Composites formed by layered double hydroxides with inorganic compounds: An overview of the synthesis methods and characteristics

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REVIEW HIGHLIGHTS

- Various synthesis routes of hydrotalcite/inorganic compounds composites are described.
- The synthesis route allows generating different morphologies of the composites.
- The composites obtained exhibit multifunctional properties for different applications.

ARTICLE INFO

ABSTRACT

Nowadays, layered double hydroxides (LDH), sometimes referred as hydrotalcite-like compounds, have gained great attention since their composition and structure can be easily modified, so that they can be implemented in multiple fields. LDH-based composite materials based on LDH exhibit tremendously improved properties such as high specific surface area, which promotes the accessibility to a greater number of LDH active sites, considerably improving their catalytic, adsorbent and biological activities. Therefore, this review summarizes and discusses the synthesis methods of composites constituted by LDH with other inorganic compounds such as zeolites, cationic clays, hydroxyapatites, among many others, and describe the resulting characteristics of the resulting composites, emphasizing the morphology. Brief descriptions of their properties and applications are also included.

Specifications Table

Subject area:	Materials Science
More specific subject area:	Clays' composites
Name of the reviewed methodology:	Methods for the synthesis of composites
Keywords:	hydrotalcites, anionic clays, synthesis, morphology, sustainable materials
Resource availability:	NA
Review question:	1. What is a layered double hydroxide?
	2. What inorganic materials have been used to form composites with layered double hydroxides?
	3. What synthesis methods have been used to obtain LDH composites with inorganic materials?
	4. What morphological characteristics do these composites present?
	5. What applications have been given to LDH composites with inorganic materials?

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Background

LDH belong to the group of clay minerals of anionic type (anionic clay) also known as hydrotalcite-like compounds, hydrotalcites, mixed metal hydroxides [1], or more recently, layered double metal hydroxides [2] or double metal hydroxides [3], and if it contains rare earth elements are known as layered rare-earth hydroxides (LRH) [4]. They are natural or synthetic clays have the ability to compact upon losing water molecules, resulting in macroscopic cracking as shown in Fig. 1. Additionally, they exhibit a two-dimensional layered structure based on brucite ($Mg(OH)_2$), with octahedral coordination around the metal ions [5]. The general chemical formula is $[M^{2+}_{(1-x)}M^{3+}_x(OH)_2](X^{m-}_{(x/m)})\cdot nH_2O$, where M^{2+} and M^{3+} are divalent and trivalent cations, x represents the metal ratio $M^{3+}/(M^{3+}+M^{2+})$, and n is the number of water molecules [6,7].

The divalent metal ions can be substituted isomorphically by trivalent ions of similar coordination properties so originating a positively charged sheet. These metal ions are strongly bonded via covalent bonds, which are electrically neutralized by compensating anions (X^{m-}) within the interlayer galleries [8]. They are bonded to the layers by Coulomb and van der Waals interactions, and by hydrogen bonds [9]. The water molecules inside of galleries are bonded to the layered OH ions and/or with the anions in a highly disorganized way [10], as represented in Fig. 2.

Frequently, divalent metal ions are those whose ionic radii vary between 65pm (Mg) and 80pm (Mn), whereas the radii of the trivalent metal ion has to be comprised between 50pm (Al) and 69pm (Cr) [11] with the most frequently divalent cations constituting the LDH structure being Fe^{2+}, Co^{2+}, Zn^{2+} or Cu^{2+} and, as trivalent cations, Al^{3+}, Fe^{3+} or Ga^{3+} [12–14]. Still, some unusual cations have been introduced in the LDH lamellae such as La^{2+} [15], Ce^{2+} [13], uranyl (UO_2^{2+}) [16], Zr^{2+} [17], Ru^{3+} [18], Rh^{2+} [19] and Sn^{2+} [20], among others. LDH composed by three or more types of cations forming the lamellae have also been reported, for example, in CuZnAl- or FeZnMgAl-LDH [21–27]. In addition, cations with a coordination number greater or lesser then 2 or 3 have also been used to form LDH, such as Zr^{4+} [28,29], Sn^{4+} [30] and Ti^{4+} [31] or Li^+ [32]. Some cations that present a Jahn-Teller effect, such as Cu^{2+} and Mn^{3+}, can only be part of the LDH lamellae when associated with other type of cation [33,34].
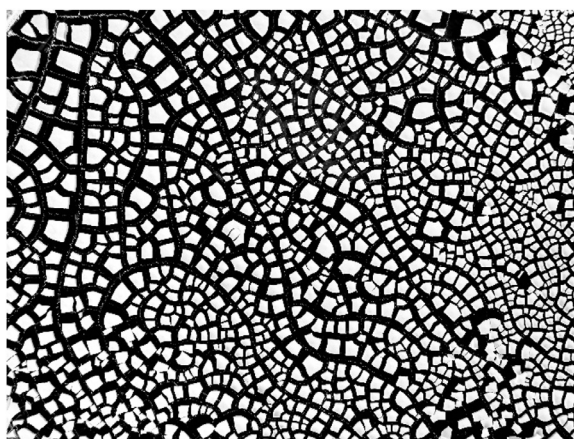

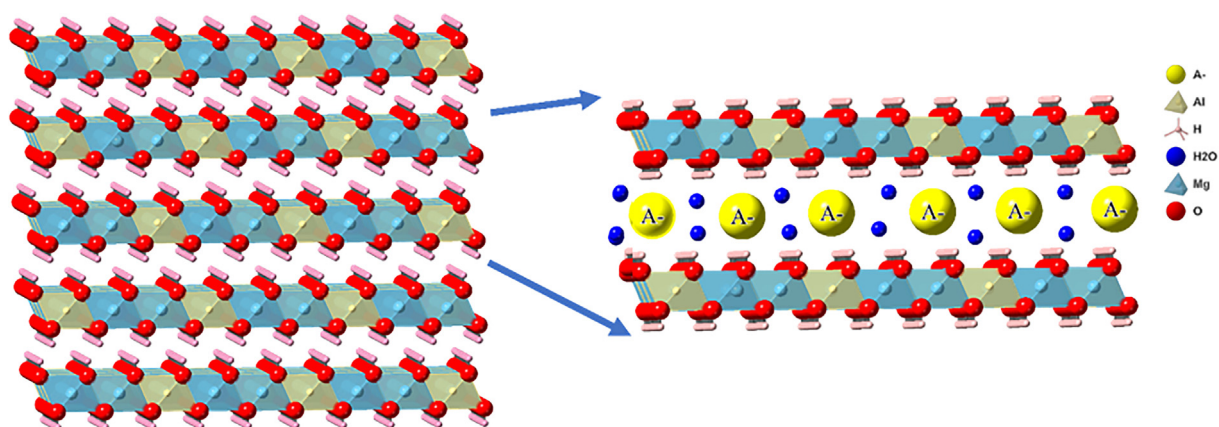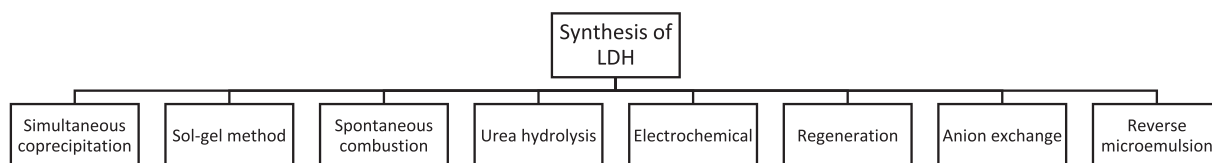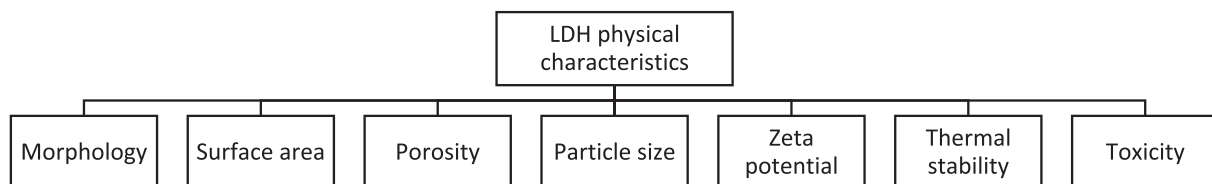**Fig. 1.** Image of a resulting dried LDH after the ultimate step of synthesis.

Fig. 2. Layered double hydroxide structure representation.

F.D. Velázquez-Herrera, Y. Zarazua-Aguilar, A.S. Garzón-Pérez et al.

MethodsX 13 (2024) 102912

Scheme 1. Synthesis methods of the LDH.

Scheme 2. LDH physical characteristics.

Regarding the anions that can be part of the LDH structure, there is practically no limits in terms of sizes and charges. Commonly, the typical compensation anions are, in order of affinity, $CO_3^{2-}>SO_4^{2-}>OH^->F^->Cl^->Br^->NO_3^->ClO_4^-$ [35–38]. Many other inorganic and organic anions, or even compounds with negative charge density, have been reported in the literature, such as $^{131}I^-$ [39], hemoglobin [40], DNA [41], siRNA [42] or papain [43]. In normal conditions of LDH synthesis, it is difficult to obtain a solid free from CO_3^{2-} cations considering that its formation is generated from the atmospheric CO_2 absorbed on the basic reaction medium. LDH free of carbonates are normally obtained only when synthesized in inert atmospheres. Recently, Velázquez-Herrera and Fetter [3] reported the possibility to form LDH containing a heterogeneous distribution of two different type of anions, these materials finding important applications in the pharmaceutical field as a vehicle of successive drug-liberation.

Pure LDH are often only obtained when the M^{2+}/M^{3+} metal molar ratio is between 2:1 and 4:1 (or $0.2 < x < 0.33$) [44]. Exceptions have been recently reported concerning metal molar ratios of 0.5 [45], 1 [46], 5 [47], 6 [48], 7 [49], 8 [50], 9 [50], and 10 [48], but, in many cases, the presence of other compounds as impurities were detected. The pH of the synthesis medium also influences the formation of pure LDH. The optimal pH values run from 8 to 10, depending on the LDH composition, but pH's lower than 6 or higher than 10 have also been reported [51–53].

LDH synthesis methods, Scheme 1, include simultaneous coprecipitation, sol-gel method, spontaneous combustion, urea hydrolysis, electrochemical, reverse microemulsion, among others [54–56]. The most widely used method is simultaneous coprecipitation, due to its process simplicity and the stoichiometric control of the cations integrated to the structure, as well as its reproducibility [54].

