

Review

Layered Double Hydroxide Materials: A Review on Their Preparation, Characterization, and Applications

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Abstract: Layered double hydroxides (LDHs), a type of synthetic clay with assorted potential applications, are deliberated upon in view of their specific properties, such as adsorbent-specific behavior, biocompatibility, fire-retardant capacity, and catalytic and anion exchange properties, among others. LDHs are materials with two-dimensional morphology, high porosity, and exceptionally tunable and exchangeable anionic particles with sensible interlayer spaces. The remarkable feature of LDHs is their flexibility in maintaining the interlayer spaces endowing them with the capacity to accommodate a variety of ionic species, suitable for many applications. Herein, some synthetic methodologies, general characterizations, and applications of LDHs are summarized, encompassing their broader appliances as a remarkable material to serve society and address several problems viz. removal of pollutants and fabrication of sensors and materials with multifaceted useful applications in the medical, electrochemical, catalytic, and agricultural fields, among others.

Keywords: layered double hydroxides; LDH; anionic clays; 2D materials; synthetic clays



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1. Introduction

The layered double hydroxides (LDHs) consist of brucite-like layers, also known as anionic clays, and are broadly utilized in the field of catalysis and anion exchange processes [1,2]. LDHs are composed of cationic metal-containing layers with charge-balancing hydrated anions in the interlayer spaces, with the general formula $[M^{2+}(1 - X)M^{3+}(X)(OH)_2]X^+(An^-)_X/n.mH_2O$, where M^{2+} and M^{3+} represent bivalent (Cu^{2+} , Ca^{2+} , Mg^{2+} , Zn^{2+} , Ni^{2+} , Co^{2+} .) and trivalent cations (Al^{3+} , Fe^{3+} , Ga^{3+} , Cr^{3+} .), respectively, whereas the An^- is the interlayer (Cl^- , Br^- , NO_3^- , I^- , OH^- , SO_4^{2-} .) anion with coulombic charge n , and x represents the molar ratio of divalent to trivalent cations, which is generally in the order of 2.0–6.0 [3]. A diagram of LDH represented is shown in Figure 1 [4]. A wide range of distinctive countercharge anions can be intercalated into the LDH structure, including natural and inorganic species, the most common being halides (chlorides) and oxyanions (carbonates, nitrates) [5].

LDHs can capture and exchange both organic and inorganic anions as well as cations. Such properties make these materials unique, as they show increased selectivity for the carbonate ions because of their flat location in the interlayer space and position parallel to the layers of the matrix [6]. Carbonate salts are reported to be the most common salt form of LDH [Figures 2 and 3].

LDHs can be synthesized by different techniques depending on the requirements of the final product, such as phase purity, high crystallinity, and porosity. The varied combinations of metals and anions can be utilized to acquire desired properties. LDHs bring about an enormous class of isostructural materials with tunable physicochemical characteristics. Various direct synthetic techniques, such as coprecipitation [7], hydrothermal [8], urea [9], sol–gel [10], and in situ, have been deployed for the preparation of LDH and LDH-containing hybrids. Coprecipitation and hydrothermal methods are the most

widely accepted methods due to ease of synthesis as well as separation. The presence of exchangeable interlayer anions in the LDH is responsible for adsorptive removal of charged contaminants of wastewater. Because of their tunable and fascinating anion exchange capacity, LDHs are exceptionally useful for various applications, such as sorbent materials for a wide variety of pollutants covering inorganic species, such as oxyanions [11], metals [12], and phosphate [13], and organic pollutants, such as pesticides [14], herbicides [15], and dyes, among others [16]. Herein, LDH materials are reviewed in the light of some commonly used synthetic methodologies, general characterization techniques, and their modern applications.

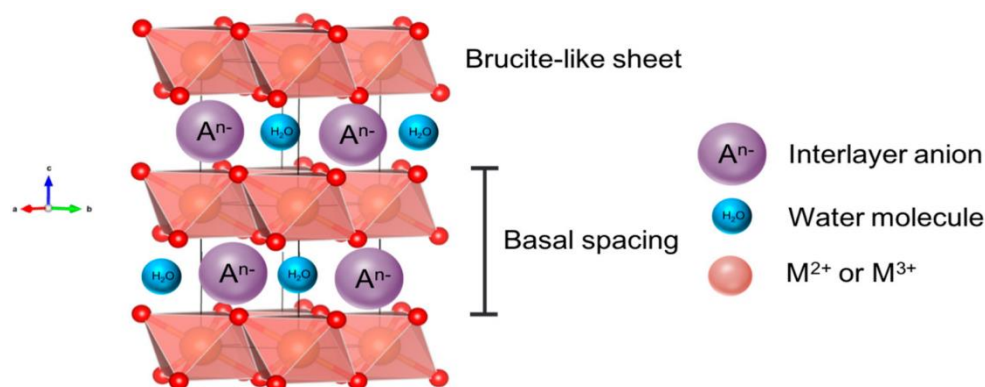


Figure 1. Diagram showing the structure of layered double hydroxide “Ref. [4]”.

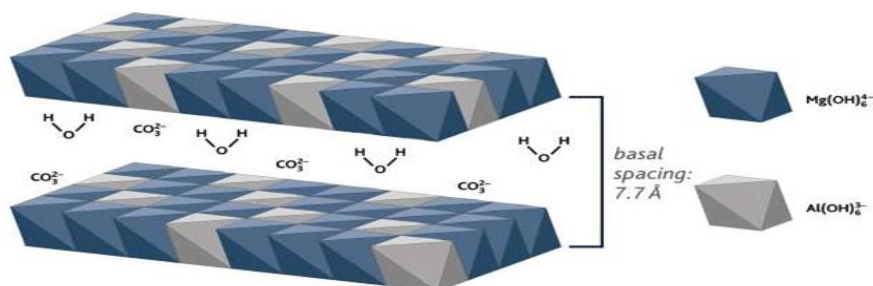


Figure 2. Schematic picture of LDH “Ref. [6]”.

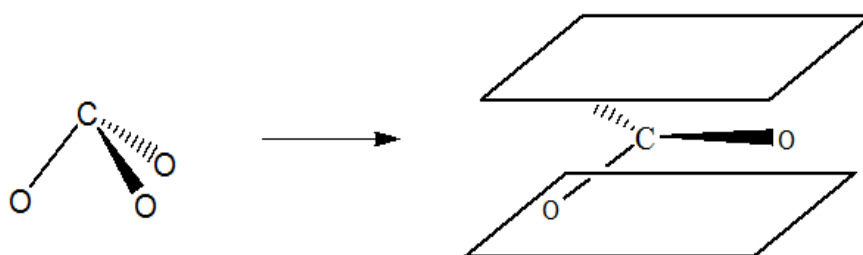


Figure 3. Plane-parallel arrangement of CO_3^{2-} ions in the inner space of LDHs “Ref. [6]”.

