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Article

Uniqueness of Layered Double Hydroxide Materials: A Critical Review of Synthesis Methods, Properties, Composites, and Remediation Mechanism

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Abstract

Layered Double Hydroxides (LDH) are anionic clays with tunable structures, high surface areas, and versatile ion-exchange properties, making them promising for environmental remediation. This review highlights recent advances in LDH synthesis methods, structural properties, composite design, and remediation mechanisms. Synthetic techniques such as coprecipitation, hydrothermal treatment, sol-gel, and urea hydrolysis are evaluated for their impact on morphology, stability, and functionality. Integration of LDH with carbon materials, metal oxides, polymers, and MOFs improves their performance in environmental remediation. A key theme is the "memory effect" of LDH, which enables reversible structural transformations, enhancing ion-exchange and adsorption capacities. The pollutant removal mechanisms involve ion exchange, adsorption, and photocatalysis. Current challenges include scalability and regeneration. Sustainable synthesis methods and characterization of multifunctional composites are discussed to advance the development of LDH for efficient water and wastewater treatment.

Keywords: *Layered Double Hydroxides (LDH); LDH composites; Advanced materials; Properties modification; Enviromental remediation*

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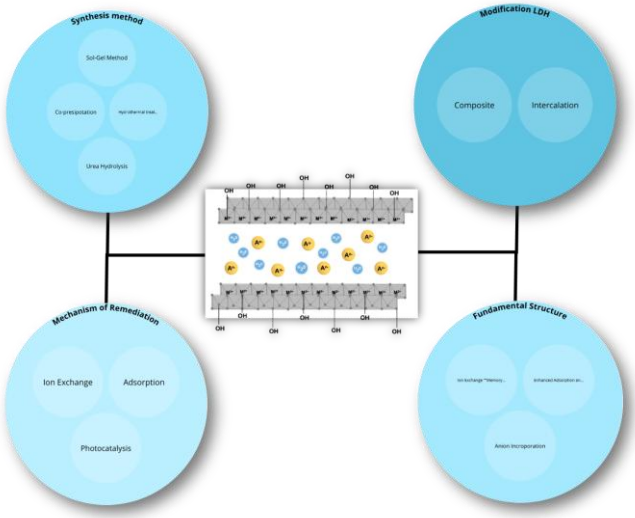
Graphical Abstract



Introduction

Layered double hydroxides (LDH), also known as anionic clay materials, have attracted attention due to their distinctive properties. Harnessing its distinctive properties, LDH has the potential to be applied in the environment, energy, and biomedical. LDH are commonly synthesized using inorganic salts and the addition of water or organic solvents. Achieving optimal results necessitates precise control of both pH and temperature to induce precipitation and crystal growth [1]. The LDH formed under these controlled conditions exhibit a distinctive 2D layered structure [2]. Layered LDH structures, synthesized through a combination of multi-metal compositions and interlayer anions, can confer desirable properties such as enhanced surface area, phase purity, porosity, and crystallinity [11–13]. Furthermore, LDH showcase unique characteristics, including high chemical stability, ion exchange capability, structure memory effect, reactive interlayer space, catalytic activity, specific surface area, biocompatibility, and modifiable hydroxide layer or structural interlayer composition [12,14–16].

The characteristics of LDH make them highly promising for applications as catalysts and adsorbents in wastewater treatment, owing to their straightforward preparation process. Beyond their role in wastewater treatment, LDH find diverse applications in photocatalysis, CO_2 capture, oxygen/hydrogen evolution, energy storage, coatings, biomedical applications, and supercapacitors [18–22]. In addition to their effectiveness in wastewater treatment, LDH demonstrate excellent adsorption capabilities for various inorganic and organic pollutants, including dyes, metals, pesticides, phosphates, and CO_2 [14].

Numerous reports in the literature delve into the modification of LDH, employing various techniques. Intercalation methods, exemplified by Xu et al. [15] and Chen et al. [16], and composite approaches, researched by Gu et al. [10], Jiang et al. [7], and Kumari et al. [17] represent key strategies in research development. In a specific application, Mg-Al LDH were modified by intercalating nitrate ions into the LDH interlayer, serving as an effective adsorbent for removing methyl orange dyes [18].

Based on the research by Elanchezhiyan & Meenakshi [19] synthesized a chitosan/Mg-Al composite using the coprecipitation method in a separate study focused on enhancing adsorption capacity. When employed as an adsorbent for removing oil particles from oil-in-water emulsions, this composite exhibited a remarkable adsorption capacity of 78%, a substantial improvement compared to LDH (30%). The mechanism of the adsorption was attributed to hydrophobic-hydrophobic interactions. In summary, these findings highlight the diverse modification techniques for LDH and showcase the promising application of the chitosan/Mg-Al composite for efficient oil particle removal through enhanced adsorption capacity.

This article explores the unique structural and chemical characteristics of LDH materials, which make them highly adaptable for various environmental applications. By selecting appropriate synthesis methods, researchers can fine-tune the morphology and interlayer chemistry of LDH and create multifunctional composites with tailored properties. These advancements open up opportunities to improve structural stability, increase surface area, and boost pollutant removal efficiency. However, most current studies are still conducted under controlled laboratory settings and have not yet been widely tested on real wastewater systems. Moving forward, research should aim to build a clearer understanding of structure–function relationships, adopt sustainable and low-cost strategies, evaluate performance under realistic environmental conditions, and develop regenerable hierarchical composites that can be applied on a larger scale.