The simultaneous coprecipitation method consists of a slow addition of a mixed solution of divalent and trivalent metal salts in adequate proportions with a second solution (alkaline solution) into a reactor containing water at a selected pH value, followed by a hydrothermal treatment of crystallization, filtering, washing, and drying [51]. Commonly, the hydrothermal treatment is used to increase yields, to promote the LDH crystallization and to control the crystal sizes. Among the hydrothermal treatments, the conventional method of stirring the reaction mixture at specified times and temperatures, is the most used [57], but much more current crystallization techniques such as microwave irradiation [58,59] and ultrasound [21,60], or simultaneous irradiations (ultrasound/microwave) [61], have improved the production of these materials. Currently, there has been a trend towards developing environmentally friendly preparation methods for LDHs, which offer significant advantages in terms of solvent usage, processing times, and environmental contamination. These methods avoid the demanding conditions such as high energy consumption and the need for working under an inert atmosphere. These techniques include: mechanochemistry [62], electrocoagulation method [63], and one-pot method [64].

After the synthesis process, most of the time, LDH are calcined to improve some properties, such as dielectric and conductive properties [65], the specific surface areas or to generate active sites [66]. Calcining at temperatures higher than 600 °C, the LDH become crystalline spinel-like oxides, but, at temperatures between 400 and 600 °C, mixed oxides or double metal oxides are formed [2,67,68]. One of the most important characteristics of the mixed oxides occurs when they are placed in contact with anions in water solution, giving rise to a LDH structure regeneration process known as the "memory effect" [69-71].

Regarding some physical characteristics of LDH (Scheme 2) such as morphology and texture they can be controlled mainly by the conventional coprecipitation method followed by hydrothermal treatment with microwaves, ultrasounds and combined treatments have been described. For example, the increase in area follows the trend of microwave < ultrasound < conventional [21,72,73]. Concerning the anion exchanger properties, LDH have an anion exchange capacity between 200 and 450 mmol/mg, which depends on the metallic cation ratio [74]. Another important property of LDHs is their particle sizes. Nanosized particles can be obtained from the delamination of the material in colloidal form [75,76]. Zeta potential of the LDHs plays an important role in the measurement of charge density and their stability in water as a function of pH [76] having an elevated positive surface charge in the range of +10 to +75 mV [77,78] in pHs between 4 and 12. However, it has been reported that it can become negative at pHs above 10 [79] or by electrostatic phenomenon [80]. The LDH thermal stability depends on the chemical composition which is dependent on the
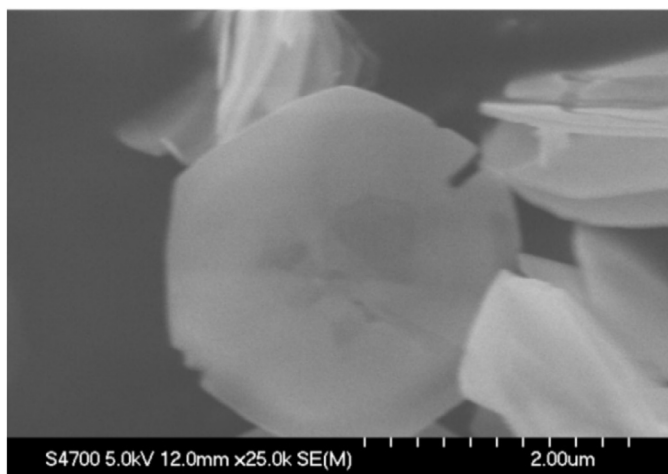
Fig. 3. SEM image of a hexagonal-shaped crystallites of Mg-Al LDH. Copyright 2011, reproduced with permission from reference [86].

cation nature and content in the lamellae as well as the nature and content of the interlayer anions [74,81,82]. Regarding to the LDH toxicity, that containing heavy metals such as Cr or Co usually present some cytotoxicity, but the major common compositions based on Mg and Al have no toxicity [83,84] and, contrarily, they have some benefits to the human health, for example, the solid milk of magnesia commercially available pills used to neutralize stomach acidity [85].

Morphologically, LDH crystallites have often a hexagonal form of stacked flakes [86], as shown in the micrograph taken at X25000 (Fig. 3). Also, LDH particles in the form of three-dimensional ordered microporous [87] or spherical shapes have been synthesized [88]. Also, rounded nanoparticles in colloidal systems have been reported [75,89].

Considering all aspects of the LDH materials mostly related to the design or modification of the chemical composition and structure, besides being environmentally friendly [90] and nontoxic to humans [36], they come into a class of multifunctional materials able to be used in many applications such as catalysts [10,91–94], adsorbents of pollutant compounds [95], antibacterial materials [96–99], containers for drug delivery [100], medical applications [101,102], analytical chemistry [103], among others [104,105]. On the other hand, they are currently being applied for environmental benefit, such as Fenton-based oxidation processes for water and wastewater treatment [106], degradation of organic and inorganic pollutants [107,108] and conversion of biomass-derived molecules [109].

In addition, considering the chemical composition and structural versatility of the LDH materials, the development of a new family of sustainable inorganic composites involving two inorganic materials has recently been reported in the literature. Usually, composites are prepared combining materials with different properties to obtain, for example, an acid-basic material, or to enhance the materials specific surface areas to improve their adsorption or catalytic properties [110]. Briefly, the presence of LDH as a component of composites forming LDH/inorganic compound composites makes it possible to obtain special materials with enhanced physicochemical and textural properties, and thus, being able to be applied in many fields from industrial to quotidian life.

In view of this, the present systematic review will disclose the principal inorganic compounds employed in the formation of LDH/inorganics composites, their synthesis methods, and the characteristics and properties resulted from their combination. Finally, it highlights the given applications of these materials. This review would be highly beneficial in encouraging future research by novice researchers seeking to delve into the field of clays and their combination with inorganic materials, thereby generating new materials that could have very innovative applications. Thus, this review stands out from others by exploring synthesis methodologies used to produce inorganic composites and explores their applications in science and technology with environmental sustainability in mind.

Composites description

At the present, the inorganic materials that have been combined with LDH to form composites are SBA-15, zeolite, hydroxyapatite, titania, montmorillonite, vermiculite, halloysite, kaolinite, sepiolite, laponite, palygorskite, silica, alumina, magnetite, and some metallic salts, as summarized in Scheme 3. Some brief description of each one, as well as the characteristics and applications of the resulting LDH/inorganic composites are described below:

LDH/SBA-15 composites

One type of mesoporous silica material is known as SBA-15, labeled by its developers and means Santa Barbara Amorphous Number 15 [111]. It is a highly ordered hexagonal mesoporous silica structure synthesized by using commercially available block-copolymer surfactants in strong acid media [112]. It possesses a specific surface area larger than 700 m^2/g and a 0.8 cm^3/g of pore volume [113] in two-dimensional hexagonal (space group p6mm) silica-block copolymer mesophases with unidirectional mesoporous channels having 6.0 – 7.0 nm of diameter [111,112]. Some characteristics that make this material unique are the high thermal and
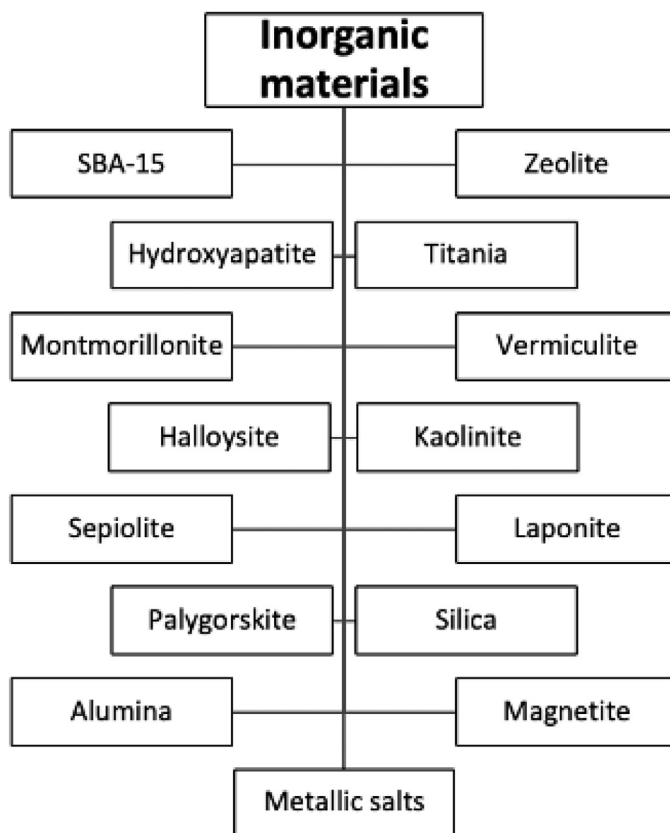
Scheme 3. Inorganic materials that have been used to form composites with LDH.

chemical stability, high specific surface area, and homogeneous pore sizes [114]. Furthermore, the SBA-15 material is of a nontoxic nature, presenting excellent biocompatibility and biodegradability, in addition to the facile functionalization with different organic-inorganic groups [115–119]. All these characteristics make SBA-15 an attractive material to combine with LDH to improve the properties of both materials.

The synthesis process of this type of composites involves many methods which are described below:

