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Research Paper

Malachite Green Dye Adsorption from Aqueous Solution using a Ni/Al Layered Double Hydroxide-Graphene Oxide Composite Material

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Abstract

Ni/Al layered double hydroxide and Ni/Al-graphene oxide composite materials were created using the coprecipitation method. The materials were successfully synthesized and prepared using XRD, FT-IR, and BET studies. The optimal pH as a result of malachite green dye adsorption is pH 4. The kinetics models of all materials follow the pseudo second order model. After being composited with graphene oxide, the maximum adsorption capacity of Ni/Al layered double hydroxide increased from 99.010 to 111.111 mg/g. All materials' isotherm models adhere to the Langmuir isotherm model. The adsorption process was endothermic and spontaneous. The Ni/Al-graphene oxide composite material has a more stable structure than the Ni/Al layered double hydroxide. The regeneration procedure was repeated five times, and the Ni/Al-graphene oxide composite material did not show a significant decrease until the fifth cycle, when it dropped from 97.561 to 77.046%, however the Ni/Al layered double hydroxide material dropped rapidly from 85.00 to 5.667%.

Keywords

Malachite Green, Graphene Oxide, Adsorption, Layered Double Hydroxide

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1. INTRODUCTION

Similar to brucite ($Mg(OH)_2$), layered double hydroxide (LDH) is a two-dimensional layered structure composed of two or more cations. LDH has the usual chemical formula $[M(II)_{1-x}M(III)_x(OH)_2]^{x+}[A_{x/m}]^m \cdot_n H_2O$, where M(II) and M(III) are metal cations and A is an anion in the interlayer (Zhang et al., 2023). LDH can be synthesized by various methods including sol-gel, hydrothermal, and coprecipitation methods (Pavlovic et al., 2022; Valeikiene et al., 2019). However, among these methods, the coprecipitation method is the most commonly used method because of the ease of the process and the ease of control such as pH parameters during the synthesis process (Wang et al., 2022).

LDH with excellent ion exchange ability, large surface area, good thermal stability, and easy to synthesize makes this material widely used in various applications such as catalysts, electrochemistry, and removal of contaminants in water or adsorption (Zhang et al., 2023; Zubair et al., 2022). When the adsorption application takes place, LDH material has the disadvantage that it cannot be used repeatedly as an adsorbent because the structure of LDH is easy to peel off (Normah et al., 2021) and agglomeration is formed during application (Zubair et al., 2022). As a result, the structure must be modified by compositing carbon-based elements in order to improve the layer structure in LDH so that it may be utilized frequently. Various carbon-based materials that are known to be used as supporting materials in improving the structure of layered double hydroxide include using biochar (Wijaya et al., 2021), humic acid (Shi et al., 2020), graphite (Hu et al., 2019), and charcoal (Ahmad et al., 2023a).

Research conducted by Wijaya et al. (2021) conducted a composite of LDH with biochar, where after compositing with biochar, LDH material can be used repeatedly for up to five cycles and an increase in surface area from 46.279 m^2/g in layered double hydroxide to 200.90 m^2/g in layered double hydroxide/biochar composite. Research conducted by Hu et al. (2019) improved the structure of LDH with graphite can increase the adsorption capacity from 5,656 mg/g to 13,441 mg/g on Cr(VI) adsorption. In addition to using the above materials, many materials can be used to improve the structure of layered double hydroxide including using graphene oxide (Rashed et al., 2022).

Graphene oxide is a single layer of graphite with sp^2 and sp^3 hybridized carbon atoms and numerous functional groups on the surface such as epoxy (-O-) and hydroxyl (-OH) and carboxyl (-COOH) and carbonyl (-C=O) towards the margins. Graphene oxide has good physical and chemical properties where it has a large surface area, outstanding conductivity, and also has good catalytic activity (Heshami et al., 2023). The characteristics of many functional groups, especially oxygen in the graphene oxide structure, which will bind to cations in the LDH surface structure are expected to improve the structure of LDH and increase its adsorption capacity (Rashed et al., 2022).