Fundamental Structure and Chemistry of LDH

LDH, often referred to as multi-metal clay materials and exhibit layered structures resembling hydrotalcite/brucite in 2D. LDH consist of divalent and trivalent metal cations, with the general formula 1.

$$[M^{II}_{1-x}M^{III}x(OH)_2]^{x+}\cdot[An^{-}x/n.mH_2O] \quad \ldots\ldots\ldots (1)$$

where, M^{II} represents divalent metallic cations (e.g., Ni, Ca, Mg, Zn, Cu), M^{III} includes trivalent metallic cations (e.g., Al, Fe, Cr), and An- indicating negatively charged interlayer LDH anions (figure 1) [1–5]. The octahedral layers, adopting a brucite-like structure reminiscent of $Mg(OH)_2$, form the fundamental framework of LDH. These positively charged layers are stacked with interlayer spaces containing charge-compensating anions and water molecules. A variety of interlayer anions can be incorporated during synthesis, including carbonate, nitrate, chloride, sulfate, or even more complex organic anions [25]. This interplay between the octahedral metal hydroxide layers and the exchangeable interlayer anions gives LDH their tunable physicochemical properties, making them versatile platforms for functional modifications [26-27] .

The structural flexibility enables anion exchange, intercalation, and post-synthesis functionalization, allowing LDH to host a wide range of inorganic and organic species within their interlayer galleries. A distinctive and scientifically significant property of LDH is their structural reconstruction behavior, widely known as the “memory effect”. When LDH are calcined at elevated temperatures (typically 500–800°C), they undergo dehydroxylation and decomposition of interlayer anions, resulting in the formation of mixed metal oxides (LDOs) [28]. Remarkably, when these LDOs are

subsequently exposed to water containing appropriate anions, they can spontaneously reconstruct their original layered structure through a rehydration process.

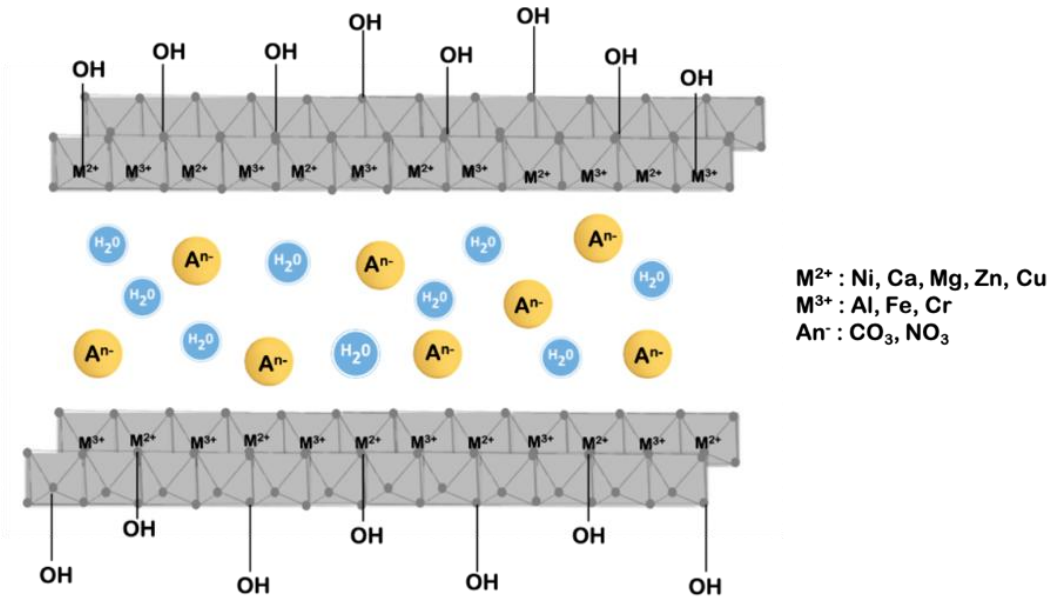


Figure 1. Schematic structure of LDH

This reconstruction phenomenon has profound implications in environmental and catalytic applications:

1. Ion-Exchange Activation through Memory Effect: When LDH is calcined, it transforms into mixed metal oxides (LDO). Upon rehydration in aqueous solutions containing appropriate anions, the LDO can spontaneously reconstruct its original layered structure, a phenomenon known as the memory effect. This reconstructed LDH phase reactivates ion-exchange processes, allowing target cations such as Cd^{2+}, Pb^{2+}, or Cu^{2+} to replace Mg^{2+} within the layers during pollutant removal [29], [30]. Recent studies have shown that this effect significantly enhances the adsorption capacity and regeneration efficiency of LDH-based materials during repeated treatment cycles.

2. Enhanced Adsorption and Regeneration: The reconstruction route also enables efficient incorporation of guest anions including organic molecules, amino acids, and peptides into the LDH interlayer, making this a valuable strategy for synthesizing functional LDH-based composites [31]. Anion Incorporation: The reconstruction route also enables efficient incorporation of guest anions including organic molecules, amino acids, and peptides into the LDH interlayer, making this a valuable strategy for synthesizing functional LDH-based composites.

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Overall, the fundamental structure and chemical versatility of LDH particularly their reconstructive structural behavior are key to understanding their synthesis, modification strategies, and environmental remediation mechanisms discussed in the following sections.

Synthesis Methods

The crystallinity, morphology, layer arrangement, and anion content of LDH are all determined by the synthesis process, and these factors ultimately affect how well they function in different applications. A vast array of synthesis methods, from traditional wet-chemical methods to more environmentally friendly and sustainable approaches, have been developed over the last few decades. Regarding structural control, scalability, cost, and environmental impact, each approach has pros and cons of its own.

