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SYNTHESIS, PROPERTIES AND PRACTICAL APPLICATIONS OF DOPED AND UNDOPED, ZINC-CONTAINING LAYERED DOUBLE HYDROXIDES – A BRIEF REVIEW

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Recently, layered double hydroxides (LDHs) have been developed for more potential applications in various industry fields. Many synthesis methods of LDHs in the literature have been considered and classified by changing properties. However, the property change by the synthesis method does not happen in the same way in all LDHs. Also, when LDH with the same composition is synthesized by the same synthesis method, it is possible to obtain LDHs with the same composition but different properties due to the change of reaction parameters. Recently, the doping of semiconductor nanocomposites with different element atoms has led to new properties. The effect of the elements used in the doping process on the properties of zinc-containing LDHs was investigated and explained in the review article. The synthesis's techniques differ from each other by the reaction parameters and they impact the crystal structure, physicochemical properties, and morphology of LDHs. Finding more additional effects of doping processes and doping elements' nature on the zinc-containing LDHs' properties, the synthesis procedures, and the determination of reaction parameters are shown in this review article. It was revealed that rare earth elements are not selected as three valence metals for the main structure of LDHs, they are mainly used as a doping element which is replaced by three valence metals (Al, Cr, Ti, Fe(III)) as dopants because of the ionic radius and various factors.

Keywords*: Layered Double Hydroxides (LDHs), synthesis procedures, methods, structural properties, photocatalyst.*

1 INTRODUCTION

In order to obtain new physical and chemical properties in nanosized materials, new nanomaterials are currently being synthesized in two-dimensional (2D) structure and modification of known nanostructures is being carried out. For layered nanostructures, atoms on the layer chemically and covalently bonded each other and some physical interactions between the layers form bulk crystals [1–3].

LDHs have been studied for their amazing applications in sorption, catalysis, thermal stability, supercapacitors, chemical resistance and lithium-ion battery (LIB) due to host cationic layered structure. They exhibit anion exchange, semiconductivity, stability, alkaline resistivity etc. LDHs are symbolized by the famous general formula $[M^{2+}_{1-n}M^{3+}_{n}(OH)_2]^{n+}[A^{z-}]_{n/z}\times mH_2O$, where $M^{2+}$is divalent cation (M = e.g. Fe, Cu, Mg, Ni, Zn or Co) and $M^{3+}$is trivalent cation (M = e.g. Al, Cr, Fe, Ga or Mn). A^{z-} shows the anions which enter the structure from the salts used in the reaction. By changing the molar ratio of M^{2+}/M^{3+} cations, the nature of the metals, type of interlayer anions, and different physicochemical properties can be obtained. This review will focus on layered double hydroxides (LDHs) which are very important representatives of 2D nanostructures containing positively charged layers and their compositional properties will be discussed in detail. LDHs are layered crystalline hydroxides with excellent conductivity, surface active, optical, electrical, mechanical, thermal, magnetic, catalytically properties which have broad application in photocatalysis [4, 5] supercapacitors [6], fire retardant [7], energy storage [8, 9], anion exchange materials [10, 11], biomedicine [12, 13], electrochemical sensors [14], batteries [15, 16] and other field.

There are many important review articles on LDH-synthesis by various methods, properties, application [17–19]. These methods have been analyzed by different authors, their similarities and differences, effects on LDH properties have been investigated, and their advantages have been determined. Some of these methods are presented in the review article. However, since there are very few review articles in the literature on the systematization and conception of the modification and doping with metals and non-metals of LDHs, there is a great need to write and study such very significant and important facts. Since this is a very general topic, in the current review article, the synthesis of Zn-containing LDHs, composition-property relations was investigated, the effect of doping agents, and applications were discussed, and important results were examined.

LDHs are synthesized by various methods like co-precipitation, urea hydrolysis, hydrothermal, anion exchange, sol-gel, sonochemical, mechanochemical, etc. which is summarized in Figure 1.

2 SYNTHESIS METHODS

Co-precipitation method. It is the simplest and most common method for the preparation of LDHs. Here, inorganic salt solutions of divalent and trivalent metals in a certain proportion are co-precipitated in the presence of an alkaline solution. If we consider that the mixed salt solution creates an acidic environment, the amount of alkaline solution is even taken in excess. Also, the dissociation rate of the salt has a significant effect on the formation. After getting the solution to the optimum pH, the pH of the solution may decrease again because of the low dissociation rate. In this case, the pH of the slurry is controlled by the increasing pH method. pH 9 and 10 are considered optimal for some metals (like Zn, Fe, Ni, Mn, etc.) [20]. Even extremely high concentration alkaline solution is used to obtain LDHs of some metals (like Ba, Ca) [21] because of the ion radii.

