

Layered double hydroxides, structures and properties

ELEONORA BUTENKO ▪ Azov Sea State Technical University, Mariupol, Ukraine ▪ butenko_e_o@pstu.edu

ALEXEY KAPUSTIN ▪ Miami University, Oxford, USA ▪ kapustinlesha@gmail.com

Érkezett: 2024. 06. 21. ▪ Received: 21. 06. 2024. ▪ <https://doi.org/10.14382/epitoanyag-jsbcm.2024.15>

Abstract

Mg-Al-LDHs with different Mg/Al mol ratio were studied. Adsorption of different organic substances on the layered double hydroxides (LDHs) of different composition was studied. Fatty alcohols, phenols and naphthalene derivatives were used as sorbates. The changes of structures, taking place because of this process, was explored by X-ray method. It has been found, that at the adsorption of such substances as alcohol and phenols the anion exchange take place. After the sorption the specific surface and inner distance of LDHs were measured. The mechanism of structure change was proposed.

Keywords: Layered double hydroxides, sorption, structure

Kulcsszavak: réteges kettős hidroxidok, szorptio, szerkezet

Prof. Eleonora BUTENKO

Donetsk National University – M.Sc. Chemical faculty, June 2007, Specialization: Analytical Chemistry, Donetsk, Ukraine. Pryazovskiy State Technical – Ph.D. Environmental Safety, October 2011, Protection of water bodies using synthetic anionic clays, Kharkov, Ukraine. Pryazovskiy State Technical University – Head of Department of Chemical Technology and Engineering, 2018 present, Mariupol, Ukraine.

Prof. Alexey KAPUSTIN

Mendeleev University of Chemical Technology – M.Sc. Chemical Engineering, May 1981, Specialization: Catalysis, Moscow, Russia. Mendeleev University of Chemical Technology – Ph.D. Chemical Engineering, January 1985, Catalysis, Moscow, Russia. Mendeleev University of Chemical Technology – Doctor of Science (Chemistry), December 1996, Catalysis, Moscow, Russia. Pryazovskiy State Technical University – Research Associate, September 1985-2015, Department of Chemical Technology and Engineering, Mariupol, Ukraine. Miami University – Professor, 2015-present, Oxford, USA.

1. Introduction

Synthetic layered double hydroxide (LDH) compounds are important industrial materials used as absorbents, base catalysts, catalysts precursor, acid neutralizing agents, and precursors in the formation of spinel ceramics [1-2]. Represented by the general formula $[M^{II}_{1-x}M^{III}_x(OH)_2]^{x+}[A^{m-}_{x/m}nH_2O]$ (abbreviated notation $M^{II}_R M^{III}/A$ with $R = (1-x)/x$) where M^{II} and M^{III} represent metallic cations and A^{m-} the interlayer anion, the layered structure of an LDH is built by the periodical stacking of positively charged $(M^{II}, M^{III})(OH)_6$ octahedral layers related to brucite and negatively charged interlayers consisting of anions and water molecules. Owing to the highly tunable LDH intralayer composition coupled with the wide possible choice of anionic organic moiety, a large variety of LDH hybrid materials has been reported, and several review articles and chapters are devoted to the intercalation chemistry of LDHs and their application [3-4].

Most synthetic LDHs resemble the naturally occurring hydrotalcite $Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$. They grow as hexagonal crystals; the structure being derived from that of brucite. Each cation is octahedrally surrounded by hydroxide groups, the octahedral sharing edges forming two-dimensional sheets. These sheets carry an excess positive charge due to the presence of triply charged cations and this is balanced by interlayer anions that bind the sheets together. Most LDHs are binary systems, i.e. with two kinds of metal cations within the hydroxide layers, but ternary LDHs have also been reported. A wide variety of cations can be incorporated into the hydroxide layers of the LDH as well as inorganic or organic anions into the interlayer space either by direct synthesis or ion exchange, and this has resulted in substantial academic and industrial interest, which is discussed below in a non-exhaustive manner. Directly related to the trivalent cation content, the anionic exchange capacity may vary in a large range between 0.1 to 5 meq/g. Even for the lower value, it corresponds to twice the layer charge density of smectite materials, resulting in