Co-presipitation Method

The co-precipitation method is one of the simplest and most commonly employed synthesis techniques for layered double hydroxides (LDH). In this approach, the synthesis is typically carried out under a controlled pH condition, which is adjusted according to the divalent and trivalent metal cations involved. Generally, solutions of divalent and trivalent metal salts are mixed in a molar ratio of 3:1. To maintain a stable pH during synthesis, a sodium hydroxide (NaOH) solution is slowly added to the mixture. According to Bünning et al. [32] and Boulaiche et al. [6], the synthesis involves maintaining a flow of nitrogen or argon gas to minimize the potential presence of carbonates within the LDH interlayer. After the solution preparation, the mixture is heated at 80°C for 20 h, by the procedures outlined [30–32]. Subsequently, the suspension is separated, and the drying process is conducted at 50°C until complete dryness, as described by [36].

The successful synthesis of LDH structures via the co-precipitation method is typically confirmed through X-ray diffraction (XRD) and Fourier-transform infrared spectroscopy (FTIR) analysis as shown in Figure 2 . In studies conducted by Edañol et al. [34], Normah et al. [37] and Wijaya et al. [38], the XRD patterns of Ni/Cr LDH matched the standard hydrotalcite-like phase (JCPDS 52-1626), with characteristic diffraction peaks at 2θ ≈ 11° (003), 23° (006), 35° (015), and 60° (110), indicating the formation of well-ordered layered structures. FTIR spectra exhibited broad O-H stretching bands at 3400-3600 cm^{-1} and a water vibration band at 1630 cm^{-1}, confirming the presence of interlayer hydroxyl groups and water molecules. A band at 1383 cm^{-1} indicated the presence of interlayer NO_3^- anions, while bands in the 500-1000 cm^{-1} region corresponded to M-O-M vibrations (M = Ni, Cr). These features collectively verify the successful formation of NiCr-LDH through co-precipitation.

Amirahmadi et al. [39], emphasized that the co-precipitation method offers several advantages, including the production of LDH with high purity and good crystallinity. However, the formation of well-crystallized hydrotalcite structures or amorphous phases is influenced by several parameters, such as solution concentration, pH, ionic strength, cation concentration, temperature, aging time, and the molar ratio of metal cations. Therefore, careful optimization of these synthesis parameters is essential to obtain LDH with well-defined structures and desirable physicochemical properties.

Hydrothermal Treatment

Because it can produce highly crystalline materials with controlled structural characteristics, the hydrothermal method is often used for the synthesis of layered double hydroxides (LDH). This process typically involves heating a metal precursor solution (M^{2+}/M^{3+}) to temperatures above 100 °C in a Teflon-lined stainless steel autoclave. Since each LDH has an optimal heating time, the temperature and duration

of the hydrothermal treatment significantly affect the crystallinity and crystal size of the LDH. These factors include crystallinity, layer density, and crystal size [20], [40], [41].

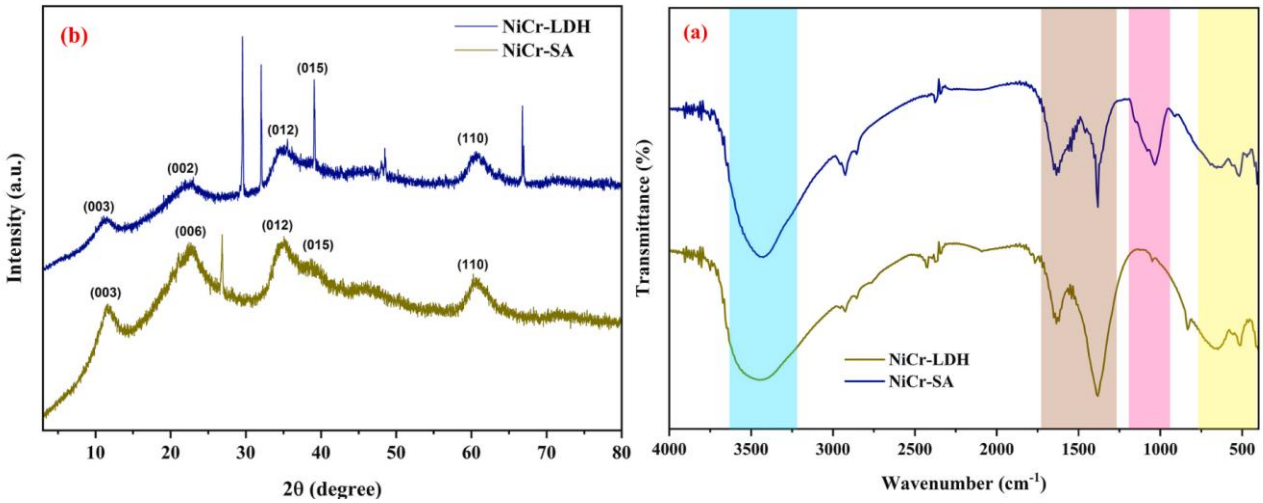

Figure 2. FT-IR Spectra (a), XRD patterns (b) of NiCr-LDH and NiCr-SA. Reproduced with permission from Ref. [38]. Copyright Elsevier, 2025.

LDH formation occurs through the precipitation of divalent and trivalent metal ions, facilitated by adding alkaline compounds such as NaOH, NH_4OH, or urea [42]. The hydrothermal methods pH, crucial in determining the growth of LDH layers, varies with different metal cations, requiring specific pH levels for precipitation [43]. The formation of LDH phases occurs within the pH range of 8.7 to 12. Furthermore, the crystallinity, morphology, and structural properties of the synthesized LDH are strongly influenced by the kinds and molar ratios of metal cations as well as the type of interlayer anions [39–41].

Sol-Gel Method

The sol-gel method is viable for LDH synthesis, offering precise control over crystal formation. A vital advantage of this method is the enhanced specific surface area achieved by creating pores within the particles by elevating their overall porosity. This results in an increased quantity of anions in the interlayers of LDH, consequently bolstering its anion adsorption capacity. Despite these benefits, the sol–gel method is often associated with drawbacks such as a relatively complex experimental procedure and extended synthesis times, which can limit its scalability [47], [48].