To carry out the formation of LDHs by co-precipitation, the divalent and trivalent metal salt solutions should be dropwise added to the distilled water to get a mixed salt solution. Co-precipitation is carried out by the subsequent dropwise addition of an alkaline solution to the mixed solution. The technology of preparing the mixed solution also affects the formation. Thus, it can be prepared in 3 ways: prepare divalent and trivalent salt solutions in the required concentration and then 1) mix them together, 2) mix them and add dropwise into distilled water at the same time, 3) or mix them and dropwise add into the alkaline solution at the. Third case results in a direct reaction. During co-precipitation, supersaturation is an important driving force for nucleation crystal growth. We need to control the supersaturation process if we want to form nanoparticles of the desired size and composition. At low-level supersaturation, crystals grow faster than they nucleate and a very slow cooling down results in it. As a result, large size crystals are formed [22]. At high-level supersaturation, the rate of nucleation is greater than the rate of crystal growth and faster cooling rates result in it. At this supersaturation, nucleation dominates crystal growth and results in smaller crystals [23].

Urea hydrolysis. Highly crystalline LDHs are formed using this method [24, 25]. The use of urea instead of NaOH as a precipitating agent is obviously advantageous because, during the precipitation reaction, a lower degree of super-saturation is achieved. The average diameter of LDH nanoparticles obtained by this method is significantly smaller and the basal distance is larger compared to other methods [26].

Hydrothermal synthesis is the most widely used method to get rapid heating at a temperature higher than 100 °C and to obtain well-crystallized oxides, sulfides, hydroxides, and ternary systems. This method is also used to perfect the LDH's particle size, disperse distribution and improve crystal structures [27, 28].

Anion exchange is also often used method for the formation of new LDHs from other LDHs. Although this method is included among the synthesis methods of LDHs, it can be considered as a modification of LDHs. Because the precursors of the anion exchange method are the LDH itself, the structure and composition of the host remain the same and only change interlayer anions. Because divalent ions are more selective than monovalent ones, nitrate ion is easily displaced in interlayers and is very suitable precursors for the anion exchange synthesis of LDHs [29]. The selectivity of counterions is compared as follow: $CO_3^{2-}>SO_4^{2-}>>OH^->F^->Cl^->Br^->NO_3^->ClO_4^-$ and it depends of ionic radii [29, 30].

Mechanochemistry and tribochemistry. Hydrotalcite-like Mg-Al-NO_3-LDHs are successfully prepared using the magnesium and aluminum hydrates obtained from their nitrate salts by manually grinding technique [31, 32]. Although this method is not yet applied to many LDHs, it stands out for its simplicity and effectiveness.

Rehydration (Reconstruction) exploits the "memory effect" of hydrotalcite [33] and is similar to anion exchange. By calcination at 100–200 °C LDHs lose water molecules between the layers, at 300–500 °C LDHs transform into a mixture of divalent and trivalent oxides because of the dehydration. It is reconstructed [34, 35] to LDHs again by heating in water or weak acid solution.

Microwave and sonication are also effective methods for the preparation of highly dispersed and crystallized LDH materials [30]. The effect of microwave irradiation during the hydrothermal process to obtain LDHs was investigated by S. Zadaviciute et al. [36] Genty et al. [37] compared these methods by co-precipitation and a significant increase of the specific surface area and catalytic activity is observed. Microwave-assisted synthesis of LDHs also stands out with its low cost and low environmental impact treatments resulting in efficiency in preparing LDHs using a low cost and low environmental impact.