layers tightly stacked via the attractive forces with the interlayer anions filling the gallery. Vacancies in the inter-sheet domains are not great considering the high packing of anions balancing the layer charge. Interplanar distance in LDHs can vary over a very wide range, from 1 to 50 Å. The size of the interlayered space (as well as anion exchange capacity) depends on the cationic composition of the matrix, the degree of isomorphous replacement, the solvation degree, but most of all on the size of the anions and molecules in the intralayer space [5]. The size of the internal space depends primarily on the size of the ion in the interlayer space.

LDHs are manufactured on an industrial scale with a manufacturing capacity that increased by at least 50,000 metric tons [6].

The heat treated or calcined caused to the destruction of LDHs structure. The thermal treatment of LDHs up to about 200 °C induces dehydration. Treatment up to 500 °C induces dehydroxylation and loss of vaporizable anions such as carbonate, nitrate, oxalate, and acetate. Heating at up to 900 °C leads to the formation of mixed metal oxides, and in many cases, spinels. These calcined LDHs possess a “structural memory effect” [7]. Remarkably, the partially or fully dehydrated mixed metal oxides produced from LDH precursors will rehydrate on contact with water, take up anions, and reform the original LDH structure with great fidelity.

Another “structural memory effect” is in effect on the structure of LDHs the anions that were present in solution during synthesis. The size of the interplanar spacing strongly correlated with size of anions in the mother liquor [1-3, 8-9]. Using this “memory effect” makes it possible to control the structure of LDHs, making adsorbents and catalysts beforehand given sieve properties. This feature makes LDHs exceptionally promising substances as catalysts and sorbents [10-16].

2. Experimental procedures

2.1 Synthesis of LDHs samples

LDHs samples were synthesized following the co-precipitation method [17], at 318 K, under magnetic stirring and constant flow of nitrogen. The precipitants NaOH/Na₂CO₃, as solution A, and the Mg and Al salts, as solution B, were added dropwise together in such a way that the pH of the synthesis medium was kept at a constant value of 9. After an aging step, the obtained precipitates were then separated by centrifugation, washed with warm deionized water, and dried under air at 393 K. The nature of the LDHs is shown in Table 1. For the LDH sample (Mg/Mg+Al = 0.52), for instance, 150 mL of aqueous solutions of MgNO₃ + Al(NO₃)₃ with a total concentration of 1 mol/l and 150 ml of a second solution containing 0.5 mol/l Na₂CO₃ and 1.0 mol/l NaOH were added together dropwise, for 40 min, in a 1 l flask. The obtained white precipitate was aged, under magnetic stirring, at 353 K, for 24 h, washed with warm deionized water and dried, under air, at 393 K, up to constant weight.

Structure of LDHs was studied by X-ray diffraction. The X-ray diffraction experiments were performed using a SIEMENS D-500 diffractometer with Co K_α - radiation. Special computer programs were used for smoothing, background correction and decomposition of overlapped diffraction peaks.

X-ray fluorescence spectroscopy (XRF), Shimadzu XRF-1700 sequential XRF spectrometer was used to determine the Mg/Al atomic ratios of the samples.

The nitrogen adsorption–desorption isotherms were recorded at 77 K on a Coulter SA 3100 automated gas adsorption system on samples previously degassed at 383 K for 7 h under vacuum. Specific surface areas (SBET) were determined using the Brunauer–Emmett–Teller (BET) method based on adsorption data. The pore volume (V_p) values were determined by using the t-plot method of De Boer.

Thermal behaviors were studied by thermo-analytical methods, the samples calcined under air (the temperature was raised with 10 K/min to 1273 K).

The basicity and acidity of synthesized LDHs were determined by Hammett indicators method [18-19].

