Ternary Layered Double Hydroxide for Remediation of Methyl Orange and Congo Red

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By

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Chapter 7: Conclusions and Future Work

7.1. Conclusion of the present investigation

In **Chapter 1,** we have discussed the current situation of wastewater produced by the textile industries and all the techniques used for wastewater treatment. This chapter shows how the textile industry's usable dye continuously affects people and the environment. In this decade, many researchers have been working on this topic. They are successfully removing the dye from the wastewater, but the problem arises with the long-term recyclable and highly efficient material preparation. I have listed all the materials that have already worked to remove the dye and found the research gap. The main goal of this thesis is to develop an efficient and highly recyclable material.

In **Chapter 2**, we have discussed all available processes used to synthesize the layered double hydroxide (LDH). All characterization techniques are mentioned to measure the chemical and physical properties of the LDH. All chemicals and instruments mentioned, are used to synthesize the ternary LDH.

The main objective of this thesis was to develop an efficient and highly recyclable Layered Double Hydroxide (LDH). To achieve this, ternary LDH was synthesised using the coprecipitation method. The synthesised materials were then analysed using various characterisation techniques. XRD was employed to confirm the material's structural properties, while FTIR and BET analyses were conducted to identify the functional groups and porosity. The surface morphology was examined using SEM and TEM. After verifying these characteristics, the synthesised materials were tested for their ability to remove pollutants from textile industry wastewater. Methyl Orange (MO) and Congo Red (CR), two commonly used dyes, were selected for these experiments. Based on the findings, the overall conclusions of the thesis were drawn.

Chapter 3 discusses how the synthesis time was adjusted to determine the optimal parameters for the ternary LDH while keeping the temperature constant for CoAlFe LDH. The ideal conditions for achieving highly crystalline LDH were identified as 60 °C for 24 hours, with a molar ration $\frac{Co^{2+}}{Al^{3+}} = 2, \frac{Co^{2+}}{Fe^{3+}} = 2$. The XRD pattern of the sample synthesised at 24 hours showed maximum crystallinity, and the absence of impurity peaks confirmed the selection of these parameters. This LDH is applied to remove CR from the aqueous solutions. The adsorption capacity of CoAlFe LDH is 82.92 mg/g.

A systematic study of the synthesis of MgZnFe LDH applied for Congo Red removal from aqueous solution is discussed in **Chapter 4**. The material was calcined by heat treatment at 500 °C for 4 h for surface property modification. Due to heat treatment surface area and the increased pore volume of calcined LDH. The calcined and uncalcined LDH are characterised by the XRD, FTIR, BET, SEM and TEM analysis. SEM images demonstrated the layered structure of synthesised material, and TEM images indicate that surface morphology was hexagonal. Adsorption isotherm data is analysed using the Langmuir and Freundlich isotherm models, while the pseudo-first-order, pseudo-second-order, and intraparticle diffusion models fit the kinetic data. The maximum adsorption capacity of calcined and uncalcined LDH were 89.76 and 205.76 mg/g.

Chapter 5 discussed the synthesis of MgZnAl LDH and its use in removing MO from aqueous solutions. The material was heat-treated at 500°C for 4 hours to enhance its surface properties. This calcination process increased the surface area and pore volume of the LDH. Both calcined and uncalcined LDH were characterised using XRD, FTIR, BET, SEM, and TEM techniques. TEM images showed a hexagonal surface structure. The adsorption isotherm data were analysed with the Langmuir and Freundlich models, while the kinetic data were applied to the pseudo-first-order, pseudo-second-order, and intraparticle diffusion models. The maximum adsorption capacities for the calcined and uncalcined LDH were 220.26 mg/g and 869.56 mg/g, respectively. The calcined LDH showed recyclability up to the 6^{th} cycle, maintaining a removal efficiency of 55% The operating cost of MO removal by the adsorption process was 0.001 US$/L, which is much less compared to other methods, such as Electro Coagulation (0.47 US$/L), Chemical Coagulation (0.2 US$/L), Fenton Oxidation (0.026-0.046 US$/L) and Photo-Fenton Oxidation (0.027-0.036 US$/L).

Chapter 6 focused on synthesising MgNiAl LDH and its application for removing CR from aqueous solutions. The material underwent calcination at 500°C for 4 hours to modify its surface properties. This heat treatment increased the surface area and pore volume of the calcined LDH. Both calcined and uncalcined LDH were characterised using XRD, FTIR, BET, SEM, and TEM analyses. TEM images revealed a hexagonal surface morphology. The adsorption isotherm data were analysed with the Langmuir and Freundlich models, while the kinetic data were fitted to the pseudo-first-order, pseudo-second-order, and intraparticle diffusion models. The maximum adsorption

capacities for the calcined and uncalcined LDH were 181.81 mg/g and 480.81 mg/g, respectively. The calcined LDH demonstrated excellent recyclability up to the 14th cycle, with a removal efficiency of 99% through the 9th cycle. After the 9th cycle, the efficiency gradually decreased to 85% by the 14th cycle. To assess the calcined LDH after CR adsorption, XRD and FTIR analyses were performed on the recycled material. XRD spectra showed no additional peaks in the recycled material, while the FTIR spectrum at 1105 cm^{-1} indicated a peak associated with the sulphonyl group, which likely contributed to the reduced degradation efficiency of the recycled material. The operating cost for CR removal through adsorption was 0.00062 US$/L, which is significantly lower compared to other methods such as Electro Coagulation (0.47 US$/L), Chemical Coagulation (0.2 US$/L), Fenton Oxidation (0.026-0.046 US$/L), and Photo-Fenton Oxidation (0.027-0.036 US$/L).

7.2. Future work

Future research can take various directions, with the following points presenting promising opportunities for further progress in this field:

- The adsorption capacity of CoAlFe-CO_3 LDH can be improved through modification, and its recyclability can be examined.
- The ternary layered double hydroxide can remove heavy metals, antibiotics, and microplastics from wastewater.
- The photocatalytic properties of the ternary LDH can be investigated.
- The adsorption capacity of ternary LDH can be evaluated in the acid medium.
- The layered double hydroxide can be applied in different fields, such as energy harvesting and environmental pollution control (CO_2 adsorption).
- Developing composites of ternary LDHs with materials like graphene, biochar, or metal-organic frameworks (MOFs) involves integrating the unique properties of these materials with those of LDHs to enhance their performance
- Investigate the effects of controlling particle size, surface area, and porosity on performance in various applications.
- Enhance photocatalytic properties for breaking down organic pollutants under visible light.
- Explore ternary LDHs in applications like batteries, supercapacitors, and electrocatalysis (e.g., hydrogen evolution reaction).

- The ternary LDH can be used as a smart senso for detecting pollutants in different pH and temperature.
- Investigate their potential in drug delivery, antibacterial coatings, and bio-imaging, leveraging their biocompatibility and ion-exchange properties.