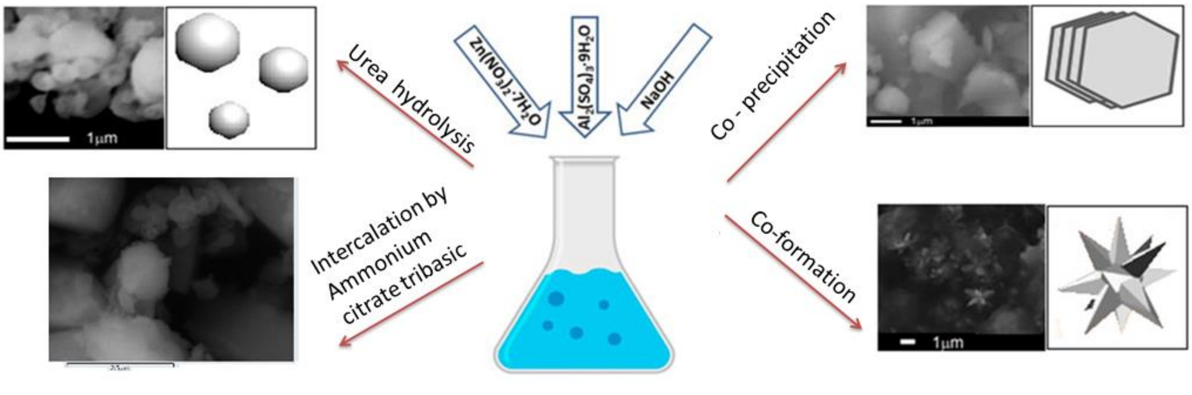


Figure 1. Synthesis of ZnAl-LDHs by different methods

3 SYNTHESIS OF ZN CONTAINING LDHS

Yanzhen Liu et al. synthesized ZnAl-SO_4-LDH by anion exchange using ZnAl-CO_3-LDH as a precursor [38]. Synthesis of ZnAl-Cl-LDH and ZnAl-SO_4-LDH is also carried out by anion exchange. Synthesis of ZnAl-SO_4-LDH was also carried out by repeating the remaining steps of using 0.1 mol Na_2SO_4 and 0.4 g ZnAl-Cl-LDH. Successfully replacing CO_3^{2-} with SO_4^{2-} is defined by FT-IR spectra and the framework of LDH is not spoiled [38]. According to the XRD pattern, the basal space increased after the anion exchange as Zn-Al-CO_3-LDH < ZnAl-Cl-LDH < ZnAl-SO_4-LDH.

In another work, Aliaksandr Mikhailau et al. presented for the first time a method for preparing ZnAl-NO_3-LDH conversion coatings on metallic zinc [39]. The ZnAl-LDH has been synthesized using 1 mM $Al(NO_3)_3$ + 0.1 M $NaNO_3$ mixed solution at 90 °C and pure zinc as a substrate. The obtained ZnAl-LDH is well attached to the substrate and uniform. To understand the mechanism of LDH growth, the effect of chemical and electrochemical processes was studied here. The kinetics of particle growth has been studied by Al and Zn concentration in the electrolyte, time, and the solution pH [39].

For the sealing treatment (ST) and Zn-Al-LDH fabrication: anodized samples immersing in boiling water for 20 min at atmospheric pressure, then vertically immersed in $Zn(NO_3)_2 \times 6H_2O$ (0.05 M) and NH_4NO_3 (0.3 M) mixture solution with the neutral pH adding diluted ammonia at 45 °C for several hours. This process leads to the arrangement of LDH nanosheets [40].

The co-precipitation method was also used to prepare Zn-Al-NO_3-LDH at the different Zn^{2+}/Al^{3+} ions molar ratios like 2, 3, 4,5, and 6 and pH = 7.5 [41]. The synthesis was carried out at 70 °C for 18 h in an oil bath shaker (50 rpm). After washing the precipitate dried at 70 °C in an oven for two days. The results show that ZnO nanoparticles are formed when the Zn/Al ratio is increased at the beginning of the reaction. Similar work was done a few years ago by E.M. Seftel et al. In all cases, as the amount of Zn metal in the mixed solution increased, the diffraction peaks corresponding to ZnO increased. The formation of diffraction peaks corresponding to ZnO with high intensity was also observed in my research [26]. Thus, when the concentration of the NaOH in a base solution was increased (from 2 M to 5 M), along with ZnAl-LDH nanocrystals, ZnO was also formed with high crystallization. This is explained by the solubility of amphoteric aluminum in the extremely high alkaline solution and the participation of more Zn^{2+} ions in crystallization.

ZnCr layered double hydroxides with F^-, Cl^-, Br^-, $I^-$and NO_3^- anions were synthesized by the co-precipitation method [42] For the synthesis of ZnCr-LDHs with different anions like F, Cl, Br, and I via the wet impregnation method. The difference in crystal structure stability of obtained LDHs was explained by the physical properties like Mulliken bonding population, states density, and hydrogen bonding calculated by Density-functional theory. These LDHs demonstrate high performance for photocatalytic degradation of hexachlorobenzene and it is related to their structural stability.

