe LDH nanoparticles	Indigo carmine	PSO	Interactions between solid and dye molecules	[73]
CaAl LDH/biochar	Crystal violet	PSO	Electrostatic interactions	[74]
ZnMgAl LDH	Crystal violet	PSO	Hydrogen bonding, electrostatic attraction, n- π interaction, π - π interaction, mesoporous filling	[75]
La-, Mo-, W-doped NiFe LDH	Methyl orange	PSO	Anion exchange and the attraction of layer charge	[25]
MIL 88B (Fe)/ZnTi LDH (MZ 30%)	Tetracycline	PSO	External mass transfer and internal particle diffusion	[57]
ZnCoFe LDH	Levofloxacin	Fits well into several models	Electrostatic interactions, hydrogen bonding, surface complexation, ion exchange, π - π stacking, and redox activity	[67]
Zn/Ce LDH	Doxycycline	PSO	Interactions of doxycycline molecules with Zn cations	[76]
LDH MgAl/MgFe composites	Tetracycline	PSO	Chemical and π - π interactions	[77]
MgAl LDH/cellulose nanocomposite	Amoxicillin	PSO	Electrostatic interactions	[21]
GLDA-ZnAl LDH	Pb(II), Cd(II)	PSO	Chemisorption, metal-LDH interactions	[78]
NiFe LDH/glycerol	Cr (VI)	Fits well into several models	Electrostatic attraction, ion exchange, complexation, hydrogen bond formation	[59]
ZnAl LDH/silica	Cr (VI)	PSO	Intercalation of Cr ₂ O ₇ ²⁻ and exchange between Cr ₂ O ₇ ²⁻ and OH ⁻	[68]
NiCo LDH/MOF nanocomposite	Pb(II), Cd(II)	Combination of PFO and PSO	Surface complexation, cation- π interactions	[58]
ZnCoFe LDH	As ³⁺ , Pb ²⁺ , Hg ²⁺	PSO	Non-electrostatic attraction, ion exchange, surface complexation	[79]
MgAl LDH calcined MgAl LDH	Cr (VI)	PSO	Electrostatic attractions, intercalation	[63]

Table 2. Cont.

LDH	Pollutant	Adsorption Kinetics	Adsorption Mechanism	Ref.
ZnAl LDH, MgAl LDH	PFOA	PSO	Electrostatic interactions	[80]
CuMgFe-LDH	PFOA, PFOS	-	Electrostatic interactions, hydrophobic interactions	[81]
nanostructured ZnAl LDHs	Polystyrene nanoplastics	-	Electrostatic interactions	[82]
calcined ZnAl LDH	Polystyrene nanoplastics	Intraparticle diffusion model	Electrostatic interactions, hydrogen bonds, pore filling and complexation	[83]

4.1.1. Adsorption of Dyes

Dyes are commonly vastly present in numerous industries and have been labeled as hazardous pollutants. Therefore, efficient dye removal from wastewater is a crucial step towards environmental remediation. Adsorption has emerged as a simple, efficient, and cost-effective method for their removal, with the potential to achieve high efficiency even at low adsorbent concentrations. The adsorbent selection is a crucial step influencing the adsorption efficiency, and materials with high surface area, stability, and reusability, such as LDHs and their derived mixed oxides, have been found to be particularly efficient in the adsorption process [66,69–71].

Around 60% of all dye pollutants are azo dyes and depending on the charge of the dissolved ion they are classified as anionic and cationic [2,5,66,69–73,84]. As previously mentioned, there are different approaches to the adsorption mechanism explanation (Table 1), but most commonly the adsorption mechanisms of dyes by LDH-based adsorbents are defined by electrostatic interactions between positively charged layers of LDHs and anionic dyes [25]. While LDHs naturally favor anionic dye adsorption, appropriate surface functionalization or adjustment of the solution pH, which can modify the surface charge of the layers, enables them to also effectively capture cationic dyes [74,75].

In their study, Bharali and Deka investigated the adsorption process of Congo red dye with sonochemically synthesized NiAl-LDH and characterized the process as physisorption that took place on the LDH surface, since no significant changes in adsorbent structure and morphology were detected after adsorption. Furthermore, they concluded that the adsorption process followed the PSO model and that there are several rate-limiting steps in the adsorption process apart from intraparticle diffusion, such as external mass transfer [65]. The pH of the solution was also considered as a key factor. Maximum adsorption was observed at pH 6, where electrostatic attraction is dominant, whereas lower pH values cause partial dissolution of the adsorbent and higher pH values induce electrostatic repulsion, both resulting in reduced removal efficiency [65]. On the contrary, a new study on the adsorptive performance of Congo red on ZnCoCr LDH determined and concluded that pH 2 was the optimal value for the adsorption process (98% efficiency), owing to strong electrostatic interactions between the positively charged surface of the LDH and sulfonate groups of Congo red [69]. It was also indicated that pH lower than the point of zero charge of the adsorbent enhanced the adsorption as a result of surface charge for anionic dyes, while pH higher than the point of zero charge favored adsorption of cationic dyes, emphasizing the importance of electrostatic attraction for overall adsorption process [66,73–75]. These findings suggest that the effect of pH is dependent on the nature of adsorbents and dyes used in the process. Mahmoud et al. investigated the affinity of ZnFe LDH nanostructure towards anionic (Methyl orange) and cationic (Methylene

blue and Malachite green) dyes. LDH used in the experiment showed higher affinity and adsorption capacity towards anionic dye in both single and ternary mixtures, resulting from electrostatic interactions between surface of LDH and dye molecules that favored anionic dye over cationic dyes [71]. A recent study by Al-Furhud et al. shed some light on the important role of interlayer anions in the LDH structure on the adsorption process. MgAl-NO₃ LDH showed higher efficiency for light green dye adsorption when compared to MgAl-CO₃ LDH because NO₃[−] can interact with the dye molecules through hydrogen bonds, thus creating additional adsorption active sites [72]. LDHs can be thermally treated and decomposed to mixed oxides that also can be used as effective adsorbents in dye removal from wastewater (Figure 8). Indigo carmine dye adsorption was more efficient with calcined mixed metal oxide than ZnAl LDH, attributable to the memory effect and intercalation that take place in metal oxides [85]. A similar conclusion regarding higher adsorption efficiency of Acid yellow 42 on calcined MgAl-CO₃-LDH was also discussed in research conducted by dos Santos et al. [86]. Notably, the recalcined adsorbents were successfully used in five adsorption cycles, with low efficiency loss, emphasizing their ecoefficiency.

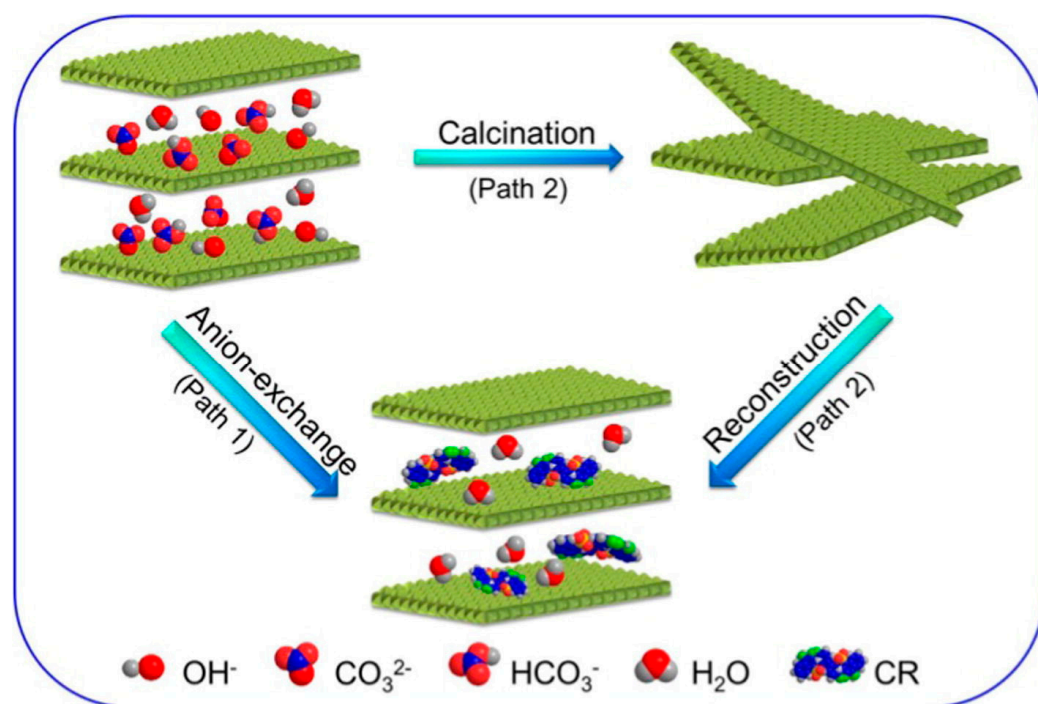


Figure 8. Possible adsorption mechanism of Congo red dye on LDH and calcined mixed oxide (reproduced from [87] with permission from Elsevier via Copyright Clearance Center).