Because of their carcogenicity, difficulties in natural degradation, and high toxicity, color pollutants such as cationic and anionic dyes that are discharged have a substantial influence on human health, living organisms, and the ecological environment. Together with the rapid development of contemporary industry, particularly in the textile, printing, and dyeing industries, it creates pollution that requires regular care. Every year, more than 1.6 million tons of dyes are produced globally, with 10-15% of them being wasted and entering the environment (Lv et al., 2022; Yan et al., 2022).

In this study, Ni/Al LDH modification was carried out by compositing graphene oxide which was applied as an adsorbent in the selective process of malachite green dye. X-Ray Diffraction (XRD), Fourier Transform Infrared (FT-IR), and Brunauer Emmet Teller (BET) were used to investigate the materials. It was also utilized to look at aspects including pH change, contact time, temperature and concentration, and regeneration.

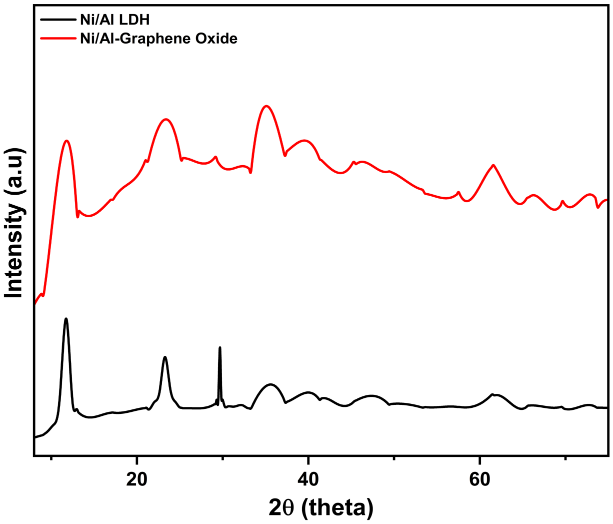

Figure 1. XRD Diffractogram of Ni/Al LDH and Ni/Al-Graphene Oxide Materials

2. EXPERIMENTAL SECTION

2.1 Chemicals and Instrumentation

The chemicals used in this study include sodium carbonate (Na_2CO_3), sulfuric acid (H_2SO_4), graphite, sodium nitrate ($NaNO_3$), sodium hydroxide (NaOH), aluminum nitrate nonahydrate ($Al(NO_3)_3.9H_2O$), distilled water (H_2O), hydrogen peroxide (H_2O_2), hydrochloric acid (HCl), nickel nitrate hexahydrate ($Ni(NO_3)_2.6H_2O$), rhodamine B ($C_{28}H_{31}N_2O_3Cl$), methylene blue ($C_{16}H_{18}ClN_3S$), and malachite green ($C_{23}H_{25}ClN_2$). Supporting instruments used were XRD (Rigaku MiniFlex-6000), FT-IR spectrophotometer (Shimadzu Prestige-21), UV-Vis spectrophotometer (Biobase BK-UV 1800 PC), and BET equipment (Quantachrome Instruments).