4 METAL- AND NON-METAL-DOPED OR MODIFIED LDHS

4.1 Transition metal-doped LDHs

Cobalt (Co) and copper (Cu)-doped LDHs. High effective Visible-Light-Driven Co (or Cu)-doped ZnAl-LDH photocatalysts were papered by coprecipitation method [43, 44]. The Co/Zn/Al (or Cu/Zn/Al) atomic ratios in the initial metal salt (precursor) in mixed solution were set as 0/2/1; 0.1/2/1; 2/2/1 and 4/2/1 (0/2/1; 0.1/2/2; 1/2/1 and 2/2/1 for Cu). It was found that Co (or Cu)-doped LDH sheets demonstrate higher light absorption capacity than pure ZnAl-LDH materials. The authors explain this effectiveness by the Cobalt ions server as the photo-generated charges separator which results in the higher photocatalytic degradation of RhB [43, 44].

Iron (Fe)-doped LDHs. Zn-Ti-LDH/montmorillonite (ZTL/MT) and Fe-doped ZTL/MT (ZTL/Fe@MT20%) were synthesized by refluxing method as effective photocatalyst for the reduction of Cr(VI) [45] The Fe-containing LDH/montmorillonite composite prevents charge recombination and ensures the distribution of Cr(III) on the surface and/or MT interlayers. As a result, the overall photoelectrochemical equilibrium changes.

Nickel (Ni)-doped LDHs. Nickel-doped ZnAl-LDHs were synthesized for modeling active sites in the application as heterogeneous catalysts for water oxidation [46]. The authors incorporated iron and nickel atoms into the non-magnetic ZnAl-LDH material and used EPR spectroscopy to extract the electronic and geometric information of the paramagnetic centers of the resulting Fe(III) and Ni(II) doped ZnAl-LDH. In the synthesis of Ni-doped ZnAl-LDH, the composition of the precursor consisted of 2 mM $Al(NO_3)_3$ and 2 mM of a variable mixture of $Zn(NO_3)_2$ and $Ni(NO_3)_2$ solutions.

Manganese (Mn)-doped LDHs. Mn-doped Zn/Al-LDHs with Mn 0.5%, 1%, and 3.0% mol with respect to the content of Zn were studied [47]. Photocatalytic activity of obtained Mn-doped Zn/Al-LDHs was studied for 4-chlorophenol. In the photocatalytic degradation mechanism, it was found that Mn acts as an electron (Mn^{3+}; Mn^{4+}) or hole (Mn^{2+}; Mn^{3+}) trap in accordance with its oxidation state and enhances the separation of charges.

Vanadium (V)-doped LDHs. ZnFe-LDH was doped with vanadium (V) to get environmentally friendly, effective, cheap, and sustainable catalysts [48]. The resulting Vanadium (V)-doped ZnFe-LDH catalyst shows high effective degradation activity for pymetrozine in 90 min ultrasonic treatment. The pymetrozine removal efficiency of V-doped and undoped ZnFe-LDH is 73% and 32%, respectively, which is explained by the high strong synergistic effect with a synergy factor of 7.17 of ultrasonic/ V-doped ZnFe-LDH [48, 49].

4.2 Rare earth elements-doped LDHs

It has been shown that the doping and incorporation with rare earth elements like La^{4+}, Eu^{4+}, Ce^{4+}, etc. improve the photocatalytic efficiency of semiconductor materials by avoiding the recombination of charges during the photocatalytic reactions [50–53].

If we look at the published articles, we can see that rare elements containing LDHs are mostly synthesized by co-precipitation and hydrothermal methods, at constant pH, and the application frequency of elements can be arranged as Ce>La>Eu>Tb>Y>Nd>Dy>Sm> Yb>Sc>Er>Pr>Lu [54].

In most cases, rare earth elements are in a +3 oxidation state which replaces the trivalent metals in the crystal lattice in a certain proportion when doping them to LDHs. Some rare earth elements like samarium, europium, thulium, and ytterbium, can be reduced to +2 under certain conditions, while others, such as cerium, praseodymium, and terbium, are oxidized to +4. In such cases, although they take replace of divalent and trivalent metals in the crystal lattice, create many lattice defects, which is one of the important factors for photocatalytic applications [54].

Cerium (Ce)-doped LDHs. ZnAlCe-LDH with different (3.5, 5.0, and 10.0% mol) Ce content were synthesized via one step co-precipitation method [55]. The incorporation of cerium into the layers of the LDH material is appreciated in the X-ray diffractograms and shows some deformations in the crystal structure of the ZnAlCe-LDHs. After calcination of cerium incorporated ZnAl-LDH at 400 °C, the absorbance spectra of UV–vis shifted toward the blue part of the spectra and the XPS analysis substantiates the co-existence Ce^{4+} and Ce^{3+} in the ZnAlCe-LDHs [55]. The photodegradation of phenol was particularly enhanced in the sample which contains 5% mol of Ce. In the photocatalytic mechanism, Ce^{4+} performs as an electron scavenger, facilitates the electron transfer by absorption of O_2, and increases the generation of radicals OH•. In another work, a Ce-doped Zn-Al-LDH nanocontainer was synthesized for the investigation of the anticorrosion activity of aluminum alloy 2024 [56]. Obtained results indicated that Ce 3+ ions were incorporated into LDH layers and exhibited high anti-corrosion properties.