This behavior can be explained by larger surface areas of mixed oxides and charge density as well as by the “memory effect” that is triggered during the rehydration process, which can also further enhance electrostatic interactions between adsorbent surface and dye molecules [3,85,88]. In order to elucidate the memory effect, Hadnađev-Kostić et al. investigated the adsorption mechanism of mixed oxides derived from ZnAl, ZnCuAl, MgAl and MgCuAl LDH. The results showed higher adsorption efficiency of mixed oxides compared to their LDH precursors [3]. XRD analysis of mixed oxides (CMO) after Methyl orange adsorption was conducted in order to confirm that the “memory effect” occurred after rehydration (Figure 9). After Methyl orange adsorption, mixed-oxide XRD patterns were not detected and only the LDH XRD patterns, corresponding to the pristine LDHs that were thermally treated in order to obtain mixed oxides, were identified, confirming that rehydration of LDH derived-mixed oxides took place during the adsorption of Methyl

orange. Since the patterns of pristine LDHs and mixed oxides after the adsorption process were almost identical, it was concluded that the adsorption process took place on the surface of adsorbents and the intercalation of dye molecules did not occur [3]. The study also revealed high stability of calcined LDHs after four consecutive adsorption cycles, suggesting scale-up potential.

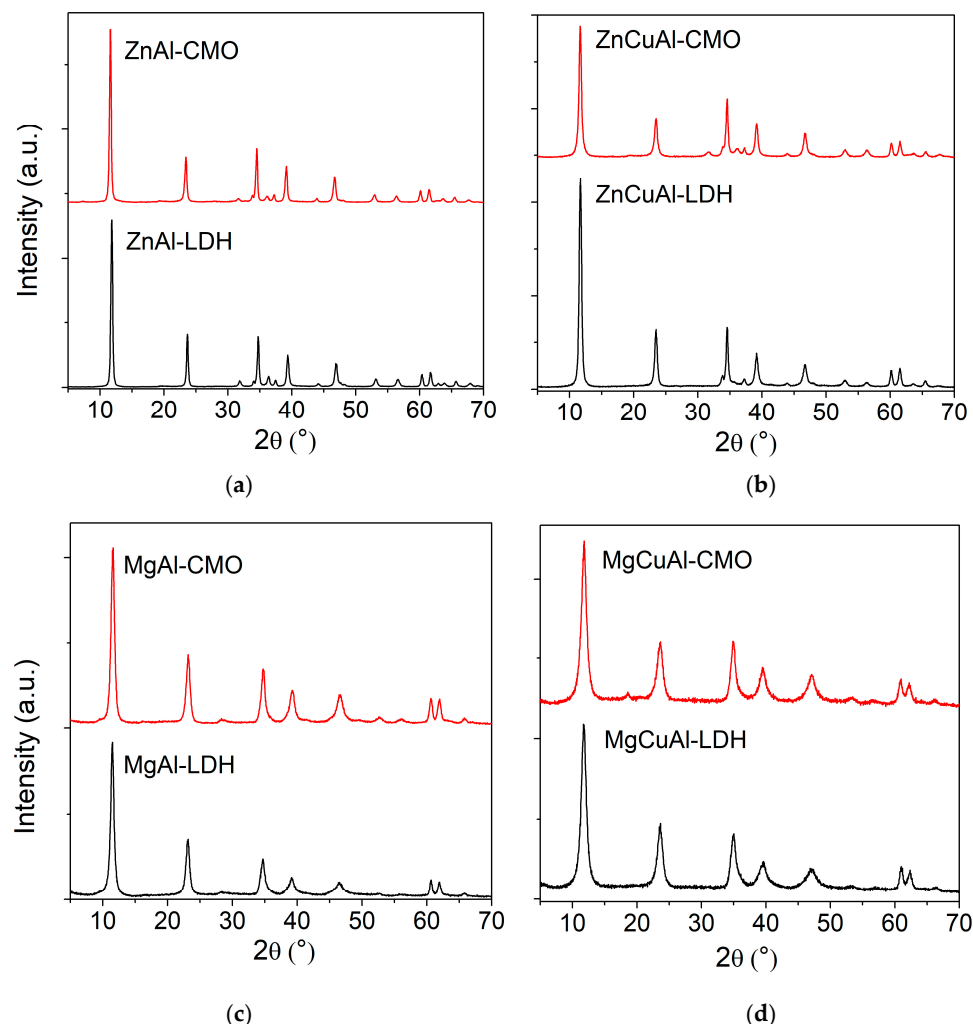


Figure 9. XRD patterns of (a) ZnAl, (b) ZnCuAl, (c) MgAl and (d) MgCuAl samples, both pristine (LDHs) and after Methyl orange adsorption on thermally treated LDH-derived mixed oxides (CMO) (reproduced from our previous publication [3] CC 2022 SCS).

4.1.2. Adsorption of Pharmaceuticals

The development of medicine and the pharmaceutical industry dictates the way the modern world integrates science, technology, and innovation into everyday life. The continuous consumption of various drugs has resulted in their frequent detection in surface water, groundwater, and even drinking water. The incomplete removal of drug residues during conventional wastewater treatment raises serious environmental and health concerns, since many of these compounds are biologically active and persistent. For example, diclofenac, one of the most commonly detected non-steroidal anti-inflammatory drugs in wastewater, is known for its photostability and toxicity even at trace levels. Careful design of LDHs with desirable properties has made these materials promising adsorbents for pharmaceutical wastewater treatment. Some of the successful applications will be further discussed. Santamaria et al. used ZnTiAl-LDH for efficient removal of diclofenac from water [89]. The adsorption mechanism involved electrostatic attraction and hydrogen bonding between

the carboxylate groups of diclofenac and the hydroxyl groups on the LDH surface, while the kinetic data fitted well to the PSO kinetic model, suggesting that chemisorption was the dominant rate-controlling step [89]. Another common drug, ibuprofen, was effectively adsorbed onto MgAl-LDHs through electrostatic interactions between its negatively charged carboxylate groups and the positively charged LDH surface, as well as by anion exchange within the interlayers, forming stable and crystalline LDH–ibuprofen composites. This efficient adsorption mechanism not only enables the removal of ibuprofen from aqueous systems but also demonstrates the potential of MgAl-LDH as a drug carrier, capable of providing controlled and sustained release of intercalated pharmaceuticals [90].

The wide use of antibiotics initiates their high presence in wastewater causing antibacterial resistance to different bacterial species. Therefore, the use of LDHs as straightforward and environmental friendly solutions for the removal of these substances has great potential and importance. Numerous studies have been conducted in order to further explain the adsorption mechanism of antibiotic removal with LDH-based materials [21,57,67,76,77]. Luo et al. showed that ZnTi LDH coupled with MOF successfully adsorbed tetracycline from aqueous solution. Fe-based MOFs interacted with functional groups of tetracycline molecules and promoted adsorption. Adsorption followed PSO kinetics and the process was mainly governed by chemisorption [57]. Successful levofloxacin removal was achieved with ZnCoFe LDH adsorbent [67]. As in previous studies, the influence of the pH was significant in intensifying electrostatic forces between adsorbent and pollutant caused by changes in the surface charge of adsorbents. The results revealed that low pH values promoted a surface positive charge and led to the repulsion of levofloxacin molecules, therefore causing lower adsorption efficiency in comparison to the results obtained at higher pH values [67]. The importance of solution pH was also underlined by Ramezani et al. for doxycycline adsorption on Zn/Ce LDH. The positively charged surface of Zn/Ce LDH at pH 6 attracted zwitterions of doxycycline through electrostatic forces, enhancing the adsorption capacity. The Zn/Ce LDH exhibited high reusability, with a slight decrease (~15%) after six cycles, outlining its potential in doxycycline adsorption from wastewater [76].