The concentration of phenols was determined photometrically by the reaction of formation of the colored compound with 4-aminoantipyrine in an alkaline medium (pH=10) in the presence of ammonium persulfate. The concentration of alcohol, naphthalene and β-naphthol was determined by chromatography on a chromatograph HP 5890.

Sorption studies were performed in periodic conditions, loading the sorbent in a solution with sorbed substances (alcohols, phenols, naphthalene derivatives), and samples were taken after vigorous mixing for some time. Standard concentrations of sorbates were as follows: alcohol = 1.0·10⁻⁵ mol/l, PhOH = 1.0·10⁻⁴ mol/l, naphthalene and β-naphthol = 1.0·10⁻⁶ mol/l.

3. Results and discussion

The properties of LDHs samples are shown in Table 1.

Mg/Mg+Al, mol/mol	0.52	0.72	0.81	0.86
d, Å	3.038	3.036	3.045	3.058
c, Å	22.62	22.67	22.81	23.40
Specific surface, m ² /g	250	200	192	180
Pore volume, ml/g	0.50	0.20	0.20	0.20
Acidity, meq/g	0.41	0.32	0.21	0.06
Basicity, meq/g	0.73	0.54	0.63	0.85
E _{din.} , meq/g	0.075	0.081	0.041	0.036
E _{stat.} , meq/g	0.38	0.41	0.10	0.08

Table 1. The properties of synthesized Mg-Al-LDHs
1.táblázat Az szintetizált Mg-Al-LDH-k tulajdonságai

Results of the thermal analysis of LDHs are shown on the Fig. 1. The first basic peak corresponds to the loss of crystallization water, the following – to transition from the hydroxide form into oxide one.

The kinetics of ion-exchange sorption of phenol on LDHs of different composition was studied.

Since phenol is an acid, so the represented reaction is an anion exchange reaction of neutralization occurs rather quickly and irreversibly.

Investigation of the kinetics of the reaction showed that the rate of sorption depends on the concentration of phenol and amount of LDH. To replace the mass of the sorbent on the concentration of active sites (C_{as}) in a volume of solution used the values of the dynamic capacity of LDHs, defined in dynamic conditions (Table 2).

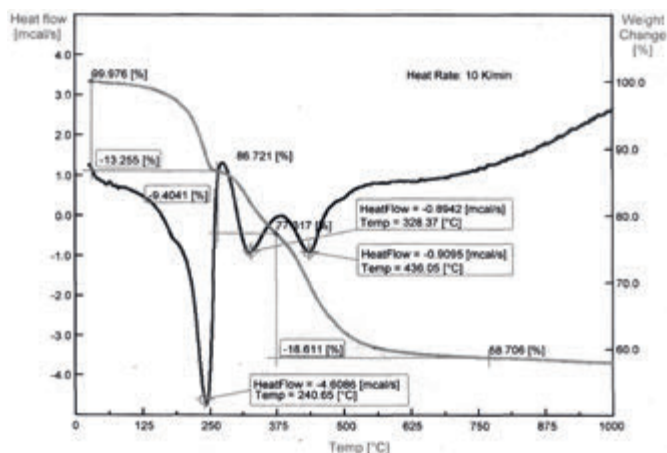


Fig. 1 Calcining of LHD
1.ábra Az LHD-k kalcinálása

Mg/(Mg+Al), mol/mol	0.52	0.72	0.81	0.86
E, meq/g	0.18	0.31	0.36	0.46

Table 2 The dynamic capacity of LDHs (phenol)
2. táblázat Az LDH-k dinamikus kapacitása (fenol)

The kinetic equation of sorption of phenol is as follows:

$$Q = k \cdot C_{PhOH} \cdot C_{as}$$

The rate constants of phenol sorption for sorbents with different molar ratio of Mg/(Mg + Al) have been calculated. The obtained values of the constants are presented in Table 3.

