

Theses of the Doctoral (Ph.D.) Dissertation

(Photo)catalytic application of layered double hydroxides with unique metal composition

Vivien Szabó

Supervisor:

Prof. Dr. Pál Sipos, *Professor*

Consultant:

Dr. Márton Szabados, *Research Fellow*

Doctoral School of Chemistry

Material and Solution Structure Research Group

Department of Molecular and Analytical Chemistry

Faculty of Science and Informatics

University of Szeged



Szeged

2025

1. Introduction and objectives

In the past two to three decades, layered double hydroxides (LDHs) have gained prominence across various fields of chemistry due to their favorable properties. Although hydrotalcite – the first naturally occurring representative of LDHs – was identified as early as 1842, significant research and industrial interest in these materials has only surged in recent decades.

LDHs are two-dimensional anionic clays that show considerable promise as both adsorbents and heterogeneous catalysts. They can be synthesized easily and cost-effectively, yielding materials with relatively high specific surface areas, and which may exhibit high catalytic activity and biocompatibility. Their structure is highly tunable via adjustment of the M(II)/M(III) cation ratio in the positively charged layers, as well as through the selection of easily exchangeable interlayer anions. These properties enable the efficient removal of a wide range of pollutants and targeted (photo)catalytic transformations, establishing LDHs as key materials in environmental and sustainable chemical technologies.

The literature describes numerous synthesis strategies for LDH-based composites, each resulting in materials with distinct structural characteristics and functionalities (e.g., incorporation of secondary metals or organic ligands, hybridization with other 2D materials). The current focus of research is the rational surface modification of LDHs to enable the selective and green conversion of biomass-derived feedstocks and the removal of organic micropollutants under environmentally benign conditions.

The primary goal of this dissertation was the synthesis of previously unreported LDH materials with novel structural, compositional, and physicochemical characteristics. The experimental work was carried out in three interrelated phases.

The first phase focused on the synthesis and fine-tuning of new Mn- and In-containing LDHs with conventional structures. Through deliberate selection of metal ion arrangements and interlayer anion compositions, we developed new materials with tunable optical, electronic, and acid-base properties.

In the second phase, we modified the less commonly studied gibbsite CoAl₄-LDH framework with Mn²⁺, Ni²⁺, Cu²⁺, and Zn²⁺ ions, then evaluated the hydrogenation performance of the resulting layered triple hydroxide (LTH) precursors in a continuous flow reactor. By investigating the catalytic activity of samples with varying compositions, we identified key structural motifs governing performance and achieved precise control over the physicochemical properties of the LDHs.

In the final phase, we explored the structural, optical, and photocatalytic properties of the scarcely investigated Ca_2Cr -LDH system, with particular attention to the effects of mechanical and sonochemical treatments, as well as its role in the degradation of lidocaine (LDC).

The overall significance of this research lies in the successful integration of sustainable and scalable synthetic and catalytic methodologies (mechanochemistry, sonochemistry, continuous flow), which enabled the preparation, detailed characterization, and (photo)catalytic application of nearly a dozen entirely novel LDH phases. Our results provide a unified framework for understanding structure–property relationships and may offer new directions for the rational design of functional LDHs, particularly in the context of catalytic and environmental applications.

2. Experimental section

2.1. Materials used and synthesis procedure

During the synthesis of MnAl -, MnSc -, MnCr -, MnFe -, MnGa -, MnIn -, and Co_xAl -type LDHs, the chloride salts of the corresponding metals and aqueous NaOH solution were used as starting reagents.

In the preparation of aluminum-rich and transition metal-deficient $\text{M(II)}\text{Al}_4$ -LDHs (where $\text{M(II)} = \text{Mn, Co, Ni, Cu, Zn}$), mechanochemically activated $\text{Al}(\text{OH})_3$ and the chloride salts of the respective M(II) cations were employed. In the case of MnAl_4 -LDH, bromide and sulfate salts were also used. To prepare aluminum-rich LTHs from these LDHs, additional chloride salts of Li^+ , Mg^{2+} , Co^{2+} , Ni^{2+} , Cu^{2+} , and Zn^{2+} ions were introduced alongside the respective LDH precursors.