In general, adsorption of pharmaceuticals on LDH-based materials involves a combination of mechanisms such as electrostatic interactions, hydrogen bonding, surface complexation with metal centers, and π – π stacking between aromatic rings of antibiotics and adsorbent surfaces. Kinetic models (PFO, PSO) are commonly applied to clarify the adsorption mechanism and surface characteristics. These findings indicate that LDH-based materials show strong potential for removal of pharmaceuticals from water; however, for practical application, further studies are required on regeneration, stability, and efficiency in complex real wastewater matrices.

4.1.3. Adsorption of Heavy Metals

Heavy metals are considered highly hazardous pollutants owing to their toxicity, persistence, and ability to bioaccumulate in living organisms. Their presence in water mainly originates from industrial activities, and unlike organic contaminants, their removal is more complex and challenging for the scientific community [2]. LDH-based materials have been successfully used for the removal of various metal cations (Pb^{2+} , Cd^{2+} , Cr^{6+} , Cu^{2+} , Ni^{2+} , Hg^{2+}) from water [59,71,79,91]. A summary of possible mechanisms for heavy metal adsorption is given in Table 1. Most commonly used explanations for heavy metal adsorption mechanisms are ion exchange (intercalation of metal cations in the interlayers), surface complexation and chemical precipitation (Figure 10) [2,92]. Surface chemical precipitation involves the formation of metal hydroxide precipitates on the LDH surface, while complexation occurs through the coordination of metal ions with surface hydroxyl groups, leading to anion–metal complex formation.

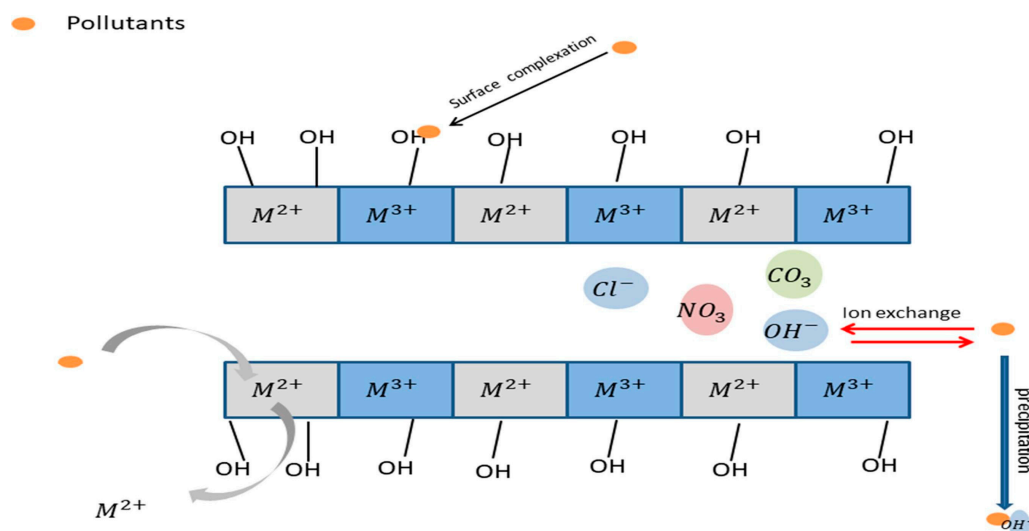


Figure 10. The adsorption mechanism of metal cations by LDHs (reproduced from [2] with permission from Elsevier via Copyright Clearance Centre).

The study of Abo El-Reesh et al. investigated Cr (VI) removal by NiFe LDH considering that Cr (VI) is one of the most common and toxic metals that can be present in water. NiFe LDH with loaded glycerol successfully adsorbed Cr (VI) from water and showed high adsorption capacity. Further, they concluded that the initial adsorption was very fast and that various kinetic models could be used for the description of the process. Considering that the pH value influences the Cr(VI) species in the solution, as well as surface charge of LDHs, this process can be defined as pH-dependent. The highest adsorption capacity of Cr(VI) was determined at pH of around 5, probably as a result of the strong electrostatic attractive forces between different charges [59,93]. The research of Zhang et al. revealed high adsorption capacity of ZnAl LDH with silica towards Cr (VI) from water solution. It was concluded that chemisorption had a significant role in the overall adsorption since the process followed the PSO kinetic model. Samples exhibited high potential for industrial application, since high efficiency was achieved after six adsorption cycles [68]. A study by Benhiti et al. [63] showed that the “memory effect” of calcined MgAl LDH adsorbent was pivotal for high Cr (VI) adsorption efficiency, due to intercalation of Cr (VI) during rehydration. High recyclability of the adsorbent with calcination/rehydration (7% decrease in efficiency after five cycles) could also be attributed to the memory effect. Lead and cadmium are toxic heavy metals that are often present in wastewater and can cause a wide range of health problems for humans and animals. Soltani et al. used NiCo LDH/MOF nanocomposites for Pb and Cd removal and concluded that high adsorption capacity can be achieved due to (i) favorable textural properties; (ii) the surface functionality and physicochemical nature of the adsorbent surface; (iii) the availability of active adsorption sites; and (iv) the morphology of the adsorbent particles. As in previous studies, it was concluded that pH directly affects the surface charge which can have a negative impact on the adsorption resulting from repulsive electrostatic forces [58]. In the study conducted by Aita et al., ternary LDH, ZnCoFe LDH, showed very high adsorption capacity for As and Hg removal, emphasizing that chemisorption played the key role in the adsorption process. Furthermore, the results indicated that the optimal pH was different for each metal, because of their chemical nature (pH = 3 for As, pH = 4.5 for Hg and pH = 3 and 5.5 for Pb). Recyclability tests for adsorption of Pb revealed that after five adsorption cycles, the efficiency decreased by 40%, owing to a low number of available adsorption sites [79].

4.1.4. Adsorption of Per- and Polyfluoroalkyl Substances

Per- and polyfluoroalkyl substances have recently become a serious focus in environmental remediation owing to their exceptional chemical stability, environmental persistence and potential health risks. These synthetic compounds, widely used in firefighting foams, textiles, food packaging, and cosmetics, have strong C–F bonds that make them highly resistant to biological and chemical degradation. As a result, PFASs, including persistent pollutants, such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS), are frequently detected in soil, sediment, and aquatic environments worldwide [80]. Among various treatment methods, adsorption using LDH-based materials has recently emerged as a promising and sustainable approach for PFAS removal. As previously mentioned, LDHs offer tunable surface charge, adjustable interlayer spacing, and anion-exchange capabilities that enable strong interactions with anionic PFAS species. Comparative studies have shown that nitrate-intercalated ZnAl and MgAl LDHs exhibit high affinity and fast adsorption kinetics for PFOA. The ZnAl LDH adsorbent showed superior performance compared to MgAl that was attributed to its higher surface charge, larger surface area, and more accessible interlayer sites [80]. Similarly, CuMgFe-LDHs have demonstrated rapid removal of PFOS and PFOA, achieving equilibrium within approximately one hour. Adsorption was found to be chain-length-dependent, with stronger affinity for longer-chain PFASs, suggesting that hydrophobic interactions play a key role together with the electrostatic attraction [81]. More recent research has explored the potential of composite and nanoscale LDH systems to enhance both adsorption and degradation mechanisms. For example, a zero-valent iron (ZVI)/LDH composite was developed to combine the high reactivity of nanoscale ZVI with the adsorption capacity of LDH. The LDH coating stabilized ZVI particles, reduced aggregation, and enabled a synergistic process involving PFAS adsorption and partial defluorination, particularly for PFOA [94]. These coupled systems could open a pathway to integrated adsorption–degradation technologies that could transform PFAS remediation under ambient conditions. Additionally, new work has focused on ultrathin LDH nanosheets prepared via green, low-temperature exfoliation methods. These nanosheets exhibit exceptional PFOS sorption capacities (up to 1502 mg g^{−1}) resulting from their defect-rich and high-surface-area structure, as well as from multiple interaction mechanisms, including electrostatic attraction, ion exchange and hydrogen bonding [95]. Although their powder form currently limits large-scale separation and reuse, these findings highlight the potential of defect engineering and morphology control to achieve efficient PFAS removal while maintaining environmental compatibility. Overall, these recent studies highlight the versatility of LDH-based materials in overcoming the challenges of persistent PFAS contamination. Future research should focus on expanding the understanding of structure–function relationships for both short- and long-chain PFAS, exploring green synthesis routes for scalable LDH production, and developing composite systems that combine adsorption with catalytic degradation.