Mg/(Mg+Al), mol/mol	0.52	0.72	0.81	0.86
$k \cdot 10^3, \text{l/mol}\cdot\text{s}$	0.83	1.29	1.67	1.70

Table 3 Second order constants of sorption
3. táblázat A szorpció másodrendű konstansai

(Studies of sorption of alcohols, naphthalene, beta-naphthol and other compounds have been carried out. Kinetics of sorption are similar. Studies have shown that the rate of sorption increases with the acidity of the adsorbate).
The reaction with phenol proceeds rapidly (Fig. 2), and after 15-20 minutes the dynamic equilibrium set.

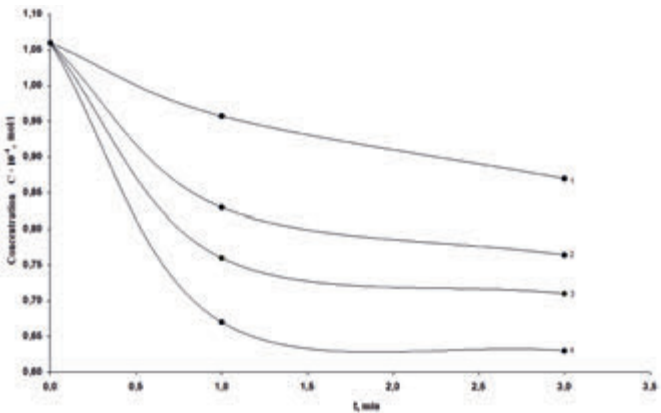


Fig. 2 Change of phenol concentration at different temperature; 1 – 21 °C, 2 – 40 °C, 3 – 60 °C, 4 – 75 °C
2. ábra A fenol koncentrációjának változása különböző hőmérsékleten; 1 – 21 °C, 2 – 40 °C, 3 – 60 °C, 4 – 75 °C

However, if at the end of the sorption to leave the LDHs in a solution of phenol in a long time (48 hours), the degree of adsorption increases, a simultaneous increase the interplanar distance from 3.03 to 5.76 Å. By our assumption, this is due to the reorientation of the aromatic rings in the inner space of LDHs (Fig. 3), with increased availability of active sites.

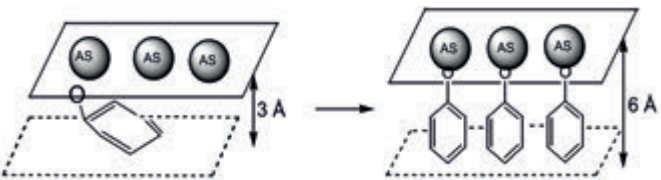


Fig. 3 Reorientation of phenol molecules of LDHs inner space
3. ábra A fenolmolekulák reorientációja az LDH-k belső terében

To test this hypothesis, studies on the sorption of alkoxides were conducted. Experiments have shown that the sorption of alcohols also occurs through a mechanism of anion exchange. To prove this, the LDH was placed into alkoxide solution and left for 48 hours. Sorbents after adsorption was washed with ethanol up to the absence of alcohol traces, and then washed with an aqueous solution containing carbonate ions. In the washing solution by chromatography established the presence of alcohol, indicating that the reverse reaction of anion exchange.

(The same phenomena are observed for phenol, and β-naphthol, but not for naphthalene. In the case of naphthalene, a complete removal during washing with ethanol and ether take place). We can assume that after a long stay in alcohol solution the hydrocarbon radicals are arranged perpendicular to the main plane of LDHs.

It is interestingly to trace the change of interlayered distances at sorption of alcohols with the different length of hydrocarbon radical [20]. These results well correlate with the changes of interlayered distances in case of adsorption of alkylsulfates.

In the linear approximation of the results described by the following dependencies:

- For alcohol: $d \text{ (Å)} = 2.43 + 1.45 C$
- For alkylsulfates: $d \text{ (Å)} = 5.41 + 1.35 C$
- 5.41 – the contribution of sulfonic group, 2.43 – the contribution of the hydroxyl group, 1.35-1.45 – the contribution of each –CH₂– group.