Terbium (Tb)-doped LDHs. Terbium doped ZnCr-LDH. Various amounts of terbium Tb^{3+} doped ZnCr-LDH (Tb-ZnCr-LDH) have been formed by a co-precipitation method [57]. In all cases, Tb doped ZnCr-LDH samples prove an effective separation of charge carriers and effective charge injection efficiency measured by photoluminescence (PL) and photo-electrochemistry (PEC), compared with the pristine ZnCr-LDH [57]. In another work Tb-doped ZnAl-NO_3-LDHs, with different molar ratio of $Zn^{2+}/(Al^{3+}+Tb^{3+})$ is 1.3, 2.0, 3.0 and 4.0, were co-precipitation synthesized characterized [58]. In the experiment firstly 12.52 g of solid Tb_2O_3 was solved in HNO_3 (63%, 100 ml) and H_2O_2 (30%, 10 ml) mixed solution prepared and used in the coprecipitation process as a doped precursor [58].

Dysprosium (Dy)-doped LDHs. Dysprosium doped LDHs has been synthesized with Zn^{2+}:Al^{3+}:Dy_{3+} molar ratios of 30:10:0, 30:9:1, and 30:0:10, and anion exchanged with Different Organics [59]. The broad photoluminescence spectrum on the yellow emission (574 nm) of Dy suggests that the Dy^{3+} has various degrees of distortion and which is matching with the XPS information. The Dy^{3+} emission is affected by ligands. At 767 nm emission, LDHs samples are useful for optical investigations and this type of light is able to transpose cell walls [59].

Europium (Eu)-doped LDHs. Eu-doped ZnAl-LDH with Zn/(Al+Eu) of 2/1 molar ratios, and Eu/(Al+Eu) of 0.06 was coprecipitation synthesized at room temperature and phase transitions have been studied by annealing temperatures [60]. At high temperatures (after 300 °C) the emissions of Eu3+ ions at 5D0–7FJ transition (J = 1, 2, 3, 4), especially (J = 1, 2), has been happened depending on the phase transition from ZnAl-LDH to mixed phases of ZnO and $ZnAl_2O_4$ of host materials which indicate that the structure of the ZnAl-Cl-LDH more favored the emissions of Eu3+ ions. This structure makes the materials a promising candidate in biology or medical diagnosis for fluorescent probes applied with high-temperature stability and less toxicity [60].

Lanthanum (La)-doped LDHs. The constant pH coprecipitation method was also applied for the synthesis of ZnAlLa-CO_3-LDHs) with different molar ratios of Zn/Al/La like 20/6/4; 20/7/3; 20/8/2 and 20/9/1 [61]. According to the results, ZnAlLa-CO_3-LDHs is a very good thermal stabilizer that enhances the long-term and initial thermal stability of PVC with 2.4 phr content. Especially ZnAlLa-CO_3-LDHs with a 20/8/2 molar ratio of Zn/Al/La demonstrate a better stabilizing effect for PVC than another ratio. The composite containing 100 g PVC, 70 g dioctylphthalate, 0.3 g calcium stearate, 0.4 g β-diketones and 2.4 g ZnAlLa-CO_3-LDHs presents a high enhancement in the thermal stability because the effective coordinating with allylic chlorine atoms of ZnAlLa-CO_3-LDHs [61].

In another work Co-precipitation synthesized Zn/Cr/La-LDHs with Zn/Cr/La = 2.0/0.7/0.3 molar ratio indicates 1.4 times higher photocatalytic activity compared to Zn/Cr-LDH for organic dye under xenon light irradiation because of the increased surface area and light harvesting ability [62].

Several multi-metal doping also gives or enhances LDHs some important properties. M. Sarkarat. et al [63] achieved multi-metal doping by substituting both divalent and trivalent metal atoms in LDH crystal lattice by hydrothermal technique. Multi-metal doping LDH is also carried out by incorporation of titania onto ZnNiAlLa-LDH at the same work (Ti^{3+}/ZnNiAlLa-LDH) [63]. According to the results, the authors concluded that doping lanthanum led to poor crystallinity in LDH structures but affected the formation of nickel zinc titanium oxide and doping nickel prevents the formation of pure zinc titanate phase. A partial phase transformation from ZnO to nickel-zinc-titanium oxide by the calcination up to 800 °C and showed lower photocatalytic activities compared to pure zinc titanate [63].