4.1.5. Adsorption of Micro- and Nanoplastics

Microplastic (MPs < 5 mm) and nanoplastic (NPDs < 1 µm) have recently emerged as major environmental concerns because of their persistence and potential toxicity to aquatic organisms and humans. These plastic particles, originating from industrial, domestic, and agricultural sources, are not only resistant to biodegradation, but also act as carriers for persistent organic pollutants and heavy metals, thereby intensifying ecological risks. Because of their high surface area, strong hydrophobicity, and variable surface charge, their removal from aqueous systems remains a challenge. LDHs and their mixed oxides have recently been engaged as adsorbents for the removal of micro- and nanoscale plastic debris. For example, Tiwari et al. [82] reported for the first time the successful adsorption

of negatively charged polystyrene nanoplastic using nanostructured ZnAl LDHs, achieving up to 96% removal through electrostatic interactions. The adsorption efficiency was influenced by competing anions and pH, being highest for low-ionic-strength and slightly acidic media, which strengthen the electrostatic affinity between negatively charged particles and the positively charged LDH surface. These findings highlight the potential of LDH-based systems for the eco-friendly and physical removal of nanoplastic contaminants from freshwater environments. Building on this, Zhang et al. [83] synthesized a hierarchically structured flower-like ZnAl LDH-derived oxide (H-ZnAl-LDO) using a one-step calcination process. This adsorbent exhibited a high polystyrene removal efficiency (85–100%) across a wide pH range (3–11), demonstrating excellent stability even under acidic and alkaline conditions. Notably, the H-ZnAl-LDO maintained over 90% removal efficiency after five regeneration cycles, confirming its reusability and structural resilience. Further improvement was achieved with the development of three-dimensional superparamagnetic magnesium–aluminum LDH-derived oxides (M-MgAl-LDOs), which enabled easy magnetic recovery after adsorption. Yang et al. [96] reported that these materials achieved a maximum adsorption capacity of 203 mg g^{-1} for polystyrene MPs through a combination of electrostatic and π - π interactions, pore filling, and hydrogen bonding. The superparamagnetic characteristic provided fast separation and reusability, emphasizing the potential of magnetic LDOs as next-generation adsorbents for microplastic removal in complex aquatic environments. Together, these findings establish LDHs and LDOs as versatile, tunable, and regenerable adsorbents for microplastic and nanoplastic remediation. The adsorption mechanism given in Figure 11 involves a combination of electrostatic attraction, hydrogen bonding, complexation, and pore filling. The layered structure, high surface charge and compositional flexibility of LDHs allow them to perform efficiently under diverse environmental conditions, while emerging modifications (hierarchical porosity, magnetic separation, or composite formation) further enhance their practical applicability.

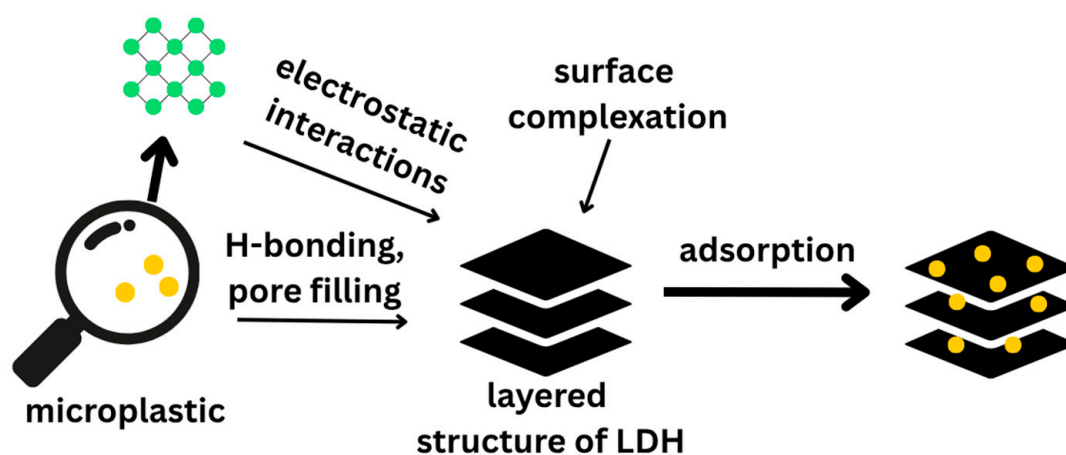


Figure 11. Proposed mechanism of microplastic adsorption.

4.2. Photocatalytic Processes

Photocatalysis is an advanced oxidation process that leads to chemical photodegradation of organic molecules. Principles of photocatalytic oxidation can be described in several steps [4,49,52,97–102]:

1. Adsorption of pollutant molecules from the medium on photocatalyst active sites through physisorption or chemisorption.
2. Photon absorption of suitable wavelength, with energy equal to or higher than the band gap energy. Electrons are promoted from the valence band (VB) to the conduction band (CB), triggering positively charged holes (h^+) in the VB. This process generates

electron–hole (e^-/h^+) pairs, which are the primary charge carriers in photocatalytic reactions (Figure 12).

3. Migration of generated e^-/h^+ pairs to the surface of the photocatalyst, where reactant molecules are adsorbed.
4. At the surface, simultaneous oxidation and reduction processes occur. The photo-generated electrons in the CB are typically transferred to molecular oxygen, yielding superoxide radical anions ($\bullet O_2^-$) as the first reactive oxygen species (ROS). Simultaneously, photogenerated holes in the VB oxidize hydroxide ions or water molecules to produce hydroxyl radicals ($\bullet OH$), which are among the most powerful oxidizing agents. These ROS, together with the direct redox activity of electrons and holes, initiate the degradation of adsorbed pollutants. The organic contaminants are gradually converted into intermediate species and, ultimately, mineralized to harmless products such as carbon dioxide (CO_2) and water (H_2O).
5. Desorption of the final products from the photocatalyst surface, thereby regenerating the active sites and allowing subsequent catalytic cycles.

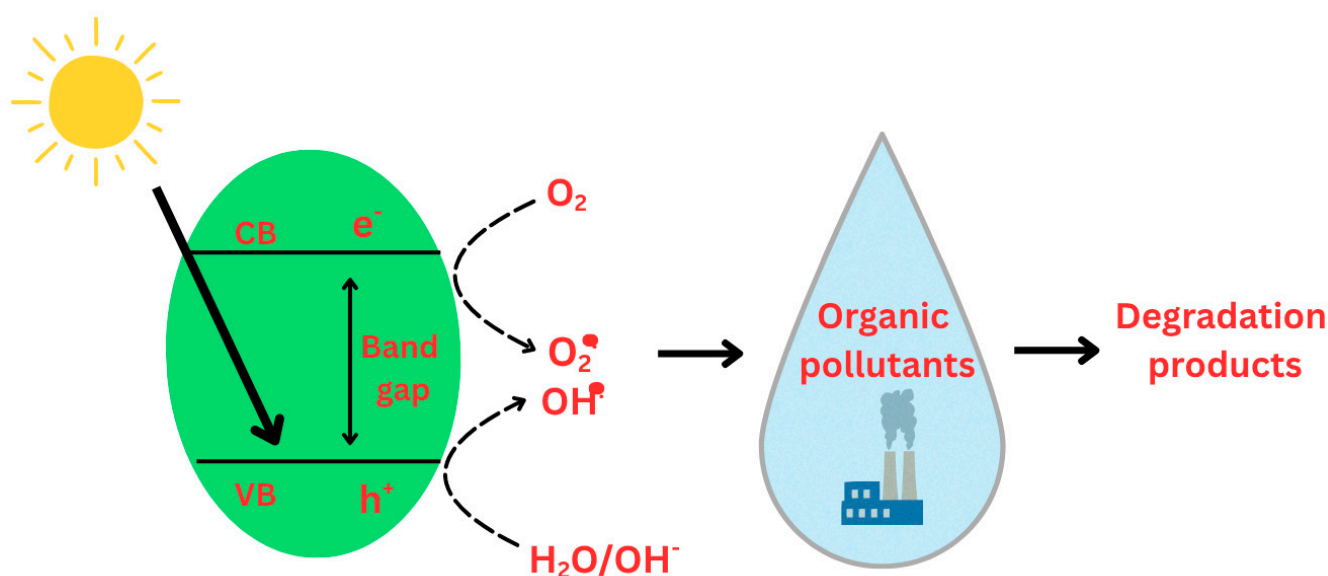


Figure 12. Proposed mechanism for photodegradation of organic pollutants.