2. Synthesis Techniques for LDH

As previously indicated, LDH materials are the combination of divalent (M^{2+}) and trivalent (M^{3+}) cationic metal-containing layers with counter charge balanced by hydrated anions in the interlayer spaces. The main factors to be considered during the synthesis of LDHs are the combination of the metallic cations, pH, temperature, aging time, and their arrangement strategy. There are many techniques used for the preparation of LDHs: (1) coprecipitation, (2) hydrothermal synthesis, (3) microwaves, (4) ion exchange, (5) salt oxide, (6) urea, (7) induced hydrolysis, (8) sol–gel, and (9) mechanochemical, among others.

2.1. Coprecipitation Method

Coprecipitation is the most common strategy for synthesis of LDH hybrids. This method consists of dissolving the inorganic salt in alkaline medium at constant pH, which permits control of molecular size and morphology of the hybrids. In general, it involves a mixture of solutions of the metal ions and the hydroxyl anions that constitute the layers in the presence of a salt of the interlayer anion. The pH is maintained above 8 while titrating with 0.1 N NaOH to speed up the coprecipitation. The precipitate is aged at room temperature for a minimum 24 h, centrifuged, washed with deionized water, and vacuum-dried to generate the desired product. Biomolecule–LDH hybrids can be set up by particle-trading interlayer anions of LDH with biomolecules [17], wherein the size of the particles can be simply adjusted by the length of maturation and temperature viz. smaller particles are obtained by less aging [18]. Miyata modified some key parameters, such as the concentration of the reactants, the washing condition, and pH, for the formation of LDHs [19]. The protocol entails the slow addition of solutions containing the divalent and trivalent cations in the appropriate molar ratio, before adding an aqueous solution of the chosen interlayer anion. An alkaline solution is then added to adjust the pH during the reaction and to promote the coprecipitation of the two metal salts [20,21]. Rouby et al. supplied a schematic flowchart for an alkaline solution, e.g., NaOH, NaHCO₃, and NaCO₃, to adjust the pH during the reaction to promote coprecipitation of the metal salt [22]. Recently, Amer et al. demonstrated a modification of ternary Mg–Cu–Al LDH (MLDH) using 3-amino-1H-1,2,4-triazole. The prearranged MLDH and LDH were assessed as proficient adsorbents during the remediation of copper and cadmium ionic species. The factors affecting adsorption interaction—mass of adsorbents, acidity of the medium, and convergence of ionic species in the medium, as well as temperature and cycle time—were evaluated [23].

2.2. Urea Hydrolysis

Urea is a weak Brønsted base that is highly soluble in water and thus can be used as a precipitating agent to increase pH through its thermal decomposition. Urea decomposition is slow and starts around 90 °C, which leads to an increase in pH up to 10 and a lower degree of supersaturation. During hydrolysis, CO₃^{2−} anions form and act as interlayer anions, where urea enables pH control and can form monodispersed LDH materials with purity and high crystallinity [24]. Most often, urea hydrolysis is combined with coprecipitation and hydrothermal synthesis. Berber et al. reported that the optimal synthesis conditions were to obtain uniform particles by altering the M²⁺:M³⁺ molar ratios, aging, and urea concentration [25]. Some other studies have been performed in parallel using coprecipitation and urea hydrolysis while applying calcination of Ni–Al–Mg hydrotalcite [26]. Inayat et al. studied Zn–Al-layered double hydroxides with nitrate as the charge-adjusting anion in the interlayer space, which were combined by precipitation from a homogeneous environment utilizing urea hydrolysis, a combination of such methods with varied synthesis times was demonstrated to distinguish the boundaries that control whether nitrate or carbonate will act as the interlayer anion. The steady presence of carbonate, which begins from the disintegration of urea, restricts the wide application of this technique [27]. To overcome this issue, treatment with mineral acid or salt and acid were introduced to substitute the carbonate particles in the LDH structure with anions such as chloride [28]. Drawbacks of such techniques are longer time involvement and extra work associated with post treatment strategies. A diminishing in crystallite size, crystallinity, and loss of substance because of the incomplete disintegration of the LDH in the acidic medium could happen if the treatment conditions are not appropriately changed [29]. An elective post blend strategy for the change in carbonate-containing LDHs into the chloride structure was presented by Costantino et al. by treating the LDHs with weak HCl gas at 150 °C to preclude any LDH disintegration. In brief, urea hydrolysis is better than coprecipitation because it is a slow reaction that leads to a low degree of superstitution and provides thin platelets of narrow particle size distribution with shorter aging. The formation of CO₂ by

urea decomposition and formation of carbonates as counter anions may be drawbacks of urea hydrolysis synthesis [30].

2.3. Sol–Gel Method

The sol–gel method is known for its low cost and fast synthesis of nanoscale particles with large specific surface area, high purity, and high homogeneity. This method permits access to the structural properties of the final products by just altering the substantial amount of the reactants and the aging time by eliminating or adding reactant species. This method deals with dissolution of the appropriate metal salts in water at room temperature [31]. Prince et al. proposed an overall sol–gel technique for the synthesis of LDHs that can be adjusted to acquire materials encompassing specific metallic cations, and characterized morphology with high specific surface areas (up to $290 \text{ m}^2 \text{ g}^{-1}$) and very narrow pore size distributions (3–4 nm). Nano capsular morphology ensues, instead of the platelet-like particles obtained by coprecipitation and other methods [32]. One of the pieces of evidence for pharmaceutical application is via a rehydrated sol–gel method wherein synthetic hydrotalcite can also be calcined and rehydrated without losing its lamellar structure [33]. In several cases, the sol–gel approach is preferred for its simplicity and the high quality of the materials acquired. The sol–gel method is useful for the synthesis of small particles with high specific surface area and purity, but this synthetic route leads to lower crystallinity, which requires additional treatment such as microwave irradiation, hydrothermal treatment, and ultrasonication.

2.4. Hydrothermal Treatment

As previously reported, hydrothermal treatment under mild conditions has been used to improve the crystallinity and size of particles during the synthesis of LDHs, where the temperature can be raised slowly to 150°C for several days [34]. Before transferring the solution into a stainless-steel reactor under hydrothermal conditions, the metallic salts are dissolved in water, where the temperature is raised from 30°C to 300°C and the steam pressure maintained accordingly for different synthesis times [35,36]. Lin et al. synthesized Cu–Zn–Al hydrotalcite catalysts for arsine abatement to test the impact of the temperature from 35°C to 140°C . However, some other parameters, such as calcination time, molar ratio, and temperature, make it difficult to interpret the results [36]. The most used starting materials are hydroxides and metal oxides as reactants. Ogawa et al. also used natural minerals such as gibbsite and brucite as starting materials to study their structural properties [37]. It has been reported that hydrothermal treatment increases the crystallinity, the purity, and increment in particle size, but on the other hand it required high energy and more time-consuming treatment. An example of a green approach that attracts researchers for the synthesis of hydrotalcite-like structure was expressed by Labuschagne et al. as they studied untreated oxides of magnesium and hydroxide of aluminum for hydrothermal precipitation [38].

2.5. Microwave-Assisted Synthesis of LDH

LDH synthesis utilizing microwave (MW) irradiation is a process where the aging occurs by microwave. Irradiation with microwaves assists in a speedy aging technique: around 15–60 min [39,40]. Homogeneous particles can be synthesized utilizing MW maturation. The particles obtained are smaller than particles created utilizing reflux maturation. Surface regions with high Mg–Al–LDH have been blended from the combination of $\text{Mg}(\text{OH})_2$ and $\text{Al}(\text{OH})_3$ in Mg/Al molar proportion of 2:1 [41].