A new type of LDH containing La^{3+}-Zn^{2+}-Al^{3+} cations and MoO_4^{2-} anions with La/Zn/Al = 1/7/2 molar ratio was synthesized by co-precipitation and anion exchange by [64]. The desulfurization effect of LaZnAl-MoO_4^{2-}-LDH is higher compared to LaZnAl-CO_3^{2-}-LDH and it is a potential photocatalyst in the degradation of thioether, thiophene, dibenzothiophene and its derivatives [64].

Neodymium (Nd)-doped LDHs. Neodymium-doped Zn-Al-LDHs were synthesized supporting by polyaniline (PANI@Nd-LDH) via an ex-situ oxidative polymerization and indicates selective fluorescence detection and adsorb the Cr(VI) pollutant in a short time [65]. The synthesized PANI@Nd-LDH shows high selectivity and adsorption capacity performance (219 mg/g) for Cr(VI) from aqueous solutions even containing metal ion mixtures.

4.3 Non-metal-doped LDHs

Carbon-doped LDHs. The composites of LDHs with carbon materials are very promising materials that enhance the surface area and increase the oxidizing groups and improve the surface performance, adsorption and catalytic activity [66]. These materials having excellent physical and chemical properties are prepared by direct mixing, self-assembly and growth in situ methods.

In the direct mixing process, Liu et al. [67] used benzoic acid as an intercalating agent for LDH, and mixed the intercalated LDH with a xylene / C_{60} solution→ ultrasonically stirred at 70 °C for 48 h. This method is not considered effective to obtain an ideal structure because of between carbon and LDH have complicated interactions. Compared to direct mixing, self-assembly controls the structure of materials with required functions [66]. In the growth in situ method, LDH forms and grows using carbon materials as a substrate.

The Characteristics of Carbon Materials and Preparation of LDH-Carbon Composites. Due to the outstanding advantages of carbon materials, such as rich types, wide sources, good pore structure, countless active sites, and stable surface charges, researchers adopt different modification methods for various carbon materials to improve the defects of carbon materials and their applications and performance. Commonly used carbon materials include active carbon (AC), active carbon fibers (ACFs), bio-carbon (BC), carbon nanotubes (CNTs), fullerene (C_{60}), graphene (GO), etc. At present, the known carbon materials used with LDHs to create composites mainly include biochar, fullerene, carbon nanotubes, and graphene [66].

As carbon dopants, biochar (BC), fullerene (C_{60}), carbon nanotubes (CNTs), graphene (GN) and graphene oxide (GO) are promising materials for potential applications in the fields of adsorption, light, electricity, magnetism, supercapacitors, catalysis, etc.

Carbon nanotubes (CNT)-doped LDHs. CNT/LDH nanocomposite was prepared by successfully assembling electrostatic interaction between Zn-Al-LDH and multiwalled carbon nanotubes (MWCNT) which were functionalized by poly acrylic acid. Poly acrylic acid enriches the surface of CNT with a negative charge and easy interaction with positively charged layers of LDH occurs [68] The resulting carbon electrode nanocomposite exhibited excellent electrochemical activity for the oxidation of catechol ($C_6H_4(OH)_2$) [69].