LDH-based materials are frequently used in photocatalytic wastewater purification processes, attributable to simple synthesis, tunable properties, facile electron transport and limited undesirable electron/hole recombination [103–105]. The kinetics of photocatalytic reactions can be described by the pseudo-first-order (PFO) Langmuir–Hinshelwood model. The fundamental assumption of this model is that the reaction rate is the rate-limiting step of the overall photocatalytic process [4,44,52,106]. In other words, the process proceeds in the kinetic regime rather than being diffusion-controlled, as is the case with simple adsorption. In addition to this primary assumption, the model also postulates that adsorption of reactants onto the photocatalyst surface is the initial step of the catalytic process, and that the reaction can occur only between adsorbed species.

For the comparison, a table that summarizes the results from numerous studies on photocatalytic degradation using various LDH-based materials is presented in Table 3.

Table 3. Summary of pollutant photodegradation by LDH-based materials.

LDH	Pollutant	Reaction Kinetics	Light Source	Energy Band Gap (eV)	Ref.
ZnAl LDH	Rhodamine B	PFO	UV light	-	[49]
ZnCoFe LDH	Methylene blue	PFO	Visible light	2.14	[100]
g-C ₃ N ₄ /NiAl-LDH/CeO ₂ NC	Rhodamine B	PFO	UV light	2.54	[55]
Mg–Al LDH@g-C ₃ N ₄ @Ag ₃ PO ₄ NC	Methylene blue	-	Visible light	2.69	[107]
ZnCr mixed oxides	Methylene blue, Brilliant cresyl blue	PFO	Simulated solar irradiation	-	[4]
ZnCr 500	Crystal violet	PFO	Simulated solar irradiation	1.98	[39]
ZnFe mixed oxides	Methylene blue	PFO	Simulated solar irradiation	-	[44]
mixed oxides ZnO/TiO ₂ /CeO ₂ /Al ₂ O ₃	Methylene blue, Methyl orange	PFO	UV light	3.19	[108]
TiO ₂ —ZnAl LDH	Rhodamine B	PFO	Solar light	-	[52]
NiAl-LDH/Polyoxometalate	Malachite green	PFO	UV light	3.22	[109]
ZnCr LDH/rGO	Rifampicin	-	Visible light	-	[8]
ZnFe mixed metal oxides	Ibuprofen	PFO	Simulated solar irradiation	-	[110]
mixed oxides ZnO/TiO ₂ /CeO ₂ /Al ₂ O ₃	Paracetamol	PFO	UV light	3.19	[108]
ZnAl LDH, ZnAl mixed oxide	Tolperisone hydrochloride	-	UV light	-	[111]
MgAl LDH/MOF	Diclofenac	PFO	Visible light	4.02	[112]
CuMgAlTi—LDH	Polyethylene and polystyrene microplastics	-	Visible light	2.82	[113]

4.2.1. Photocatalytic Degradation of Dyes

As mentioned before, dye-polluted wastewater is of great concern in environmental protection. Therefore, numerous research studies on photodegradation of cationic azo dyes, such as Methylene blue, Rhodamine B, Crystal violet, and Brilliant cresyl blue, have been conducted [4,39,44,49,52,55,100,107,108].

Wu et al. investigated Zn–Al LDHs with high crystallinity and demonstrated that the photocatalytic performance of Rhodamine B strongly depends on the Zn/Al molar ratio, with an optimal ratio of 3 achieving up to 97.7% degradation efficiency. The photocatalytic performance of the catalyst was strongly influenced by the solution pH. The degradation efficiency increased with pH from acidic to neutral conditions, reaching a maximum at pH 7, and then decreased under more alkaline conditions. This trend is closely related to the behavior of electron–hole pairs in the catalyst. In alkaline environments, although abundant hydroxyl groups are available to react with metal sites in the hydrotalcite, excessive OH[−] can hinder electron capture by the metal atoms, promoting recombination of electrons and holes and reducing activity [114]. Conversely, in acidic conditions, hydrogen ions can scavenge hydroxyl radicals and partially dissolve the catalyst, also limiting degradation. Therefore, it was concluded that a neutral environment provides the optimal balance,

maximizing the generation and utilization of reactive species and leading to the highest photocatalytic efficiency. Even though high photocatalytic activity was mainly achieved by the photo-generated holes from LDH photocatalysts, the authors stated that the generated $O_2^{\bullet-}$ and $\bullet OH$ species also contributed to the photodegradation in the following order: $h^+ > O_2^{\bullet-} > \bullet OH$ [49]. Moreover, in a different study ternary ZnCoFe LDH was reported to exhibit visible-light activity towards photodegradation of Methylene blue and its high photocatalytic activity was attributed to the production of radical species $OH^\bullet$ and $O_2^{\bullet-}$. However, in this study, the pH value did not have much impact on degradation efficiency and the photocatalyst was deemed efficient in a wide pH range (3–8) [100].

Mixed oxides derived from LDHs exhibit great potential for successful application in photocatalytic degradation of dyes in wastewater thanks to their favorable textural (usually large surface area and wide pore size distribution) and structural (presence of photocatalytic active phases, i.e., ZnO) properties. Several studies emphasized that calcination of ZnCr LDH improves the photocatalytic efficiency for degradation of various dyes such as Crystal violet, Methylene blue, Orange II and Brilliant cresyl blue, as a result of the formation of photocatalytic active phases [4,24,39,44]. Thermal treatment at temperatures higher than 500 °C triggered heterojunctions between crystal phases that further promote photocatalytic efficiency. The presence of hydroxyl radicals and photogenerated holes is considered crucial for photocatalytic mechanisms of wastewater purification [4,24,39,44]. It was also noted that pH value can have a great effect on photocatalytic process, depending on the type of dye used. The difference in photocatalytic degradation between Methylene blue and Brilliant cresyl blue on ZnCr calcined at 900 °C is mainly governed by pH-dependent surface charge effects. Methylene blue solution, being more alkaline than its pK_a , carries a negative charge that promotes strong electrostatic attraction to the catalyst, enhancing adsorption and photodegradation. In contrast, Brilliant cresyl blue, with a nearly neutral pH relative to its pK_a , interacts weakly with the catalyst surface. Consequently, Methylene blue shows higher photocatalytic efficiency compared to Brilliant cresyl blue under the same conditions [4].

Combinations of different constituent metal salts during synthesis of LDH can lead to formation of very photocatalytically efficient mixed oxides, as demonstrated in the study by Castro et al., where quaternary mixed oxides successfully degraded 95% of Methylene blue and 99.3% of Methyl orange. High efficiency of the obtained photocatalysts is ascribed to the effective charge separation that is caused by synergistic interactions between mixed oxide phases in the photocatalyst [108]. Furthermore, relatively high reusability of LDH-based mixed oxides was observed in several studies, indicating a great potential for scale-up experiments [4,39,108].

4.2.2. Photocatalytic Degradation of Pharmaceuticals

With the increasing consumption of pharmaceuticals, drug contamination in wastewater has emerged as a significant environmental challenge [115]. The photodegradation of ibuprofen, as one of the most commonly used drugs, was investigated by Di et al., using ZnFe mixed metal oxides (MMOs) under simulated solar light irradiation. The photocatalytic performance of ZnFe-MMOs was strongly influenced by Zn/Fe molar ratio and calcination temperature. Increasing the Zn/Fe ratio improved Ibuprofen photodegradation, while calcination at 300 °C triggered formation of ZnFe-MMOs with interconnected micropores, laminar structure, and high metal oxide dispersion, resulting in the highest rate constant. Lower calcination temperatures led to less efficient catalysts, whereas higher temperatures caused layer collapse, reduced surface area, and decreased light absorption, diminishing photocatalytic activity. Consequently, ZnFe-MMOs with a Zn/Fe ratio of 4 calcined at 300 °C were selected as the optimal photocatalysts in the study. The mechanism of

the photodegradation reaction indicates that photogenerated holes govern the process [110]. Paracetamol, also a very common drug, was partially degraded (60%) in a photocatalytic reaction with quaternary ZnO/TiO₂/CeO₂/Al₂O₃ mixed metal oxides under UV light irradiation, in a study by Castro et al. The synergistic interactions among the incorporated oxides promote efficient charge separation and transfer, thereby improving the overall photocatalytic performance [108]. ZnAl LDH and derived mixed oxides exhibited high efficiency towards degradation of muscle relaxant tolperisone hydrochloride under UV light. Despotovic et al. concluded that their photocatalytic activity is a result of favorable textural properties, as well as phase composition and the presence of ZnO phases [111].