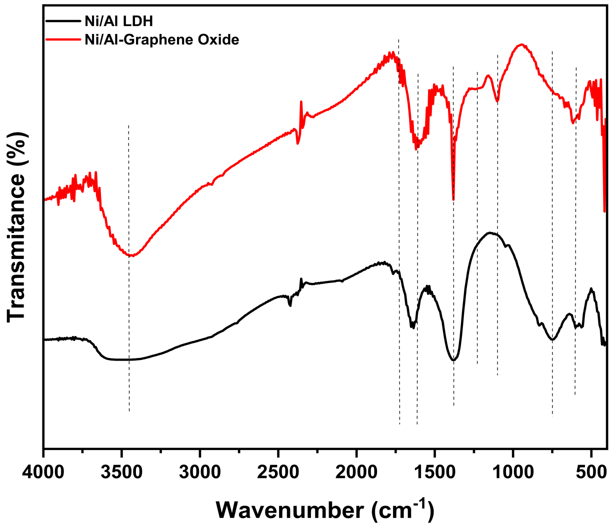

Figure 2. IR Spectra of Ni/Al LDH and Ni/Al-Graphene Oxide Materials

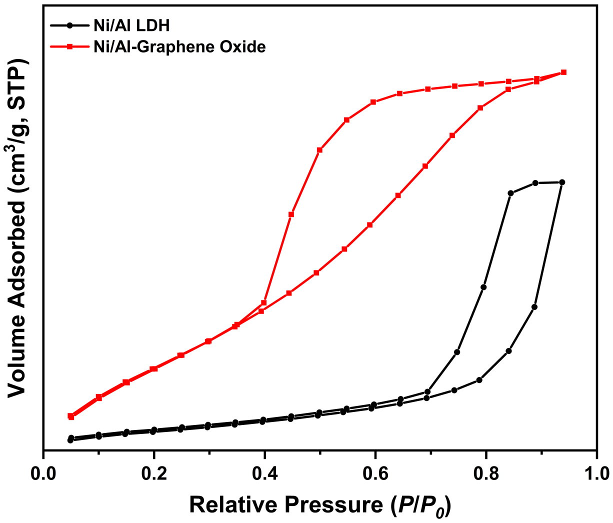

Figure 3. N_2 Adsorption–Desorption Isotherms of Ni/Al LDH and Ni/Al-Graphene Oxide Materials

2.2 Synthesis of Ni/Al LDH

The coprecipitation approach was used to synthesize Ni/Al LDH. In a glass becker, 100 mL of 0.75 M Ni^{2+} and 0.25 M Al^{3+} solution was first combined. The solution was then carefully dripped with 50 mL of 2 M NaOH and 100 mL of

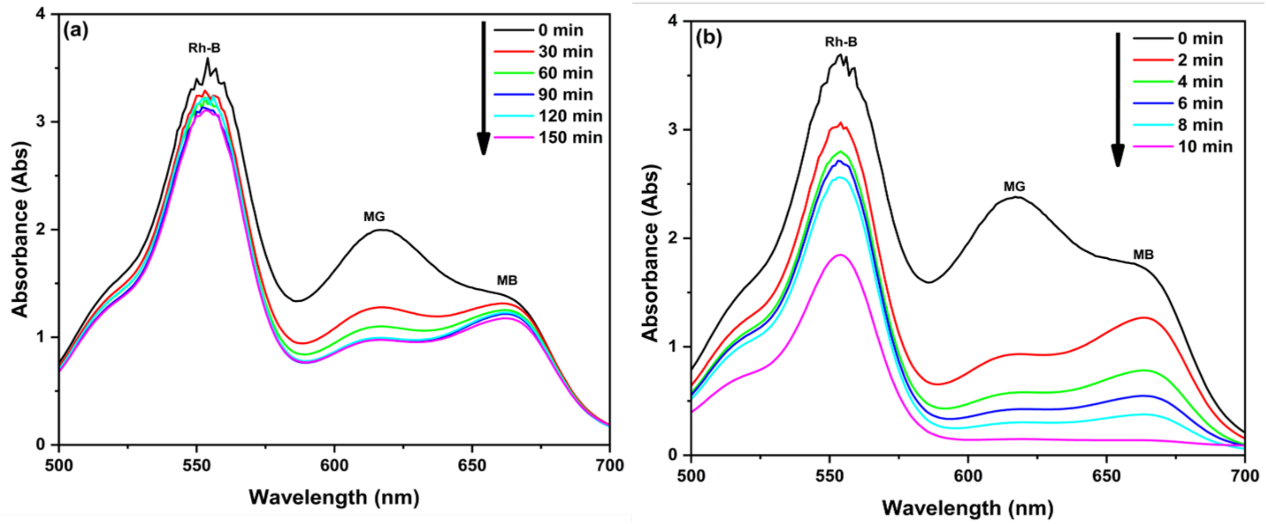

Figure 4. Selectivity of Dye Mixture on Ni/Al LDH (a) and Ni/Al-Graphene Oxide (b)

Table 1. Brunauer Emmet Teller of Ni/Al LDH and Ni/Al-Graphene Oxide Materials

Material	Surface Area (m^2/g)	Pore Size (nm), BJH	Pore Volume (cm^3/g), BJH
Ni/Al LDH	40.912	7.460	0.153
Ni/Al-Graphene Oxide	78.348	2.414	0.095

0.3 M Na_2CO_3. By adding 2 M NaOH, the pH of the metal combination was adjusted to pH 10. Additionally, stirring was performed for 17 hours at an 80°C temperature. After getting the precipitate, it was filtered and washed with distilled water. The precipitate that formed was then dried and analyzed using XRD, FT-IR, and BET.