1. The most commonly used method is known as *in situ* method [113,116,117,120–124]. It consists of dispersing a certain amount of calcined SBA-15 in water. Subsequently, the reactants to form a LDH are added to the SBA-15 aqueous dispersion followed by a hydrothermal treatment with microwaves, ultrasound, or by a conventional method to promote the LDH crystallization. Finally, the obtained solids are filtered, washed with deionized water, and dried. This method promotes the formation of LDH particles located inside the SBA-15 pores and over its external surface [117,120].
2. Another synthesis method consists in preparing the LDH gel separately, mixing it with a SBA-15 dispersion, and then treating the mixture with ultrasound to promote the integration of one material with the other [125]. The resulting composites were formed with different LDH proportions, but, in all cases, the LDH component was dispersed in the form of nanocrystals over the external surface of the SBA-15 material. No destruction of the SBA-15 framework was detected.
3. Another synthesis method, known as gel mixing method, is based on preparing a gel of both compounds, mixing them and bringing under a crystallization stage [126,127]. The gels mixing method results in composites with a homogeneous distribution of both components.
4. The simultaneous synthesis method comprises preparing an SBA-15/LDH composite by mixing the Pluronic P123 with the LDH precursors salt solutions in diluted HCl solution, and then adding the TEOS reactant. The resulting mixture is then stirred, followed by a conventional crystallization stage [118,128,129]. This method generates composites with uniform distribution of the LDH crystals incorporated into the pores of the SBA-15 component. Fig. 4 shows the TEM micrographs of the composite obtained by this method. In it, the hexagonally arranged cylindrical pores of SBA-15 can be observed, with no evidence of LDH presence, suggesting that it is located inside the pore channels [118].
5. One more method is by the addition of a magnesium methoxide solution on a previously synthesized Al-SBA-15 and stirring for a short time [130]. The resulting composite was formed by flake-shaped SBA crystallites, while the LDH component remains as crystallites supported on the surface of the SBA-15 particles.
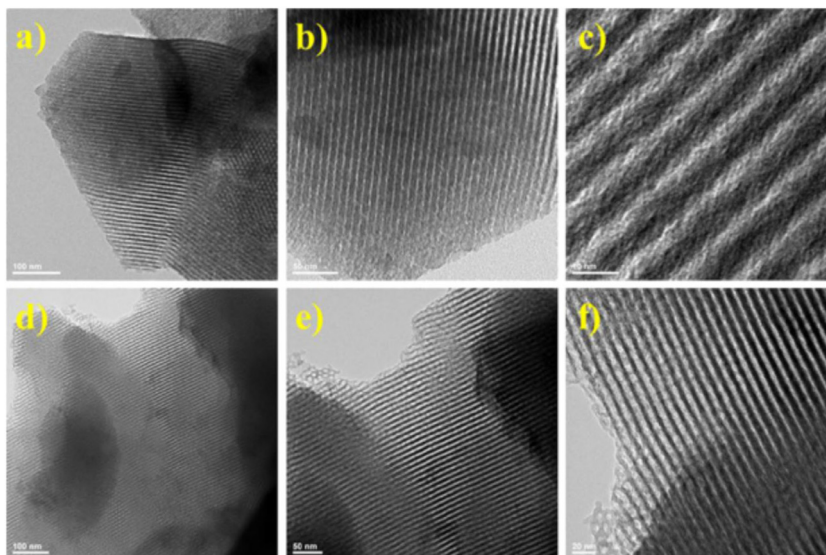
Fig. 4. TEM micrographs of $(a - c)$ SBA-15 and $(d - f)$ MAZ-1/SBA-15 nanocomposites. Copyright 2019, reproduced with permission from reference [118].

All these synthesis methods generate acid-basic composites with improved LDH textural properties. This kind of composites have been used as adsorbents of CO_2 [116,120], water vapor [110], BTEX [113], heteropoly acids [117], organic compounds [113,127], and heavy metals [121]. In catalysis, they promote many reactions such as chromenes [125] and pyrans [117] syntheses, biodiesel production [118] and hydroisomerization of olefins [126].

LDH/zeolite composites

Zeolites are natural or synthetic crystalline aluminosilicate microporous solids having the general formula $M_{x/n}(AlO_2)_x(SiO_2)_y$, where n is the oxidation number of the cation M, x and y are the total number of tetrahedra per unit cell, and y/x the Si/Al atomic ratio [131,132]. Zeolites are formed by a three dimensional negatively charged framework structure built from TO_4 tetrahedra (with $T = $ Si, Al, P, Ge, B, Zn, Mg) in structures that enclose channels and/or cavities with nanometric diameters [131]. These cavities are connected by channels that contain hydrated cations, which neutralize the negative charged zeolite framework [133]. Due to their regular microporous structures, zeolites have "molecular sieving" properties [132,134,135].

Zeolites can be classified according to different criteria: the dimensions of the pore apertures [132], the dimensionality of their channels, and the possibility to modify the hydrophilic character [136]. Their synthesis can be carried out under different conditions, with the Si/Al ratio, the water amount, the cations type, the temperature, pressure, and time being the most frequent variables. With these variations, different kinds of zeolites with different compositions, crystal sizes and morphology can be obtained [131].

Commonly, zeolites present very high surface areas and pore volumes that can host a great amount of reversibly adsorbed water. Furthermore, due to their cationic exchange property, the cations can generally be easily exchanged for others of a similar nature [134]. Besides, the cation exchange properties of zeolites can also be useful in terms of catalytic applications [137]. The replacement of the original cations by protons or transition metals results in the creation of strong acidic sites for acid or redox catalyzed processes [131].

Considering that zeolites are acidic solids [138], the combination with a material with basic character, such as LDH, could result in interesting composites having the dual acid-basic character. The most common zeolites used in combination with LDH are ZSM-5, Y, A, and X.

The LDH/zeolite composites can be synthesized by different methods. They are:

1. Synthesizing and crystallizing LDH on a zeolite dispersion. In one case, the resulting composite are constituted by NiAl-LDH lamellae uniformly dispersed over the Y zeolite particles, exhibiting a typical spheroidal rose-like arrangement [139]. In another case, the MgAl or CaAl-LDH are dispersed in the form of small lamellae over a ZSM-5 zeolite. The Fig. 5 shows the morphology of the composite obtained in this latest methodology, where the presence of zeolite is identified by large prismatic crystals, while the LDH is identified by the observation of uniformly flower-like shaped edges [140].
2. Powdered zeolite mixed with the LDH precursor metal salts and then mixed with a NaOH solution [141,142] or a urea solution [143–145], generates composites where the LDH in the form of rough layers covering the zeolite surfaces. The arrangement of the components in the composite is independent of the LDH or zeolite type.
3. Simultaneous synthesis of both components, *i.e.*, mixing the precursor solutions of both components [146]. This method generates a composite where the MgAl-LDH crystals are attached to the ZSM-5 zeolite particles.
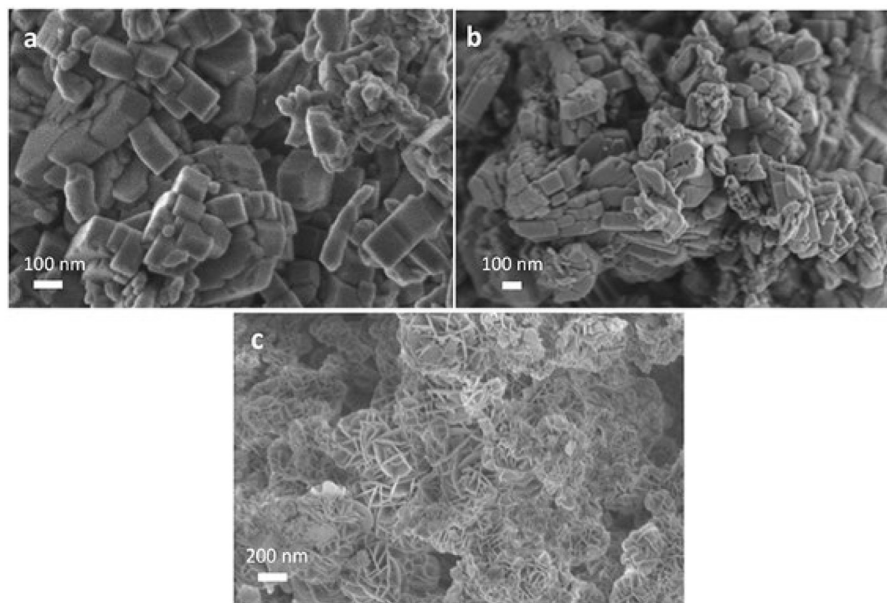
Fig. 5. SEM pictures of (A) ZSM-5 calcined, (B) ZSM-5-NH$_3$, and (C) LDH/ZSM-5 composite. Copyright 2019, reproduced with permission from reference [140].

4. Mixing the powdered zeolite and the powdered LDH in water, evaporate the solution to form a dry gel and treat it with water steam [146]. The resulting composite exhibits MgAl-LDH crystals on the surface of the ZSM-5 zeolite particles.

5. Direct synthesis of LDH/zeolite from red mud waste of the alumina obtention process [147] or mine waste from the production of copper [148]. This method favors plate shaped crystals of LDH located on the LTA zeolite surface.

6. Mixing the powdered zeolite with non-crystallized LDH and bringing them under a conventional hydrothermal treatment [149,150]. The resulting solids consist of Y zeolite particles covered by NiAl- or CoAl-LDHs in the form of a flower-like arrangement.

7. Crystallizing LDH and zeolite at the same time from a mother liquor of an X or A zeolite precursor solution [151,152]. This method generates composites where the zeolite exhibits a morphology of cubic crystals with defined edges, while the LDH shows a series of hexagonally plate-like particles as shown in the Fig. 6 [152].

8. Dipping zeolite particles on a colloidal suspension of LDH and treating with ultrasound irradiation [153] or conventional hydrothermal treatment [154]. The resulting composites are constituted by monolayers of MgAl-LDHs located on the ZSM-5 zeolite surface.

9. Mixing a dispersion of LDH with a dispersion of zeolite Y, followed by a hydrothermal treatment to well-disperse the materials, generating solids where the LDH is dispersed on the surface of the zeolite [155].

Some of the LDH/zeolite composites have found applications as adsorbers of CO$_2$ [140,151,154], toluene [156] or dyes [147]. As catalysts, some of them are used in CO$_2$ methanation [139], cyanobacteria pyrolysis [143], bio-oil production [144], reduction of NOx [150], Glaser Homocoupling reaction [155] and m-phenylenediamine obtaining [149]. In addition, some LDH/zeolite composites catalysed reactions of synthesis of dyes [141], benzoin ethyl ether [142] and 5-hydroxymethylfurfural [146].

LDH/hydroxyapatite composites

Hydroxyapatite (HAp) or hydroxylapatite [157] is the most usual form of calcium phosphate biocrystal [158] with a hexagonal structure (P63/m space group) and the lattice parameters a = 9.37 Å and c = 6.88 Å [159]. It is formed by calcium, phosphate and hydroxyl groups according to the formula Ca$_{10}$(PO$_4$)$_6$(OH)$_2$ having a Ca:P molar ratio of 10:6 [157]. In this material, the hydroxyls can be replaced by other anions such as Cl$^-$ or F$^-$, originating compounds named as chlorapatite or fluorapatite [157]. Otherwise, they can also retain cations such as Co^{2+},Cd^{2+}, Sr^{2+}, and Pb^{2+} in their structure if these cations are present in acidic solutions [158,160,161].

HAp is the inorganic component of mineral bone that can be successfully obtained from various sources, such as mineral rocks or animal bones. They can also be obtained from biogenic products such as eggshells and mussel shells, among others [159]. Furthermore, it is the most attractive material for bone implantation, due to its similarity in composition with human bones.

