

Selectivity of Ni/Al LDH Material Supported by Green Tea Leaf (*Camellia sinensis*) Extract as an Adsorbent for Removing Cationic Dyes in Wastewater Treatment

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Abstract

Water pollution due to synthetic dye waste is a serious environmental problem, so an effective method is needed to overcome it, including using adsorption. In this study, Ni/Al-CS materials were synthesized and characterized to evaluate their selectivity towards Rhodamine B (RhB), Malachite Green (MG), and Methylene Blue (MB) dyes. Adsorption selectivity tests were conducted using UV-Vis spectroscopy to determine changes in dye concentration before and after interaction with the material. Results showed that Ni/Al-CS had the highest adsorption capacity towards MG, followed by MB and RhB. With its selective adsorption properties, Ni/Al-CS material has the potential to be used as an effective adsorbent in the treatment of dye effluents in aquatic environments, especially MG dye.

Keywords

Layered Double Hydroxide, Selectivity, *Camellia sinensis*, Extract, Cationic Dyes

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1. INTRODUCTION

Layered Double Hydroxide (LDH) is a layered structured material capable of forming organic-inorganic composites due to its anion exchange properties. They are often referred to as “anionic clays” or hydrotalcite-like compounds, with the general formula $[M^{2+}_{(1-x)}M^{3+x}(OH)_2]^{x+}(A^{n-})_{x/n} \cdot mH_2O$ (Jamshidi et al., 2025; Li et al., 2025). In its structure, LDH contains divalent cations (such as Mg^{2+} , Co^{2+} , Cu^{2+} , Ni^{2+} , Zn^{2+}) and trivalent cations (such as Al^{3+} , Co^{3+} , Cr^{3+} , Fe^{3+}), as well as anions such as Cl^- , NO_3^- , OH^- , CO_3^{2-} , SO_4^{2-} , and PO_4^{3-} (Khlyustova et al., 2025; Ma et al., 2024). This structure consists of positively charged layers stabilized by balancing anions (A^{n-}) and hydration water molecules (mH_2O) (Tamura et al., 2025).

LDH with its distinctive layered structure and host-guest interaction properties allows it to have a large surface area and high ion exchange capacity. These characteristics make LDH a highly effective adsorbent for pollutants in the aquatic environment (Xu et al., 2025). The use of additional support materials is necessary to increase the adsorption capacity of LDH (Wang et al., 2025). This is due to the often limited surface activity of LDH, resulting in a relatively small number of adsorption sites as well as a tendency to aggregate, which can reduce the overall adsorption effectiveness (Huang et al., 2025). Ahmad et al. (2024a) conducted the addition of supporting materials, namely lignin,

in the removal of anionic dyes. Green tea (*Camellia sinensis*) contains polyphenols, which mostly consist of flavonoids and can be further classified into flavones, flavanones, isoflavones, flavanol-flavandiols, anthocyanins, as well as phenolic acids (Loto et al., 2014).

Water pollution is a serious global problem due to the discharge of large amounts of industrial effluents. Among various pollutants, organic dyes are of major concern due to their detrimental impact on aquatic ecosystems. Industries such as food, cosmetics, paper, textiles, leather, pharmaceuticals and printing use dyes in large quantities, resulting in contaminated wastewater that pollutes the surrounding aquatic environment (Sowmya and Joseph, 2025). Cationic organic dyes, such as Malachite Green (MG), Rhodamine B (RhB), and Methylene Blue (MB), are major contaminants in textile and dyeing industry wastewater (Rakcho et al., 2025; Wen et al., 2025; Yilmaz et al., 2021). In addition to polluting water resources, these compounds have potential carcinogenic, teratogenic and mutagenic effects on humans (Wen et al., 2025). Therefore, it is necessary to remove the presence of these dyes from the environment (Wijaya et al., 2024).

This study aims to modify Ni/Al LDH with green tea (*Camellia sinensis*) leaf extract to improve its performance as an adsorbent. The modified materials were analyzed using fourier transform infrared spectroscopy (FT-IR) to identify functional

groups. In addition, the material was tested for selectivity against various cationic dyes namely RhB, MG, and MB to determine the type of dye most effectively adsorbed by Ni/Al-CS.