2.3 Preparation of Ni/Al-Graphene Oxide

Composite materials were prepared by preparing a solution of 30 mL each of a mixture of Ni^{2+} and Al^{3+} as treated in procedure 3.1 to be carried out with the addition of NaOH until pH 10. The mixture was then stirred for 1 hour and then 3 grams of graphene oxide synthesized by the Hummers method was added. After mixing, the solution was stirred for up to 3 days at 80°C until a precipitate formed. The resulting precipitate was then filtered, washed using distilled water and dried. The material was then characterized using XRD, FT-IR, and BET.

2.4 Selectivity of Cationic Dyes

The same concentrations of methylene blue (MB), rhodamine B (RhB), and malachite green (MG) were used to conduct selectivity studies. 20 mL of the combined dye solution was mixed with 0.02 gram of Ni/Al LDH material and 0.02 gram of Ni/Al-graphene oxide composite. Wavelength measurements were then made in the 500-700 nm range on the Ni/Al-graphene oxide composite material at time variations of 0, 2, 4, 6, 8, and 10 minutes and on the Ni/Al LDH material at time variations of 0, 30, 60, 90, 120, and 150 minutes. The dye that had been most absorbed was then used in the adsorption procedure.

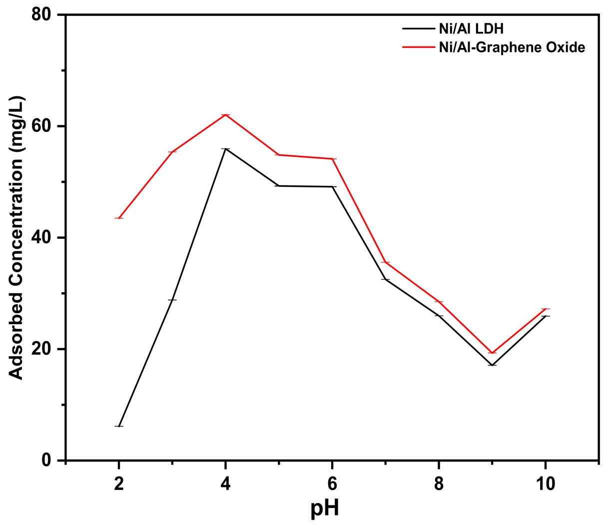

Figure 5. pH Optimum of Ni/Al LDH and Ni/Al-Graphene Oxide

2.5 Adsorption of Phenol

The adsorption process was carried out by studying the effect of pH; time; temperature and concentration. The effect of pH was carried out to determine the pH conditions that absorb the most dyes by varying pH 2-10. The effect of time was carried