Synthetic HAp can be produced by different methods, such as co-precipitation or sol-gel followed by a crystallization step that consists in a treatment with ultrasound, microwave irradiation, or by conventional heating [162,163]. The co-precipitation method with calcium and phosphorous precursor solutions at controlled temperature and pH is the most used [164]. By varying the synthesis
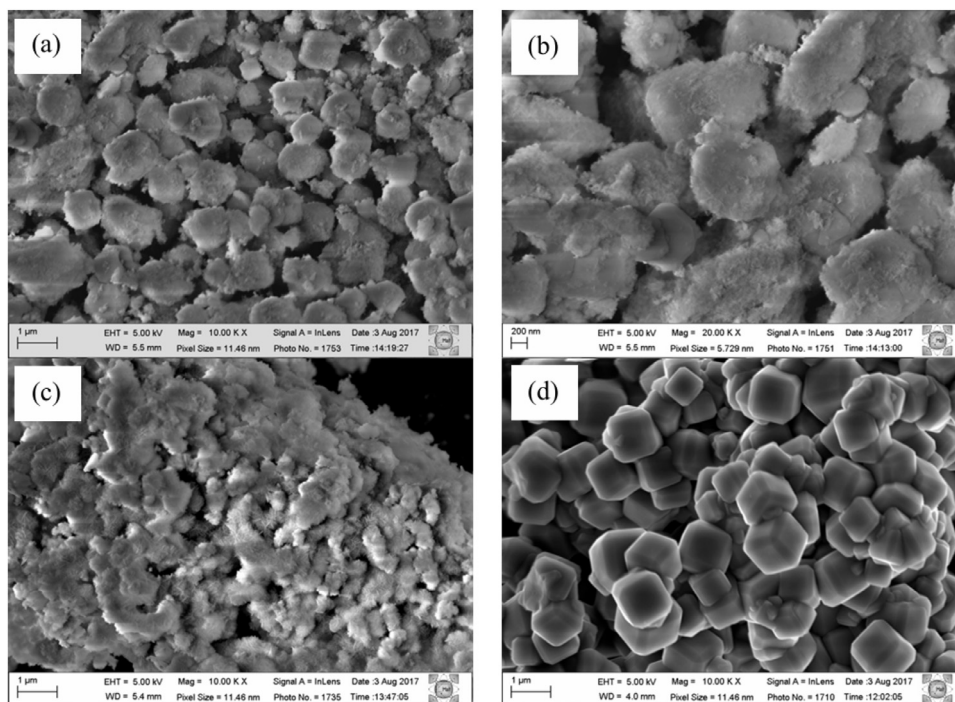
Fig. 6. Micrograph of the composite ZAHD (a,b) material compared to zeolite A (c) and LDH (d). Copyright 2019, reproduced with permission from reference [152].

methods and their conditions, HA with different characteristics can be obtained, thus originating materials with high surface area and porosity, so promoting their adsorption capacities. Also, the acid-base character, thermal stability and mechanical resistance can be adjusted by changing the synthesis conditions [157,159,161,165].

Considering this, hydroxyapatite is an excellent material to form composites with LDH, improving the characteristics and properties of both initial materials.

LDH/HAp composites can be obtained by different methods, as summarized below:

1. Co-precipitation of HAp over a dispersion of powder LDH [166]. By this method, the formation of large HAp particles over a crystallized LDH is obtained.
2. Crystallizing LDH on previously synthesized HAp [71,166,167]. The resulting composites are formed by small LDH particles well dispersed over the HAp surface.
3. Simultaneous synthesis of both materials by mixture of the precursors [90,166,167]. The resulting composites are characterized by a homogeneous combination of very small particles of both components forming a cloud-like network arrangement.
4. Simultaneous hydrothermal treatment of the gels of LDH and HAp [164,168–170]. The resulting composites present micrometer-sized agglomerates of poorly crystallized LDH and HAp. The Fig. 7 shows the micrograph of the composite obtained by this method, where low crystallinity is observed with particles ranging in size from 1 to 5 nm [169].
5. Mechanical mixture of nano-powder LDH with a dispersion of powder hydroxyapatite [2,171]. This method generates a physical mixture of both materials in a heterogeneous distribution of particles, where the LDH component is dispersed in the form of small particles located in the intergranular spaces of the large HAp grains.
6. Mixing the HAp with LDH gels, followed by a hydrothermal treatment [172,173]. By this method, the HAp particles are covered by sheet-like crystals of LDH uniformly dispersed in terms of quantity and size.
7. Generating HAp over the LDH by reacting Na_2HPO_4 with CaFe-LDH [174] or Na_3PO_4 with CaAl-LDH [160,175,176]. By this method, the LDH surface is covered by needle-like crystals of hydroxyapatite distributed uniformly over the lateral surface of the platelet-like LDH particles.

Thus, the composites constituted by the combination of LDH with HAp result in multi-functional materials [166] that have found many applications in different fields. As adsorbents they are used in $(Cr_2O_7)^{2-}$ [164], F^- [71], Cl^- [167,177], $(UO_3)^{6-}$ [168,175], PO_4^{3-} [174], or dyes [2] retention. They also present properties as drug delivery carriers [160] and as catalyst in the transesterification of soybean oil [169,171,178], oxidation of glucose [176], chromene synthesis [166] and a cross-linking reaction of epoxy resins [172]. They also show high activity as antibacterial materials [90] with potential use as implants for osteo-reconstitution that prevent bacterial infections at the same time [173].
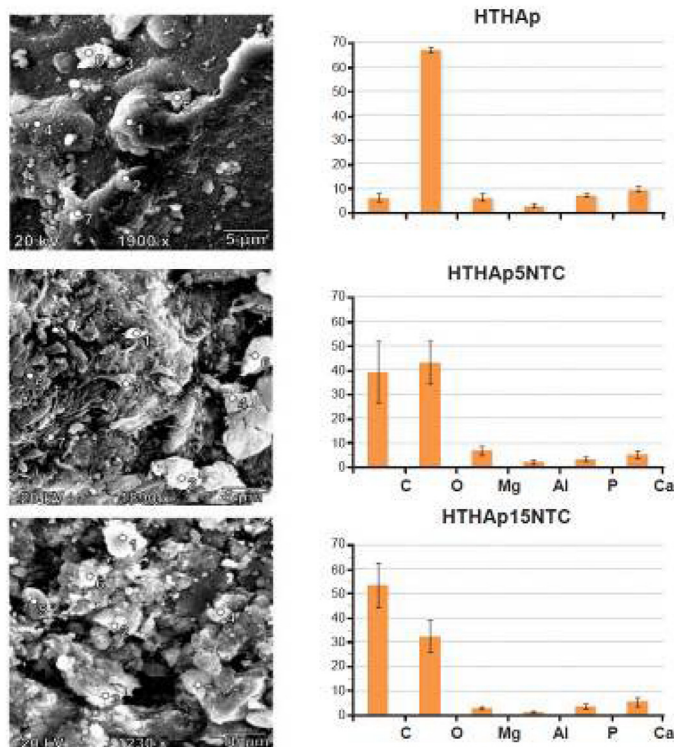
Fig. 7. Micrographs and semi-quantitative elemental analysis (at%) of the synthesized LDH-HAp samples (LDH-HAp). Copyright 2018, reproduced with permission from reference [169].

LDH/titania composites

Titania (TiO$_2$) is a low cost, nontoxic, low cost, negative conduction band, naturally abundant, and highly stable compound with a semiconducting property [179,180]. This material has been used in photocatalytic processes, for example, for removal and oxidation of various organic pollutants in waste water [181]. TiO$_2$ crystallizes in three systems: anatase, rutile, and brookite. Its photocatalytic activity is attributed to the presence of a wide band-gap from 3.03 eV for the rutile phase, to 3.18 eV for the anatase phase. In particular, the anatase form is excellent for photocatalytic processes, although this material suffers from electron recombination [182]. Titania also presents a great ability to produce powerful oxidants (holes in the valence band) and reductants (electrons in the conduction band) by absorbing photons from the UV region [182,183]. Besides, titania is a high hydrophobic material [184] and, due to this property, the reactions catalyzed by titania have the disadvantage of reducing its catalytic activity when being processed in an aqueous medium [185]. Also, as TiO$_2$ nanoparticles are difficult to separate from the reaction medium, it has been proposed by many researchers [184] to immobilize it on appropriate materials, such as LDH, to promote their separation by filtration after a photocatalytic process. Supporting nanoparticles of TiO$_2$ also contribute to have better regeneration of the photocatalyst [185], by hindering the recombination of e$^-$-h^+, and thus, increasing the efficiency of the photocatalytic degradation of contaminants [184,186].

Several methods have been proposed to combine titania with LDH:

1. Simultaneous co-precipitation of both components from the metal salts solutions in the presence of a basic solution, followed by a conventional crystallization step [181,182,184,186–194]. In general, the resulting composites are characterized by a random dispersion of TiO$_2$ nanoparticles on the LDH surface forming agglomerates with a spongy appearance.
2. Mixing powder LDH previously dispersed in a basic solution and mixed with powder TiO$_2$, followed by a hydrothermal treatment to well-disperse the materials [185,195,196]. A variation of this method consists of mixing the powder TiO$_2$ with a LDH in its colloidal form [197], generating solids where the titania phase is dispersed on the surface of the LDH.
3. Mixing a titania dispersion with a LDH gel and treating the mixture by conventional heating [179,198–200]. This method generates the agglomeration of LDH nanoflakes on TiO$_2$ nanotube particles, as shown in the Fig. 8 [179].
4. Crystallizing titania on LDH previously dispersed in water or ethanol [183,201–203]. The resulting composites are formed by TiO$_2$ nanoparticles randomly distributed on the surface of the LDH platelets.
5. Mechanical mixture of LDH with TiO$_2$ powders by a kneading method [204]. In this case, a composite was constituted by a heterogeneous mixture of both components.
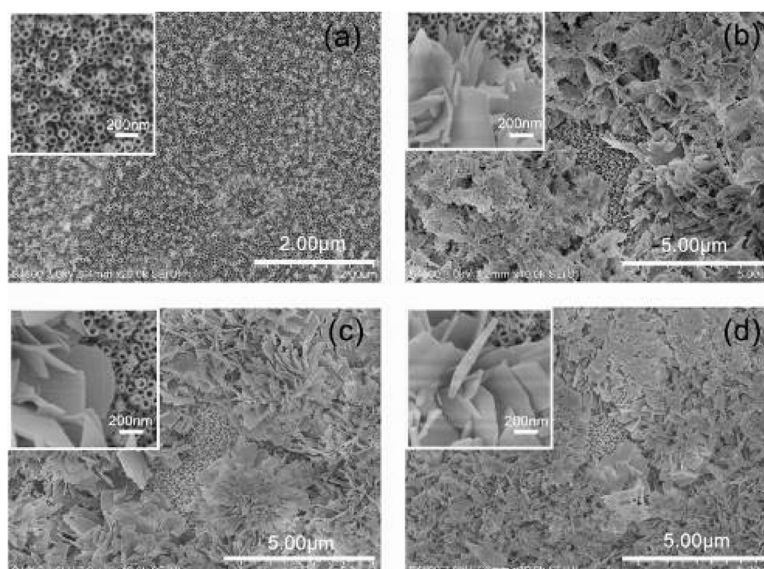
Fig. 8. (a) SEM image of TiO_2, (b, c, d) SEM images of LDH/TiO_2 composite. The figures in the upper left corner are magnified views. Copyright 2018, reproduced with permission from reference [179].