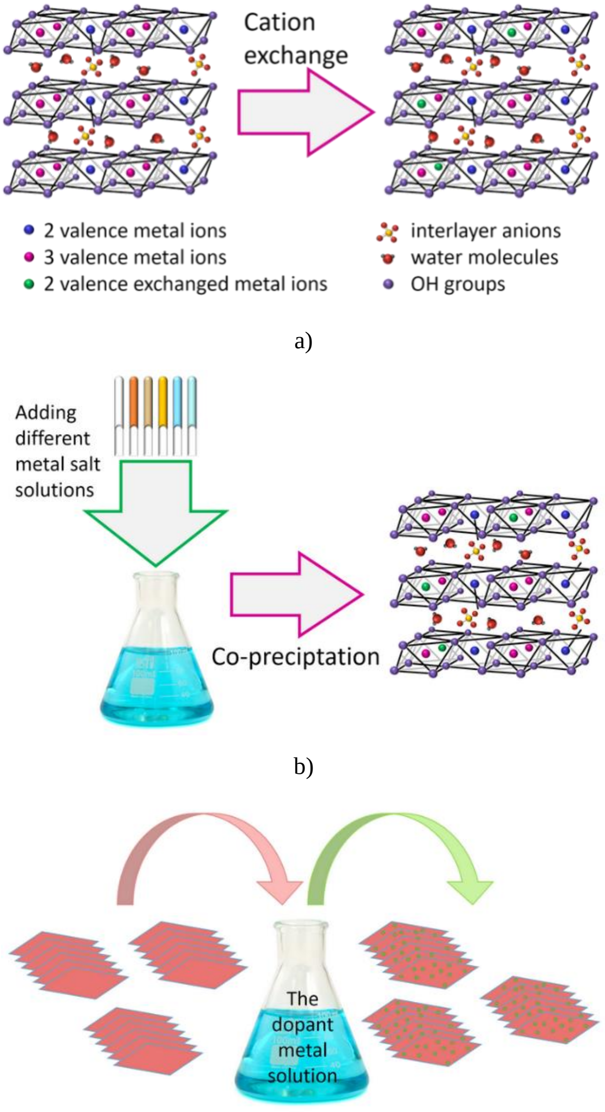
ZnCr-LDH/carbon nanotube (CNT) was synthesized with great crystallinity and characterized by various analyses [91]. BET analysis shows a high specific surface area for ZnCr-LDH / CNT compared to pure ZnCr-LDH.

Graphene oxide (GO)-doped LDHs. GO doped Zn-Al-LDHs with the combination of N and S were synthesized to get a low-cost and efficient electrocatalyst for Oxygen Reduction Reaction (ORR) [70]. According to the results of the electrochemical effect, ZnAl-LDH/N, S-rGO with a 1/1 wt ratio shows the best electrochemical activity among the synthesized electrocatalyst because synergistic and interaction effect between N, S-rGO and ZnAl-LDH [70].

Fullerene (C_{60})-doped LDH. Xiao Lei Liu [67] achieved the noncovalent intercalation of C_{60} into the benzoic acid (BA) modified LDH for the potential application in the photoelectron chemistry process and in the field of optical limiting equipment. ZnAlTi-LDO supported C60-AgCl nanoparticles have been obtained by coprecipitation and light-induced method and photodegradation of organics like Bisphenol A was studied quantitatively by high-performance liquid chromatography (HPLC) [71]. The C60 is used in there as supporting materials to improve the Ag-based photocatalyst stability. The C60@AgCl reduces the recombination of e-h pairs and supplements the efficiency of photocatalysis. The degradation rate is determined as 90% and found that C60@AgCl-LDO nanoparticles can be used for considerable environmental remediation [71].

Black phosphorus (BP)-doped LDHs. Black phosphorus nanosheets were firstly prepared by liquid exfoliation method and used for the preparation of environmental-friendly BPNs / ZnAl-LDH composite by electrostatic self-assembly consisting of p-type (BP) and n-type (ZnAl-LDH) semiconductors [72]. The as-obtained BP/LDH composite with Zn/Al/P =73.01/26.88/0.11 atomic ratio shows high effective methylene blue (MB) degradation under visible light irradiation, which is greater than pristine BP (11%) and ZnAl-LDH because [72].

As can be seen, element doping is one of the most important modification methods applied to change the physical, chemical, mechanical, and biological properties of LDHs, which are obviously very important [73–75]. Dopping of the above-mentioned elements and substances to LDHs has been applied in the literature by various methods [42, 76]. Several of those methods are presented with a graphical description (see Figure 2).



a)

b)

c)

Figure 2. Schematic illustration of LDH modification: Doping by: ion exchange (a), co-precipitation (b), impregnation (c) methods

5 CONCLUSIONS

The synthesis of Zn-containing LDHs and doping with different element atoms by various methods were considered in the review article. It turned out that the main line of different methods is co-precipitation. Although co-precipitation is considered a separate method, co-precipitation of divalent and trivalent metals is carried out in all methods. Although controlling the pH is the main factor in the formation of LDHs and it is somewhat difficult in urea hydrolysis and urea-based hydrothermal method. Here the amount of urea plays an important role in determining the pH of the alkaline environment. It turned out that since LDHs contain both divalent and trivalent metals, when the metals in the LDH are partially replaced by other metals, there is no fundamental change in the crystal lattice (host structure), and only crystal structure defects appear, which in most cases have a positive effect on the optical properties and photoactivity. In most cases, metal-doped Zn-containing LDHs prove an effective charge carrier separation and effective charge injection efficiency compared with the LDH. In the formation of LDHs, rare earth elements are not selected as a 3-valent metals for the main structure, they are mainly replaced to 3-valent metals (Al, Cr, Ti, Fe(III)) as a dopant because of the ionic radius and various factors.