2. EXPERIMENTAL SECTION

2.1 Chemicals and Instrumentation

This study used various chemicals, including cationic dyes (methylene blue, malachite green, and rhodamine B) and ethanol 96% (C₂H₅OH). Nickel nitrate hexahydrate (Ni(NO₃)₂·6H₂O), aluminium nitrate nonahydrate (Al(NO₃)₃·9H₂O), and sodium hydroxide (NaOH) were obtained from Sigma-Aldrich. Hydrochloric acid (HCl) was obtained from Mallinckrodt. Distilled water (H₂O) was used as a solvent and obtained from BrataChem Indonesia. Green tea leaves as a natural ingredient in this study were obtained from a supermarket. Characterization of functional groups in the modified material was carried out using Fourier Transform Infrared (FTIR) spectroscopy with a Shimadzu Prestige-21 instrument. Measurement of the concentration of adsorbed substances was carried out using a UV-Vis spectrophotometer type EMC-18PC.

2.2 Extraction of *Camellia sinensis* and Preparation of Ni/Al-CS

Dried green tea (*Camellia sinensis*/CS) leaves were pulverized, saturated in 96% ethanol, and stirred for 1 h. After filtering, the filtrate obtained was evaporated using a rotary evaporator at 48°C to produce green tea leaf extract. Ni/Al-CS materials were synthesized through a co-precipitation method based on Luo et al. (2023). A total of 0.5 gram of green tea leaf extract was added to a solution of metal ions Ni²⁺ and Al³⁺ (5:1 ratio) in 200 mL of distilled water. The pH of the solution was adjusted to 10 using 0.5 M NaOH, and then stirred at room temperature for 24 h. The materials obtained were washed repeatedly and the pH was adjusted to 10 using 0.5 M NaOH. The obtained material was washed repeatedly and dried at 60°C. Characterization was carried out using FT-IR analysis.

2.3 Cationic Dye Selectivity

A total of 20 ppm solutions of dyes RhB, MG, and MB were mixed homogeneously. From the mixture, 20 mL of solution was taken and 0.02 gram of each adsorbent was added. The mixture was then stirred according to the predetermined contact time for each material. After the adsorption process, the solution was tested using a UV-VIS spectrophotometer to measure the decrease in intensity of the absorption peak of each dye.

3. RESULTS AND DISCUSSION

Figure 1 shows the FT-IR spectrum of the Ni/Al LDH and Ni/Al-CS material with some characteristic absorption bands, which provide information related to the functional groups in the material. The absorption band observed at around 3440 cm⁻¹ is associated with O–H stretching vibrations of hydroxyl groups (–OH), which originate from hydroxyl groups on the surface of the material and OH groups of polyphenols (alcohols and phenols) in catechin compounds (Gao et al., 2025; Rohmatullaili

et al., 2024; Tamang et al., 2025). The absorption band observed at 1380 cm⁻¹ can be attributed to the presence of nitrate anion (NO₃⁻) in the LDH material interlayer (Mansour et al., 2025; Oktiyanti et al., 2019). Significant absorption bands are also seen in the 800-400 cm⁻¹ range, which are generally associated with metal-oxygen (M–O) or metal-hydroxide (M–OH) bond vibrations, which in this case can be attributed to the interaction between Ni/Al in the material structure (Ahmad et al., 2024b; Liu et al., 2025). In the Ni/Al-CS material there is an absorption band at 1620-1650 cm⁻¹ which indicates the presence of carbonyl groups (C=O) from polyphenols such as CG, ECG, EGC, EGCG, GCG which are characteristic of green tea leaf extract (*Camellia sinensis*) (Tamang et al., 2025). The peaks generated from the spectra indicate the successful formation of the Ni/Al-CS composite material.

