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Comparative study on removal of Monodyes by using Ni-Al layered double hydroxides

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Abstract. The toxic and carcinogenic effluents discharged from the textile and garment industry represent a major concern to the environment and public health. Treatment technologies for these effluents to reduce toxicity and dye concentration are therefore crucial. The objective of this study was to focus on eliminating four monoazo dyes including Congo (CR), Methyl orange (MO), Methylene blue (MB) and Crystal violet (CV) using adsorbents, Ni /Al LDHs. The results proved that Ni/Al LDHs exhibit excellent adsorption capacity on anionic dyes (CR, MO) instead of cationic dyes (CV, MB). The maximum adsorption capacity of Methyl Orange on Ni/Al LDHs was found to be 93.517 mg/g and Congo red at 87.975 mg/g. In addition, the study of leaching test of adsorbents show stable synthetic materials in aqueous media. The analyses mentioned above indicates that Ni/Al LDHs may be a promising adsorbent to remove anion dyes from aqueous media.

1. Introduction

The development of organic pollutants stems from the growth of the textile and apparel industry, posing a threat to the ecosystem and human life. Most of the dyes used are azo reactive dyes that are light in color due to the presence of one or several azo groups ($-N=N-$) related to the replacement aromatic structure. Methyl orange (MO), Crystal violet (CV), Congo red (CR) and Metyl blue (MB) are monoazo dyes commonly used in textiles and other commercial products [1]. They are present in colored industrial wastewater which can cause health problems and environmental pollution. Health problems stemming from dyes can be naturally toxic, causing mutations if inhaled or swallowed or wearing dyes for a long time can lead to skin cancer. Meanwhile, environmental problems stem from the stability of the aromatic ring structure, which characterizes the persistence properties of monoazo dyes. Stemming from the toxic nature and the serious impact on ecosystems and humans, it has led to the development of engineering technologies to eliminate water-soluble organic matter [2–7]. Although existing technologies including



UV/ozone treatment, photocatalytic attenuation and electrochemical reduction have been shown to be effective in dye removal, their high cost can create inconsistencies. Satisfied for the dyeing industry to develop, promote further development in efficient processing techniques. Adsorption is a separation technique that can meet the stringent requirements for water treatment processes applied to dyeing plants including low cost, insensitivity and simplicity in implementation and operation [8–13].

Layer double hydroxides (LDHs), commonly referred to as Hydrotalcite or anions, are recognized as effective and potential adsorbents for removing organic pollutants in wastewater due to their outstanding properties such as porosity and surface area, relatively large surface, thermal stability, ion exchange capacity in alternating layers [14–19]. In this study, Ni/Al LDHs materials were synthesized by hydrothermal method. The main objective of the study was to identify and compare the adsorption capacity of Ni/Al LDHs with various organic dyes including Congo red (CR), Methyl orange (MO), Methylene blue (MB) and Crystal violet (CV). The results will serve as a precursor to further research into the practical application of LDHs in dyeing wastewater treatment.

2. Materials and Methods

2.1. Experimental

The chemicals used in this study include: Nickel sulfate ($\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$), Ammonium sulfate ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) purchased from Sigma-Aldrich. Ammonium hydroxide (NH_4OH), Sodium hydroxide ($\text{NaOH} \geq 96\%$), hydrochloric acid ($\text{HCl} \geq 98\%$) were purchased from Xilong, China. The hydrothermal method was adopted to prepare Ni-Al-LDHs. To be specific, $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ and $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ at $\text{Ni}^{\text{II}}/\text{Al}^{\text{III}}$ molar ratio of 3/1 were dissolved in distilled water. The solution then had its pH 10 value adjusted using NH_4OH solution. The suspension was then introduced into a 50 mL stainless Teflon-lined autoclave, which was subjected to heating at 180 °C for a predetermined duration and allowed to cool to ambient temperature naturally. Following that, distilled water and absolute ethanol were used to wash the filtered products. Final apple-green solids was dried at room temperature for 12 hours [20].