In the study conducted by Takanashi et al. [47] the sol–gel method was implemented by initially mixing divalent and trivalent metal cations in a 3:1 molar ratio in solution. After adding citric acid as a complexing agent, the mixture was agitated at 80 °C for an hour. After adding ethylene glycol, the mixture was constantly swirled at 150 °C until all of the solvent had evaporated, creating a viscous gel. To get the finished product, this gel was then calcined at 650 °C for 4 h and dried at 105 °C for twenty-four h. This process shows how the sol-gel method can create LDH materials with specific structural and textural characteristics that are appropriate for cutting-edge uses.

Urea Hydrolysis

The urea hydrolysis method is a well-established approach for synthesizing layered double hydroxides (LDH), offering

advantages such as high crystallinity and uniform particle size distribution .Based on the research results Viscusia et al. [44] and Brahma et al. [45], the detailed synthesis of LDH using the urea hydrolysis method involves mixing magnesium nitrate salt ($Mg(NO_3)_2{\cdot}6H_2O$) as a divalent metal and aluminum nitrate salt ($Al(NO_3)_3{\cdot}9H_2O$) as a trivalent metal in a 1:2 ratio, stirring until homogeneous. Subsequently, a solution comprising 0.035 urea (NH_2CONH_2) and 150 mL deionized water is added continuously to the nitrate salt mixture over 30 min. The pH is adjusted to 10 by introducing a sodium hydroxide (NaOH) solution.

According to Aladpoosh & Montazer. [51], the combined use of urea and NaOH helps prevent the formation of locally inhomogeneous concentrated particles during precipitation, which in turn promotes the formation of LDH with high crystallinity, larger crystal size, and uniform particle size distribution [52]. The homogeneous solution is then transferred into a tightly sealed hydrothermal autoclave and heated in an oven at 110°C for 24 h. Guo et al. [54] reported that the optimal temperature range for LDH synthesis using this method is typically 80-130 °C, with a synthesis time of 4-12 h. Higher synthesis temperatures have been shown to accelerate crystal growth rates in accordance with crystal growth theory. After hydrothermal treatment, the system is allowed to cool to room temperature. The resulting precipitate is then separated by centrifugation and washed repeatedly with deionized water (up to five times) to remove residual ions. Finally, the product is dried at 70 °C for 12 h to obtain the final LDH material [50].

Liu et al. [55] conducted X-ray diffraction (XRD) analysis on ZnAl LDH synthesized via the urea hydrolysis method and observed distinct diffraction peaks corresponding to the (003) and (006) planes, as well as additional peaks for the (012), (015), and (018) planes, indicating a well-defined layered structure. Furthermore, the presence of sharp and symmetrical peaks corresponding to the [110] and [113] planes suggests excellent dispersion of metal ions within the hydroxide layers. These observations are consistent with the findings of Viscusi. [49] and Liu et al. [44] confirming the high crystallinity of ZnAl LDH produced through the urea hydrolysis method.

Overall, the crystallinity, morphology, and functional performance of LDH materials are influenced by the distinct benefits and drawbacks of each synthesis technique. Although co-precipitation is straightforward and scalable, it is extremely sensitive to reaction conditions and pH, which can result in amorphous phases [38], [56]. Although it necessitates longer synthesis times and specialized equipment, the hydrothermal method produces highly crystalline LDH [57] and provides excellent control over layer growth and morphology [58]. Although sol-gel processes are complicated and have limited scalability, they yield materials with high porosity and surface area that are appropriate for adsorption [59]. Although it takes longer to react, urea hydrolysis produces high crystallinity and a uniform particle size distribution

Composite of LDH

In exploring LDH characteristics, structural flexibility emerges as a pivotal factor driving numerous modifications, enabling LDH to take on various forms and structures to achieve optimal conditions. Several vital aspects come into play when considering the modification of LDH characteristics. Firstly, the ease of modification involves altering the structural or interlayer composition, expanding beyond binary metal cation combinations to include more complex ternary and quaternary metal combinations.

Secondly, incorporating various guest anion species between LDH layers is a crucial avenue for modification. This includes a wide range of inorganic anions, such as halides, non-metal oxoanions, polyoxometalate anions, complex anionic transition metal species, and organic anions like biomolecules and polymers [60,61].

Moreover, LDH structural modifications can be precisely customized for specific purposes by employing composite synthesis methods. This involves the strategic selection of support materials for composite formation and the utilization of synthesis methods capable of achieving desired chemical and physical property characteristics, including surface area and basal layer spacing. Research results demonstrate that LDH has undergone diverse modifications by incorporating various carbon types or organic compounds such as cellulose, chitosan, biochar, and graphite. A comprehensive summary of the data from these diverse studies is available in Table 1.

Table 1. Composite of LDH by various research groups.

Materials	Surfaces area (m^2)	Synthesis methods	Typical Application	References
Zn/Al Cellulose	-	Urea Hydrolysis	-	[49]
ZrO_2/MgAl-LDH	-	Urea Hydrolysis	Adsorption	[50]
MgCoAl-CO_3-LDH	55.08	Co-precipitation	Adsorption	[36]
UVSA-ZNAL/PP composite fiber		Co-precipitation	-	[44]
Ni/Al functionalized humic acid and magnetite	62.966	Co-precipitation	Adsorption	[62]
NiCo-LDH/NiFe-LDH	-	Hydrothermal treatment	Catalyst	[11]
Pistachio biochar/$CoFe_2O_4$/Mn-Fe-LDH	-	Co-precipitation	Photocatalytic	[9]
CNFs/ZnAl-LDH Composite films	-		Photothermal	[13]
LDH-Albumin-WO_3 composite	-	Co-precipitation	Coating	[63]
FeMg-LDH@bentonite	-	Co-deposition	Adsorption	[64]
Graphene/LDH	-	-	Energy storage	[10]