A product prepared by MW heating analyzed by XRD is represented in Figure 4. The powder product after the drying process followed by hydrothermal treatment showed explicit XRD peaks for Mg–Al–LDH, but the product in the state of slurry before the drying process did not show any crystal structure at all [42].

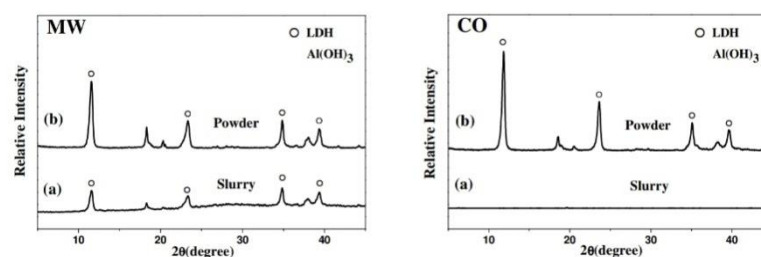


Figure 4. XRD patterns of products with microwave (MW) irradiation for 20 min and conventional heating (CO) for 2 h. (a) Before and (b) after drying at 100 °C “Ref. [42]”.

2.6. Ion Exchange Process

The ion exchange process is used when the desired interlayer anions are not fit to be incorporated utilizing urea or coprecipitation strategies. The preferred anion replaces the anions existing in a formerly arranged layer double hydroxide. Nitrate and chloride anions are routinely used as a part of layered double hydroxides, as they can be successfully exchanged with a broad assortment of regular organic and inorganic anions, e.g., Ni-Al-NO₃ layered double hydroxide (LDH) compound has been intercalated with benzoate anion through an anion exchange process used for amoxicillin drug adsorption [43].

2.7. Reconstruction

In this method, metal salts are calcinated at 500 °C for 4 h in a nitrogen environment at a heating rate of 5 °C/min. The prepared material is added to a guest molecule containing distilled water solution at a maintained pH, and the resulting precipitate is aged at room temperature, filtered, washed thoroughly with deionized water, and dried under vacuum [44]. Microspheres of carbon have been incorporated by the calcination-reconstruction strategy. It has been revealed that the morphology of methotrexate (MTX) intercalated layered double hydroxide (MTX/LDH) nanohybrids without template of carbon microspheres is highly irregular, while MTX/LDH nanohybrids arranged with the layout of carbon spheres with monodispersed template provided it a regular morphology [45].

2.8. Oxide Method

In this strategy, divalent metal oxide and anion intercalate with a trivalent metallic cation arrangement. The pH should be kept marginally acidic for moderate hydrolysis of the divalent cation oxide. This strategy has a few prerequisites, such as the need for the divalent cation oxide to undergo slow hydrolysis, and the anion to be intercalated must be able to form a soluble salt with the trivalent cation and be stable in an acidic medium [46].

2.9. Mechanochemical Methods

The synthesis of various LDHs has been accomplished by mechanochemical means wherein grinding, milling, or crushing techniques were adopted by researchers [47–54]. These types of syntheses were carried out by simply grinding the solid precursors in agate/ceramic mortars [47,48] or crushing in planetary ball mills [49,50]. However, some researchers have combined wet synthesis techniques with this protocol. Mei-Gui et al. performed hydrothermal treatment with a ground mixture of reactants to increase the crystallinity and obtain a monodispersed phase of LDH [51]. Bhojaraj et al. deployed a mechanochemical approach to perform decarbonating intercalation of anions [52] to carry out anion exchange from carbonate to nitrates. Brenda et al. constructed a specially designed wet grinding mill for continuous batch synthesis of LDHs [53]. Nanocomposites of LDH bearing Ni-Cr with PbS have been prepared by grinding and stirring the mixture in aqueous medium [54]. The superiority of this approach over other conventional methods has been affirmed by several examples viz. during the oxidation of cyclohexanone and Claisen–Schmidt condensation, mechanochemically prepared yttrium-modified LDH catalysts reportedly could activate reagents well relative to catalysts prepared by coprecipitation method [55]. Similar results were obtained for the superior adsorption

of naphtholate AS dye on mechano-Zn LDHs than coprecipitated-Zn LDHs [56]. The mechanochemically prepared diclofenac-LDH dispersed in the hydrogels expressed the largest anion retention capability compared to conventional coprecipitation, direct anion exchange, and dehydroxylation–rehydration routes [57]. Mechanochemical methods have advantages of being environmentally friendly with the minimal requirements of separating and drying of the ensued products.

3. Characterization Techniques Used for LDHs

Various techniques are applied to understand the properties of LDHs and to identify the competence of LDHs as an anion adsorbent. To study structural features of the LDH, electronic and spectroscopic techniques, such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), Fourier-transform infrared (FTIR) spectroscopy, powder X-ray diffraction (PXRD), and energy-dispersive X-ray (EDX) spectroscopy, and thermal investigation methods, such as differential thermal analysis (DTA), differential scanning calorimetry (DSC), and powder X-ray diffraction (PXRD), are deployed [58–65]. Further specific characterizations are as follows:

A variety of spectroscopic techniques, such as XAS (X-ray absorption spectroscopy), extended X-ray absorption fine structure (EXAFS), X-ray absorption near-edge structure (XANES), and MAS-NMR (magic angle spinning—nuclear magnetic spectroscopy) are used to identify the oxidation state and coordination geometry of the cations in the layers [61]. In some cases, other explicit methods have been applied, such as Mossbauer spectroscopy for hydrotalcite containing iron [62]. XANES spectroscopy is used to detect the geometry of cations, which reveals the most preferred geometry of the metal cations with octahedral climate in the brucite-like layers; however, sometimes Zn^{2+} cations are reported to be arranged tetrahedrally, which after calcination retain octahedral geometry in the layers of mixed metal hydrotalcite containing carbonate [63]. Ultraviolet-visible (UV-vis) spectroscopy is applied to confirm the arrangement of cations as in-fluid arrangements in the layers. Sometimes when strongly colored anions (e.g., decavanadate) exist in the interlayer, their retention groups cover them [64]. FT-IR spectroscopy has been utilized to follow the functional group changes in LDHs containing carbonate anions. Electrostatic associations along with the presence of water molecules in the interlayer space caused two significant changes in the FT-IR spectra when compared to reference carbonate materials. Importantly, a main band is due to hydrogen-bonded hydroxyl groups with a broad νOH band (centered at $3500\text{--}3650\text{ cm}^{-1}$), while a weak band close to 1080 cm^{-1} was recorded as an additional band in shoulder form [65]. Benicio et al. determined internal surfaces and mesoporous structure of Zn-Al CO_3^{-2} LDH, typical BET-specific surface area measurements varied up to $20\text{--}100\text{ m}^2\text{ g}^{-1}$ [66].

4. Thermal Properties of LDHs

Thermal properties of LDHs are studied using thermogravimetric analysis (TGA) and differential thermogravimetric analysis (DTA). These properties are found to depend on the type of the cation, interlayer anion, and experimental conditions kept during thermal decomposition.