Synthesis of MnAl -, MnSc -, MnCr -, MnFe -, MnGa -, MnIn -, and Co_xAl -LDHs

For the synthesis of Mn(II) -based LDHs, the commonly used co-precipitation method was applied. Manganese and indium chlorides were dissolved in distilled water and the resulting solution was added dropwise to an aqueous NaOH solution under mechanical stirring. As the synthesis required detailed optimization, various parameters were systematically altered: stirring duration (1–4 days), temperature (5–75 °C), base concentration (1–4 M), molar ratio of manganese to indium salts (Mn:In between 4:2 and 10:2), and reaction atmosphere (air, N_2 , H_2).

Syntheses performed in air were conducted in open vessels, and filtration, washing, drying, and storage were also carried out under ambient conditions. Working under N_2 or H_2 atmospheres required special conditions: the metal salt and base solutions were mixed in glass tubes inside a nitrogen-filled glovebox, after which the sealed tubes were transferred into high-

pressure, vertically aligned autoclaves. The autoclaves were sealed under N₂ and thoroughly purged with high-purity H₂ before being pressurized and closed. The sealed vessels were then placed into an oil bath on a magnetic stirrer.

For the synthesis of other Mn(II)-based LDHs, the optimized conditions established for MnIn-LDH (H₂ atmosphere, 4 days stirring at 50 °C, and 2 M NaOH, with a 4:1 Mn(II):M(III) molar ratio) were applied. Co_xAl-LDHs were also synthesized via co-precipitation, based on our experience and literature precedent (N₂ atmosphere, 2 days stirring at room temperature, pH ~12, 3 M NaOH), using Co(II):Al(III) molar ratios of 2:1 and 3:1.

Synthesis of M(II)Al₄-LDHs

The process began with the mechanochemical activation of Al(OH)₃ in a vibratory mill (Retsch MM400) under the following fixed parameters: 12 Hz grinding frequency, 6 hours dry milling, 100:1 ball-to-sample mass ratio. The activated Al(OH)₃ was then mixed with aqueous solutions of metal chlorides at room temperature for 6 days in air. For the incorporation of Co²⁺, Zn²⁺, Ni²⁺, and Cu²⁺, initial M(II):Al molar ratios of 2:1, 1:2, and 1:3, respectively, were used. In the case of Mn(II), the Mn:Al ratio and reaction temperatures were varied across a wide range. The resulting samples were washed with distilled water, filtered, and dried overnight at 70 °C. Syntheses under N₂ or H₂ atmospheres were conducted as described previously.

Synthesis of aluminum-rich LTHs

The Co_xM_{1-x}Al₄ layered triple hydroxides were synthesized using the above-described mechanochemical route, by co-incorporating M(II) cations via mixing the required metal salt solutions with Al(OH)₃. The molar ratio of the incorporated M(II) ions was adjusted by varying the initial molar ratio of metal chlorides while maintaining a fixed 2:1 Co:Al ratio. These ratios were selected based on the different impregnation abilities of the M(II) ions, quantified in our previous study. Washing, filtration, and drying of the LTHs followed the previously described procedures.

Preparation of reduced catalysts

Reduced LDH/LTH-derived catalysts (R-Co₂Al, R-Co₃Al, R-CoAl₄, or R-Co_xM_{1-x}Al₄, where M = Mn, Ni, Cu, Zn) were obtained via direct reduction of the solid materials. The hydroxides were placed in a tubular furnace and heated to 650 °C at a rate of 25 °C/min, holding this temperature for 1 hour. The gas atmosphere consisted of 150 cm³/min H₂ and 50 cm³/min N₂ (purchased from Messer Ltd., with H₂ purity 2.5 and N₂ purity 4.5).