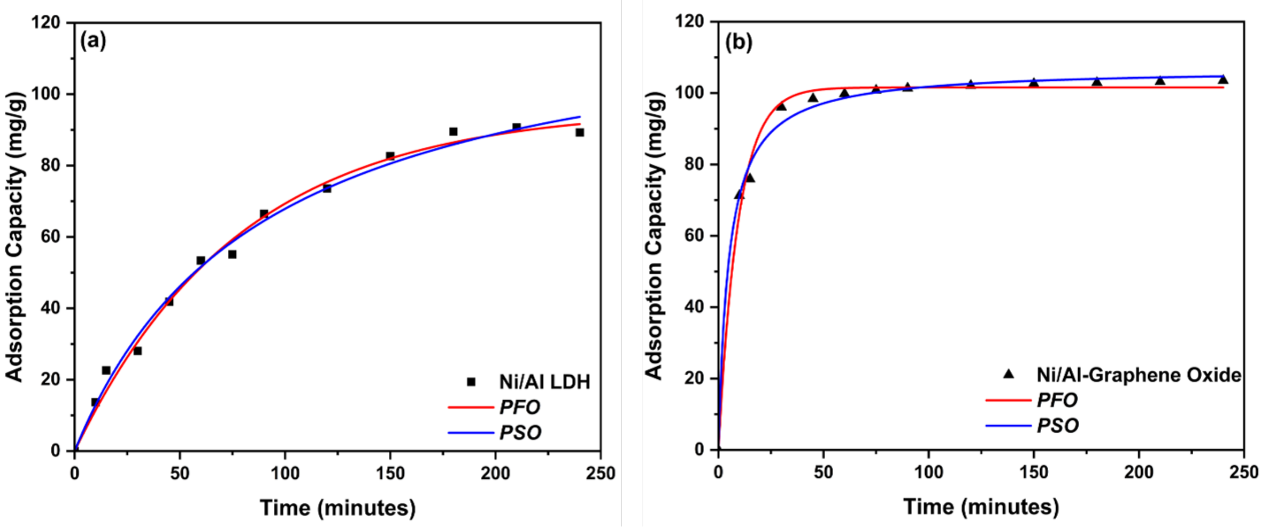

Figure 6. Effect of Time Variation on Adsorption of Malachite Green on Ni/Al LDH (a) and Ni/Al-Graphene Oxide (b) Materials

Table 2. Kinetics Parameters of Malachite Green Dye Adsorption Process

Kinetic Model	Parameter	Materials	
		Ni/Al LDH	Ni/Al-Graphene Oxide
pseudo first order	Qe_{exp} (mg/g)	89.276	103.498
	Qe_{calc} (mg/g)	94.363	27.676
	k_1 (min^{-1})	0.016	0.024
	R^2	0.977	0.883
pseudo second order	Qe_{exp} (mg/g)	89.276	103.498
	Qe_{calc} (mg/g)	125.000	105.263
	k_2 (min^{-1})	0.0001	0.002
	R^2	0.982	0.999

out to determine the parameters of adsorption kinetics with time variations of 0, 10, 15, 30, 45, 60, 75, 90, 120, 150, 180, 210, and 240 minutes. The effect of temperature and concentration was used to determine the adsorption isotherm and thermodynamic parameters using temperature variations of 30-70°C at various concentrations of 70, 80, 90, 100, and 110 mg/L. Observations were made with a UV-VIS spectrophotometer at a wavelength of 617 nm by mixing 20 mL of dye with 0.02 grams of material.

The regeneration process is carried out by performing the adsorption-desorption process repeatedly. The desorption process aims to release the dye bound to the adsorbent material by ultrasonic washing of distilled water. The regeneration process was carried out with 5 repetition cycles.

3. RESULTS AND DISCUSSION

3.1 XRD Analysis of Ni/Al LDH and Ni/Al-GO Materials

In Figure 1(a), the diffraction peaks in the Ni/Al LDH material are obtained at angles 2θ= 11.80° (003); 23.16° (006); 35.32° (012); 39° (015); and 62.48° (113). According to Li et al. (2017), the resulting diffraction peaks are in accordance with JCPDS No. 15-0087 data which is characteristic of diffraction peaks in Ni/Al LDH materials. In the Ni/Al-graphene oxide composite material, peaks were obtained at angles of 11.58° (003); 23.26° (006); 35.12° (012); 39.5° (015); and 66.28° (113) as shown in Figure 1(b). At a diffraction angle of 11.58° (003), a less sharp peak is obtained and there is a shift in the diffraction angle towards a lower angle from 11.80° to 11.58°. According to Rashed et al. (2022) the occurrence of a shift in the diffraction angle towards the lower Ni/Al-graphene oxide composite material is due to the addition of graphene oxide material to the Ni/Al LDH material. Based on the above information, it can be concluded that the synthesis of Ni/Al LDH material and the preparation of Ni/Al-graphene oxide composite material were successfully carried out.

3.2 FT-IR Analysis of Ni/Al LDH and Ni/Al-GO Materials

Figures 2(a) and 2(b) show the results of FT-IR analysis on Ni/Al LDH material and Ni/Al-graphene oxide. The spectrum peak at wave number 3448 cm^{-1} is obtained in the Ni/Al LDH material, indicating the presence of O-H stretching vibrations. The presence of NO_3^- functional groups in the intelayer is shown by the spectrum peak at wave number 1381 cm^{-1} (Santosa et al., 2021). The FT-IR spectrum peak at wave number 1620 cm^{-1} indicates the bending vibrations in water molecules. In addition, there is a peak in the area around 400-800 cm^{-1} indicating the presence of metal groups that bind to oxygen (M-O) (Mallakpour et al., 2023).