Generally, all synthesis methods lead to LDH/TiO_2 composites having improved properties, useful for photocatalytic purposes such as the photodegradation of dyes [186,188–191,203], drugs [185,205], surfactants [184,187], and organic compounds [181–183,202,204,206]. Furthermore, they have been successful applied in photocatalytic reduction of heavy metals [196], NOx [201], and CO_2 [194,197,207]. Moreover, LDH/TiO_2 composites are active as bactericide materials [195] or in photocathodic protection [179] as well as in photoanode construction [198].

LDH/montmorillonite composites

Montmorillonite (MMT) is a natural clay that belongs to the smectite group (2:1 dioctahedral smectite) [208,209], thus being composed of two layers of tetrahedra silicon oxide combined with a central octahedral aluminum oxi-hydroxide layer [210,211] having the chemical formula $M_x(Al_{4-x}Mg_x)Si_8O_{20}(OH)_4$, where M is a monovalent cation and x the degree of isomorphous substitution which can range between 0.5 and 1.3 [212]. The surface of the MMT is negatively charged due to isomorphous replacement of Mg^{2+} for Al^{3+} in the octahedral layers. This negative charge is usually balanced by alkaline earth cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, etc.) which can be exchanged with inorganic and/or organic polycations [208,213]. The presence of cations create micropores between the layers allowing for the entrance of water [210]. MMT particles are typically plate-shaped with thickness of 1 nm and diameter of 0.2 – 2 μm [214] and their surface is highly hydrophilic [210]. The principal types of MMT clays are named based on the dominant exchangeable cation, for instance, sodium montmorillonite and calcium MMT [209,210].

In many MMT deposits, the MMT clay is found mixed with other compounds such as gypsum, quartz, calcium, and sodium feldspar [($CaAl_2Si_2O_8$), ($NaAl_3Si_2O_8$)] [209,211,213], originating a mineral known as bentonite. To be considered as a bentonite, the percentage of MMT must be greater than 80 % [215]. Depending on the treatment type to modify their structure, bentonites are usually classified into native bentonite, active bentonite (alkaline or acid bentonite), and organic bentonite (presence of organic compounds) [216]. These terms also apply to the mineral MMT.

Considering the excellent physicochemical properties of this kind of clay, such as possessing large specific surface area, high cation exchange capacity, strong adsorptive affinity, low cost, swelling capacity, and low hydraulic conductivity, among others [211,214], MMT or bentonite are suitable materials to combine with LDHs.

The reported LDH/MMT composites synthesis methods are described below:

1. Mechanical mixing of both components is the most used method for generating LDH/MMT composites. This method consists of a mixture of LDH and MMT in powder form, dispersed in water and mixed by stirring [217], or adding the MMT in powder to a LDH dispersion [218]. The method presents some variations of the conditions as specified below:
   1.1. Mixing of both components in the colloidal form and stirring at room temperature [219] or treated with ultrasound [220]. The resulting composites are characterized by a heterogeneous mixing of MMT and LDH particles, this being independent of the treatment used.
   1.2. Formation of mixed thin films of LDH and MMT from colloidal suspensions employing glass [221], quartz [222], and polystyrene [223] as supports. Independently of the support used, the LDH/MMT nanocomposites are characterized by a periodic alternation of MMT and LDH nanosheets. This methodology involves evaporation processes to alternate layers of material, as depicted in the Fig. 9 [221].
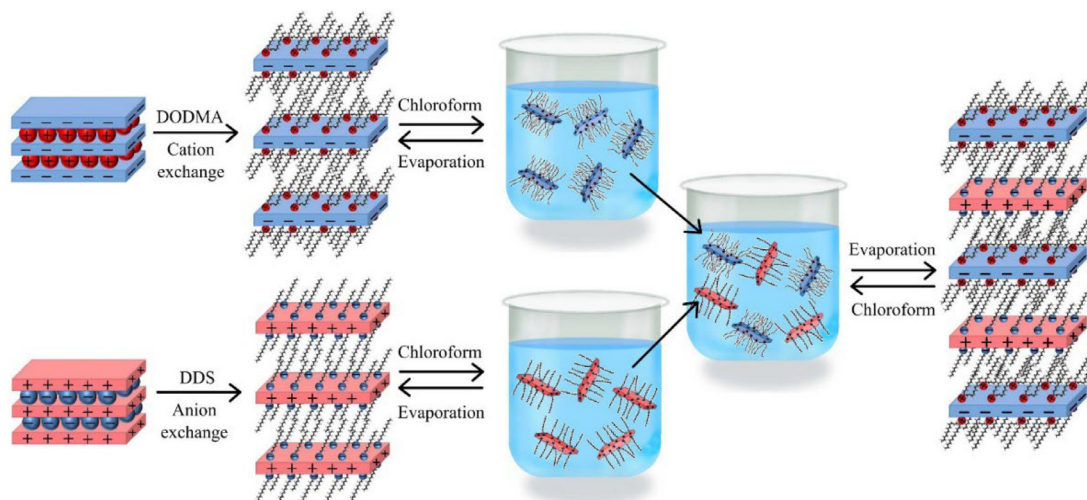
Fig. 9. Schematic illustration of the formation of layered hybrids with periodically alternating positive and negatively charged inorganic sheets. Copyright 2013, reproduced with permission from reference [221].

    1.3. Mixing LDH and MMT delaminated dispersions by stirring. In this case, the resulting composite is assembled layer by layer of each component. Their lamellar structures result in a slight difference in terms of thickness and sizes compared to the initial materials [224].

    1.4. Ultra-stirring of LDH with MMT dispersions [225–227]. This synthesis procedure promotes the formation of LDH/MMT composites characterized by small round plates of LDH well distributed over the irregular large thin flakes of the MMT.

2. Crystallizing LDH over a dispersion of powder MMT. In this case, the metal salts of LDH precursors and a basic solution are added dropwise to an aqueous dispersion of MMT, with some variations in the treatment conditions as described below:

    2.1. Crystallizing LDH over MMT dispersion followed by an ultrasound irradiation treatment [228]. This LDH crystallization mode generates a composite with micrometer-sized particles where no interaction between particles were observed.

    2.2. Crystallizing LDH over MMT dispersion over stirring without hydrothermal treatment [229,230]. The resulting composite is characterized by a corrugated and scrolled morphology, with the LDH crystals being located on the external surface of MMT clay. This type of morphology exhibits the flower-like structure with spherical agglomerates, as shown in the Fig. 10 [230].

    2.3. Coprecipitating LDH on colloidal MMT dispersion without a conventional hydrothermal treatment [231]. The resulting composite is composed by the dispersion of LDH flakes on the MMT surface.

    2.4. Adding colloidal suspension of MMT (in the form of bentonite) to the LDH precursor salts, followed by a conventional hydrothermal treatment [232]. The generated composite is formed by the combination of bentonite nanoparticles homogeneously embedded in the layers of the LDH material, producing a fluffy nanostructure with higher interparticle porosity.

3. Crystallizing LDH by the urea hydrolysis method over a dispersion of powder MMT. The urea hydrolysis method consists of using a urea solution instead of hydroxides as a precipitating agent. Some modifications on the conditions of this method are presented below:

    3.1. The LDH is prepared by the urea hydrolysis over a dispersion of powder MMT. In this composite, the LDH compound is adhered to the bentonite surface and pores [233].

    3.2. The LDH is prepared by urea hydrolysis on a colloidal MMT. The composite is constituted by homogeneously grafted LDH particles on the MMT surface [234].

    The presented LDH/MMT composites can be characterized by being of low cost [233], environment friendly, and having a high adsorption capacity, that makes them useful as adsorbents of phosphates for waste water remediation [226], uranyl ions from natural and polluted waters [229], or to purify waste water from Pb^{2+}, Cu^{2+}, and methyl orange [233]. Other applications of these composites are as antibacterial materials against *Escherichia coli* [217], in corrosion protection [235], and in CO_2/N_2 separation [223].

LDH/vermiculite composites

    Vermiculite is a naturally abundant clay mineral, an inexpensive hydrous phyllosilicate 2:1 composed of the combination of two silica tetrahedral sheets with an octahedral sheet [236]. Generally, this clay is formed by weathering or hydrothermal alteration of biotite or phlogopite [237]. The vermiculite presents negatively charged layers which are neutralized by cations such as Ca^{2+} or Mg^{2+}. This clay exhibits high cation-exchange capacity and high charge density, generated by isomorphic substitution of Si^{4+} by Al^{3+} [236,238]. The vermiculite´s mainly properties are a large surface area, high rehydration, and exfoliation capabilities, and chemical and mechanical stability. Furthermore it is of relatively low cost, as well as harmless to the environment [237,239,240]. A particularity of this material is an easy delamination process which can be accomplished when it is rapidly heated, generating particles in the form
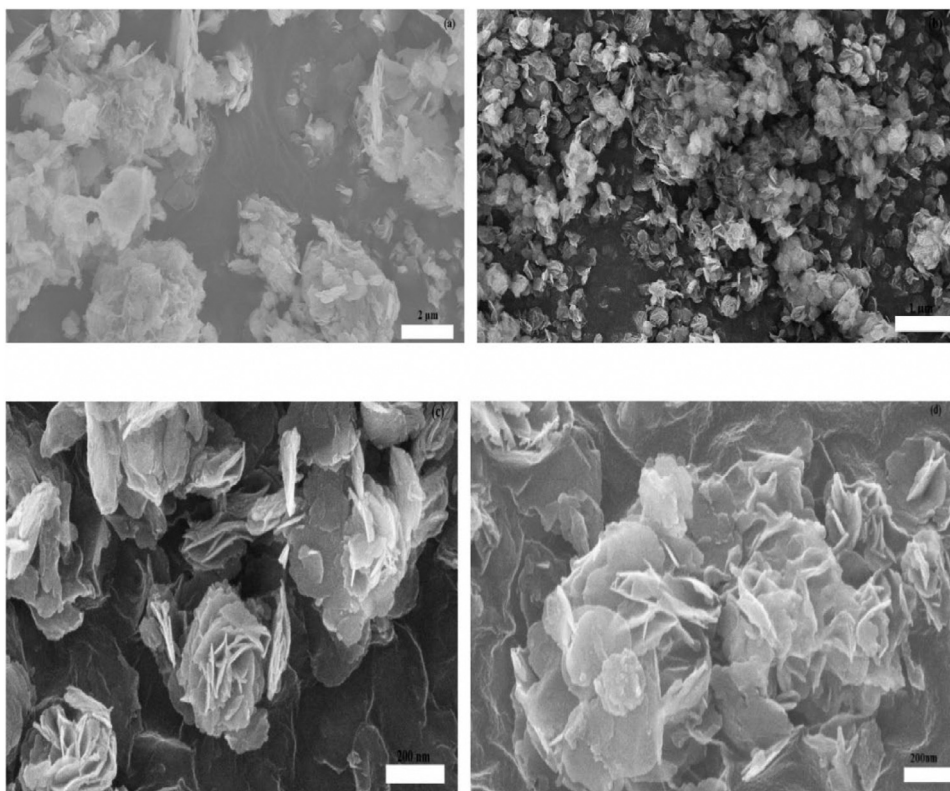
Fig. 10. SEM analysis of S2-LDH (a), S2-MMT (b), S3-MMT (c), S4-MMT (d). Copyright 2023, reproduced with permission from reference [230].