Synthesis of hydrocalumite and Ca-containing hydrotalcites

Ca₂Cr-LDHs were synthesized via co-precipitation by slowly adding 20 cm³ of a calcium and chromium chloride solution (0.3 mol/dm³ Ca²⁺, 0.15 mol/dm³ Cr³⁺; Ca:Cr = 2:1) to 7.1 cm³ of NaOH solution, followed by stirring for 4 days (1000 rpm) under N₂ atmosphere. At the end of the synthesis, the LDHs were washed with 150 cm³ distilled water, filtered through 0.45 µm pore-size membranes, dried, and stored at 70 °C in a desiccator flushed with N₂.

Ca₂Al-, Ca₂Sc-, Ca₂V-, Ca₂Fe-, Ca₂Ga-, Ca₂In-, Mg₂Cr-, and Zn₂Cr-LDHs were prepared following the previously optimized synthesis parameters for CaIn-LDH with minimal modifications [M. Szabados et al., *J. Catal.* **391** (2020) 282]. Stirring was carried out at 50 °C for 4 days, except for Ca₂V-LDH, which was synthesized at room temperature to avoid oxidation of V³⁺. Filtration, washing, and drying followed the same protocol as for Ca₂Cr-LDH.

Mechanochemical structure modification

Ca₂Cr-, Mg₂Cr-, and Zn₂Cr-LDHs were ground under N₂ atmosphere using the conditions described in Section 4.2.1 (12 Hz for 60, 90, and 120 minutes, and 15 Hz for 60 and 90 minutes). Grinding vessels were sealed under nitrogen.

Ultrasound-assisted structural reconstruction

Ground Cr(III)-containing samples (0.15 g) were placed in sealed glass tubes with 2 cm³ CO₂ free distilled water. The resulting dispersions were subjected to ultrasonic treatment (Hielscher UP200Ht, 30 W) in a thermostated glass vessel at various temperatures (25, 40, 60, and 80 °C) and durations (1–96 hours). Control experiments were conducted using mechanical stirring (1000 rpm) under identical conditions. The solid phase was separated by filtration through a 0.45 µm membrane without washing and dried under N₂ at 70 °C for 24 hours.

2.2. Catalytic reactions

Catalytic transformation of hydroquinone

Test reactions were conducted both in the absence of light and under UV-Vis irradiation using a temperature-controlled photoreactor (15 W germicidal lamp, Lighttech GCL307T5L/CELL). For each experiment, 50 mg of catalyst (MnCl₂×4H₂O, InCl₃×4H₂O, LDHs, ZnO, TiO₂, or oxidized Mn(OH)₂ - comprising a mixture of M₃O₄, Mn₂O₃, and MnO₂) and 20 mg of hydroquinone were used (~2:3 molar ratio of LDH catalyst to substrate), dispersed/dissolved in 3 cm³ of acetonitrile. Reactions were carried out in open quartz glass tubes (5 cm³ volume) under magnetic stirring (500 rpm), at 25 ± 2 °C, in ambient air and atmospheric pressure, for 5 hours. Acetonitrile evaporation was monitored gravimetrically and found to be in the range of 5–10%. The quartz tubes were positioned at a fixed distance of ~10

mm around the light source. After the reaction, the mixtures were centrifuged, and the supernatants evaporated under vacuum. The resulting solids were washed repeatedly with acetonitrile, dried at room temperature, and stored under N₂ atmosphere. Reaction products were monitored quantitatively by ¹H-NMR spectroscopy (Bruker Avance NEO 500, 500 MHz, DMSO-d₆ solvent, ethylene carbonate as internal standard). Side products were identified using ¹H-NMR and GC-MS (Agilent 8890-5977B EI MSD).