Functionalization of LDHs with different agents such as graphene oxide and metal–organic frameworks can improve the photocatalytic efficiency of LDHs. Recent studies emphasize the possibility of achieving high photodegradation efficiency with different graphene-based composites, since graphene oxide (GO) and reduced graphene oxide (rGO) can improve light absorption of LDH, as well as adsorption of dye molecules on the photocatalyst surface [8,97,116,117]. The study by Sadeghi Rad et al. showed that the combination of ZnCr LDH with GO and rGO significantly improved the rifampicine photocatalytic degradation efficiency, increasing it from 31.2% for ZnCr LDH to 36.6% for ZnCr LDH/GO and 39.5% for ZnCr LDH/rGO. This enhancement can be attributed primarily to more efficient separation of photogenerated electron–hole pairs, as both GO and rGO facilitate charge transfer and inhibit fast recombination. Additionally, the incorporation of GO and rGO reduces aggregation of the LDH nanosheets, increasing the number of available reactive sites for pollutant degradation. Since adsorption of pollutant molecules on the catalyst surface is a prerequisite for photocatalytic activity, the higher specific surface area of the composites further contributes to their improved performance. The superior efficiency of ZnCr LDH/rGO compared to ZnCr LDH/GO is likely due to the higher electrical conductivity of rGO, which enhances charge mobility and overall photocatalytic activity [8]. Furthermore, diclofenac was efficiently degraded (100% after 20 min) under visible light with a MgAl LDH-functionalized composite with a Zr-based organic framework. The study indicated that photogenerated holes are dominant active species and the reaction followed a radical pathway [112].

4.2.3. Photocatalytic Degradation and Photoaging of Microplastic

The use of photocatalysis to degrade or transform microplastic into smaller, less persistent molecules is gaining considerable attention as a sustainable strategy for plastic pollution control. LDH-based materials and their derived mixed oxides offer unique advantages for this purpose, including tunable energy band gap, high surface area and the ability to generate ROS under light irradiation. In a pioneering study, Jiang et al. [113] synthesized a quaternary CuMgAlTi-LDH photocatalyst capable of degrading polyethylene (PE) and polystyrene (PS) microplastic under visible light. The composite induced significant reductions in particle size (by 33–54%) and surface oxidation of MPs after prolonged irradiation, producing oxygen-containing intermediates (ketones, aldehydes, esters) and visible morphological damage confirmed by FTIR, SEM, and GC–MS analyses. Hydroxyl (•OH) and superoxide (•O₂[−]) radicals were identified as the dominant active species responsible for polymer chain scission. This study demonstrated, for the first time, the feasibility of visible-light-driven photocatalytic oxidation of microplastic using LDH-based materials. Su et al. [118] revealed that MgAl-LDH accelerated the photooxidation of polyethylene microplastic under simulated sunlight, increasing surface oxidation and fragmentation through ROS generation. Both experimental results and density functional theory (DFT) calculations indicated that van der Waals and electrostatic interactions between LDH and polymer surfaces facilitated ROS attack on specific C–H and C–C bonds, thereby enhancing

degradation. These findings not only expand the understanding of MP aging mechanisms, but also suggest potential applications of LDHs as natural photoactive material in the environment. More advanced photocatalytic systems are emerging through composition and defect engineering. Wu et al. [119] designed a high-entropy CoNiFe(VZn–Al)-LDH for peroxymonosulfate (PMS)-assisted photocatalytic degradation of various microplastics. The creation of high-valence metal sites ($\text{Fe}^{3+\delta}$, $\text{Co}^{2+\epsilon}$, $\text{Ni}^{2+\zeta}$) significantly enhanced solar absorption and charge separation, lowering the energy barrier for PMS activation and improving ROS generation. This atomic-level design strategy provides a new path for developing LDH-based photocatalysts capable of treating plastic waste under mild conditions. Collectively, these studies demonstrate that LDH-derived photocatalysts are not only capable of promoting photoaging and oxidative breakdown of microplastic, but also open pathways toward combined remediation and resource recovery. Future research should focus on improving degradation kinetics, understanding byproduct formation, and designing visible-light-active, stable composites for large-scale water treatment applications. Recent advances in the adsorption and photocatalytic degradation of micro- and nanoplastic reveal the vast potential of LDH-based materials as multifunctional tools for plastic pollution mitigation. Adsorption studies highlight their tunable surface chemistry and structural stability, while photocatalytic approaches demonstrate their capacity to initiate polymer oxidation and fragmentation under visible light. Continued development of hierarchical, magnetic, and defect-engineered LDH composites, coupled with in-depth mechanistic investigations, will be key to achieving scalable, sustainable solutions for the removal and degradation of plastic debris from aquatic systems.

4.3. Photocatalytically Induced Antimicrobial Activity

In addition to their well-established functions in adsorption and photocatalytic pollutant degradation, LDH-based materials have recently emerged as promising agents for microbial control in wastewater treatment. Thanks to their flexible structure with adjustable metal content, the ability to exchange anions between layers, and a large surface area, LDH-based materials have been successfully developed to combine strong antimicrobial effects with efficient photocatalytic function. These dual or even multifunctional characteristics position LDHs as valuable candidates in sustainable water purification technologies, especially where both chemical and microbiological pollutants coexist [14,120].

Based on available literature and conducted studies, the mechanism of photocatalytically induced antibacterial activity can be proposed [4,121–123]. The process, presented in Figure 13, is induced by adsorption of photons that have appropriate energy (sufficient for an electron to jump from the valence band to the conduction band) by the photocatalyst, initiating a chain reaction that generates ROS through reactions with water and oxygen from the air. Photocatalysts attach to the convoluted bacterial cell wall via electrostatic attraction, hydrophobic interactions, or van der Waals forces, after which the generated ROS attack the cell membrane and disrupt basal metabolism. However, the exact reason for bacterial cell death is still unknown, but it is assumed to involve one of the following mechanisms:

1. Oxidative stress: Free radicals produced by photocatalysis initiate the oxidation of membrane lipids causing the damage to proteins and mRNA [121,124]. The ultimate result is lipid peroxidation and protein denaturation due to permanent damage to nucleic acids. The crystallinity and the phase composition of photocatalysts can significantly affect their photocatalytic properties, such as the ability to generate higher amounts of ROS in the presence of sunlight, leading to oxidative stress in the cell [121–123]. When exposed to UV or visible light, LDH-based composites coupled with semiconductors such as TiO_2 , ZnO , $\text{g-C}_3\text{N}_4$, or Ag nanoparticles are capable of

generating ROS that inflict oxidative damage on microbial membranes, nucleic acids, and intracellular proteins, leading to rapid inactivation of pathogens [4,122]. In such systems, the LDH structure is crucial for separating photogenerated charge carriers, prolonging their lifetimes and increasing ROS yield.

2. Release of metal ions from photocatalysts: Metal ions penetrate the cell membrane and react with -SH, -NH, and -COOH groups of nucleic acids and proteins, causing damage. In systems based on Zn, Cu, Co, or Fe, divalent or trivalent metal ions are gradually released into the aqueous environment, disrupting essential microbial processes such as membrane transport, enzyme activity and oxidative balance [125]. Zn^{2+} ions are recognized for their antibacterial properties, which include disrupting the integrity of the cell membrane and enzyme functions [126,127]. However, this mechanism is insufficiently studied and is mostly mentioned in the literature as an additional reason for cell death. Elrafey et al. synthesized a NiFe LDH and evaluated its bactericidal activity against *Proteus mirabilis*, *Salmonella Typhimurium*, *Escherichia coli*, *Pseudomonas aeruginosa* and *Klebsiella pneumoniae*. They reported that Fe- and Ni-containing LDHs strongly interact with bacterial proteins, disrupting regulated transport across the plasma membrane by altering its permeability, which triggers a stress response in the transport system and ultimately leads to bacterial death [128]. ZnFe-LDHs synthesized via co-precipitation and intercalated with nitrate have been reported to eliminate both *Escherichia coli* and *Staphylococcus aureus* effectively, while simultaneously removing heavy metals and organic dyes from solution [129]. Similarly, CoFe-LDHs treated with gamma irradiation showed notable antibacterial activity coupled with significant removal of Malachite green and Methylene blue dyes [130].