The peaks of the base material, namely the peak in the Ni/Al layered double hydroxide material and the peak in the

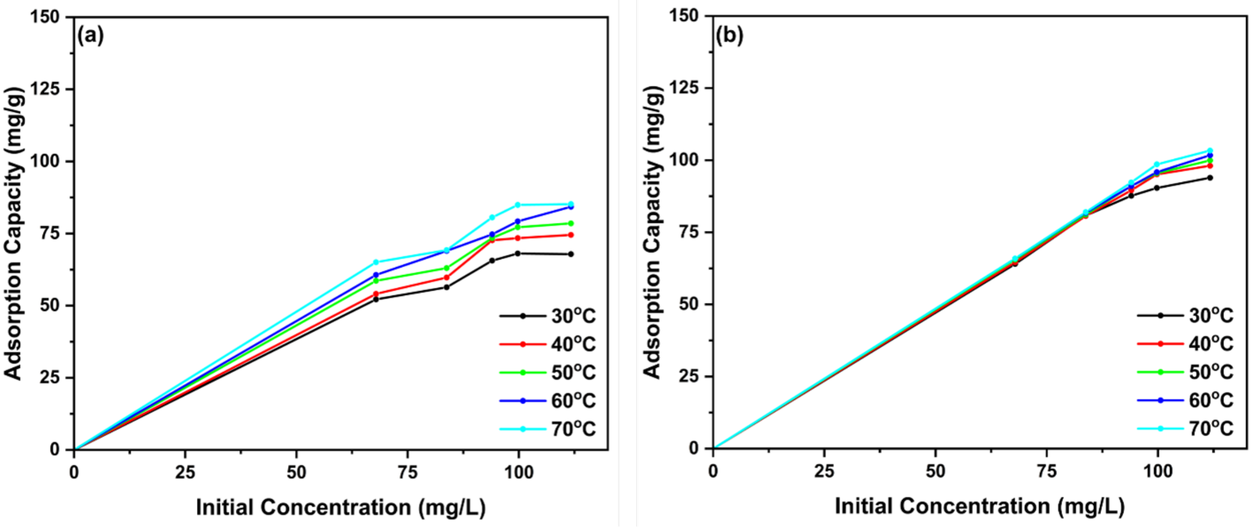

Figure 7. Effect of Concentration and Temperature Variations on the Adsorption of Malachite Green on Ni/Al LDH (a) and Ni/Al-Graphene Oxide (b) Materials

Table 3. Isoterm Parameters the Adsorption Process

Materials	Temperature (°C)	Model Isoterm Adsorpsi					
		Langmuir			Freundlich		
		Q_m	KL	R^2	n	KF	R^2
Ni/Al LDH	30	84.034	0.104	0.948	3.573	24.300	0.737
	40	94.340	0.105	0.896	3.180	24.643	0.604
	50	91.743	0.171	0.946	4.239	34.506	0.710
	60	99.010	0.184	0.987	4.016	36.475	0.963
	70	88.496	0.624	0.974	8.591	57.531	0.628
Ni/Al-Graphene Oxide	30	101.010	0.786	0.991	6.649	62.345	0.501
	40	107.527	0.838	0.982	5.348	62.936	0.445
	50	108.696	1.122	0.991	5.408	66.773	0.528
	60	111.111	1.169	0.992	4.924	67.313	0.627
	70	109.890	1.750	0.974	12.853	82.035	0.109

graphene oxide material, must be visible during the successful fabrication of the Ni/Al-graphene oxide composite. The FT-IR spectrum of the Ni/Al-graphene oxide composite material peaks at wave number 3410 cm^{-1}, indicating the existence of O-H groups, according to the results of the FT-IR spectra. The presence of nitrate groups in the Ni/Al LDH interlayer and O-H bending vibrations in graphene oxide is shown by the FT-IR spectra peak at wave number 1381 cm^{-1}. The presence of the C=C group is indicated by the peak of the FT-IR spectra at wave number 1620 cm^{-1}. The presence of C=O and C-O-C groups in graphene oxide is shown by the FT-IR spectra peaks around the wave numbers of 1720 and 1080 cm^{-1} (Zhang et al., 2020). Metal oxide (M-O) groups in the Ni/Al LDH material were also discovered in the FT-IR spectra of the Ni/Al-graphene oxide composite material at 400-800 cm^{-1}.