Catalytic hydrogenation of cinnamaldehyde

Continuous-flow hydrogenation in the liquid phase was performed using an H-Cube[®] reactor system, where H₂ gas was generated in situ via water electrolysis. Catalysts were packed into heated stainless steel HPLC columns (dimensions: 4.6×100 mm), diluted with 1.9 g of SiO₂ (1 cm³ total bed volume, 0.1–0.8 mm particle size) to reduce aggregation. Before and after each experiment, the reactor was flushed with isopropanol (IPA) for 30 minutes at a flow rate of 0.5 cm³/min. Cinnamaldehyde (CAL) was dissolved in 4 cm³ IPA, passed through the catalyst bed, and product solutions were collected under steady-state conditions. For optimization, reaction parameters such as temperature, H₂ pressure, flow rate, CAL concentration, and catalyst composition and loading were systematically varied. Products were identified by ¹H-NMR spectroscopy (Bruker Avance NEO 500, 500 MHz) and GC-MS (Thermo Scientific Trace 1310, ISQ QD).

Photocatalytic degradation of lidocaine

The photocatalytic potential of both pristine and mechanochemically modified hydrocalumite and hydrotalcites was assessed through the degradation of lidocaine, employed as a model pharmaceutical contaminant. The experimental basis was adapted from the photocatalytic degradation of naproxen over CuZnAl-LDH [B. Peng et al., *CrystEngComm* **24** (2022) 5080], with modifications inspired by the degradation of other xenobiotics (e.g., lidocaine, diclofenac) using ZnO, WO₃, and g-C₃N₄ catalysts. Experiments were performed in a thermostated photoreactor: 100 ml dispersions of lidocaine, water, and LDH were irradiated in open quartz tubes with UV-Vis light (Lighttech GCL307T5L/CELL, 15 W), with the light source positioned axially ~50 mm from the center of the reactor. The catalyst dosage, lidocaine concentration, and pH were systematically varied over a wide range. Prior to irradiation, dispersions were stirred in the dark for 1 hour at room temperature under ambient air to establish adsorption–desorption equilibrium, followed by 5 hours of irradiation at 500 rpm. During the reaction, 2 ml samples were collected hourly. Lidocaine degradation was monitored via gas chromatography (Thermo Scientific Trace 1310, ISQ QD MS detector, TG-SQC column), by

determining initial (C_0) and time-dependent (C_t) concentrations using dimethyl sulfoxide as internal standard and commercial calibration solutions.

2.3. Structural characterization methods

Powder X-ray diffraction (XRD) measurements were performed using a Rigaku Miniflex II diffractometer. Normalized diffractograms were recorded with a scintillation detector at a scanning rate of 2°/min in continuous mode over a 2θ range of 3–80°. Identification of LDH/LTH phases and assignment of characteristic reflections and Miller indices were carried out using the JCPDS-ICDD database (Joint Committee on Powder Diffraction Standards – International Centre for Diffraction Data – 1998). Average crystallite sizes of LDHs were calculated from the first diffraction peaks using Gaussian fitting and the Scherrer equation, while for metallic phases, calculations were based on the most intense (111) reflection.

The thermal stability and decomposition behavior of the samples were examined by thermogravimetry (TG), differential thermogravimetry (DTG), and differential thermal analysis (DTA) using a Netzsch STA 409 PC Luxx derivatograph. Measurements were performed in synthetic air at a constant flow rate of 60 mL/min with a heating rate of 10 °C/min. For each analysis, 50–60 mg of sample was placed in high-purity α -alumina crucibles.

For Al-rich samples, a TA Instruments Discovery TGA thermobalance was operated under a continuous Ar flow containing 10% O₂ (60 mL/min) at a heating rate of 10 °C/min. A Faraday cup detector was used in the mass spectrometer to detect ions in the 1–300 m/z range, in full scan mode with electron impact ionization (70 eV). The gas connection between the thermobalance and the mass spectrometer (inert quartz capillary) was heated to 200 °C.