Materials	Surfaces area (m^2)	Synthesis methods	Typical Application	References
GO/Mg-Al LDH	-	Hydrothermal treatment	Coating	[65]
NiAl-Biochar	438.942	Co-precipitation	Adsorption	[33]
NiAl-Graphite	21.595	Co-precipitation	Adsorption	[33]
NiCo-LDH/C/GF composite	-	-	-	[66]
LDH nanomaterials	-	-	Photocatalytic	[67]
Fe_3O_4@SiO_2@NiAl-LDH	-	Solvothermal	Biomedical	[39]
Pomelo peel Biochar/MgFe-LDH	20.995	Co-Precipitation	Adsorption	[68]

Numerous studies consistently affirm that LDH composite materials showcase superior characteristics compared to pure LDH. According to Gu et al. [10], graphene/LDH composites display exceptional properties, including high electrical conductivity, mechanical strength, and robust chemical reactivity, facilitating electron transfer. These findings significantly bolster the potential applications of LDH in electrochemistry.

For example, research conducted by Zheng et al. [70] on the hierarchy of NiCo-LDH core/shell structures unveils a notable combination of large surface area and open electrochemical active sites. The study reports a maximum capacity reaching 2640.2 F/g, emphasizing that LDH composite materials exhibit outstanding electrochemical performance, particularly the hierarchy of NiCo-LDH core/shell structures. These findings further fortify the argument for the broad and impactful applications of LDH composites in electrochemistry.

Based on the research by Bian et al. [71], it is evident that MgFe-LDH@biochars stand out by exhibiting the largest specific surface area, coupled with excellent magnetic responsiveness and increased adsorption capacity. This observation aligns with the findings from Fang et al. [72], the enhancement of physicochemical properties in LDH-BC composites was demonstrated through various treatments. These treatments include magnetic treatment, acid treatment, alkali treatment, control of metal ion ratios, LDH intercalation, and calcination processes. Such comprehensive approaches highlight the versatility and potential of these treatments in tailoring LDH-BC composite properties for specific applications [9].

The research highlights the significant influence of the synthesis method on the success of LDH composite formation. Bian et al. [73] reported that employing the co-pyrolysis method in LDH/BC synthesis can significantly enhance adsorption and catalytic performance by generating a larger

surface area. This improvement is attributed to the release of water molecules between layers, which promotes the formation of LDH/BC composites with abundant functional groups, mixed metal oxides, and LDH particles exhibiting high crystallinity and uniform particle size.

SEM analysis, as reported by several researchers, is commonly used to examine the surface morphology of LDH composites. For example, Figure 3 illustrates the typical morphological transformation of rambutan peel-derived carbon (Hc) before and after LDH incorporation [73]. Because lignin and cellulose degrade during carbonization, the carbonized rambutan peel (Figure 3b) displays more spherical and homogeneous particles than the raw peel (Figure 3a), which has a flat surface with uneven particle sizes and rough textures. LDH–Hc composites, including Ni/Al-Hc (c), Cu/Al-Hc (d), and Zn/Al-Hc (e), exhibit a high degree of aggregation along with rough, uneven surface textures, irregular pores, and large pore sizes. These morphological characteristics are advantageous for improving pollutant adsorption and surface reactivity.

To provide a clearer overview of the various LDH composite systems reported in the literature, Table 2 summarizes the types of support materials, synthesis methods, enhanced properties, and their typical applications.

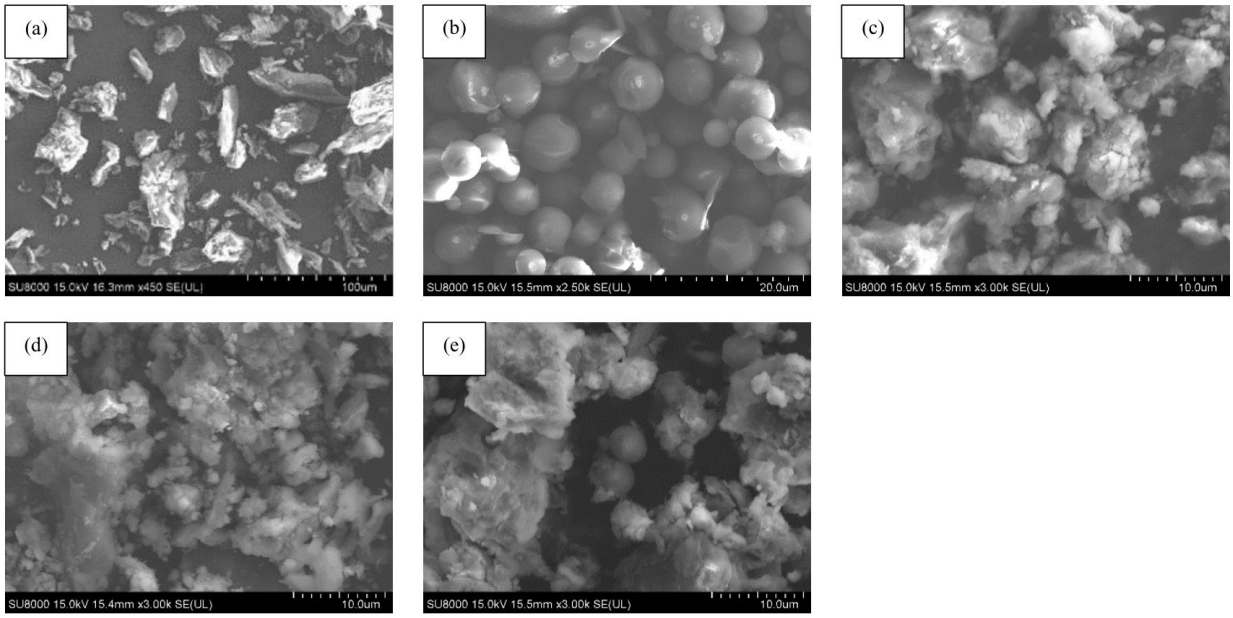

Figure 3. SEM image of Rambutan peel (a), Hc (b), Ni/Al-Hc (c), Cu/Al-Hc (d), and Zn/Al-Hc (e). Reproduced with permission from Ref. [74]. Copyright Environment and Natural Resources Journal 2022.