of flakes assembled in an accordion-like mode [239]. According to the vermiculite characteristics and properties, its combination with LDHs to form composites could bring advantages with respect to the original components. The methods to obtain the LDH/vermiculite composites are described below:

1. A particular method involves the use of spent liquor from acid treatment of vermiculite that is alkalinized with NaOH solution to form the LDH [241,242]. The obtained products are composed of small crystals of LDH confined into the pores of the partially leached vermiculite, generating a plate-like structure, which is visualized in the Fig. 11.
2. Impregnation of powder LDH in a vermiculite dispersion [243]. The resulting composites are characterized by uniform sizes of LDH crystals densely located on the surface of the vermiculite component, generating a rough surface and an increased porous structure. Their adsorption capacities are considerably increased.

The composites originating from both methods present high activity as catalysts in the CO methanation [242] or in the adsorption of dyes or heavy metals [241,243].

LDH/halloysite composites

Halloysite is an ecological and biocompatible clay of low cost and high availability that has cation exchange capacity. The most common and interesting type is that of structurally formed aluminosilicate nanotubes (halloysite nano-tubular, HNT) with a chemical formula $Al_2Si_2O_5(OH)_4 \cdot 2H_2O$. Due to its kind of tubular structure this material presents a high surface area and porosity [244,245] and makes it an excellent material to form composites [246]. The LDH/HNT composites synthesis methods are based on:

1. LDHs precursors added over a powder HNT dispersion followed by a conventional hydrothermal treatment [247,248]. The resulting LDH/ HNT composites are composed by nano-sized LDH particles that surround the large nanotubular particles of halloysite, Fig. 12.
2. LDH synthesized by the urea hydrolysis method added on a HNT dispersion [249,250]. In this case, the composite is formed by random particles of HNT embedded within the LDH platelets that looks like a sandwich structure.
3. Mechanical mixing of LDH with HNT. This method generates a composite characterized only by the dispersion of both halloysite and layered double hydroxide particles [247].

The applications of these LDH/HNT composites consist of the adsorption of $(Cr_2O_7)^2$, $(HAsO_4)^{2-}$ and $(SO_4)^{2-}$ from waste water [247,248], and as a lysozyme reservoir for pharmacological bactericide material [249].
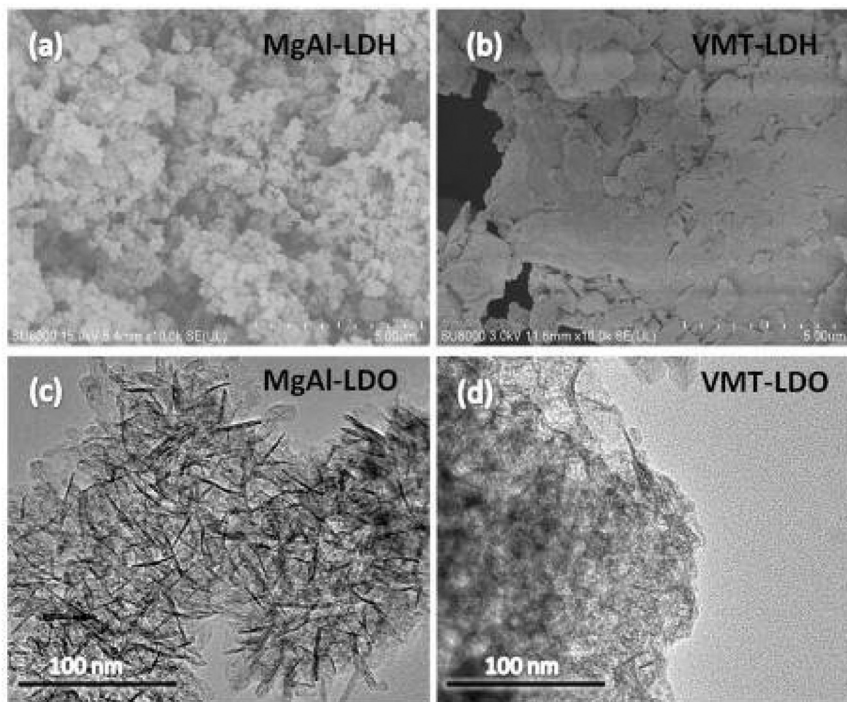
Fig. 11. SEM images of (a) MgAl-LDH; (b) LDH/vermiculite (VMT-LDH); TEM images of (c) MgAl-LDO and (d) VMT-LDO. Copyright 2017, reproduced with permission from reference [242].
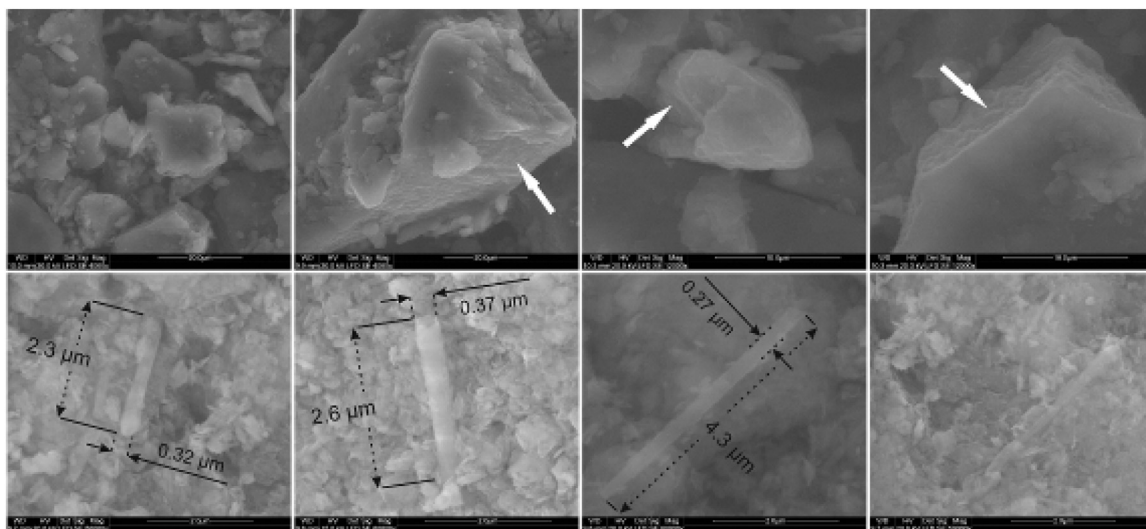
Fig. 12. SEM microphotographs of the LDH sample (4 upper images) and LDH/HNT (Hall-LDH) material (4 bottom images). The white arrows indicate cross sections of the agglomerates with visible stacking of LDH particles. Copyright 2019, reproduced with permission from reference [247].

LDH/kaolinite composites

Kaolinite is a worldwide abundant clay with the chemical formula $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$. Its structure is formed by one alumina octahedral sheet and one silica tetrahedral sheet, classified by a 1:1 type [251,252]. Kaolinite is a natural mineral with a high adsorption capacity [253] being a low cost and environmentally friendly material, thus being a good candidate to combine with the LDH.

The only reported method to combine kaolinite with LDH is by crystallizing LDH from salt precursors on a kaolinite dispersion [254–256]. The resulting composites are formed by loosely packed aggregates of LDH clusters with enlarged inter-particle voids of the kaolinite, producing high porosity, necessary to catalyze the Fischer–Tropsch synthesis [256], or adsorb PO_4^{3-} [254], and $(Cr_2O_7)^{2-}$ [255].

LDH/sepiolite composites

Sepiolite is a natural fibrous mineral clay with a chemical formula $Si_6O_{15}Mg_4(OH)_2 \cdot 6H_2O$ [257]. Its structure is composed of blocks of two sheets of tetrahedral silica that are interspersed with an octahedral sheet of magnesium oxide hydroxide corresponding to the 2:1 type clay. The structure is formed by layers in the form of parallel ribbons with interior channels of 0.36 to 1.1 nm [258,259]. This type of structure leads to a material having a high specific surface area and porosity, becoming an interesting material to be used in catalytic or adsorptive processes. They represent an optimal support of the LDH compounds to enhance the LDH surface areas.

Some methods to generate LDH/sepiolite composites consist of:

1. Adding LDH salt precursors and basic solutions dropwise together to a sepiolite dispersion, followed by a conventional hydrothermal treatment [260,261]. This generates composites where the LDH particles are located on the sepiolite surface with a good distribution.
2. Reconstructing the LDH structure from the LDH derived mixed oxides present in the surface of a sepiolite. Although this method is efficient in the reconstruction of the LDH structure, it does not favor the assemblage of LDH with the sepiolite, resulting in a rather heterogeneous mixture of the two types of nanoparticles [262].
3. The mixing of both components is done layer by layer sequentially under suction vacuum [263]. This methodology generates a porous structure with a uniform platelet-like morphology corresponding to LDH, while Sepiolite exhibits a rod-like structure, as shown in the Fig. 13.

These kind of composites have been used efficiently as catalysts in the photodegradation of methyl orange and methylene blue dyes [261], or in the production of proton conducting membranes [260].

LDH/palygorskite composites

Palygorskite (or attapulgite) is a naturally hydrated magnesium silicate clay mineral with the formula $Si_8Mg_8O_{20}(OH)_2(H_2O)_4 \cdot 4H_2O$ [264]. It is made up of talc-like tapes of 2:1 phyllosilicate units. Each ribbon is connected to the next by the inversion of SiO_4 tetrahedron along a set of Si–O–Si bonds. The rotation of these structural units creates tunnels of 0.64 nm × 0.37 nm [264–266]. In the process of mineralization, some Mg^{2+} ions in octahedral sites of the palygorskite crystal are substituted by trivalent cations (Al^{3+} or Fe^{3+}) creating an excess of positive charges [267]. Properties such as a high specific surface area, low cost, natural abundance, nontoxic nature, and environmental friendliness [268] and a special pore structure make the material suitable for combination with LDH. This material is characterized by having a morphology of rod-like crystal bundles and agglomerates, as shown in the Fig. 14 [268].