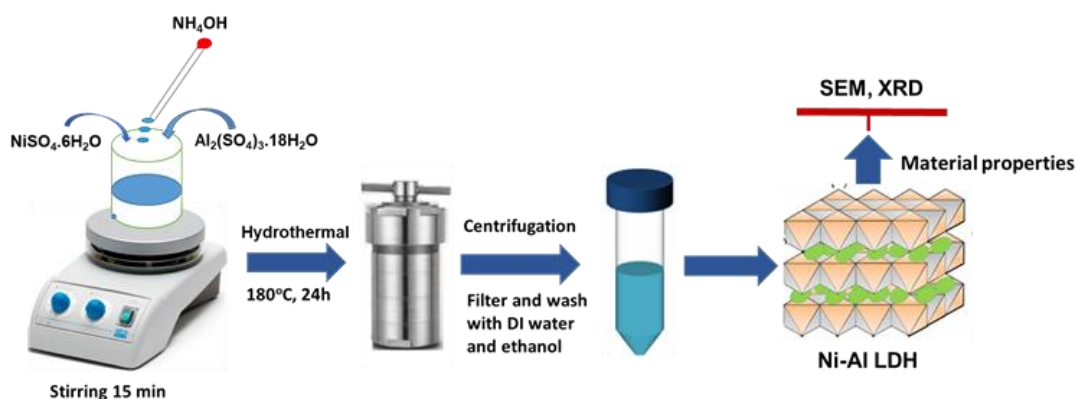


Figure 1. Synthesis process of Ni /Al LDHs

2.2. Experimental Procedure

The Synthesised Ni/Al LDHs was added to 100 mL solution of known dye concentration at neutral pH. The adsorbent concentration was changed within the range of 10-40 mg at constant CR concentration of 20 mg/L to investigate the effect of adsorbent dosage and immobilized at 40 mg for testing to evaluate adsorption behavior of monoazodye group (CR, MO, MB, CV). All experiments were performed at ambient temperature (30°C) with an orbital shaker operating at 200 rpm. At a time interval, about 4 mL of solution was withdrawn and centrifuged for 10 minutes at 7000 rpm. UV Vis Spectroscopy (EVOLNING 60S) was used to determine dye concentrations in residues. The percentage removal of pollutant was calculated as follows:

$$\text{Removal efficiency (\%)} = \left(1 - \frac{C}{C_0}\right) \times 100 \quad (1)$$

Where C_0 and C are the initial and residual dye concentration (mg/L), respectively.

The dye uptake at equilibrium q_e (mg dye/g sorbent) was calculated as follow:

$$Q_t = \left(\frac{C_0 - C_t}{m}\right) \times V \quad (2)$$

Where, C_0 and C_e are the initial and equilibrium dye concentrations (mg/L), respectively, V is the volume of solution (L), and m is the adsorbent dosage (g).

3. Results and discussions

3.1. Removal of CR from aqueous media by adsorption onto the efficient Ni-Al-LDHs

Optimizing the dosage of materials is of significance to boost the cost-effectiveness in any treatment process. Herein, the Ni/Al LDHs are used as representative adsorbents to demonstrate the CR removal efficiency of Hydrotalcite. Accordingly, the effect of adsorption dose (10-40 mg) was studied at constant concentration of 20 mg/L, shaking time in 210 min. The results in Figure 2 show that the increased adsorption dose (10-40 mg) leads to enhance the CR removal efficiency, reaching the peak of capacity (98.762%) at 40 mg of adsorbent dosage. It is conceivable that the improvement of the CR removal efficiency when adding adsorbents may be attributable to the availability of active “adsorption sites” enhanced [21]. Therefore, the optimal dose that affects the effect of CR adsorption on Ni/Al LDHs were fixed at 40 mg.

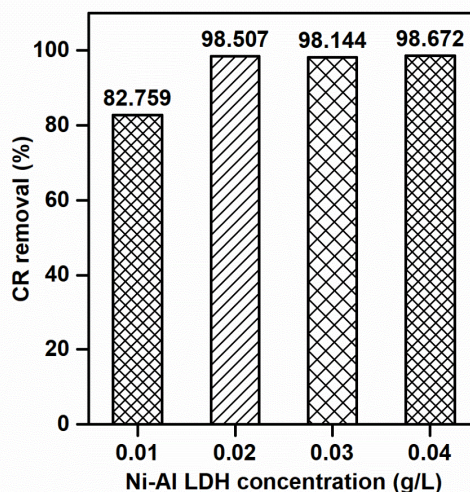


Figure 2. Effect of adsorbent dosage on the removal efficiency of CR

3.2. Leaching test study

The heterogeneity and stability of the adsorbent in the reaction medium is assessed by the leaching test [8]. The purpose of this experiment is to evaluate the adsorption sites on the surface of materials that contribute to CR adsorption after being separated from the mixture. 40 mg of adsorbent was added to 100 mL of CR solution 40 mg/L at neutral pH. After 10 minutes, the solids were completely separated by centrifugation. The residual solution was stirred for 210 min and the dye concentration was determined by UV-Vis spectroscopy at different intervals. The C_t/C_0 curve over time (t) was shown in Figure 3. The results showed that the C_t/C_0 ratio decreased to about 0.38 after the first 10 minutes, however no changes were observed after separating the adsorbent from the solution. This result shows that Ni/Al LDHs was chemically stable and the presence of Ni/Al LDHs initiated the adsorption process and that the process is irrelevant to the leaching of adsorbent into the aqueous phase. This result is consistent with previous research results [22].