Such patterns suggest a perpendicular arrangement of hydrocarbon radicals of alcohols relative to the layers of inorganic matrix. Similar patterns were observed for dicarboxylic acids, which bind two oxide layers to each other, while the interplanar distance is proportional to the length of the hydrocarbon radical. For ethoxylated products radicals initially arranged in a plane perpendicular to the layers of monomolecular hydroxide, and after drying the angle is 56 degrees.

Alcohols with long hydrocarbon radicals, settling initially parallel to the layers of the matrix in the inner space of LDH, should block the active sites. To test this hypothesis, we determined the concentration of the basic sites remaining in the LDH after 15 minutes (Table 4).

Alkoxide	Amount of sorbed alcohol, mmol/g	Non-active sites, meq/g
CH ₃ O ⁻	0.86	-
C ₂ H ₅ O ⁻	0.51	0.34
n-C ₃ H ₇ O ⁻	0.36	0.49
i-C ₃ H ₇ O ⁻	0.32	0.53
i-C ₄ H ₉ O ⁻	0.31	0.54
n-C ₄ H ₉ O ⁻	0.27	0.58
n-C ₅ H ₁₁ O ⁻	0.11	0.74
i-C ₅ H ₁₁ O ⁻	0.09	0.76

Table 4 Basicity of LDH after the sorption of alkoxides (E₀ = 0.85 meq/g)
4. táblázat Az LDH bázikussága az alkoksizidok szorpciója után (E₀ = 0.85 meq/g)

At short adsorption time the increase of interlayered distance does not take place and the hydrocarbon radicals blocking the access to the basic sites, the degree of block is proportional to the length of the hydrocarbon radical. At long stay of LDHs in the alkoxides solution it will be the radical reorientation, accompanied by an increase in the interlayered distances.

Enthalpy of moving apart the layers were determined from kinetic experiments. LDH was placed in a solution containing ions C₇H₁₅O⁻ and kept some time at various temperatures,

and then the ethoxylation in the presence of LDH as basic heterogeneous catalyst was carried out. The reaction rate increased with an increase in the interlayered distances, respectively with the increasing availability of basic sites. The obtained value of the enthalpy is $\Delta H = 2$ kcal/mol. Change of the reaction rate after LDH heating (1 – original LDH, 2 – 110 °C, 3 – 130 °C, 4 – 150 °C).

Similar experiments we carried out, examining the sorption of naphthalene and β -naphthol. These substances were chosen for study because of the sorption of these substances occurs by different mechanisms: physical sorption for naphthalene, ion exchange – for β -naphthol). After completion of the sorption process the specific surface area was measured and interlayered distances was determined. Measuring the size of internal space was performed using X-ray diffraction analysis.

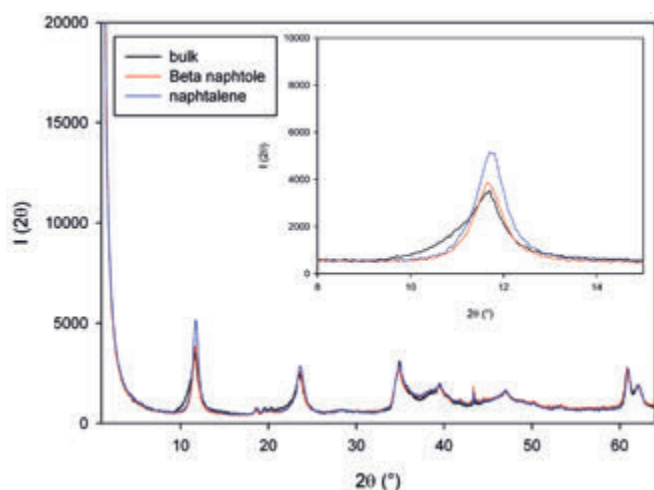


Fig. 4 XRD patterns of LDH $Mg/(Mg+Al) = 0,72$ mol/mol (as-prepared, with naphthalene, with β -naphthol)