Sample morphology was studied using scanning electron microscopy (SEM, Hitachi S-4700). Samples were mounted on double-sided carbon adhesive tape. To improve image clarity, a thin gold coating (a few nanometers thick) was applied using a sputter coater. Elemental composition and mapping were performed using a Röntec QX2 energy-dispersive X-ray spectrometer (EDX) attached to the SEM.

The structural properties of LDHs were characterized by Fourier-transform infrared (FT-IR) spectroscopy using a JASCO FT/IR-4700 spectrophotometer, with a resolution of 2 cm⁻¹ and averaging over 256 scans. Measurements were carried out using a ZnSe ATR accessory and a DTGS detector.

Raman spectroscopy was performed using a Bruker Senterra II Raman microscope with a 2.5 mW laser operating at an excitation wavelength of 532 nm. Spectra were collected using 32 scans with an exposure time of 8 seconds and a 50× objective.

UV–Vis spectra of the LDHs were recorded using diffuse reflectance spectroscopy (DRS) on an Ocean Optics USB4000 spectrometer equipped with a DH-2000-BAL light source. BaSO₄ was used as the white reference in the 220–850 nm wavelength range. Optical band gaps were determined by extrapolating the linear region of the modified Schuster–Kubelka–Munk function plotted as a function of photon energy.

Elemental analysis of both solution and solid samples was carried out by inductively coupled plasma mass spectrometry (ICP-MS) using an Agilent 7900 instrument and ICP Multielement Standard Solution IV (CertiPUR) as reference.

The solvodynamic diameter, particle size distribution, and zeta potential (surface charge) of the LDHs were determined by dynamic light scattering (DLS) using a Malvern NanoZS instrument. Measurements were conducted using a 4 mW He–Ne laser ($\lambda = 633$ nm) in backscattering mode at 173°. Samples were dispersed in cyclohexanone via 2 hours ultrasonic treatment at a concentration of 0.1 g/dm³.

Acidity and basicity were investigated by NH₃-TPD and CO₂-TPD, respectively. Measurements were performed using a BELCAT-A instrument, applying CO₂/He and NH₃/He gas mixtures. Prior to analysis, surface contaminants were removed by preheating the samples to 150 °C. The amount of desorbed gases was monitored using a thermal conductivity detector (TCD).

Specific surface area was determined via N₂ adsorption–desorption measurements using a Quantachrome Autosorb iQ instrument. Prior to measurements, samples were pretreated at 150 °C for 2 hours to remove any surface contaminants. Surface areas were calculated from the adsorption branches using the Brunauer–Emmett–Teller (BET) equation.

3. New scientific results

T1. To the best of our knowledge, we were the first to report the synthesis of Mn(OH)₂-based, In(III)-containing layered double hydroxides (LDHs) in the literature. Phase-pure MnIn-LDHs with chloride as the interlayer anion were successfully synthesized over a wide Mn:In molar ratio range (between 5:2 and 8:2). Comprehensive materials characterization (XRD, IR, TG, SEM-EDX) confirmed the formation of the LDH phase.

The incorporation of In(III) into the layered structure of Mn(II) hydroxide – and thus the synthesis of MnIn-LDHs – was successfully achieved using the co-precipitation method, which is commonly applied for LDH preparation. Achieving a practically phase-pure product required a reductive (hydrogen) atmosphere; although the LDH phase also formed under air or nitrogen, significant amounts of Mn₃O₄ by-product were observed in both cases. The amount of

incorporated In(III) also proved to be a critical parameter. At Mn:In molar ratios below 5:2, part of the indium precipitated as separate In(OH)_3 , while above 8:2, intensive oxidation of Mn(OH)_2 became unavoidable.