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ЛЕГИРЛЕНГЕН ЖӘНЕ ҚОСЫЛМАҒАН, ҚҰРАМЫНДА МЫРЫШ БАР ҚАБАТТЫ ҚОС ГИДРОКСИДТЕРДІҢ СИНТЕЗІ, ҚАСИЕТТЕРІ ЖӘНЕ ПРАКТИКАЛЫҚ ҚОЛДАНЫЛУЫ – ҚЫСҚАША ШОЛУ

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Жақында әртүрлі өнеркәсіп салаларында әлеуетті қолдану үшін қабатты қос гидроксидтер (ҚҚГ) әзірленді. Әдебиетте ҚҚГ синтезінің көптеген әдістері қарастырылған және қасиеттерін өзгерту арқылы жіктелген. Бірақ синтез әдісімен қасиет өзгеруі барлық ҚҚГ-де бірдей бола бермейді. Сондай-ақ құрамы бірдей ҚҚГ бірдей синтез әдісімен синтезделгенде, реакция параметрлерінің өзгеруіне байланысты құрамы бірдей, бірақ қасиеттері әртүрлі ҚҚГ алуға болады. Соңғы уақытта әртүрлі элемент атомдары бар жартылай өткізгіш нанокомпозиттерді легирлеу жаңа қасиеттерге әкелді. Допинг процесінде қолданылатын элементтердің құрамында мырыш бар ҚҚГ қасиеттеріне әсері шолу мақаласында зерттелді және түсіндірілді. Синтез әдістері бір-бірінен реакция параметрлері бойынша ерекшеленеді және олар ҚҚГ-ның кристалдық құрылымына, физика-химиялық қасиеттеріне және морфологиясына әсер етеді. Допингтік процестердің және допинг элементтерінің құрамында мырыш бар ҚҚГ қасиеттеріне қосымша әсерлерін табу, синтездеу процедуралары және реакция параметрлерін анықтау осы шолу мақаласында көрсетілген. Сирек жер элементтері ҚҚГ негізгі құрылымы үшін үш валентті металдар ретінде таңдалмайтыны анықталды, олар негізінен үш валентті металдармен (Al, Cr, Ti, Fe(III)) қоспалар ретінде ауыстырылатын қоспа элементі ретінде пайдаланылады, өйткені иондық радиус және әртүрлі факторлар.

Түйін сөздер: *қабатты қос гидроксидтер (ҚҚГ), синтез процедуралары, әдістері, құрылымдық қасиеттері, фотокатализатор.*

КРАТКИЙ ОБЗОР СИНТЕЗА, СВОЙСТВ И ПРАКТИЧЕСКОГО ПРИМЕНЕНИЯ ЛЕГИРОВАННЫХ И НЕЛЕГИРОВАННЫХ ЦИНКСОДЕРЖАЩИХ СЛОИСТЫХ ДВОЙНЫХ ГИДРОКСИДОВ

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В последнее время слоистые двойные гидроксиды (СДГ) были разработаны для применений в различных областях промышленности. В литературе рассмотрено множество методов синтеза СДГ и их классификация по изменению свойств. Однако изменение свойства методом синтеза происходит не во всех СДГ одинаково. Также при синтезе СДГ одинакового состава одним и тем же методом синтеза возможно получение СДГ с одинаковым составом, но разными свойствами за счет изменения параметров реакции. В последнее время легирование полупроводниковых нанокомпозитов атомами различных элементов привело к появлению новых свойств. Влияние элементов, используемых в процессе легирования, на свойства цинксодержащих СДГ исследовано и объяснено в обзорной статье. Методы синтеза отличаются друг от друга параметрами реакций и влияют на кристаллическую структуру, физико-химические свойства и морфологию СДГ. В обзорной статье показано обнаружение дополнительных влияний процессов легирования и природы легирующих элементов на свойства цинксодержащих СДГ, процедуры синтеза и определение параметров реакции. Редкоземельные элементы не выбираются в качестве трехвалентных металлов для основной структуры СДГ, их в основном замещают трехвалентными металлами (Al, Cr, Ti, Fe(III)) в качестве легирующих примесей из-за ионного радиуса и различных свойств.

Ключевые слова: *слоистые двойные гидроксиды (СДГ), способы синтеза, методы, структурные свойства, фотокатализатор.*