4. ábra 4. Az LDH XRD mintázatai $Mg/(Mg+Al) = 0,72$ mol/mol (előállított állapot, naftalinnal, β -naftol-lal)

The X-ray diffraction patterns shown in Fig. 7 are very similar that indicates a minor change of the structure. The insert shows that intensity decreases and a reflection corresponding to 11.8° diffraction angle shifts to smaller angle. This means increasing interplanar distance resulted from penetration of organic substances inside the lattice (see also shifts of the reflection at 43, 61 and 62°). The shift of some reflections, appearance and increase of new reflections with unchangeable other reflections indicate a change of the structure. The structure evolution occurs in a row: as-prepared – with naphthalene – with β -naphthol.

(Mg/(Al + Mg), mol/mol)	As prepared	with β -naphthol	with naphthalene
0.52	162.4	155.7	156
0.72	14.4	14.4	14.4
0.81	9.2	12.0	19.59
0.86	22.0	28.0	34.7

Table 5 The value of specific surface area (S , m^2/g) for different LDHs
5. táblázat A fajlagos felület értéke (S , m^2/g) különböző LDH-k esetében

Anion exchange with β -naphthol leads not only to change the distance between layers, but also to a change in specific surface area. The specific surface area of sorbents with absorbed organic compounds were determined by the BET method. The results are presented in Table 5.

Presented in Table 5 data shows that at low ratios of $Mg/(Al + Mg)$ the sorption of naphthalene and β -naphthol has virtually no effect on the value of specific surface, but at high ratios of $Mg/(Al + Mg)$ – changes much more.

Aromatic rings of adsorbed naphthalene are arranged in parallel position to inorganic layers, regardless of the concentration of active sites. The interlayered distance in LGHs is about 7 Å. The effective area for β -naphthol is 9.89 Å. This value is comparable to the «thickness» of the aromatic ring, constituting ~ 3.5 Å, whereas the diameter of the benzene molecule is 7.1 Å. For β -naphthol plane-parallel arrangement is observed only in the initial stage of ion exchange.

After then the equilibrium, accompanied by an increase in the interplanar spacing and increasing the number of active sites available for the following molecules β -naphthol take place. This process is accompanied by an increase in specific surface area of LDHs.

Dependence of the specific surface after sorption of naphthalene and β -naphthol, depending on the composition of layered double hydroxides are shown in Fig. 9. It is seen that with increasing of degree of isomorphous substitution, and sorption of organic compounds leads to large changes in specific surface area.

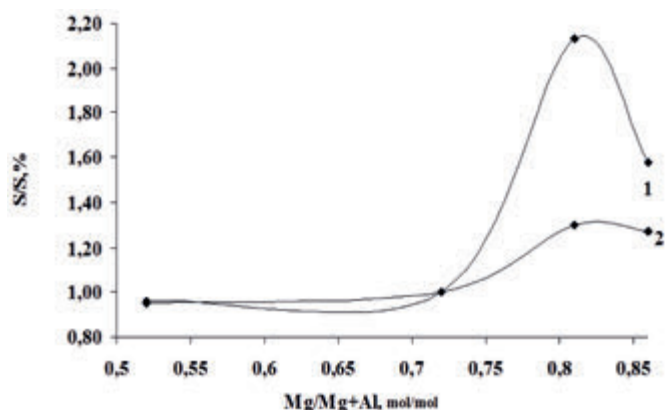


Fig. 5 The relative change in specific surface, depending on the composition of LDHs; 1 – with naphthalene, 2 – with β -naphthol

5. ábra A fajlagos felület relatív változása, az LDH-k összetételétől függően; 1 – naftalinnal, 2 – β -naftollal

Thus, our work revealed a difference in the behavior of LDHs during the sorption of organic compounds by ion-exchange mechanism and by physical sorption. As well patterns, confirming the assumption that changes in the structure of LDHs in the process of sorption of compounds having dimensions that exceed the size of the interlayer space of LDHs was established.