Only one synthesis method for this composite has been reported. It consists in adding the LDH precursors salts solutions over a palygorskite dispersion, followed by a conventional hydrothermal treatment [269,270]. This synthesis method leads to the formation of small crystals of LDH located between the fibers of the palygorskite, implying that the spaces between the fibers are partially or completely filled.

The LDH/palygorskite composites have found application as photocatalysts in the discoloration of yellow eosin [269] or as catalysts for ultra-high molecular weight polyethylene [270].

LDH/laponite composites

Laponite is a synthetic clay of smectite type similar to hectorite, characterized by 2:1 layers arrangement in which two tetrahedral silica sheets sandwich one Mg^{2+} containing octahedral sheet [271]. Its chemical formula is $Si[Mg_{5.5}Li_{0.4}H_{4.0}O_{24.0}]^{0.7-}Na^{0.7+}$. This clay exhibits a characteristic distribution of surface electrical charges that promotes its easy dispersion in water [272]. Due to its favorable physicochemical properties such as high cation exchange property and high specific surface area, laponite constitutes an optimal candidate to combine with LDH.

The only method reported to obtain LDH/laponite composites involves the mixture of mixing microemulsions of both compounds by stirring [273–275]. The obtained composites are characterized by a good intermixture composed of very fine platy particles of LDH crystals with Laponite particles as shown in the Fig. 15 [275]. They have been used as catalysts in the total oxidation of toluene [275] as well as in combustion of VOC with good performance [273,274].

LDH/silica composites

Silica (SiO_2) is an inactive material that presents a very high specific surface area and tunable porosity, being thermally and chemically stable [121].
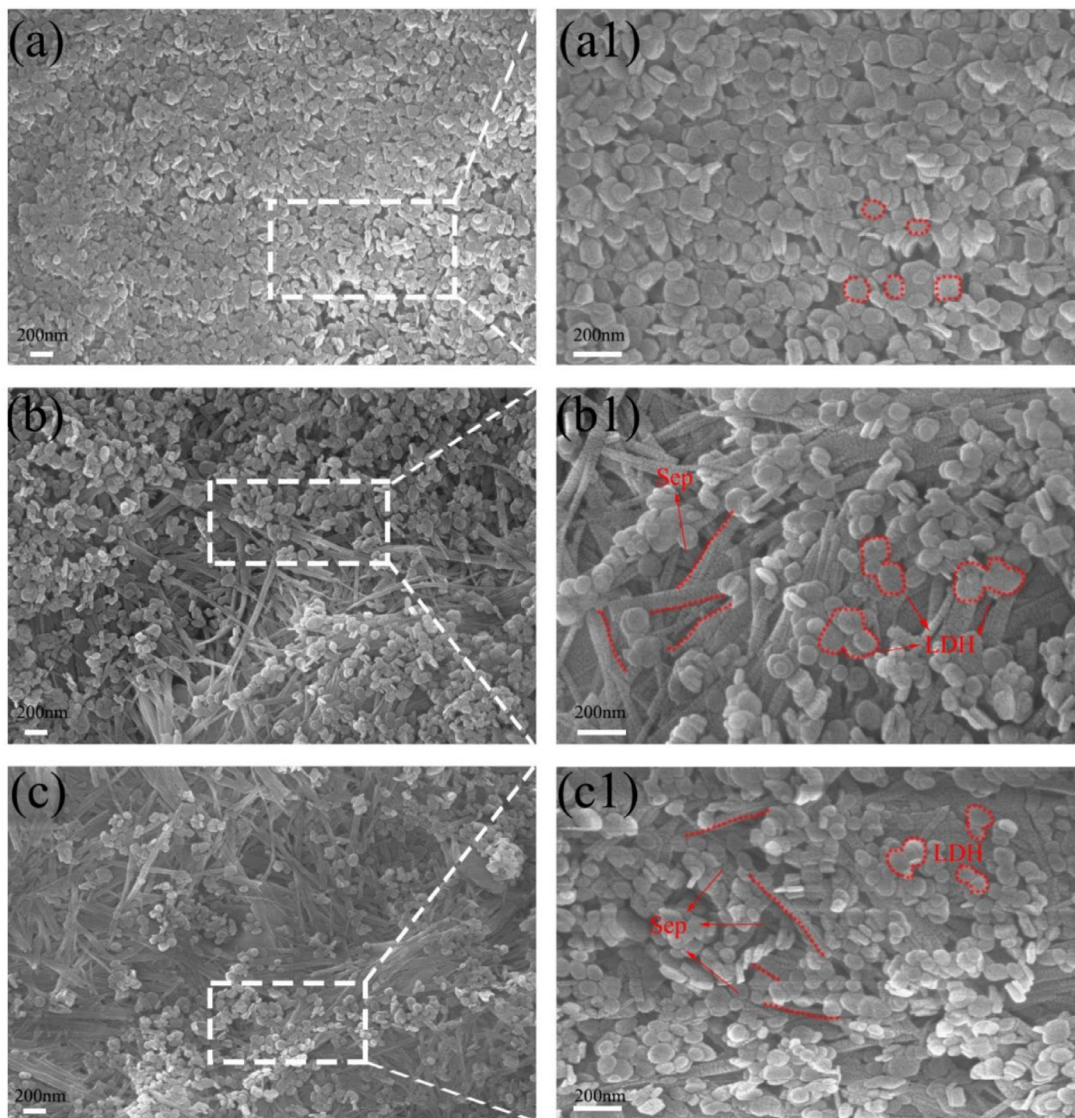
Fig. 13. Scanning electron microscope (SEM) image for LDH-M (a), (a1), M1 (b), (b1) and M2 (c), (c1) of composite membrane surface. Copyright 2019, reproduced with permission from reference [263].

2.12.1. One synthesis method to obtain this kind of composite is by coprecipitating the precursors of LDH on a SBA-15 material followed by a hydrothermal treatment carried out by microwave irradiation [121]. The resulting composite is characterized by nanolayers of LDH well dispersed on the amorphous silica surface. The SBA-15 support is degraded to amorphous silica during the synthesis reaction of LDH formation. This composite has been used as a Cr (VI) adsorbent.

2.12.2. Adding a LDH dispersion over a microemulsion of SiO_2 in a nanodot-coating approach [276]. This composite is formed by uniform SiO_2 nanodots attached on the surface of LDH nanoparticles maintaining a hexagonal plate-like morphology. This composite has been employed as methotrexate delivery systems for diagnostic and therapeutic applications.

2.12.3. Coprecipitating the LDH precursor solutions over a silica nanoparticles dispersion, followed by a conventional hydrothermal treatment [277–279]. This method generates composites constituted by SiO_2 nanoparticles extended on the LDH lamellar surface, showing a denser dimensional core-shell architecture as shown in Fig. 16 [277]. These composites have been employed as nanocarriers for clinical drugs of topoisomerase inhibitor VP16 [277] or of model DNA based on supercoiled plasmid pEGFP-N2 [278] .
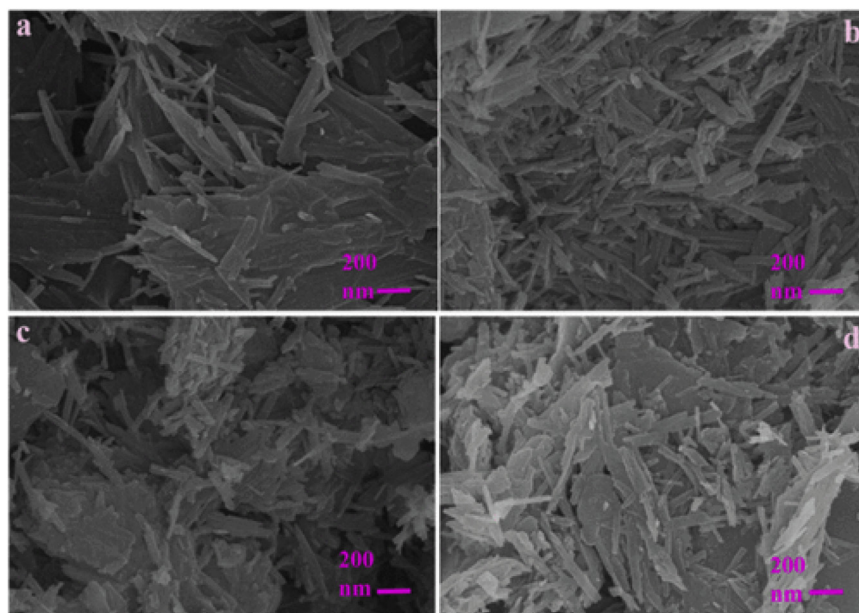
Fig. 14. FESEM images of (a) UAPT, (b) SA(0.01)-APT, (c) SA(0.05)-APT, and (d) SA(0.2)-APT. Copyright 2021, reproduced with permission from reference [268].
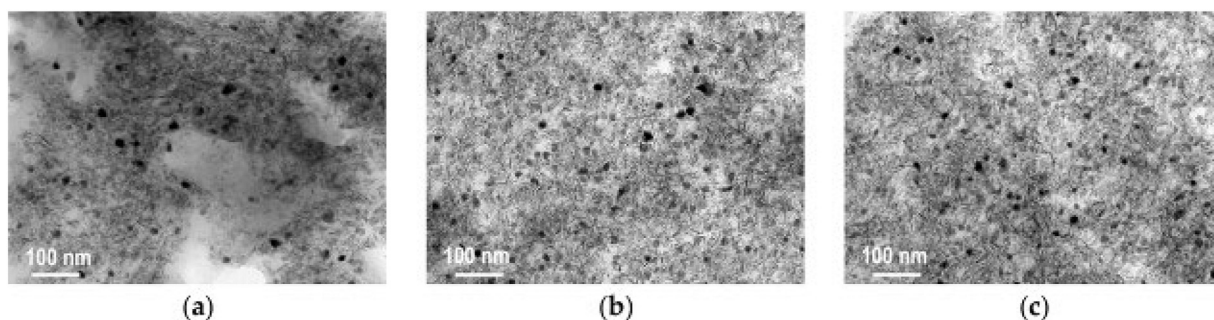
Fig. 15. TEM images of (a) CuMnAl-LDH-Laponite-NaOH; (b) CuMnAl-LDH-Laponite-TBAOH; (c) CuMnZrCeAl-LDH-Laponite composites calcined at 600 °C. TBOAH: tetrabutylammonium hydroxide. Copyright 2018, reproduced with permission from reference [275].