3.3 BET Analysis of Ni/Al LDH and Ni/Al-GO Materials

The N_2 adsorption-desorption of Ni/Al LDH and Ni/Al-graphene oxide is depicted in Figure 3. Based on the N_2 adsorption-desorption, all materials exhibit a type IV isotherm, indicating that the materials have a mesoporous size (2-50 nm) (Ahmad et al., 2023a). Table 1 shows the surface area, pore size, and pore volume of each material. The surface areas of the Ni/Al LDH material and the Ni/Al-graphene oxide composite are 40.912 and 78.348 m^2/g, respectively. The surface area of the Ni/Al-graphene oxide material is twice that of the Ni/Al LDH material affected by graphene oxide addition. Ni/Al LDH and Ni/Al-graphene oxide composite have pore volumes of 0.153 and 0.095 cm^3/g, respectively. Ni/Al LDH material and Ni/Al-graphene oxide composite have pore diameters of 7.460 and 2.414 nm, respectively. Mesoporosity describes all materials.

Table 4. Maximum Adsorption Capacity of Phenol and Comparison with Other Researches

Adsorbent	Q_{max} (mg/g)	Reference
Saal (*Shorea robusta*) flower	14.45	(Dey et al., 2018)
$ZnCl_2$ activated *Ricinus communis* stem powder	12.64	(NirmalaDevi et al., 2018)
S@TP biochar	29.277	(Vigneshwaran et al., 2021)
Potato peel	35.61	(Guechi and Hamdaoui, 2016)
Chinese fan palm fruit biochar	21.41	(Giri et al., 2022)
Ni/Al LDH	99.010	This work
Ni/Al-Graphene Oxide	111.111	This work

3.4 Adsorption Selectivity of Cationic Dyes

Figure 4 depicts a spectra graph of the selectivity measurement findings of the cationic dyes methylene blue (MB), rhodamin-B (RhB), and malachite green (MG). According to the graph, all materials are more selective towards MG dye. This is evidenced by the significant decrease in MG dye. The significant decrease in MG dye is due to the structure of MG dye which is less complex than MB and RhB so that it is more easily adsorbed. In addition, Ni/Al-graphene oxide composite material can easily absorb MG dye in a relatively short time compared to Ni/Al LDH material.

3.5 pH Optimum and Kinetic Studies in Adsorption Process

The effect of pH on the adsorption process of malachite green can be seen in Figure 5. Measurement of absorbance at each pH before adsorption of malachite green dye is very important to know the initial concentration after adjusting the pH. This is because malachite green dye is very unstable at pH<2 and in alkaline conditions (pH>7) which causes discoloration (Hong et al., 2023). Based on Figure 5, it can be seen that the adsorption capacity of all materials experiences optimum absorption at pH 4. At pH<4 there is a decrease in adsorption ability due to electrostatic repulsion due to the rich positive charge on the material and the positive charge on malachite green.

Kinetic studies were conducted at ideal pH levels for 10, 15, 30, 45, 60, 75, 90, 120, 150, 180, 210, and 240 minutes. As indicated in Table 2 and Figure 6, the data gathered to evaluate the adsorption kinetics model parameters are pseudo first order (PFO) and pseudo second order (PSO). The R^2 value in the pseudo second order (PSO) model is closer to 1 than in the pseudo first order (PFO) model, based on the data. In addition, the Qe_{calc} and Qe_{exp} values in PSO are closer to those in the pseudo first order model. As a result, we may conclude that the pseudo second order kinetics model is more appropriate for studying malachite green dye adsorption.