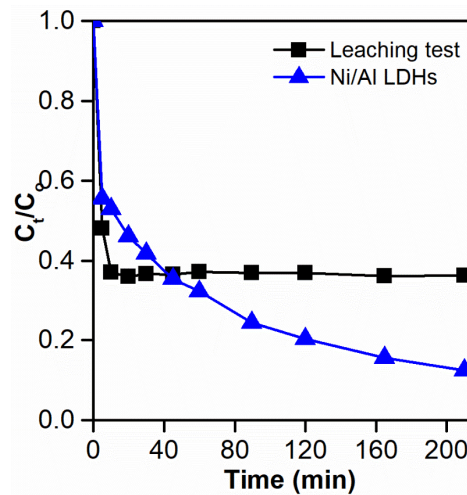


Figure 3. Leaching test study for Ni/Al LDHs

3.3. Congo Red adsorption efficiency of Ni/Al LDHs adsorbents

The adsorption capacity on different pigments of Ni/Al LDHs is shown in Figure 4. The results show that this material shows superior adsorption capacity for anionic dyes instead of cationic dyes. For methyl orange, the adsorbed CR was 93.573 mg/g and 87.975 mg/g for Congo red. Meanwhile, Ni/Al LDHs exhibited poor adsorption capacity for Crystal violet and Methylene blue. These results are explained based on the charge on the adsorbent surface and the nature of the dye. Hydrotalcite material is a positively charged material, so through electrostatic interaction they exhibit superior adsorption capacity for anionic dyes.

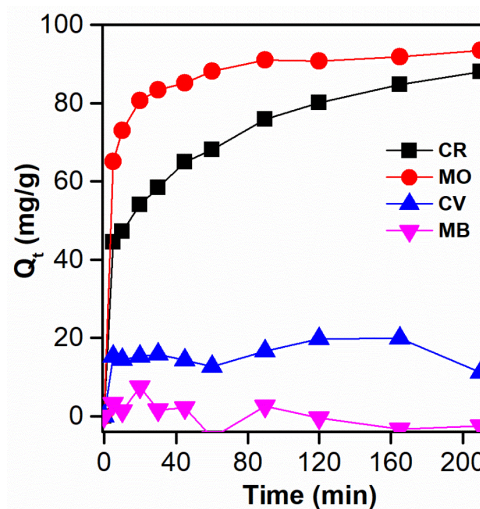


Figure 4. Adsorption capacity of Ni-Al-LDH onto different dyes

Table 1 displayed some values related to the maximum capacities (Q_m) of various adsorbents used to remove dyes. The comparison results show that the adsorption capacity of Ni/Al LDHs in this study is outperforms to other studies. This suggests the application potential for Ni/Al LDHs in treatment of other dyes from wastewater.

Table 1. The parameters of different dyes effect the adsorption capacity.

No.	Adsorbents	Maximum adsorption capacity (mg/g)	Ref.
1	Ni/Al LDHs	93.57	This work
2	NaBentonite	35.84	[23]
3	Neem leaf powder	41.20	[24]
4	EG@MnFe ₂ O ₄	71.79	[25]
5	Bentonite	40.40	[26]
6	Gold-magnetic nanocomposite	43.88	[27]
7	NiAl-LDHs	77.70	[28]

4. Conclusion

The adsorption capacity of Ni/Al LDHs compared to four monoazo dyes, methyl orange (MO), crystal violet (CV), red congo (CR), and methyl blue (MB), has been studied. The results obtained were shown good adsorption capacity for anions (CR, MO) instead of cations (CV, MB). The highest adsorption efficiency for CR and MO corresponds to greater than 87 mg/g. Specifically, 87.975 mg/g for CR and 93.517 mg/g for MO. The adsorption test results show that the optimal adsorption dose at 40 mg and the study of leaching test of adsorbents show stable synthetic materials in aqueous media. Therefore, these findings suggest that the biological adsorbent Ni/Al LDHs has potential applications for removing anion dyes from industrial wastewater.

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