From Table 2, it is evident that the choice of support material and synthesis method plays a decisive role in determining the structure and functionality of LDH composites. Carbon-based supports, such as graphene, biochar, and cellulose, significantly enhance the surface area [78], introduce abundant functional groups, and improve electron transfer, making them ideal for adsorption and electrochemical applications. Inorganic supports like metal oxides and bentonite contribute to improved thermal stability and catalytic activity, whereas polymeric matrices provide

better processability and mechanical flexibility. These trends clearly indicate that tailoring LDH composites requires not only the careful selection of support materials but also precise control of synthesis methods to achieve enhanced multifunctional performance for environmental applications.

Table 2. Summary of LDH Composites: Support Types, Synthesis Methods, Properties, and Applications

Support Material	Synthesis methods	Key Improved Properties	References
Cellulose	Urea Hydrolysis, Co-precipitation	Biocompatibility, structural flexibility, increased dispersion of LDH layers	[75]
Metal Oxides (e.g. ZrO_2, WO_3, SiO_2)	Urea Hydrolysis, Co-precipitation	Enhanced catalytic activity, improved thermal stability	[50], [76]
Biochar/Hydrochar	Co-precipitation	Increased surface area, additional functional groups, magnetic responsiveness	[77], [78]
Graphene/GO	Co-precipitation, Hydrothermal	High electrical conductivity, enhanced surface area, improved structural stability	[79], [79]
Polymeric matrices	Co-precipitation	Improved processability and mechanical strength	[80], [81]
Hierarchical LDH structures (e.g., NiCo-LDH)	Hydrothermal method	Large surface area, open electrochemical active sites, enhanced conductivity	[66], [82]
Hybrid LDH-Carbon-metal composites	Solvothermal	Multifunctionality: adsorption + photocatalysis + redox activity	[83], [84]

Mechanism of Remediation

With the ongoing industrial developments, numerous environmental impacts require careful consideration. Among the primary concerns is the direct discharge of waste into the environment, particularly when it leads to water pollution. In the literature, various organic and inorganic pollutants have been reported, posing environmental threats due to their toxic and mutagenic nature, affecting aquatic ecosystems and human health.

Various remediation methods have been proposed to address this issue, encompassing adsorption, ion exchange, biological treatment, membrane filtration, and photocatalysis. LDH emerges as a material with promising applications in these remediation processes, given its unique properties that have captured the interest of researchers in the environmental field. In

pollutant remediation using LDH, some of the most frequently reported interactions involve ion exchange, adsorption, and photocatalysis [85]. In this section, three major remediation mechanisms involving LDH are discussed in detail, namely ion exchange, adsorption, and photocatalysis. These mechanisms are strongly influenced by LDH's structural characteristics, interlayer chemistry, and surface properties, which collectively determine its performance in pollutant removal.

Ion Exchange

LDH, a type of anionic clay material with a layered structure, finds diverse applications in pollutant remediation. The primary advantages of LDH stem from its interlayer anions, which can be easily replaced, exhibit a memory effect, and allow for the adjustment of the distance between layers. These properties endow LDH with excellent adsorption capabilities toward a wide range of pollutants [86]. As illustrated in Figure 4, the remediation process typically begins with the ion-exchange mechanism, in which interlayer anions (A^-) and water molecules are gradually replaced by target pollutant anions when LDH are introduced into contaminated solutions. This exchange is driven by electrostatic interactions between the positively charged brucite-like layers and the incoming anions, leading to the stabilization of pollutant species within the interlayer galleries and their effective removal from solution. The efficiency of this process is governed by several factors associated with the physicochemical properties of the interlayer anions, including the type and charge density of the exchanged anions, the strength of electrostatic interactions, the ratio of divalent to trivalent cations in the layers, the ability of interlayer anions to stabilize the structure, and the molecular masses of both cations and anions within the LDH framework [87-88].