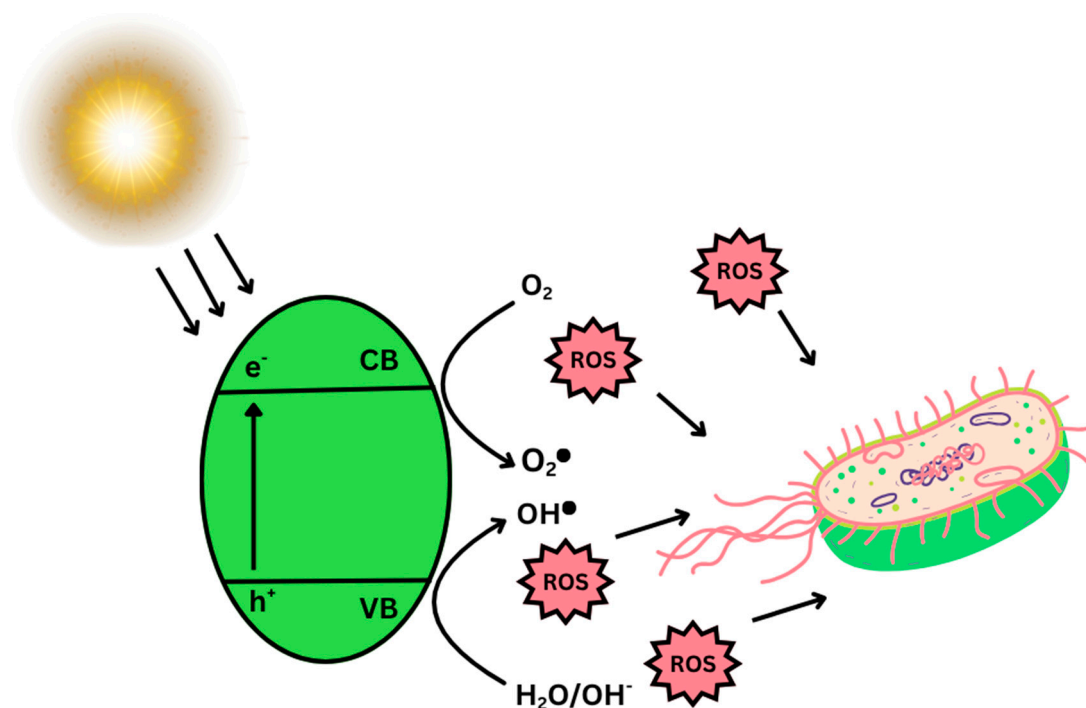


Figure 13. Proposed mechanism of photocatalytically induced antibacterial activity of LDH-based materials.

Several recent studies provide strong evidence for the effectiveness of light-activated antimicrobial LDH systems. Kotta et al. [131] synthesized silver nanoparticle-embedded LDH nanohybrids, which demonstrated significant antibacterial and antifungal activity under UV illumination. Similarly, Cardinale et al. [14] investigated a ZnAl-SO₄ LDH composite after removal of Cr(VI) from wastewater and found that it retained its antimicrobial

and photocatalytic functions under visible light, highlighting its potential for continuous dual-function applications in wastewater matrices (Table 4). Balcik et al. reported that the incorporation of ZnFeCe LDH into membrane enabled antibacterial activity, effectively inhibiting *Escherichia coli* [132].

To enhance visible-light responsiveness and overall efficiency, several engineering strategies have been explored. These include metal doping, heterojunction formation, and surface functionalization with plasmonic or carbon-based materials. For example, Fe-doped NiAl-LDH films deposited on conductive substrates and subsequently modified with Ag nanoparticles showed superior photocatalytic degradation of Methyl orange and enhanced microbial inactivation through improved charge separation and ROS generation [133]. Dual functionality, simultaneous dye degradation and bacterial inhibition, enhances treatment efficiency and safety, making it advantageous for environmental remediation and public health purposes. The NiAl-LDH/Cu-MOF photocatalyst showed dual functionality: degradation of Methyl orange dye and antibacterial activity against Gram-positive (*Staphylococcus aureus*, *Enterococcus faecalis*) and Gram-negative (*Escherichia coli*, *Pseudomonas fluorescens*) bacteria [134]. In another recent study, a Cu-cyanoguanidine-ZnCoFe/LDH composite achieved approximately 90% removal of the antibiotic levofloxacin from simulated wastewater, while simultaneously exhibiting broad-spectrum antibacterial activity and low cytotoxicity in mammalian cells [67]. This dual function is particularly relevant for pharmaceutical wastewater, where both residual drugs and resistant bacteria pose serious environmental risks.

Importantly, light-activated LDH systems offer several advantages over conventional disinfectants. The ability to activate antimicrobial function on-demand using natural or artificial light minimizes long-term leaching of biocidal ions, reducing environmental toxicity. Oxidative stress generated by ROS leads to non-specific cellular damage, which significantly lowers the risk of resistance development compared to traditional antibiotics or disinfectants [67,129]. These attributes make photocatalytic LDHs especially attractive for decentralized water treatment systems where energy input must be minimal and reliable disinfection is essential.

Despite these promising developments, several challenges remain to be addressed before widespread application. In real wastewater matrices, natural organic matter, suspended solids, and high ionic strength can inhibit both microbial contact with the active surface and the propagation of ROS, reducing disinfection efficacy [135]. The reusability and regeneration of antimicrobial activity over multiple cycles has also been poorly studied. Additionally, while silver- and copper-enhanced LDH systems show excellent activity, their cost and potential for secondary pollution through metal leaching must be carefully evaluated, especially for use in large-scale systems.

Table 4. Applications of LDH-based materials for photo-induced antimicrobial effect with suggested mechanisms and performance of used materials.

LDH	Light Source	Microorganisms	Antimicrobial Mechanism	Performance	Ref.
Zn-Fe LDH	Ambient light (field conditions)	<i>E. coli</i> , <i>S. aureus</i> , total coliforms	Bactericidal surface interactions, possible ROS activity under ambient light	High disinfection efficiency (>90%) of real wastewater; performance robust in complex matrix	[129]

Table 4. Cont.

LDH	Light Source	Microorganisms	Antimicrobial Mechanism	Performance	Ref.
Co-Fe LDH nanosheets	Visible light	<i>E. coli</i> , <i>S. aureus</i>	Synergistic effect of gamma-induced structural changes and ROS generation; enhanced surface reactivity	Gamma irradiation increased antibacterial activity and dye removal; improved magnetization and dielectric properties	[130]
ZnAl-SO ₄ LDH	Solar-simulated light	<i>E. coli</i> , <i>S. aureus</i>	ROS generation ($\bullet\text{OH}$, $\bullet\text{O}_2^-$) from photoactivated ZnO-like phases; surface interactions	Thermal annealing improved both antimicrobial and photocatalytic activity; post-treatment materials showed higher bacterial growth inhibition	[14]
Ag-LDH embedded in polymeric hydrogel	Visible light (simulated daylight)	<i>E. coli</i> , <i>S. aureus</i> , <i>P. aeruginosa</i> , <i>Candida albicans</i>	Sustained Ag ⁺ ion release from LDH matrix; ROS generation under visible light; enhanced contact killing via hydrogel network	Strong antimicrobial activity against Gram-positive, Gram-negative, and fungal strains; hydrogel improved dispersion and release control	[131]
ZnCr LDH mixed oxides	Simulated solar irradiation	<i>E. coli</i> , <i>S. aureus</i>	ROS-mediated cell damage	ZnCr900 achieved ~100% bacterial growth inhibition and complete dye removal; activity enhanced by high crystallinity and optimal band structure	[4]
ZnO nanoparticles (various sizes)	Ambient visible light (no UV lamp)	<i>S. aureus</i> , <i>E. coli</i> , other clinical isolates	ROS production (H_2O_2 , $\bullet\text{OH}$, $^1\text{O}_2$); direct contact cell wall/membrane disruption;	Smaller particles led to higher antibacterial activity; broad-spectrum effect without UV pre-activation	[136]
ZnO nanomaterials (different morphologies)	Light conditions not specified for antimicrobial tests; photocatalysis under visible light	<i>B. manliponensis</i> , <i>M. luteus</i> , <i>S. aureus</i> , <i>E. coli</i>	$\bullet\text{O}_2^-$ radical generation > $\bullet\text{OH}$; ROS-induced oxidative stress	ZnO-4M had strongest antibacterial and photocatalytic activity; morphology less critical than ROS generation profile	[137]
NiAl-LDH/Cu-MOF	Solar light	<i>S. aureus</i> , <i>E. faecalis</i> , <i>E. coli</i> , <i>P. fluorescens</i>	ROS-mediated cell damage	Strong antimicrobial activity against bacteria, inhibition zone between 12 and 14 mm, very efficient ROS generation, high surface area, and stability	[134]