Typically, the decomposition happens in four stages:

- (i) in the first stage, physically adsorbed water molecules available on the outside surface of the crystallites are removed;
- (ii) the second step follows removal of the interlayer water molecules;
- (iii) the third step occurs as removal of the hydroxyl groups from the layers as water vapors; and
- (iv) the fourth and final step is responsible for the removal of the interlayer anion.

Among these, usually overlapping of the (iii) and (iv) steps is observed; however, a precise identification of the different steps may be followed using differential thermogravimetric trace analysis. For carbonate-containing LDHs, interlayer water dehydration causes 15% loss of the total initial weight and decomposition by CO_2 evolution leads to the removal

of 40% of the underlying weight. Examination by mass spectrometry, gas chromatography, or temperature-customized decay/desorption confirmed the gases evolved during warm deterioration and supported such disintegration steps (Figure 5) [67,68].

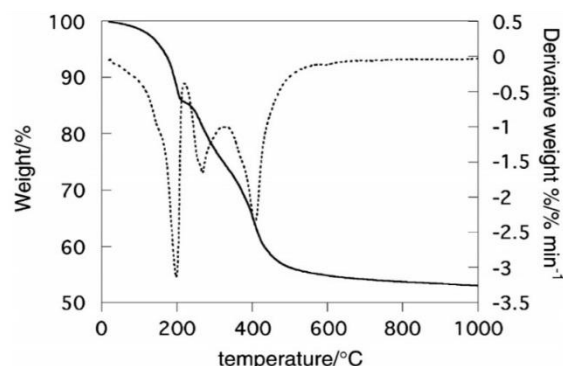


Figure 5. Thermogravimetric (solid line) and differential thermogravimetric (dotted line) analysis curves for a commercial MgAl LDH with intercalated carbonate “Ref. [67]”.

5. Applications of LDHs

LDHs have garnered a lot of attraction for industrial researchers and scientists due to their specific properties, such as nontoxicity, low cost, ease of synthesis, high chemical and thermal stability, tunability in composition, high biocompatibility, and many more. Some of the applications are discussed in this section.

5.1. LDHs as Catalysts

LDHs prepared by conventional methods have attracted considerable interest in the fields of solid-phase catalysis as a support media. In earlier studies, LDHs were reported to be effective supports for Ziegler catalysts during olefin polymerization viz. LDHs with a series of possible combinations of metal ions mainly of Ni-Al, Mg-Zn-Al, Mg-Mn-Al, Mg-Co-Mn, Cu-Al, Co-Cr, Mg-Al-Cr and Mg-Al, calcined at 200 °C to 450 °C. Among these, Mg-Mn-Al- CO_3 -LDH heated at 473 K showed the highest catalytic activity of polyethylene production [69,70]. In another report, integrated vanadium oxide catalysts supported by calcined Mg-Al-LDHs were used for oxidative dehydrogenation of butane [71]. Similarly, vanadium oxide (V_2O_5)-impregnated Mg/Al hydrotalcite have been used for the synthesis of isobutyraldehyde from methanol and n-propanol in the vapor phase [72]. It has been demonstrated that calcined as well as uncalcined LDHs may be used as supports for noble metal catalysts [73]. With enhanced attention towards environmental and economic concerns, heterogeneous solid-base catalysts such as calcined LDHs have gain importance among scientists due to their ease of separation, recyclability, simple handling, and low cost [74]. Specifically, mixed oxide LDHs, due to their excellent ability to provide Bronsted base sites, have received attention, and are apt to replace homogeneous base catalysts, as they are more recyclable and environmentally benign. These are used for numerous organic reactions, such as various condensation reactions (Knoevenagel, Aldol, and Claisen-Schmidt) and addition reactions (Michael additions and Henry reactions), among others. Rock-salt-type LDHs possess additional weightage due to the availability of both acid and base sites, the strength, and relative amounts of which depend on the molar ratio of cations and calcination temperature. Concise reviews on the development of LDHs as precursors of multifunctional catalysts and catalytic materials has also been presented in the literature [75–77]. Copper (Cu)-containing LDH materials accompanying N-arylation have shown good yield at 100 °C to 160 °C. Likhar and coworkers examined the N-arylation of benzylamine and cycloalkyl amines with some chlorobenzenes substituted with an electron-withdrawing group over catalyst Cu-Al-LDH- K_2CO_3 . Normally, the reaction yield is good to excellent (45–93%) at 100–160 °C for 8–16 h. They also noted that the presence of the electron-withdrawing group in chlorobenzene was a key for this reaction to

take place [78]. It has been reported that secondary and tertiary amines can be oxidized with various oxidizing reagents over the catalyst with specifically increased alkalinity: intercalating OBU[−] anion in Mg-Al-LDH converts it from a weak base to stronger [79].

5.2. Photocatalysis

The unique characteristic of LDHs and similarity to semiconductors [80] has attracted the attention of many researchers. Improved performance in photocatalytic activity was demonstrated by Fu et al. in doping Zn-Cr-LDHs with terbium cations [81], wherein double the catalytic activity was observed on doping with an optimum composition of 0.5%. Gomes Silva et al. carried out studies on photocatalytic water splitting using visible light irradiation with a series of Zn-to-metal atomic ratios at different metallic ratios of Ti, Ce, and Cr to generate mixed metallic LDHs [82]. Zn-Cr LDH (Figure 6) showed high quantum yields of 60.9% at 410 nm and 12.2% at 570 nm, with higher oxygen generation capacity than tungstate. The conversion of carbon dioxide to alcohol via a photochemical pathway is one of the most pursued topics in modern chemistry. It not only facilitates the removal of the potent greenhouse gas but also provides an alternative pathway to convert chemically less reactive CO₂ to liquid fuel. Iguchi et al. observed that the photoreduction of CO₂ into methanol was achieved by trimetallic carbonate of ZnCuGa-CO₃ LDHs with a promising result of >97% [83].

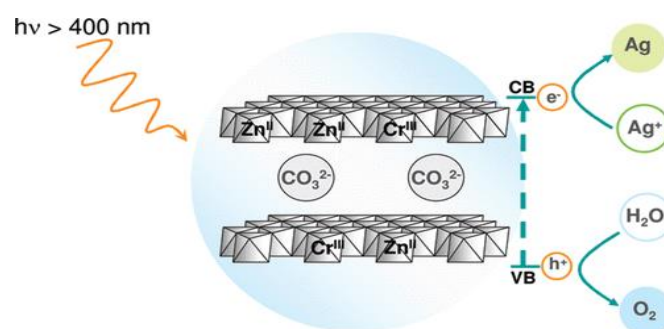


Figure 6. Water splitting by mixed metallic LDHs “Reprinted with permission from Ref. [82]. 2009, American Chemical Society”.