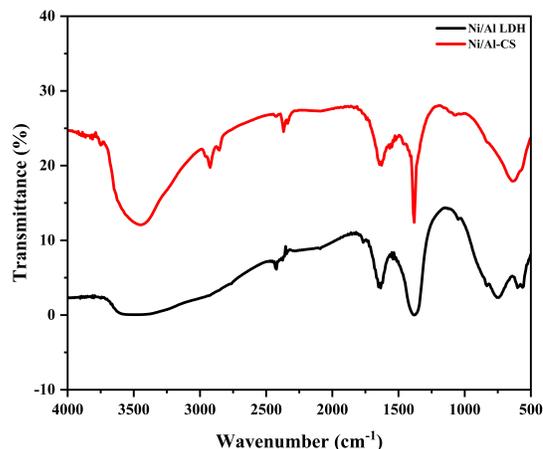


Figure 1. FTIR spectra of NiAl LDH and Ni/Al-CS

The selectivity of dye adsorption by Ni/Al-CS materials was evaluated using three different types of cationic dyes, namely Rhodamine B (RhB), Malachite Green (MG), and Methylene Blue (MB). The evaluation was carried out by analyzing the change in absorbance at certain wavelengths, as well as by measuring the concentration of dyes adsorbed by the Ni/Al-CS material. Based on the absorbance spectra Figure 2(a), there are differences in the absorbance peak intensity measured against the wavelength for the three dyes tested, both before and after the adsorption process using Ni/Al-CS. The absorbance peaks for each dye were observed at different wavelengths, which were around 554 nm for RhB, 617 nm for MG, and 664 nm for MB. It can be seen that after the adsorption process, there is a decrease in absorbance intensity that varies for each dye. The largest decrease in absorbance intensity occurred for MG, followed by MB, while the decrease for RhB was relatively smaller. This indicates that the Ni/Al-CS material is more effective in adsorbing MG and MB compared to RhB.

This result is confirmed by the adsorption capacity data presented in Figure 2(b). From the graph, it shows that MG has the

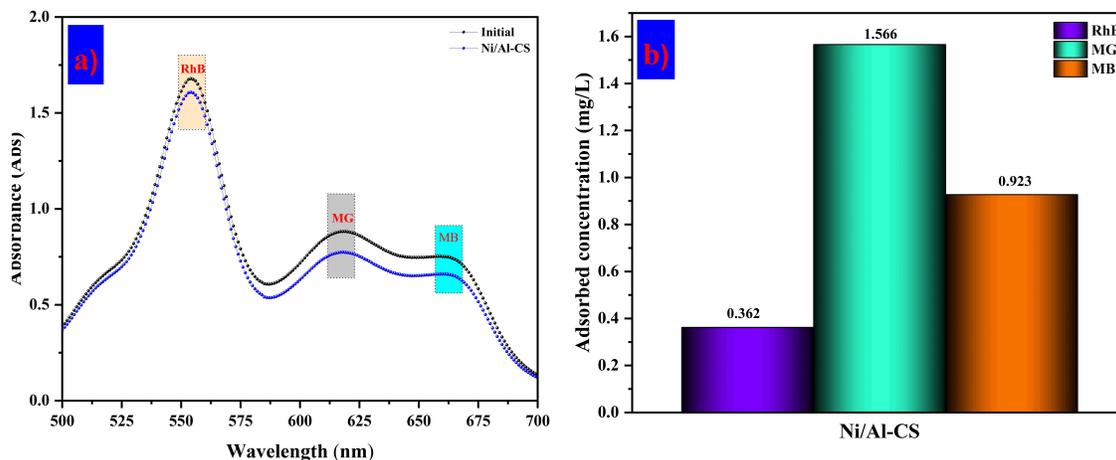


Figure 2. Selectivity of Ni/Al-CS Materials to RhB, MG, and MB Dyes Based on (a) UV-Vis Spectra Before and After Interaction with Ni/Al-CS; (b) Concentration of Dyes Adsorbed by Ni/Al-CS

highest adsorption capacity, which is 1.566 mg/L, indicating that the Ni/Al-CS material is highly selective towards MG. Methylene blue was also adsorbed in a fairly large amount, 0.923 mg/L, although not as much as MG. In contrast, RhB was adsorbed with a much lower capacity, which was only 0.362 mg/L. This difference in adsorption capacity indicates that Ni/Al-CS is more selective towards cationic dyes, especially MG and MB, compared to RhB.