T2. Mn_xIn_2 -LDHs ($x = 5-8$) were successfully applied in the catalytic and photocatalytic degradation of hydroquinone. Among them, Mn_4In -LDH exhibited the highest activity. The results were compared with the performance of other Mn_4M -LDHs ($\text{M} = \text{Al, Sc, Cr, Fe, Ga}$). The greatest photocatalytic contribution was observed upon incorporation of Cr(III), In(III), and Fe(III), and the extent of benzoquinone formation and further degradation was found to strongly depend on the chemical nature of the trivalent metal cation.

Under UV-Vis irradiation, the catalytic performance of MnIn-LDHs in hydroquinone conversion improved with increasing Mn:In molar ratios. This behavior was attributed to the change in optical band gap, which was found to decrease with increasing Mn:In ratio. The catalytic performance of the various Mn(II)-based LDHs in hydroquinone oxidation, expressed in terms of benzoquinone yield and selectivity, showed significant variation, depending on the chemical identity of the trivalent metal cation under both illuminated and dark conditions. In addition to the observed photocatalytic contribution, it is assumed that the basic character of the hydrotalcites, their water/hydroxide ion content, and the presence of redox-active manganese played a key role in the mineralization of quinone compounds in acetonitrile. Under identical conditions, commonly used photocatalysts such as ZnO and TiO₂ (Degussa P25) exhibited no photocatalytic activity in the oxidation of hydroquinone.

T3. We were the first to demonstrate that aluminum-rich layered double hydroxides (LDHs) can also be formed by incorporating Mn(II) ions. Within the lyotropic series describing the incorporation affinity of metal cations, manganese ions were positioned between Co(II) and Mg(II).

Mn(II) ions were successfully incorporated into the octahedral sites of gibbsite layers to synthesize unique Al-rich LDHs containing chloride, bromide, and sulfate as interlayer anions. Despite extensive optimization of the synthesis parameters, phase-pure MnAl₄-LDHs could not be obtained. However, the position of Mn(II) in the incorporation affinity series could be determined relative to Ni(II), Cu(II), Zn(II), Co(II), and Mg(II) ions. This study also highlighted the critical importance of the mechanochemical pre-treatment of gibbsite. Post-synthetic filling of residual octahedral vacancies for the preparation of Mn(II)-containing layered trivalent hydroxides (LTHs) proved unsuccessful due to the cation exchange capacity of these

nanostructures. However, by applying the co-incorporation approach, several $\text{Co}_x\text{M}_{1-x}\text{Al}_4$ -LTHs were successfully synthesized, where $\text{M(II)} = \text{Mn, Ni, Cu, or Zn}$.

T4. For the first time, we investigated the performance of Co-deficient/Al-rich LDH/LTH precursors under flow conditions in the catalytic reduction of cinnamaldehyde. We demonstrated that partial substitution of Co(II) with Mn^{2+} , Ni^{2+} , Cu^{2+} , or Zn^{2+} significantly enhanced the catalytic activity of the resulting CoAl_4 -LDH-derived catalysts.

The catalytic activity of H_2 reduced Al-rich LDH/LTH materials was tested in the hydrogenation of cinnamaldehyde under continuous flow, and the results were compared with those obtained from Co-rich samples (prepared via the reduction of conventional Co_3Al - and Co_2Al -LDHs). The Al-rich catalysts performed competitively with the Co-rich analogues, exhibiting high selectivity toward HCAL (3-phenylpropanal). In contrast, the Co_3Al - and Co_2Al -LDH-derived catalysts led to complete reduction, resulting primarily in the formation of HCOL (3-phenylpropanol). The Al-rich LTH precursors modified with Mn, Ni, Cu, or Zn displayed enhanced activity compared to the reduced CoAl_4 -LDH, while maintaining high HCAL selectivity – except in the case of Mn incorporation, where increased formation of HCOL was observed.