5.3. LDHs for Water Treatment and Environmental Remediation

LDH materials have acquired extraordinary interest in numerous potential applications, such as water treatment [84], drug delivery [85], catalysis [86]. Their application in water treatment as adsorbents has great potential because of their minimal expense, high surface area, profoundly tunable inside engineering [87], nontoxicity [88], and interchangeable anions [89]. Recently, the modification of LDH hybrids has gained interest in wastewater treatment. It originates from the hybridization of LDH with other materials, such as graphene (G), carbon nanofibers (CNFs), and carbon nanotubes (CNTs) [90] for the availability of freshwater without harmful substances and microorganisms [91]. Fast industrial growth and urbanization lead to water pollution due to organic dyes used in paints, plastic, and textiles, and it is increasing day by day along with other organic and inorganic pollutants. The low biodegradability, profound shading, and complex sweet-smelling development of colors make the dye-containing industrial waste a harmful, aesthetic contaminant dangerous to aquatic life [92,93]. Other potent nonbiodegradable pollutants are heavy metals that are harmful at even low concentrations and contain zinc (Zn), lead (Pb), copper (Cu), cadmium (Cd), chromium (Cr), mercury (Hg), nickel (Ni), arsenic (As), and thallium (Tl). These contaminants are present in the effluent of oil refining, coal mining, metal plating, agricultural assets [94–97]. Some inorganic anions and oxyanions, such as arsenite, arsenate, selenite, selenate, chromate, phosphate, and nitrate, as well as monoatomic anions such as fluoride, chloride, bromide, and iodide also exist in water, causing water pollution [97,98]. In water and wastewater, a few oxyanions, along with such constituents as cations and humic substances conceivably impact the science of LDHs and the speciation

of oxyanions in the fluid framework. Subsequently, a comprehensive adsorptive execution of LDHs in the multi-oxyanion framework comprising different oxyanions and lattices of interest has been identified. Lazaridis et al. studied the sorptive flotation framework for promising arsenate and chromate evacuation and successful particle separation [99,100], but satisfactory results were obtained by Gilman et al. via the “porous pot” technique for removal of arsenic (As) and illustrated that LDH might be an attractive adsorbent for wastewater treatment [98]. LDHs may be used in powder and granular forms, in a scattered powder structure, or in a fixed-bed granular structure. Granular LDHs have been found superior to powdered LDHs because they retain their intrinsic sorption properties with sufficient mechanical strength and toughness. Gillman et al. found that granular LDHs can reduce high concentrations of oxyanions [101]. After the sorption process, desorption is required to check recyclability of LDHs. Kuzawa et al. proposed a plan where the phosphate PO_4^{3-} that is sorbed could be recovered as calcium phosphate by adding CaCl_2 as eluent [102]. Wang et al. showed that Cr(VI) could be recuperated from Cr(VI)-stacked Li–Al LDHs by resuspending them in steaming hot water to upgrade synchronous Li^+ deintercalation from Li–Al LDH structures in fluid arrangement and Cr(VI) desorption [103]. Murayama et al. investigated column operations for the removal of low-concentration harmful species. Pelletized granular LDH and 5% polyvinyl alcohol solution can remove low concentrations of harmful anionic species of As(III), As(V), Se(IV), and Cr(VI) from wastewater. Among the LDHs examined, Mg–Al– NO_3 LDH showed the ideal immigration of anionic species, and it showed brilliant evacuation of anions of As(V), Se(IV), and Cr(VI) at very low anionic concentration [104]. Recently, Sari et al. applied Mn–Fe LDHs as adsorbent material for the removal of arsenic from synthetic acid mine drainage (AMD) containing many heavy metals, such as Zn, Pb, Ni, Co, and Cd [105]. Kameda et al. reported the use of acid media and higher HNO_3 uptake capacity for calcined LDHs, in which the acid media helps to increase the sorption process and proton utilization throughout reconstruction reactions [106]. On the other hand, Socias-Viciano et al. also observed that in neutral solutions, uptake capacity of nitrate can be improved above 99% by enhancing calcination and sorption temperature [107]. Significant improvement in CO_3^{2-} removal capacity of Mg–Al– CO_3 LDHs on calcination and replacement of carbonate by NO_3 as Mg–Al– NO_3 was reported by Goh et al. with an additional increase in uptake capacity to 170 mg/g, associated with a surface area increment due to a decrease in the average particle size of nitrate-intercalated LDHs to 122 nm [108]. Yoshida et al. proposed a mechanism of higher AsO_4^{3-} (arsenate) uptake capacity and affinity for Mg–Fe LDHs in comparison to that of Mg–Al LDHs on the basis of formation of inner-sphere complexes by Mg–Fe LDHs, as arsenate anions once adsorbed were not easily desorbed from Fe^{3+} -containing LDHs. By similar mechanisms, chromate uptake was also explained: pH of the solution was the main factor in determining the effectiveness of the different removal mechanisms [109]. Goswamee et al. demonstrated the sorption of dichromate at low pH by calcined and uncalcined Ni–Al– CO_3 , Mg–Al– CO_3 , and Zn–Cr– CO_3 LDHs [110]. Koilraj and Kannan explained the surface precipitation mechanism that increased phosphate sorption with decreasing pH and sorbent crystallinity [111]. The mechanism of water remediation for inorganic pollutants has been summarized in Table 1.

Hydrotalcite, a natural layered mineral, has been used for adsorptive removal of anions [112,113] as well as cations such as Cu^{2+} , Cd^{2+} and Pb^{2+} using Zn–Al–EDTA hydrotalcite by chelation [114]. Dyes released in the atmosphere leave genuine negative effects on the ecosystem and furthermore on human well-being. Consequently, these should be expelled from water bodies. LDHs have also been utilized for colored wastewater treatment. Zang et al. demonstrated a mesoporous magnetic NiFe_2O_4 –Zn Cu Cr–LDH composite and its potential for pragmatic adsorption of Congo red from wastewater under various conditions by utilizing single-factor tests, which offered the ideal adsorption conditions. Under these conditions, the adsorbent displayed high evacuation effectiveness over 97% with an exceptionally wide introductory Congo red focus of 150–450 mg/L [115]. Recently,

a superabsorbent nanocomposite, $\text{Fe}_3\text{O}_4/\text{PEG-Mg-Al-LDH}$, was reported for ultrahigh efficiency in the removal of organic dyes [116].

Table 1. LDH materials: mechanisms proposed and maximum removal capacity (C_m) proposed by various research groups.

Anion	Metal Ions	Pollutant	Removal Mechanism	C_m (mg/g)	Reference
CO_3^{2-}	Mg-Al	CrO_4^{2-}	Reconstruction	276	[108]
CO_3^{2-}	Mg-Al	AsO_4^{3-}	Anion exchange	276	[109]
CO_3^{2-}	Mg-Al	NO_3^-	Reconstruction adsorption	147	[103]
CO_3^{2-}	Mg-Al	AsO_4^{3-}	Reconstruction adsorption	116	[104]
CO_3^{2-}	Mg-Al	CrO_4^{2-}	Reconstruction	248	[106]
CO_3^{2-}	Mg-Al	CrO_4^{2-}	Reconstruction	280	[106]
CO_3^{2-}	Ni-Al	CrO_4^{2-}	Reconstruction	86	[106]
CO_3^{2-}	Zn-Al	PO_4^{3-}	Weathering/precipitation	273	[107]
CO_3^{2-}	Zn-Al	PO_4^{3-}	Weathering/precipitation	189	[107]
CO_3^{2-}	Zn-Al	PO_4^{3-}	Weathering/precipitation	75	[107]
NO_3^-	Mg-Al	NO_3^-	Reconstruction adsorption	236	[102]
NO_3^-	Mg-Al	AsO_4^{3-}	Anion exchange	31	[104]
Cl^-	Mg-Al	SO_3^{2-}	Anion exchange	495	[110]
Cl^-	Mg-Al	SO_3^{2-}	Anion exchange	415	[110]
Cl^-	Mn-Fe	SO_3^{2-}	Coprecipitation	310	[110]
Cl^-	Mg-Al	AsO_4^{3-}	Anion exchange	222	[105]
Cl^-	Mg-Fe	AsO_4^{3-}	Anion exchange	373	[105]