Calcing of LDHs containing various organic compounds leads to the formation of sporous solids. The size of the

cavities is determined by the size of removed molecules. This phenomenon can create sorbents and catalysts with beforehand set structure of internal cavities, making LDHs even more attractive precursors of catalysts and sorbents.

4. Conclusions

We investigated the structure, morphology and anion-exchange capacities of Mg-Al layered double hydroxides with varying Mg/Al+Mg molar ratio and inner space composition.

It has been founded that the size of internal cavities is determined by the size of the molecules and anions presented in the inner space of LDHs. The kinetics of sorption of organic anions has been studied. It is shown that it depends on the concentration of active sites, which, in turn, blocking by radicals in the process of adsorption. It is proved that molecules with long hydrocarbon radicals change their configuration at the end of the sorption process, which leads to a change in the interplanar spacing and physico-chemical properties of the studied LDHs.

References

- [1] Jitendra Kameliya, Aazad Verma, Partha Dutta, Charu Arora. Layered Double Hydroxide Materials: A Review on Their Preparation, Characterization, and Applications. *Inorganics*, 2023, 11, (3), 121; <https://doi.org/10.3390/inorganics11030121>
- [2] Ahmad Farhan, Aman Khalid, Nimra Maqsood, Sidra Iftikhar, Hafiz Muhammad, Adeel Sharif, Fei Qi, Mika Sillanpää, Muhammad, Bilal Asif. Progress in layered double hydroxides (LDHs): Synthesis and application in adsorption, catalysis and photoreduction. *Science of The Total Environment*. V. 912, 20 February 2024, 169160. <https://doi.org/10.1016/j.scitotenv.2023.169160>
- [3] Aamir I. Khan, Dermot O'Hare. Intercalation chemistry of layered double hydroxides: recent developments and applications. *J. Mater. Chem.*, 2002, 12, 3191–3198; <https://doi.org/10.1039/B204076J>
- [4] Lei Li, Anastasia Sevcic, Patric van Rijn. Layered Double Hydroxides as an Intercalation System for Hydrophobic Molecules. 2023, 13 (24), 3145. <https://doi.org/10.3390/nano13243145>
- [5] Eiman Alibakhshi, Ebrahim Ghasemi Mohammad Mahdavian Bahram Ramezanzadeh. The effect of interlayer spacing on the inhibitor release capability of layered double hydroxide based nanocontainers. *Aladdin Scientific. Journal of Cleaner of Production*, 251, 2019. <https://doi.org/10.1016/j.jclepro.2019.119676>
- [6] Guixiang Ding, Chunxue Li, Yonghao Ni, Lihui Chen, Li Shuai and Guangfu Liao. Layered double hydroxides and their composites as high-performance photocatalysts for CO₂ reduction. *College of Materials Engineering, College of Ecological Environment and Urban Construction, Fujian University of Technology, China, Limerick Pulp and Paper Centre, Department of Chemical Engineering, University of New Brunswick, Canada. EES. Catal.*, 2023, 1, 369-391. <https://doi.org/10.1039/D3EY00080J>
- [7] Li Jin, Xiaoyuan Zhou, Fang Wang. Insights into memory effect mechanisms of layered double hydroxides with solid-state NMR spectroscopy. *Nature Communications*, volume 13, Article number: 6093 (2022). <https://doi.org/10.1038/s41467-022-33912-7>
- [8] Peng Wang, Xueqian Zhang, Bin Zhou, Fanpeng Meng, Yishan Wang, Guangwu Wen. Recent advance of layered double hydroxides materials: Structure, properties, synthesis, modification, and applications of wastewater treatment. *Journal of Environmental Chemical Engineering*, Volume 11, Issue 6, December 2023, 111191. <https://doi.org/10.1016/j.jece.2023.111191>