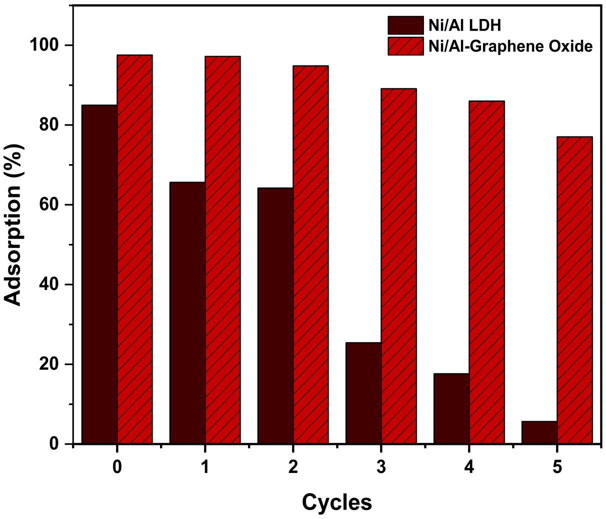

Figure 8. Adsorbent Regeneration of Ni/Al LDH and Ni/Al-Graphene Oxide Materials

3.6 Study of Adsorption Isotherms and Thermodynamics

The adsorption mechanism, interaction between the material and the adsorbate, and maximum adsorption capacity (Q_m) were all investigated using isotherms . To find the isotherm model that is compatible with the adsorption process, the Langmuir and Freundlich isotherm models are used. Figure 7 and Table 3 exhibit the study's findings regarding MG adsorption behavior. According to the data, all materials obey the Langmuir isotherm model. The R^2 value in the Langmuir isotherm is closer to one than the R^2 value in the Freundlich isotherm. According to Putranto et al. (2022), the Langmuir isotherm model assumes a monolayer adsorption process, limited capacity at each adsorption site, and a homogeneous surface. Ni/Al LDH and Ni/Al-GO materials have Q_m values of 99.010 and 111.111 mg/g, respectively. Table 4 compares the greatest adsorption capacity to that of other research.

Thermodynamic studies were conducted to review the spontaneity, the adsorption process taking place between endothermic and exothermic, and the resulting entropy values. Based on Table 5, it can be seen that all materials show positive ΔH values, indicating that the adsorption process takes place endothermically. The ΔG value is negative which indicates the adsorption process takes place spontaneously. The ΔS value with a small positive price indicates the degree of freedom in the adsorption process is not so significant.

3.7 Regeneration

Regeneration is the process of reutilizing adsorbent materials that have undergone adsorption and desorption processes (Ahmad et al., 2023b). In general, the adsorption process is very beneficial in terms of cost because the material can be used

Table 5. Adsorption Thermodynamic Parameter

Materials	Concentration (mg/L)	ΔH (kJ/mol)	ΔS (kJ/mol)	ΔG (kJ/mol)				
				303 K	313 K	323 K	333 K	343 K
Ni/Al LDH	99.737	19.693	0.071	-1.899	-2.611	-3.324	-4.037	-4.749
Ni/Al-Graphene Oxide	99.737	38.811	0.147	-5.752	-7.223	-8.694	-10.164	-11.635

several times in the adsorption process. Figure 8 shows the regeneration of Ni/Al LDH and Ni/Al-graphene oxide adsorbent materials under MG adsorption for up to five cycles. Based on the results obtained, the Ni/Al-graphene oxide material has a greater percent adsorption value and does not experience a significant decrease until the fifth cycle than the Ni/Al LDH material. This is evidenced in the Ni/Al-graphene oxide material which decreased from 97.561 to 77.046%, while the Ni/Al LDH material decreased from 85.00 to 5.667%. Thus it can be concluded that Ni/Al LDH material composited with graphene oxide can improve the material structure so that it has good stability.

4. CONCLUSION

Graphene oxide composite and Ni/Al LDH material were successfully manufactured. Using XRD, FT-IR, and BET, each material was characterized. The optimum pH of all materials was at pH 4. The kinetics and isotherm models of all materials followed PSO and Langmuir. The maximum adsorption capacity of LDH after composite with graphene oxide increased from 99.010 to 111.111 mg/g. Enthalpy and Giibs free energy values show that the adsorption takes place endothermically and spontaneously. The regeneration process was carried out up to five cycles where the composite material did not experience a significant decrease until the fifth cycle compared to Ni/Al LDH material.

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