Due to their exceptional physical and electronic properties, reasonable cost, high adaptability, and convenience, polymer chemistry and polymer-based materials are perceived as key segments in numerous significant businesses, such as automobiles, electronics, and aviation. One serious issue with numerous polymers is that they are profoundly combustible and can produce a lot of toxic smoke during ignition, which represents an incredible danger to human security and altogether confines their applications in numerous areas [117–119]. To resolve this issue, viable strategies are expected to add some nanosized fire-resistant filler into the polymer matrices. Layered double hydroxides have potential applications as flame-retardant polymer composites, and there has been rapid development in the research field for introducing the combination of fire-resistant polymer LDH nanocomposites. However, the mechanism behind the flame-retardant behavior of LDH is still not completely understood. Briefly, it may be summarized that during thermal decomposition, LDHs may lose the interlayer water molecules and disintegration of the intercalated anions as carbonates (CO_3^{2-}), along with metal hydroxide complexes occurring. This results in generation of water and fire extinguisher gases such as CO_2 , which may decrease the fuel available for combustion and bring down the heat discharge to stop the burning and ultimately stop the combustion. When not enough fuel remains to propagate the reaction further, this would initiate the formation of an expanded carbonaceous coating or char over the polymer surface, which hinders contact with the air to decrease heat release during the combustion and reduce smoke production as well [120,121]. A summary of the flame-retardant mechanism of LDHs may be attributed to the combination of the following three functions:

- (1) The endothermic decomposition of LDH works as a heat sink.

- (2) Decomposition of LDH leads to formation of mixed metal oxides, which act as an insulating film on the surface.
- (3) Generation of bound water and carbon dioxide thereby diluting the flammable gases.

The flame-retardant property of LDHs can be improved by intercalating suitable anions, such as borate or phosphate, into the interlamellar region of LDH [122]. LDHs have been melt-blended into polypropylene (PP) with intumescent fire retardant (IFR). A flame-retardant mechanism proposed by researchers is illustrated in Figure 7 [123].

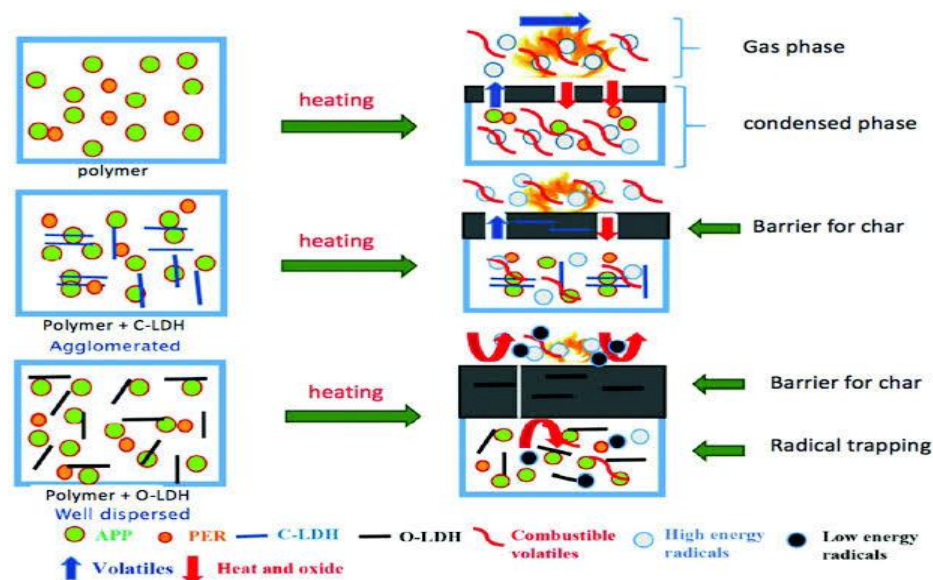


Figure 7. Dual fire-retardant action “Reprinted with permission from Ref. [123]. 2017, American Chemical Society”.

The advanced flame-retardant behavior and smoke-suppression properties of LDHs are derived from their distinctive layered structure with exchangeable anions. LDHs may be customized by intercalating organic anions into the interlamellar spaces. These organically modified LDHs can be utilized in the form of nanofillers for the synthesis of polymer-LDH nanocomposites [124,125]. Many LDH nanocomposites have been reported for flame-retardant applications to polymers such as polypropylene (PP) [126], polyethylene (PE), polymethyl methacrylate (PMMA) [127,128], acrylonitrile-butadiene-styrene (ABS), polystyrene (PS) [129], polyvinyl chloride (PVC), polylactic acid (PLA), polyamide 6 (PA 6), [130], ethylene-propylene-diene terpolymer (EPDM) [131] and polyethylene-vinyl acetate (EVA), [132]. On the other hand, most of these polymers are very combustible and can discharge a lot of smoke on burning. Thus, LDHs have been explored as fire retardants to diminish the flammability of these polymers. An outline of all the flame-retardant polymer-LDH nanocomposites reported in the literature is presented in Table 2.

Various polymers, including PMMA, PP, PVC, PE, EVA, PS, PLA, ABS, UP, PA 6, EVA [132] and EPDM, have been examined as fire-resistant polymer-LDH nanocomposites [133]. It has been mentioned that LDHs are proficient for all the previously mentioned polymers. To acquire the best fire-resistant property, various polymers require various sorts of LDHs, and among a variety of divalent cations, Zn and Mg appear more suited for flame-retardant execution over others. There is still no systematic understanding of the methods by which the varieties of divalent and trivalent metals are valuable. This should be identified, along with the thermal stability and distribution of LDHs.

Table 2. Flame retardant polymer-LDH nanocomposites and reduction in peak heat release rate (PHRR) described by various research groups.

Organic Anions	LDH Metal Ions	Polymer *	Reduction in PHRR	Reference
Undecenoate	Zn-Al	PMMA	46% (10 wt %)	[119]
Undecenoate	Ni-Al	PMMA	16% (10 wt %) 41% (10 wt %) 25.7% (10 wt %)	[119]
Undecenoate	Ca-Al	PMMA	36% (6 wt %) 24% (6 wt %) 16% (6 wt %) 36% (6 wt %) 7% (6 wt %)	[126]
Oleate	Mg-Al	PP	5% (2 wt %)	[129]
Oleate	Zn-Al	PP PMMA PE EVA	25% (4 wt %) 28% (10 wt %) 58% (10 wt %) 33% (10 wt %)	[129]
Oleate	Zn-Mg-Al	PP	38% (4 wt %)	[129]
ASA	Mg-Al	EVA	39% (3 wt %)	[132]
DBS	Mg-Al	EPDM	25% (40 wt %)	[133]
DBS	Mg-Al	PMMA	45% (10 wt %)	[129]
Cn	Mg-Al	PS	20–55% (10 wt %)	[127]

* PMMA, polymethyl methacrylate; PE, polyethylene; PP, polypropylene; ABS, acrylonitrile butadiene styrene; PS, polystyrene; PVC, polyvinyl chloride; PA6, polyamide6; EVA, ethylene–vinyl acetate, EPDM, ethylene–propylene–dieneterpolymer.