- [9] Leticia Santamaría, Antonio Gil, Miguel Angel Vicente, Sophia A. Korili. Nano-Materials as Photocatalysts for Degradation of Environmental Pollutants. *Challenges and Possibilities*, 2020, Pages 79-90. <https://doi.org/10.1016/B978-0-12-818598-8.00005-5>
- [10] Paranthaman Vijayakumar, Syama Lenus, K. Pradeeswari. In Situ Reconstructed Layered Double Hydroxides via MOF Engineering and Ru Doping for Decoupled Acidic Water Oxidation Enhancement. *Energy Fuels*, 2024, 38, 5, 4504–4515. <https://doi.org/10.1021/acs.energyfuels.3c04641>
- [11] You X., Chen L., He S., Zhang G. Solid Acid-Base Catalysts Based on Layered Double Hydroxides Applied for Green Catalytic Transformations. *Catalysts*, 2024, 14, 28. <https://doi.org/10.20944/preprints202312.0068.v1>
- [12] Dalma Sofia Argüello, Lady Catalina Cabana Saavedra, Sandra Marina Mendoza, Marcos Iván Oliva. Layered double hydroxides modified by transition metals for sustainable glycerol valorization to glycerol carbonate. *Catalysis Today*. Volume 427, 1 February 2024, 114415. <https://doi.org/10.1016/j.cattod.2023.114415>
- [13] Ming Xu, Min Wei. Layered Double Hydroxide-Based Catalysts: Recent Advances in Preparation, Structure, and Applications. *Advanced Functional Materials*, 2018, 28 (47). <https://doi.org/10.1002/adfm.201802943>
- [14] Evgeniy S. Seliverstov, Sergei N. Golovin, Olga E. Lebedeva. Layered Double Hydroxides Containing Rare Earth Cations: Synthesis and Application. *Front. Chem. Eng., Sec. Catalytic Engineering*, Volume 4, 2022. <https://doi.org/10.3389/fceng.2022.867615>
- [15] Diana Gilea, Radu G. Ciocarlan, Elena M. Seftel. Engineering Heterostructures of Layered Double Hydroxides and Metal Nanoparticles for Plasmon-Enhanced Catalysis. *Catalysts*, 2022, 12, (10), 1210; <https://doi.org/10.3390/catal12101210>
- [16] Mohsen S. Mostafa, Lan Chen, Mohamed S. Selim. Recent Breakthrough in Layered Double Hydroxides and Their Applications in Petroleum, Green Energy, and Environmental Remediation. *Catalysts*, 2022, 12, (7), 792; <https://doi.org/10.3390/catal12070792>
- [17] Li Jin, Yunhua Sun, Xiaoyuan Zhou. One-Step Synthesis of Layered Double Hydroxides by a Solvent-Free Method. *ACS Sustainable Chem. Eng.* 2022, 10, 39, 12955–12961. <https://doi.org/10.1021/acssuschemeng.2c04436>
- [18] Wei Chen, Xianfeng Yi, Ling Huang. Can Hammett indicators accurately measure the acidity of zeolite catalysts with confined space? Insights into the mechanism of coloration. *Catal. Sci. Technol.*, 2019, 9, 5045-5057, DOI <https://doi.org/10.1039/C9CY01392J>
- [19] D. W. Justin Leung, Chunping Chen, Jean-Charles Buffet and Dermot O'Hare. Correlations of acidity-basicity of solvent treated layered double hydroxides/oxides and their CO₂ capture performance. *Dalton Trans.*, 2020, 49, 9306-9311. <https://doi.org/10.1039/D0DT01587C>
- [20] Yanli Fu, Xiaoqian Fu, Wen Song, Yanfei Li, Xuguang Li, and Liangguo Yan. Recent Progress of Layered Double Hydroxide-Based Materials in Wastewater Treatment. 2023; 16 (16): 5723. <https://doi.org/10.3390/ma16165723>

Ref.:

Butenko, Eleonora – Kapustin, Alexey: *Layered double hydroxides, structures and properties*
Építőanyag – Journal of Silicate Based and Composite Materials,
Vol. 76, No. 4 (2024), 128–132 p.
<https://doi.org/10.14382/epitoanyag-jsbcm.2024.15>