T5. For the first time, Ca_2Cr -LDH was applied in the photocatalytic degradation of lidocaine molecules in aqueous media. Under neutral pH conditions, hydrocalumite dissolved and acted as a sacrificial template, resulting in the formation of catalytically active, nanometer-sized Cr(OH)_3 particles. By adding base to prevent LDH dissolution, it was demonstrated that the photocatalytic activity of the catalysts could be effectively enhanced through mechanochemical and sonochemical post-treatments.

Investigation of the lesser-known Ca_2Cr -hydrocalumite's photochemical properties in lidocaine photomineralization revealed that the pH of the medium played a crucial role due to the enhanced aqueous solubility of this LDH type. The study was extended from the frequently synthesized Ca_2Al - and Ca_2Fe -LDHs to the scarcely reported Ca_2Sc -, Ca_2V -, Ca_2Ga -, and Ca_2In -LDHs. These potentially bio-based hydrocalumites behaved as sacrificial templates below pH 12, predominantly forming small and amorphous M(III) -hydroxide/oxide particles, most of which exhibited catalytic activity. Under strongly alkaline conditions, structurally reconstructed samples showed superior catalytic performance compared to both the original and the milled LDHs. Interestingly, in the case of Ca_2Cr -LDH, only mild treatment was sufficient to obtain enhanced activity, whereas for Mg_2Cr - and Zn_2Cr -hydrotalcites, longer regeneration times were required to produce more active catalysts. Overall, it can be concluded that mechanochemical

treatment followed by mechanical or ultrasonic regeneration steps positively influence the photocatalytic performance of LDHs.

4. Practical significance of the results

The results presented in this dissertation were obtained in the context of fundamental research; nevertheless, taken together, they contribute to the development of advanced LDH-based catalysts and photocatalysts, which may play a significant role in environmental protection, improving the efficiency of industrial processes, and optimizing energy usage.

At the beginning of my experimental work, novel MnIn-LDHs were developed, followed by the synthesis of further variants incorporating other metal ions. Their catalytic and photocatalytic activity in the oxidation of hydroquinone was compared. Such materials can contribute to the effective degradation of pollutants in water and promote environmentally friendly chemical processes. In the second research topic, LDH-based catalyst precursors were developed for continuous-flow hydrogenation reactions. These materials were modified with various metal ions to enhance reaction efficiency. The resulting catalysts can support more sustainable and energy-efficient organic reduction syntheses – particularly relevant for applications in the cosmetic and pharmaceutical industries. The most recent results demonstrate the structural regeneration of LDHs via ultrasonic treatment following mechanical amorphization, with special attention to their photocatalytic activity in the degradation of lidocaine – a common contaminant in hospital wastewater. This post-synthetic LDH structural modification approach may improve the removal of pharmaceutical residues from water, contributing to the preservation of cleaner water resources.

Although each of the three studies is fundamentally rooted in basic research, together they may facilitate the development of sustainable technologies, reduce the environmental burden of industrial processes, and promote the broader adoption of renewable chemical methodologies.

5. Publications and conference presentations

The Hungarian Scientific Bibliography (MTMT) identification number: 10068946

5.1. Publications in peer-reviewed journals related to the dissertation

[1] **Vivien Szabó**, Rebeka Mészáros, Zoltán Kónya, Ákos Kukovecz, István Pálinkó, Pál Sipos, Márton Szabados: *Preparation and characterization of MnIn-layered double hydroxides (LDHs), extension of the synthesis to fabricate MnM(III)-LDHs (M = Al, Sc, Cr, Fe, Ga), and the comparison of their photocatalytic and catalytic activities in the oxidation of hydroquinone*

JOURNAL OF MOLECULAR STRUCTURE 1261 (2022) 132966.

Impact factor: 3,8 (Q2)

Independent citations: 7

[2] Rebeka Mészáros, **Vivien Szabó**, Bence Kutus, Kornélia Baán, Zoltán Kónya, Ákos Kukovecz, Pál Sipos, Márton Szabados: *Continuous-flow hydrogenation of cinnamaldehyde over catalysts derived from modified $CoAl_4$ layered double hydroxides incorporating Mn, Ni, Cu and Zn ions*

APPLIED CATALYSIS A-GENERAL 679 (2024) 119738.