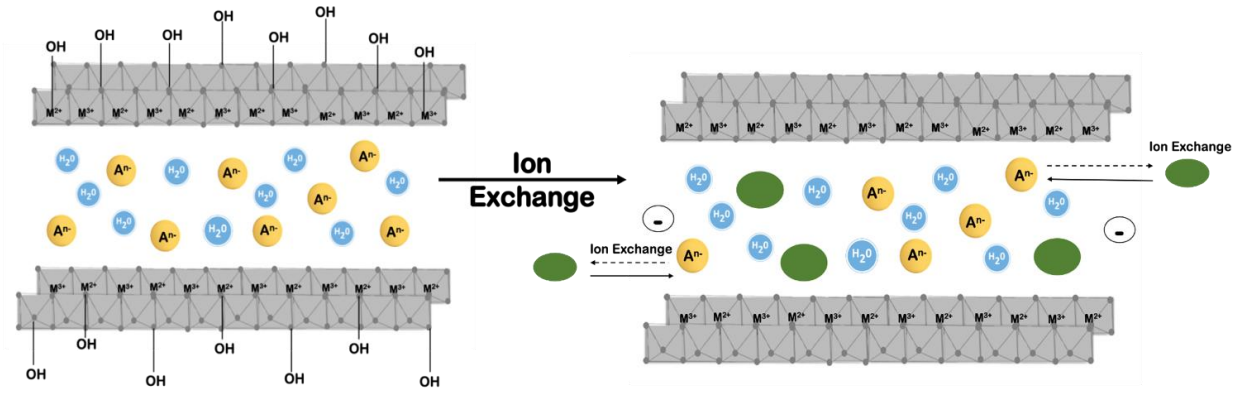

Figure 4. The illustration mechanism of pollutant ion exchange by LDH

Research conducted by Zhang et al. [90] explained that MgAl-LDH functions as an adsorbent to remove Congo Red dye using an ion exchange mechanism. Through an ion exchange process, this mechanism replaces CO_3 ions in the LDH interlayer with SO_3^- ions from Congo Red molecules. Another study by Li et al. [92] concluded that metal ion adsorption on LDH occurs through two primary mechanisms: interlayer ion exchange and external surface adsorption. In the ion exchange mechanism, the HCrO4- complex ion derived from Cr(VI) metal displaces NO_3^-, Cl^-, SO_4^{2-}, and CO_3^{2-} ions, serving as anions within the LDH interlayer.

Adsorption

Layered Double Hydroxide (LDH) is a multifunctional material for pollutant remediation due to its structural composition, positively charged surface, high chemical stability, environmental friendliness, ease of synthesis, and biocompatibility [93], [94]. LDH has the advantages of large pores and surface area, as well as active adsorption sites in the aqueous phase, which increases its adsorption capacity. Adsorption involves chemical and physical interactions [93], [94].

As reported by Song et al. [75] and Wang et al. [77], in Figure 5, the main mechanisms occurring in the LDH adsorption process include electrostatic interactions, anion substitution or intercalation, physical adsorption, and surface complexation [60], [98], [99]. Electrostatic interactions between the positively charged hydroxide layer and if the pollutant species are negatively charged, while hydrogen bonding and surface complexation occur between functional groups on the LDH surface and pollutant molecules [78]. In addition, some pollutants can replace interlayer anions or intercalate into the LDH interlayer [21]. Adsorption efficiency is greatly influenced by several factors such as the number, type, and strength of available adsorption sites, the physicochemical properties of pollutants.

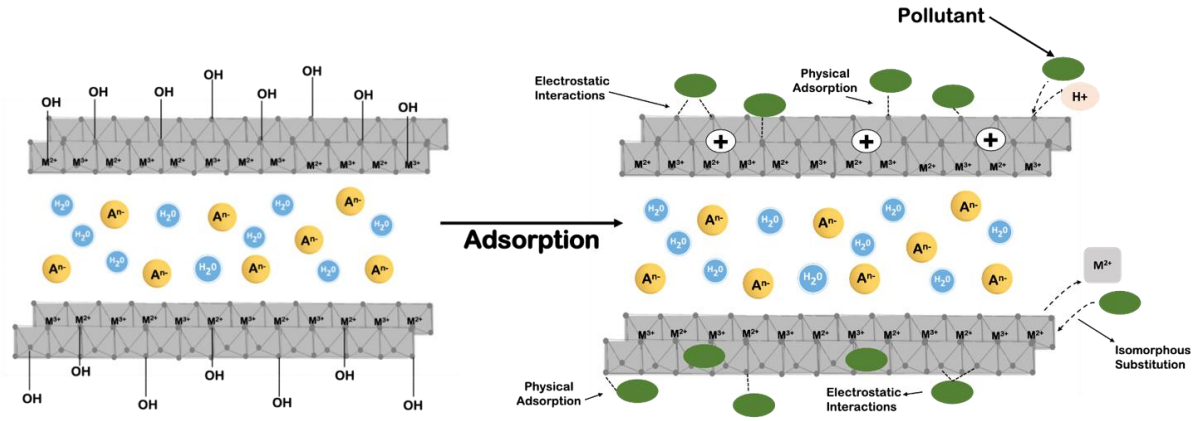

Figure 5. The illustration mechanism of pollutant adsorption by LDH

The data in Table 3 clearly demonstrate the high capacity of LDH-based materials for a wide range of pollutants, including dyes, heavy metals, anions, and gases. This performance is closely related to their adjustable interlayer chemistry, large surface area, and structural stability, making them competitive with or superior to many conventional adsorbents.

Overall, the remediation mechanisms of LDH are governed by their layered structure, tunable interlayer chemistry, and the presence of abundant active sites. Ion exchange primarily contributes to anion removal, adsorption is dominant for both cationic and organic pollutants, while photocatalysis extends LDH functionality to degrade persistent contaminants under light irradiation. Recent studies increasingly focus on integrating these mechanisms within LDH-based composites, such as combining adsorption with photocatalysis, to to improve remediation efficiency under realistic environmental conditions.

LDH can serve as an adsorbent in either powder or granular form. Kameliya et al. [100], LDH in granular form exhibits superiority due to its ability to maintain

adsorption properties and structural stability. Song et al. [95] demonstrated that MgAl-LDH effectively removed heavy metals such as Pb(II), Cu(II), and Cd(II), achieving capacities of 72%, 37%, and 18%, respectively. Furthermore, in a study by Feng et al. [101], LDH modification with 2D metal carbides (MXenes) significantly increased Ni^{2+} adsorption efficiency to an impressive 97.35%, with an adsorption capacity reaching 222.717 mg/g.