Based on these results, it can be concluded that the Ni/Al-CS material has high selectivity to cationic dyes, especially MG, followed by MB, and the lowest to RhB. This suggests that this material has the potential to be further developed as a selective adsorbent in the treatment of industrial waste containing cationic dyes.

4. CONCLUSIONS

Based on the analysis, the Ni/Al-CS material showed different adsorption ability towards Rhodamin B (RhB), Malachite Green (MG), and Methylene Blue (MB) dyes. The decrease in absorbance intensity after interaction with Ni/Al-CS, showed the largest decrease occurred for MG, followed by MB and RhB. These results were corroborated by quantitative analysis which showed that the material had the highest adsorption capacity towards MG compared to MB and RhB. Thus, Ni/Al-CS materials have potential as selective adsorbents in the treatment of effluents containing certain dyes, especially Malachite Green.

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REFERENCES

Ahmad, N., A. R. Marlan, and S. P. J. Negara (2024a). Insight of Anionic Dyes Adsorption from Their Aqueous Solutions onto

- MgAl. *Indonesian Journal of Material Research*, 2(2); 40–46
- Ahmad, N., A. Wijaya, F. S. Arsyad, I. Royani, and A. Lesbani (2024b). Layered Double Hydroxide-Functionalized Humic Acid and Magnetite by Hydrothermal Synthesis for Optimized Adsorption of Malachite Green. *Kuwait Journal of Science*, 51(2); 100206
- Gao, X., X. W. Han, J. Sun, S. Wang, C. Lu, and H. Zou (2025). (Mg-Fe) LDHs Decorated 3D Reduced Graphene Oxide Nanocomposite for Adsorption of Ciprofloxacin and Norfloxacin. *Diamond and Related Materials*, 153; 112085
- Huang, A., N. Zhang, Q.-B. Wang, B.-H. Zhao, R.-B. Zhang, M. Cheng, C. Shi, and X.-D. Hao (2025). Enhanced Wastewater Treatment Using Biochar-Supported Layered-Double-Hydroxide Composites. *Chemical Engineering Journal*, 506; 159969
- Jamshidi, E., F. Fathabadi, F. Manteghi, and R. Eshaghimalekshah (2025). Adsorption Characteristics of Metronidazole on CoZr-LDH and Its GO Nanocomposite: Experimental and Theoretical Study. *Heliyon*, 11(3); e42396
- Khlyustova, A., N. Sirotkin, M. Shipko, Z. Kalazhokov, H. Kalazhokov, M. Stepovich, E. Savchenko, and A. Agafonov (2025). Plasma Chemical Synthesis of Magnetic Fe₂O₃-Ni-Cr Layered Double Hydroxide Composites for Environmental Applications. *Solid State Communications*, 399; 115886
- Li, X., Z. Huang, B. Wan, L. Yang, Z. Chen, S. Zhang, X. Liu, C. Cheng, Y. Zhu, C. Lu, J. Zou, L. Chen, and S. Luo (2025). Oxygen Vacancy-Enriching Co₃O₄/FeNi-LDH Boosts Rapid Degradation of Formaldehyde Under Natural Light at Room Temperature. *Separation and Purification Technology*, 362; 131964
- Liu, N., T.-B. Fang, L.-X. Zhao, N. Li, X.-F. Chen, and R.-S. Zhao (2025). Efficient Adsorption Behavior of Fe-Based Ternary Magnetic LDHs for Naphthalene Acetic Acid: Role of Fe Element. *Environmental Research*, 269; 120848
- Loto, C. A., O. O. Joseph, and R. T. Loto (2014). Adsorption and