5.4. LDHs for Removal of Greenhouse Gases

Numerous articles and patents have illustrated the utilization of calcined LDHs for the effective adsorption of polluting gases, such as carbon dioxide and sulfur oxide [134]. Calcined LDHs (hydrotalcite) show solid fundamental properties that make them proficient scavengers for acidic gas recuperation from hot gas streams. The recuperation of CO₂ and SO₂ from power-plant-produced gases is considered the initial phase in decreasing absolute carbon and sulfur oxide emissions. [Ca₆Fe₂(OH)₁₆](CO₃).xH₂O, [Mg₆Fe(OH)₁₆](CO₃).xH₂O and [Ca₂Al(OH)₂](NO₃).xH₂O are found to be effective sorbents for mitigating SO₂ from tube gas and maintaining frosty sides of coal-burning power plants. Graphene oxide-incorporated layered double hydroxides (GO-LDHs) were identified for adsorbing carbon dioxide with increased efficiency. The adsorption efficiency of LDH has been enhanced by more than 60% just by increasing 7 wt% GO concentration [135]. Additional emphasis ought to be placed on preparing more combinations of LDHs to mitigate pollution problems.

5.5. LDHs for Removal of Pesticides and Related Persistent Organic Pollutants (POP)

The US Environmental Protection Agency (EPA) has classified phenols as carcinogenic pollutants due to their high toxicity. In many developing countries, phenols are still playing an important role in disinfectant production, plastic production, and pesticides, presenting many health risks and environmental pollution. Phenols are fairly soluble in water in the form of phenolates, their pK_a being quite variable according to their substituent and thus leaving them in water as persistent organic pollutants. Ulibarri and coworkers studied the sorption of trinitrophenol and trichlorophenol (2, 4, 6-trinitrophenol and 2, 4, 5-trichlorophenol) by LDHs (calcined and uncalcined), at every pH value, trinitrophenol (TNP) was exchanged more easily than trichlorophenol (TCP). TNP displaced CO₃^{2−} anions from the particle surface of uncalcined LDHs in neutral as well as in alkaline media [136–138]. The higher uptake capacity of 4-nitrophenol than phenol was studied by Chen et al.

by reconstruction of calcined Mg-Al LDHs and interactions between the layers and the $-\text{NO}_2$ group of 4-nitro phenol, which was removed by intercalation between the layers of LDHs while phenol slowly adhered at the sorbent surface [139]. The sorption of 2, 4-dinitrophenol and 2-methyl-4, 6-dinitrophenol by calcined and uncalcined Mg-Al LDHs was also studied by Chaara et al. [140]. El Shafei et al. reported that the compensating anion has a noticeable influence on the adsorption properties of LDHs toward 4-chlorophenol. At low equilibrium concentration, adsorption of 4-chlorophenol at pH above 10 occurred at the ends of the layered structure accompanied by Cl/OH substitute with an increasing order in the interlayer spaces due to substitution [141]. Other than phenols, contamination of soils and groundwater by pesticides in modern agriculture systems is a matter of big concern. Pesticide molecules with ionizable functional groups such as $-\text{OH}$, $-\text{COOH}$, $-\text{SO}_3\text{H}$ produce highly soluble anionic species in water by acidic dissociation. Anionic clays with hydrophilic and positive characters on their surface are found to be effective sorbents for anionic and highly polar organic pesticides. Besse et al. studied the adsorption of pesticides belonging to the phenoxyacetic acids family of pollutants by Mg-Al LDHs, and the adsorption capacity was found to be increased with increased layer charge density [142]. Adsorption on LDHs is proposed by an anion exchange mechanism via two steps: anion exchange at the surface followed by an interlayer anion exchange process. The adsorption capacity has been found to be dependent on the nature of the starting anions, mostly following the affinity order $\text{NO}_3^- < \text{Cl}^- < \text{CO}_3^{2-}$, as proposed by Miyata [143]. The possible mechanism and removal capacity of some organic pollutants explained by various researchers are summarized in Table 3.

Table 3. LDH materials: proposed mechanism and maximum removal capacity (Cm) proposed by different research groups.

Anion	Metal Ion	Pollutant	Removal Mechanism	Cm (mg/g)	Reference
CO_3^{2-}	Mg-Al	TNP	Anion exchange	185	[137]
CO_3^{2-}	Mg-Al	TNP	Reconstruction	1330	[137]
CO_3^{2-}	Mg-Al	TCP	Anion exchange	2	[138]
CO_3^{2-}	Mg-Al	TCP	Reconstruction	8	[138]
CO_3^{2-}	Mg-Al	4-NP	Reconstruction	370	[139]
CO_3^{2-}	Mg-Al	Phenol	Reconstruction	47	[139]
Cl^-	Mg-Al	DNP	Anion exchange	714	[140]
Cl^-	Mg-Al	DNOP	Anion exchange	503	[140]
Cl^-	Mg-Al	DNOP	Anion exchange	440	[140]

TNP, trinitrophenol; TCP, trichlorophenol; 2-CP, 2-dichlorophenol; 4-NP, 4-nitrophenol; DNP, 2, 4-dinitrophenol; DNOP, 2-methyl-4, 6-dinitrophenol.

5.6. Source of Nutrient Storage for Plants

The use of fertilizers, especially nitrogen (N)- and phosphorus (P)-containing ones, is inevitable in obtaining high agricultural yields. It is not an easy task to maintain the proportion of these elements, and it is necessary to introduce a more efficient and sustainable way to carry out the requirement. The intercalation of N and P within LDH came as an interesting option to optimize N and P supply to plants and some reports in this domain suggested LDH as a slow-release source of these nutrients. Mg-Al-Cl-LDH has been shown high potential to be used as a nutrient exchanger [144]. Berbler et al. (2014) [145] synthesized and characterized Mg-Al- NO_3^- LDH and deployed it for sustainable release of nitrates (NO_3^-) in the soil; nitrate release was monitored for acidic soil and basic one at different pH and temperatures. The release of NO_3^- intercalated with LDH showed that slow releasing process worked better for basic soil media as in the acidic soil the sustainable process worked for 16 days (15°C), whereas in basic soil at the same temperature, this

process could be observed for maximum 20 days. The results for the different pH and temperature conditions encouraged the use of these materials as sources for the slow release of NO_3^- in the soil. A new class of inorganic fertilizers may be explored by introducing least harmful anionic clays.