The resistance of microbial biofilms to LDH systems represents another critical point. Most published studies evaluate antimicrobial efficiency using planktonic bacterial strains under controlled laboratory conditions. There are just a few studies employing LDH systems to combat biofilm-forming pathogens. For example, Omonmhenle and Ifijen [15] have reported that curcuminoid-incorporated LDHs (CC-LDH) reduced biofilm formation by *Pseudomonas aeruginosa*, *Staphylococcus aureus* (MRSA) and *Candida albicans* by 54–58% compared to pure curcuminoids, as shown by broth dilution and in vitro biofilm assays. However, the behavior of biofilm-forming pathogens and microbial consortia in real ecosystems in the presence of LDH materials remains largely unexplored, despite their relevance in real-world treatment settings. It should be noted that long-term photostability and performance of these materials under solar-driven or low-intensity visible light systems require more systematic investigation.

Future research should therefore prioritize the development of biofilm-targeted LDH composites, integration with natural supports such as chitosan or alginate to improve biocompatibility, and the design of solar-driven photocatalytic reactors capable of simultaneously removing chemical pollutants and inactivating pathogens. Real wastewater studies, continuous-flow evaluations, and life-cycle assessments will also be essential to confirm the long-term stability, environmental safety, and economic viability of these materials. With continued material innovation and a stronger focus on real environment validation, these multifunctional systems hold significant promise as effective and eco-friendly solutions for integrated wastewater management.

5. Future Perspectives and Conclusions

LDH-based materials have advanced from laboratory interests to highly promising multifunctional systems for wastewater remediation. Their structural tunability, compositional flexibility and transformation into mixed oxides with superior textural and electronic properties have broadened their potential in adsorption, photocatalysis and antimicrobial applications. Despite the notable progress achieved in the synthesis, modification and application of LDH-based materials for wastewater treatment, several scientific and practical challenges remain before these materials can be implemented at an industrial scale. In order to accelerate into low-cost and environmentally safe technologies, future efforts must therefore be focused on advancing several complementary aspects of LDH-based systems, described below.

- The development of LDH materials with tailored properties requires a more detailed insight into the relationship between composition, microstructure and performance. Future research should prioritize controlled synthesis at the nanoscale, enabling fine-tuning of metal cation ratios, interlayer structure, and defect sites. Coupling LDHs with semiconductors, carbon-based materials, or other metals of interest could enhance textural properties and charge separation and extend light absorption into the visible and near-infrared regions. This would improve not only the photocatalytic efficiency under natural sunlight, but also adsorption capacity for targeted pollutants. Therefore, by controlled functional modification of pure-phase LDHs through interlayer variation, surface compounding, and calcination LDH-based materials can effectively remove emerging contaminants from water, owing to their obtained excellent electronic structures and crystal structures. Also, complex synthesis of hybrid materials such as metal–organic frameworks (MOFs) and LDHs should be thoroughly investigated and promoted considering that this combination of systems highly influences structural and functional properties, favorable particularly in catalysis and environmental remediation. Future study should also be directed to research on the relationship between structural properties and functional capabilities of these hybrid

materials, forwarding the design of more efficient and versatile materials for desired applications. Additionally, 2D/2D heterojunctions and core-shell nanostructures represent promising systems for achieving synergistic adsorption, photocatalytic and antimicrobial effects. Understanding the correlation between structure, electronic properties and performance will be essential for optimizing multifunctional efficiency.

- In order to fully elucidate the multifunctional behavior of LDH-based materials in pollutant removal, there is a general consensus that a more comprehensive understanding of their adsorption and photocatalytic mechanisms is required. Despite extensive studies, the processes governing adsorption dynamics, electron transfer, radical formation and active-site evolution remain only partially understood, highlighting the need for advanced and integrated characterization approaches.
- The reversible transformation between LDHs, their mixed oxides, and the rehydrated forms, defined as the memory effect, is a unique feature that allows fine-tuning of surface area, porosity and the distribution of active sites, while also enabling material regeneration. However, as mentioned in the review, this behavior is sensitive to calcination temperature, as excessive heating can cause irreversible phase transitions that diminish the material's ability to recover its layered structure. To ensure long-term stability and consistent performance, future studies should focus on mapping out the temperature and compositional reconstruction windows across different metal systems and correlate them to catalytic and adsorptive durability. The effect of incorporating structure-stabilizing dopants, promoters, or protective surface layers should be further investigated because it may widen the thermal tolerance and prevent degradation during repeated use. This in-depth research would contribute to mild regeneration protocols that could lead to activity resilience and stability over multiple operational cycles. A deeper understanding of the memory effect in mixed oxides will be essential for designing LDH-based materials with improved resilience, recyclability, and sustained efficiency in practical applications.
- A promising route for future research should also be directed to the development of multifunctional LDH-based materials efficient in pollutant degradation, antimicrobial protection, and resource recovery. As emphasized in the review, because of their structural versatility, LDHs can effectively couple adsorption, photocatalysis, and light-driven antimicrobial action through ion release and reactive oxygen species generation. Therefore, further work should be focused on future materials engineered for targeted multifunctionality, such as heterojunction LDH/semiconductor/carbon hybrids that enhance charge separation, offer efficient anion-exchange sites for contaminant capture, and allow controlled metal release for microbial control. Combining these effects would increase their environmental performance and versatility.
- Emerging multifunctional LDH-based systems that combine pollutant degradation, antimicrobial protection and resource recovery represent a particularly promising research direction. Recent studies have shown their capability to degrade persistent contaminants such as pharmaceuticals, per- and polyfluoroalkyl substances (PFAS), and even micro- and nanoplastic. These systems can also provide photo-induced antibacterial activity through ROS generation and controlled ion release, offering integrated approaches for chemical and biological pollutant removal. The next generation of LDH-based materials should thus be engineered for targeted multifunctionality that would be capable not only of removing pollutants, but also contributing to circular resource flows.
- Considering the environmental motivation of wastewater remediation, the synthesis of LDH-based materials should highly consider sustainable, low-waste, and resource-efficient approaches. Upcoming research should emphasize on developing green

synthesis routes that would replace hazardous reagents with eco-friendly solvents, while also incorporating waste-derived precursors such as industrial residues, slags, and biomass ashes. These strategies not only reduce production costs and the environmental footprint but also align LDH manufacturing with circular economy principles. Recent studies have shown that high-performance LDHs can be successfully obtained from industrial by-products, demonstrating both feasibility and efficiency. However, to ensure true sustainability, future work must include assessments concerning economic aspects and life-cycle analyses (LCA) to evaluate scalability, energy demand, and environmental impact, thereby guiding the transition from laboratory-scale synthesis to practical, green production of LDH materials.

- One of the main obstacles to advancing LDH-based technologies is the lack of standardized testing and reporting practices. Numerous studies report results with differences in experimental procedure, such as light sources, pollutant concentrations, solution composition, and regeneration protocols, limiting the possibility to compare results among published studies. In order to enable meaningful evaluation and accelerate progress, the research community should establish unified testing standards that include experiments with real wastewater effluents and consistent reporting of key parameters that influence the efficiency of LDH-based materials. This would lead to more reliable cross-study comparisons and faster innovation.

In summary, LDH-based materials represent one of the most promising next-generation multifunctional systems for sustainable wastewater treatment. Their tunable composition, structural versatility, and ability to transform into highly active mixed oxides enhances their efficiency in pollutant removal, antimicrobial activity and resource recovery. Nevertheless, the research focus must be shifted from laboratory-scale studies to practical applications that will require an interdisciplinary approach correlating material properties, environmental engineering, and process design with green synthesis routes as well as with thorough assessments concerning circular economy and environmental aspects. Correspondingly, pilot-scale research and evaluation of environmental and economic performance are crucial to confirm long-term stability and feasibility. By combining mechanistic insight with green synthesis, standardized evaluation and pilot-scale validation, LDH-based technologies can evolve into durable, cost-effective, and environmentally responsible solutions for the global challenge of wastewater purification.

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