LDH/alumina composites

Gamma-alumina (γ-Al$_2$O$_3$) is a mineral with a high mechanical strength and surface area [280]. It presents high chemical and physical stability, as well as acid/base characteristics. It has been the most commonly used support for various catalysts in the automotive and petroleum industries [281].

1. LDH/γ-alumina composites are generated by coprecipitating LDH from the precursor solutions over a γ-Al$_2$O$_3$ dispersion. The LDH crystallization is promoted by a conventional hydrothermal treatment [282]. In this composite, the LDH component is located on the surface and the pores of the alumina component. This composite has been used as a catalyst in the ultradeep hydrodesulfurization process.
2. Kneading of both materials in dried powder conditions [283]. The resulting composite is characterized by the mixture of both components. It has been employed for H$_2$/CO$_2$ separation [283].

LDH/magnetite composites

Magnetite (Fe$_3$O$_4$) is a natural mineral consisting of a spinel-like structure, its most relevant property being magnetism [284]. Only two synthesis methods have been found describing the LDH/magnetite composites:

1. The first method proceeds by coprecipitating LDH over a magnetite suspension, followed by a conventional hydrothermal treatment. The composite is characterized by having a good dispersion of the LDH crystals on the magnetite surface. It has been used

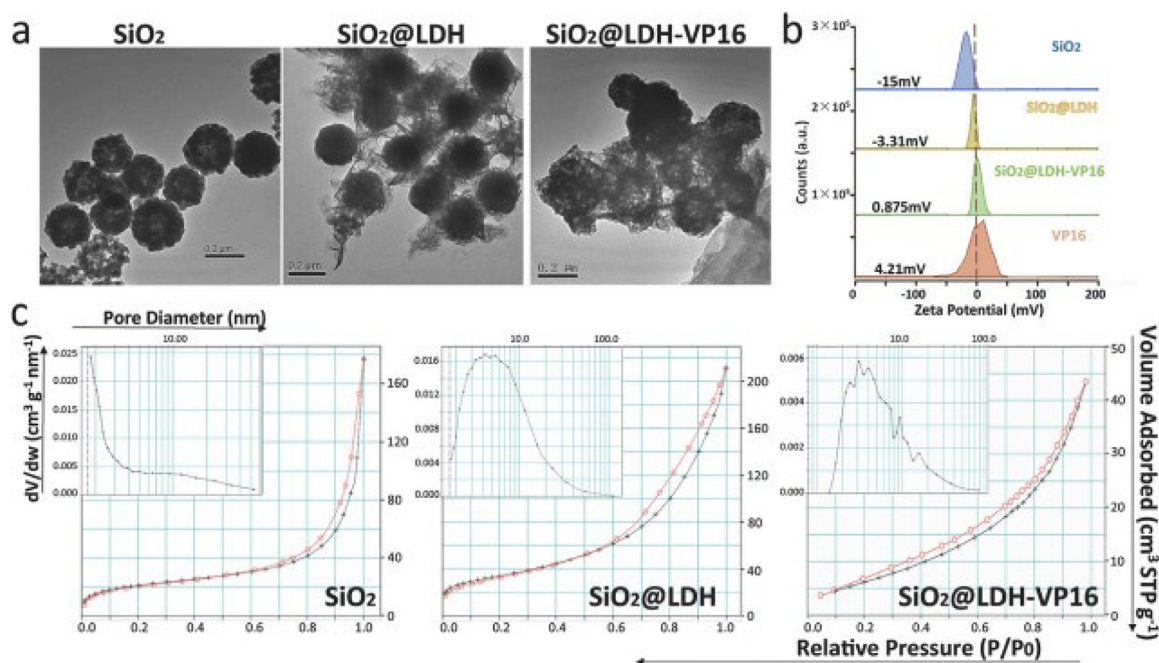**Fig. 16.** a) TEM images of SiO_2, SiO_2@LDH, and SiO_2@LDH- etoposide. b) Zeta potential distribution for SiO_2, SiO_2@LDH, SiO_2@LDH- etoposide, and etoposide. c) N_2 adsorption/desorption isotherms and BJH pore-size distribution curve obtained from the adsorption branch of SiO_2, SiO_2@LDH, and SiO_2@LDH- etoposide. Copyright 2016, reproduced with permission from reference [277].

as photocatalytic material [285], or as a material of controlled drug delivery of salicylic acid [286], l-Dopa [287], or anti-cancer agents such as doxifluridine [288], and 5-Fluorouracil [289].

2. The other method consists of coprecipitating the magnetite over a LDH dispersion, followed by a conventional hydrothermal treatment. The obtained composite presents a very porous structure composed of particles with rough surface and irregular edges. It has been applied in fluoride adsorption ions for water potabilization [290].

LDH/salts composites

Such composites are formed by the impregnation of some simple or complex salts on the LDH material. The salts used are K_2CO_3 [291], KF, KI, KNO_3 [292], $CoFe_2O_4$ [293], and Bi_2WO_6 [294]. In all composites, the LDH surface is partially covered by nanocrystals of the salts.

These composites have been employed as catalysts for biodiesel production [291,292], as photocatalyst in the degradation of rhodamine B, safranine T, and tetracycline in the presence of visible light [294] and as a methyl orange dye adsorber [293].

Waste management of applied inorganic-LDH compounds

Another important aspect to consider is the management of waste after using LDH. One of the ways to reuse LDH waste is its application as an electrocatalyst in direct methanol fuel cells [295]. Some authors report that LDH nanocages can be regenerated successfully using a solvothermal method when adsorbing anionic organic contaminants onto clay [296]. However, if heavy metals such as arsenic are absorbed, the first step for regeneration involves desorbing them from the clay with alkaline and salt solutions or mixtures of solutions, followed by a second step to regenerate the clay with NaOH [297]. Regarding reuse cycles, some authors demonstrate that after four cycles of reuse, the absorption efficiency of LDH is less than 10 %, whereas when LDH is functionalized with absorbents, the efficiency is more than 80 % [298].

As described, among the many important properties that LDH possess is the ability to regenerate through various pathways such as ion exchange or structure reconstruction. Inorganic-LDH compounds have also been characterized by retaining these characteristics, especially in their application as adsorbent materials. Although high operational efficiency has been achieved in its different applications, it is necessary to take into account the applicability on a larger scale, and the simple and lower cost preparation of LDH composite materials [299]. Furthermore, waste recovery offers a sustainable approach to mitigate the adverse environmental impacts of materials and has been recognized as one of the optimal routes to achieve and implement circular economy strategies [300]. One of the challenging sources of industrial waste is spent adsorbents saturated with various types of contaminants. Therefore, currently there are still some limitations for its reuse after the various cycles of operation and it is necessary to consider the challenge

of applying new methods for the regeneration of LDH materials, which should be the motivation for future research to seek advanced modifications, easier to operate and environmentally friendly in the industry.

Finally, when it comes to using materials based on LDH, their final use depends on the final state of the material. If its structure has collapsed, the clay can be used as filler, or its structure can be regenerated through a chemical process. If the structure remains intact, it can be calcined for reuse.

Overview of future LDH-composites

In summary, LDH composites have been applied to construct different promising sorbents and catalysts by incorporating nanoparticles or other substances. LDHs can offer their unique properties and possibly induce new performance. The main advantages of LDHs over conventional anion exchange resins include their higher anion exchange capacity for certain anions and good thermal stability [305].

The research presented focuses on the various methods developed for obtaining composites based on LDH and inorganic materials, considering the way of joining or generating each reviewed material. Since not all the reviewed inorganic materials can be obtained synthetically, their combination could range from a simple physical mixture to a complex synthesis that improves the physicochemical properties of the material, the variation of the available LDH composites and the morphology achieved by the different methods are still limited; therefore, it is worth investigating further the optimization of the synthesis condition for a variety of LDH.

Summary and perspectives

Nowadays, LDH have found applications in many fields, taking advantage of their multiple properties, such as basic character, ion exchange capacity, or enabling the inclusion of oligometals in their compositions. LDH are environmentally friendly, nontoxic, biocompatible, and low-cost multifunctional materials that present a special characteristic where their structures and chemical compositions can be easily designed to a needed application.

If we focus on the industrial aspect, production costs play a very important role. Some authors have reported that the laboratory production process of LDH is estimated at 3.12 USD/g of the produced LDH [301], while the lowest-cost production method is the sol-gel method [302]. However, depending on the required properties, the cost will vary. For instance, if a defined particle size is needed, a crystallization technique such as microwave or ultrasound will be used, which will increase the cost. If a specific element is required in the LDH layers, such as transition elements, the cost will increase significantly. Therefore, depending on the application and LDH design, the final production cost may increase or decrease. Regarding inorganic materials, we find that some materials are generally expensive to produce, such as SBA-15 [303], while others like montmorillonite, natural zeolites, or kaolinite are naturally occurring and therefore, their cost is lower.

Although LDH have an easy and kind structure modification property, their lamellar structure is constituted by big blocks of stacked layers. This kind of arrangement makes the accessibility to the active sites that are locked inside the blocks difficult, leading to materials with low ion exchange capacity or catalytic activity. Thus, the LDH conjugation with inorganic materials can promote the formation of small or nano-LDH crystals, where the active sites turn out to be much more exposed, so facilitating its accessibility and improving their adsorption and catalytic properties. In this sense, LDH have been combined with the most varied types of inorganic compounds, such as zeolites, oxides, or clays, to generate materials with enhanced physico-chemical properties. The LDH/inorganic compounds composites can originate dual basic-acid sites when combined with an acidic component, such as montmorillonite, zeolite, or halloysite, or promote biological activity when combined with iron oxide or silica, as powerful nanosystems for diverse cancer therapies [304].

Thus, the LDH/inorganic compounds composites studied in this paper can offer a wide opportunity to explore, in a deep way, their suitability and benefits for future applications in many different fields in materials science.

Ethics statements

The methods used in the study did not involve any human or animal subjects. No data was used or collected for this work.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Franchescoli Didier Velázquez-Herrera: Conceptualization, Investigation, Writing – review & editing, Visualization, Supervision. **Yohuali Zarazua-Aguilar:** Investigation, Writing – original draft. **Amanda S. Garzón-Pérez:** Investigation, Writing – original draft. **Karin Monserrat Álvarez-Gómez:** Investigation, Writing – original draft. **Geolar Fetter:** Writing – review & editing, Visualization, Supervision.

Data availability

No specific data is used.
No data was used for the research described in the article.

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F.D. Velázquez-Herrera, Y. Zarazua-Aguilar, A.S. Garzón-Pérez et al.

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F.D. Velázquez-Herrera, Y. Zarazua-Aguilar, A.S. Garzón-Pérez et al.

MethodsX 13 (2024) 102912

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