- Inhibitive Properties of *Camellia Sinensis* for Aluminium Alloy in HCl. *International Journal of Electrochemical Science*, **9**(7); 3637–3649
- Luo, Y., Y. Wang, F. Hua, M. Xue, X. Xie, Y. Xie, S. Yu, L. Zhang, Z. Yin, C. Xie, and Z. Hong (2023). Adsorption and Photodegradation of Reactive Red 120 with Nickel-Iron-Layered Double Hydroxide/Biochar Composites. *Journal of Hazardous Materials*, **443**(PB); 130300
- Ma, Y., Q. Luo, J. Li, X. Wang, F. Ma, M. Li, X. Ye, K. Li, X. Du, X. Wang, and Z. Wu (2024). One-Pot Synthesis of Transition Metals-Doped LiAl-LDHs to Improve Lithium Adsorption Performance and Stability. *Process Safety and Environmental Protection*, **192**; 878–886
- Mansour, E. A., M. Taha, R. K. Mahmoud, N. Shehata, and R. M. Abdelhameed (2025). A Combined Experimental and Computational Studies on Thiophene Adsorption from Liquid Fuels over ZIF-67@ZnFe LDH Composites. *Materials Chemistry and Physics*, **334**; 130499
- Oktriyanti, M., N. R. Palapa, R. Mohadi, and A. Lesbani (2019). Modification of Zn-Cr Layered Double Hydroxide with Keggin Ion. *Indonesian Journal of Environmental Management and Sustainability*, **3**(3); 93–99
- Rakcho, Y., M. Baidou, A. Naboulsi, A. Bouazizi, M. Mouiya, H. Sehaqui, Y. Abouliatim, A. Benhammou, M. Ouammou, A. Abourriche, and J. Alami (2025). Fabrication of Low-Cost Ceramic Nanofiltration Membrane from Natural Resources for the Removal of Cationic and Anionic Dyes: Experimental and DFT Investigations. *Chemical Engineering Journal*, **505**; 159779
- Rohmatullaili, N. Ahmad, D. Erviana, Zultriana, D. Savira, R. Mohadi, and A. Lesbani (2024). ZnAlLDH-Based Derivative Materials as Photocatalysts: Synthesis, Characterization, and Catalytic Performance in Tetracycline Degradation. *Science and Technology Indonesia*, **9**(2); 457–469
- Sowmya, P. and A. Joseph (2025). Effective Removal of Cationic Dyes in a Single Aliquot Using Thiophene-Based Bis-Chalcone Polymers. *Reactive and Functional Polymers*, **210**; 106189
- Tamang, R., A. K. Mehata, V. Singh, M. S. Muthu, and B. Koch (2025). Sustainable Synthesis of *Camellia Sinensis*-Mediated Silver Nanoparticles (CsAgNP) and Their Anticancer Mechanisms in Breast Cancer Cells. *Biomaterials Advances*, **166**; 214072
- Tamura, K., J. Tanks, D. Niizeki, Y. Watanabe, T. Kogure, H. Sakuma, and M. Kamon (2025). Heavy Metal Ion Adsorption Mechanism of Polysulfide-Modified Layered Double Hydroxide. *Applied Clay Science*, **269**; 107759
- Wang, X., H. Li, X. Li, J. Wu, Z. Pang, Z. Ji, and S. Wang (2025). Separation of Ethylparaben Using Cellulose Nanocrystals/Mg-Al Layered Double Hydroxides Composite Films and Mechanistic Investigation. *Journal of Environmental Chemical Engineering*, **13**(1); 115157
- Wen, C., Y. Huang, W. Zhang, J. Tian, C. Dong, C. Yang, and W. Liang (2025). Efficient and Convenient Purification Strategy Using Maltodextrin-Based Nanosponges for Rapid Removal of Cationic Dyes. *Separation and Purification Technology*, **362**; 131702
- Wijaya, A., R. Ardiansyah, E. Augustiara, and S. Fajri (2024). Preparation of Hydrochar from Longan Peel: A Promising Adsorbent for Cationic Dye Removal in Aqueous Solutions. *Indonesian Journal of Material Research*, **2**(3); 79–85
- Xu, T., C. Fei, H. Zhou, Y. Fan, K. Tang, C. Chen, D. Ju, Z. Zhu, and R. Mao (2025). The Formation and Adsorption Mechanism Studies of Three-Dimensional (3D Flower-Like) Layered Double Hydroxide. *Applied Surface Science*, **691**; 162671
- Yilmaz, P., D. Gunduzm, and B. Ozbek (2021). Utilization of Low-Cost Bio-Waste Adsorbent for Methylene Blue Dye Removal from Aqueous Solutions and Optimization of Process Variables by Response Surface Methodology Approach. *Desalination and Water Treatment*, **224**; 367–388