5.7. LDHs as Adsorbents for Anionic Pollutants

The multifunctionality of LDHs permits them to adsorb both cationic and anionic contaminants. The most common anionic species are utilized in agriculture sectors viz. nitrate, phosphates, and other agrochemicals. These pollutants can be removed by anionic clays by means of the adsorption processes and then slowly released back to the soil for plant growth or pest control, in a process of recycling or reuse. Terry (2009) [146] studied a clay mineral with a structure identical to LDH $[\text{Mg}_2\text{Al}(\text{OH})_6]_2\text{CO}_3 \cdot 3\text{H}_2\text{O}$, as low-cost anion exchangers to remove nitrate and phosphate from the solution. The residual concentration of measured anions in the solution is found to be lower than the levels recommended by the Environmental Protection Agency (EPA) for drinking water. Experiments were also conducted to study the mutual effect of one anion on the removal of another. It was concluded that nitrate (NO_3^-) did not affect phosphate (PO_4^{2-}) removal and vice versa even in a wide range of concentrations. Li et al. (2005) [147] prepared MgAl-LDH intercalated with number of anions viz. nitrate (NO_3^-), carbonate (CO_3^{2-}) and chloride (Cl^-). The adsorption capacities of materials were evaluated for glyphosate removal. The adsorption experiments examined that glyphosate removal by MgAl-LDH occurred in two ways: adsorption on outer surface as preliminary step and later on via interlamellar anion exchange. Glyphosate at low concentrations is singly adsorbed on the LDH outer surface, while with increasing concentrations the interlamellar anion exchange occurred. The glyphosate adsorption capacity of MgAl-LDH is reported to be increased with increasing lamellar charge density ($\text{Mg}^{2+}/\text{Al}^{3+}$ molar ratio) in LDH which resulted in increased electrostatic attraction also. The influence of interlamellar anions on the amount of agrochemicals retained by MgAl-LDH decreases in the order of $\text{Cl}^- > \text{NO}_3^- > \text{CO}_3^{2-}$ anions as exchangeable ions.

5.8. LDHs for Biomedical Applications

In the biomedical field, several inorganic materials have been investigated, such as silicon oxide, calcium phosphate, gold, iron oxide, and layered double hydroxides (LDHs) to examine their efficacy in target drug delivery. These inorganic materials show efficient drug delivery through sufficient availability, easy surface functionality, good biocompatibility, the potential for target delivery, and controlled release from inorganic nanomaterials. LDHs have garnered the attention of researchers due to their nontoxicity. In vivo and in vitro biocompatibility is utilized for gene delivery, drug delivery, bioimaging, and biosensing areas [148–150]. LDHs can participate by exchanging anions with nucleic acids (DNA, RNA), drugs, enzymes, and the specific abilities of layered double hydroxides in executing the task of targeted drug delivery to the location in a controlled and sustained way at a particular pH make them important for drug delivery applications. A suspension test has been conducted by researchers to examine the drug release abilities of LDH materials in a simulated intestinal fluid buffer at pH 7–8 and to deliver RNA and DNA to mammalian cells in vivo by incorporating them with LDH only or LDH with a drug for treating diseases [150,151]. The drug-LDH hybrids are utilized as superior anticancer drug delivery systems [152] without any side effects (Figure 8) that are clearly revealing the increasing importance of LDH in biomedical applications.

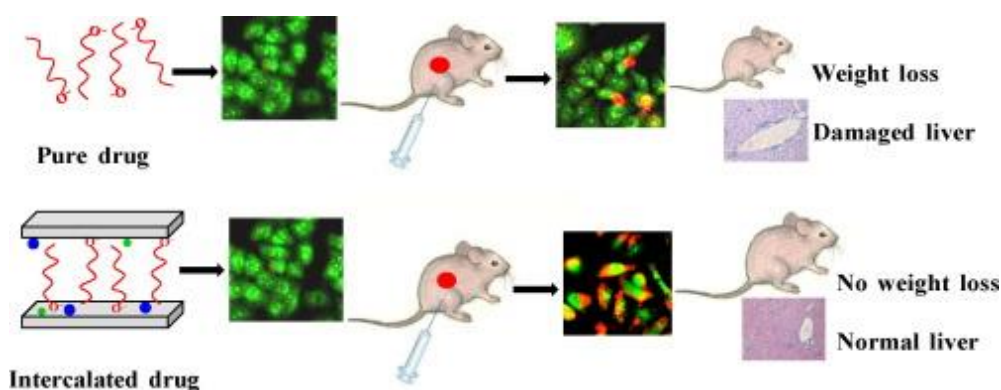


Figure 8. Drug-LDH hybrids as superior anticancer drug delivery systems “Reprinted with permission from [152]. 2016, Elsevier B.V.”.

5.9. LDH as Biosensors

The nontoxicity, biocompatibility and excellent biocatalytic properties make LDHs efficient biosensors. Urea biosensors work on the idea of immobilization of urease enzyme into oppositely charged clays [153]. More typical enzymes include the fabrication of oxidoreductase enzymes/LDH amperometric biosensors such as trans-ketolase, acetylcholinesterase, horseradish peroxidase, and glucose oxidase [154]. Unfortunately, enzyme-based biosensors do not show versatility due to their low stability and potential to be affected by temperature, pH, and ionic strength [155]. Hence, most researchers in recent times have focused on the development of enzyme-free biosensors based on the functionalization of electrodes using nanomaterials that provide them with high sensitivity [156].

5.10. LDH as Supercapacitors

LDH composites based on nickel (Ni) and combined with carbon-based nanomaterials have been used as electrodes for supercapacitors [157,158]. Ni-Al LDH nanosheets grown in situ on carbon nanotubes (CNT) exhibited promising capacitive performance. The introduction of CNTs assisted in better performance that hindered the restacking of LDHs during synthesis and provided a surface for conduction. In another example, honeycomb-like cobalt-based LDHs were deposited in situ on multilayer graphene for an energy storage device that showed a high capacitance of 883.5 Fg⁻¹ [159]. Therefore, LDH composites open the pathway for utilizing hybrid supercapacitors at a comparatively low cost.

5.11. Applications of LDHs in Display and Sensing

LDH-based photo functional materials have been obtained through controlled variations of the guest species. The fluorescence performance and stability of the guest species have been effectively enhanced by the introduction of LDHs. For displays and polarized emission, some intercalated LDH composites with a specifically tailored arrangement have been reported in the literature [160,161]. Multifunctional materials have been constructed by immobilization of QDs on the surface of LDH by self-assembly that respond to changes in pH, temperature, pressure, and light in the field of sensors. Multicolored luminescence materials have been obtained by introducing a diverse variety of chromophores (organic dyes, polymers, and quantum dots) in LDHs [162]. LDHs have also been designed to detect heavy-metal ions (HMIs), biomolecules, and chemosensors for environmental pollutants [163]. LDHs have unlimited application with ease of tunability.

6. Conclusions

LDHs were reviewed with a brief introduction on synthetic methodologies, most-used characterization techniques, and a broad range of assorted applications. The simple synthetic strategies make LDHs an alluring material for future applications, as they offer unique ion exchange properties. There is a possibility of inducing new structural features

and adjusting physicochemical performance and tunable synthesis strategies. Thus, they can open new pathways in the fields of wastewater treatment, greenhouse gas mitigation, biomedical applications, and fire-retardant materials for safeguarding the environment and our biosphere in an eco-friendly manner.

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