Impact factor: 4,8 (Q2)

Independent citations: 1

[3] **Vivien Szabó**, Rebeka Mészáros, Bence Kutus, Gergely Ferenc Samu, Zoltán Kónya, Ákos Kukovecz, Pál Sipos, Márton Szabados: *Sonochemically improved regeneration of mechanically amorphized Ca_2Cr -layered double hydroxides – Synthesis, characterization and photocatalytic lidocaine decomposition*

APPLIED CLAY SCIENCE 265 (2025) 107684.

Impact factor (2024): 5,8 (Q1)

Independent citations: 0

5.2. Publications related to the dissertation, published in conference proceedings and other volumes

[1] **Vivien Szabó**, Márton Szabados, Pál Sipos: *Synthesis of In(III)-containing Mn(II)-based layered double hydroxides with NO_3^- , Cl^- and SO_4^{2-} interlayer anions and a comparative study of their catalytic decomposition of hydroquinone*, Recent Progress in Coordination, Bioinorganic and Applied Inorganic Chemistry, Bratislava, Slovakia, Slovak Chemical Society (2022) 63–72, ISBN 978-80-8208-081-3.

5.3. Full conference presentations related to the dissertation

[1] **Vivien Szabó**, Márton Szabados, Pál Sipos: *Synthesis of In(III)-containing Mn(II)-based layered double hydroxides with NO_3^- , Cl^- and SO_4^{2-} interlayer anions and a comparative study for their catalytic decomposition of hydroquinone*, XXVIII. International Conference on Coordination and Bioinorganic Chemistry, Presentation, Bratislava, Slovakia, June 5–10, 2022.

[2] **Vivien Szabó**, Rebeka Mészáros, Zoltán Kónya, Ákos Kukovecz, István Pálinkó, Pál Sipos, Márton Szabados: *Transformation of hydroquinone with various M(III) metal ion containing Mn(II)-based layered double hydroxides (LDHs)*, 1st Forum of Young Researchers on Heterogeneous Catalysis, Presentation, Szeged, Hungary, July 11–13, 2022.

[3] **Vivien Szabó**, Rebeka Mészáros, Pál Sipos, Márton Szabados: *Synthesis of $Al(OH)_3$ based layered double and triple hydroxides containing manganese, cobalt, nickel, copper and zinc*,

catalytic use under continuous flow conditions in hydrogenation reaction, 4th Young Researchers' International Conference on Chemistry and Chemical Engineering, Poster, Debrecen, Hungary, June 1–3, 2023.

[4] **Vivien Szabó**, Rebeka Mészáros, Ákos Serfőző, Kornélia Baán, Zoltán Kónya, Ákos Kukovecz, Pál Sipos, Márton Szabados: *Incorporation of Mn, Ni, Cu and Zn ions into CoAl₄ layered double hydroxide catalyst precursors*, XXIX. International Conference on Coordination and Bioinorganic Chemistry, Presentation, Bratislava, Slovakia, June 2–7, 2024.

5.4. Publications in peer-reviewed journals not related to the dissertation

[1] Attila Kunfi, **Vivien Szabó**, Dr. Ágnes Mastalir, Dr. Imre Bucsi, Dr. Miklós Mohai, Dr. Péter Németh, Dr. Imre Bertóti, Dr. Gábor London: *Palladium on polydopamine: its true potential in catalytic transfer hydrogenations and Heck coupling reactions*

CHEMCATCHEM 9 (2017) 3236–3244.

Impact factor: 4,674 (Q1)

Independent citations: 22

Summary

Publications in Journals: 4 related to the dissertation: 3

Cumulative Impact Factor: 19.074 related to the dissertation: 14.4

Independent Citations: 30 related to the dissertation: 8