A recent investigation by Tang et al. [102] highlighted the threat of phosphate in water to human health and the environment. Efficient phosphate adsorption was achieved using the LDH/FeOOH composite adsorbent, displaying promising results and recyclability up to three times. Additionally, ZnFe-LDH exhibited commendable adsorption performance against anionic dyes such as Congo Red and Orange Yellow II, with maximum adsorption capacities of 867 mg/g and 225 mg/g, respectively. This underscores the versatile applications of pure LDH and modified LDH in water remediation. A concise summary of various pollutants that LDH can adsorb is provided in Table 3.

Photocatalysis

Photocatalysis is a promising method for degrading pollutants in water using light energy. Recent advancements combine photocatalysis with adsorption to enhance contaminant removal efficiency. One example research is the use of a TiO_2@Mg/Fe-LDH composite, which degrades toxic selenocyanate ($SeCN^-$) into oxidized forms that are then adsorbed by LDH. This dual-function system achieved up to 78.5% removal efficiency and followed pseudo-second-order kinetics [103].

Table 3. Comparison of adsorbents for adsorption of some pollutants and maximum adsorption capacity by various research groups

Adsorbent	Pollutant	Maximum Capacity	References
NiCo-LDH	Congo red	90,7%	[45]
	Cr(VI)	82%	[45]
SA/MgFe-LDH	As (III)	1.56 mmol/g	[107]
	As (IV)	1.31 mmol/g	[107]
Amino Modified Mg-Al LDH	CO_2	1.7424 mmol/g	[108]
MgAl-LDH	Congo red	769.23 mg/g	[40]
Ni-Al/LDH intercalated sodium dodecyl sulfate	Methyl orange	808.8 mg/g	[109]
CNC/MgAl-LDH composite	Tetracycline	153.3 mg/g	[75]
Fe_x/Ca-Al-LDH	PO_4^{3-}	71.81%	[110]
ZnMgAl-LDH	Methyl orange	1251 mg/g	[111]
ZnFe-LDH@Alg	Phosphate	46.894 mg/g	[112]
	Nitrate	37.864 mg/g	[112]
Alkaline sludge-LDH	Cd	901.5 mg/g	[113]
	Cu	231.3 mg/g	[113]

Another study used ZnCr-LDH with different interlayer anions (Cl^-, SO_4^{2-}, CO_3^{2-}) to adsorb the dye AO7. ZnCr–SO_4–LDH exhibited the best photocatalytic activity under visible light with good stability over reuse cycles [104]. A hybrid LDH/MOF composite, supported on

hydrochar and MIL-53(Al), showed >99% degradation of various pharmaceuticals and pesticides within 1 h under LED light, using only 20 mg of material per 50 mL solution [105]. Furthermore, a 2D NiAl-LDH/V_2C/PCN Z-scheme heterojunction effectively converted CO_2 into fuels. MXene provided high conductivity, while PCN improved light absorption and charge separation [106]. Overall, combining LDH with semiconductors (e.g., TiO_2, MOFs, MXene, g-C_3N_4) enhances photocatalytic performance and supports sustainable wastewater treatment applications. Table 3 summarizes several representative studies reporting maximum adsorption capacities of LDH-based adsorbents toward various pollutants, highlighting the versatility and tunability of LDH systems in environmental applications.

The data in Table 3 clearly demonstrate the high capacity of LDH-based materials for a wide range of pollutants, including dyes, heavy metals, anions, and gases. This performance is closely related to their adjustable interlayer chemistry, large surface area, and structural stability, making them competitive with or superior to many conventional adsorbents.

Overall, the remediation mechanisms of LDH are governed by their layered structure, tunable interlayer chemistry, and the presence of abundant active sites. Ion exchange primarily contributes to anion removal, adsorption is dominant for both cationic and organic pollutants, while photocatalysis extends LDH functionality to degrade persistent contaminants under light irradiation. Recent studies increasingly focus on integrating these mechanisms within LDH-based composites, such as combining adsorption with photocatalysis, to to improve remediation efficiency under realistic environmental conditions.

Future Trends

Layered double hydroxides (LDH) possess a range of unique structural and chemical properties, including high ion-exchange capacity, tunable interlayer chemistry, stable layered structures, and the ability to form multifunctional composites. These characteristics make LDH highly versatile materials that can be engineered through various modification strategies, such as composite formation, intercalation of functional species, and porosity enhancement. In addition, LDH can be combined with other organic or inorganic materials to to create desired structures. Looking ahead, future research on LDH is expected to focus on developing more advanced synthesis strategies and innovative composite designs to further improve their functional performance in environmental remediation and related technologies. Indonesia holds abundant natural resources such as minerals, biomass, and agricultural waste that can be transformed into environmentally friendly materials. Integrating these local resources into the development of advanced materials not only supports sustainable innovation but also adds economic value at the national level. Among emerging techniques, microwave-assisted hydrothermal synthesis stands out as a promising method, offering shorter synthesis times, better energy efficiency, and more uniform LDH structures compared to conventional approaches. By combining the synthetic method approach with rational material structure, future LDH research can be a bridge between LDH structure control as a key material for next-generation environmental remediation technologies.

Conclusion

Layered double hydroxides (LDH) exhibit unique structural characteristics that make them highly versatile for various applications. Different synthesis methods—including coprecipitation, hydrothermal treatment, sol–gel processing, and urea hydrolysis—offer distinct advantages in controlling crystallinity, morphology, and functionality. Future research is expected to focus on developing more environmentally friendly and efficient synthesis approaches, such as microwave-assisted hydrothermal techniques, to enhance scalability and energy efficiency. Indonesia's abundant natural resources, including minerals, biomass, and agricultural waste, offer valuable opportunities for producing eco-friendly and sustainable LDH-based materials. Integrating these local resources into the development of advanced materials can foster innovation while simultaneously supporting environmental sustainability.

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Author Contributions

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Conflict of Interest

The authors declare no conflict of interest

Ethical Standards

This article does not contain any studies involving human or animal subjects.

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