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Research Paper

High Efficient of Ca/Al-Graphite for Removal of Direct Orange

Yusuf Mathiinul Hakim[1,3]*, Rananda Vinsiah[2], Shahibul Fajri[3]

[1] *Graduate School, Faculty of Mathematics and Natural Sciences, Sriwijaya University, Palembang, South Sumatera, 30139, Indonesia*
[2] *Department of Chemistry, Faculty of Mathematics and Natural Science, Sriwijaya University, Palembang, South Sumatera, 30139, Indonesia*
[3] *Research Center of Inorganic Materials and Coordination Complexes, Faculty of Mathematics and Natural Sciences, Sriwijaya University, Palembang, South Sumatera, 30139, Indonesia*
*__Corresponding author__: yusufmathiinul191@gmail.com

Abstract
The layered double hydroxide-based material features of Ca/Al have improved according to the adsorption capacity and structure stabilization by transforming into composite Ca/Al-graphite. The composite was synthesized by co-precipitation method, and the chemical structure was characterized using X-ray Diffraction (XRD), Fourier Transform Infra-red (FT-IR), Brunauer-Emmet-Teller (BET) Surface Area, and Thermo-Gravimetry Differential Analyse (TG-DTA). The XRD analysis of Ca/Al-Graphite composite was noticed in 10.205°(003), 18.083°(012), 20.45°(004), 26.532°(002), 44.52°(101), 54.52°(004), and 77.38°(006). The TG-DTA analysis of the composite was noticed at 100°C as water molecule decomposition, 270°C as nitrate decomposition, and 700 and 760°C as graphite decomposition to the oxide form. BET surface area analysis of Ca/Al-Graphite composite achieved the highest surface area at 16.795 m^2/g. According to the kinetic parameter, the adsorption of direct orange to composite follows the pseudo-second-order model. The isotherm parameter of direct orange adsorption onto the composite followed the Langmuir model and occurred spontaneously and endothermically. The regeneration study proved the composite effective in 3 cycles by adsorption percentage at the third cycle reached 73.559%.

Keywords
Ca/Al Layered Double Hydroxide, Graphite, Composite, Adsorption, Direct Orange

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1. INTRODUCTION

Nowadays, industry activity has a rapid development. The industries included in the mass-production process are mining, textile, food and beverages, and pharmacy (Coimbra et al., 2019; Dutta et al., 2021; Hussain et al., 2020; Qasem et al., 2021). The textile industry impacted economic development and decreased environmental quality due to the high volume of dye waste released (Czatzkowska et al., 2022). Dye waste containing different reactive and dissolved dye structures, can spread widely causing the carcinogenic-mutagenic effect on living organisms. Direct orange is a representative dye commonly used in the textile until the food industry has toxic characteristics due to azo structure (Krewski et al., 2019).

Different wastewater treatment methods were studied, and adsorption was chosen as simple, effective, and economically better than conventional removal methods (Rashid et al., 2021). As an essential part of adsorption, the adsorbent was modified to be effective in the reusable mechanism. Adsorbent was utilized due to the active site to catch the pollutant of wastewater and the unique characteristics of adsorbent, including the high surface area, small size, and ion exchange from layered material (Lim et al., 2019). Popular adsorbent include charcoal, biochar, zeolite, and layered double hydroxide (LDH). LDH is a group of two-dimensional layered inorganic materials. LDH has unique characteristics of adjustable composition, high surface area, structural memory effect, and reusability (Siregar et al., 2022).

The adjustable composition of LDH becomes a shining issue since it can be compiled as an $M_{1-x}^{2+}M_x^{3+}$ structure, where the M^{2+} and M^{3+} are divalent metal cations (Ca, Mg, Co, Fe, Cu, Ni, Mn) and trivalent metal cation (Al, Fe, Cr), respectively (Hanifah et al., 2023). The two-dimensional divalent-trivalent metal cation piled up in several layered and provided a basal space that exchangeable anion can fill up. In aims to provide a high-efficient adsorbent, the LDH modification mechanism based on memory effects such are self-assembly, ion exchange, and delamination. It is considered an effective method to achieve a multifunctional material based on LDH using host materials (Ye et al., 2022). The host materials generally used in LDH modification include semiconductors, carbon, metal, and other LDH materials (Hong et al., 2014; Malak-Polaczyk et al., 2010; Yang et al., 2019).

Graphite is classified as carbon-based material, usually used

in the composite matrix. Recently, graphite has also been considered an efficient adsorbent. Normah et al. (2021b) recently modified the LDH of Ni-Al composited to graphite and achieved stability structure of LDH materials with adsorption capacity at 72.464 mg/g. The graphite structure of the carbon web provides growth space for LDH particles upon the graphite surfaces. This combination significantly improves the adsorption ability. In addition, Hu et al. (2020) summarized the information about the agglomeration-avoid feature from carbon-based material, which is composited to LDH material. It triggers an increase of specific surface area in the adsorbent.

According to Hu et al. (2020), the composite of Ni/Fe-LDH to carbon-based material significantly drives the maximum capacity to 323.6 and 448.4 mg/g to methyl orange and congo red dyes, respectively. Dinari et al. (2020) have reported the successful preparation of Ni/Co-LDH composited to carbon nanotube material and increased the surface area from 5.19 to 52.63 m^2/g. Additionally, the composite material has stabilized the LDH structure and increased the reusability. This statement was proven by Suh et al. (2020), which examined the Mg/Al-LDH composited with TiO_2 and achieved 97% removal of methyl orange for five cycles.

This work reported the successfully formed Ca/Al-graphite composite using the co-precipitation method. The effectiveness of material ability in dye waste removal was examined directly to remove the direct orange (DO) contaminant. The final material was characterized using XRD, FT-IR, BET, and TGA analysis. The regeneration adsorption of this material was also observed in three cycles in different wash solutions to know the effectiveness of the composite in DO removal.

2. EXPERIMENTAL SECTION

2.1 Chemicals and Instrumentation

All chemicals in pure grade condition, including calcium nitrate tetrahydrate ($Ca(NO_3)_2.4H_2O$) purchased from Sigma Aldrich, aluminum nitrate nonahydrate ($Al(NO_3)_3.9H_2O$) purchased from Sigma Aldrich, graphite powder purchased from Merck, sodium hydroxide (NaOH) purchased from Merck, chloride acid (HCl) purchased from Ajax Finechem, ethanol purchased from Avantor, and distilled water. Instrumentations were used in this work, including X-Ray Diffraction (XRD) type Rigaku MiniFlex 600 in a scan range 2θ 5-80°, Fourier Transform Infra-Red (FT-IR) type NOVA 4200e in a scan range wavenumber 400-3800 cm^{-1}, ASAP 2020 Surface Area Analyzer based on BET calculation, and TGA analyzer type Shimadzu DTG-60H with range analysis temperature of 27-900°C. Dye concentration reduction was analyzed by Bio-base BK-UV 1800 PC Spectrophotometer UV-Vis at 497 nm.

2.2 Synthesis of Ca/Al (LDH)

Ca/Al (LDH) was synthesized by homogenizing 100 mL $Ca(NO_3)_2.4H_2O$ 0.75 M and 100 mL $Al(NO_3)_3.9H_2O$ 0.25 M in 30 minutes of stirring. Then the pH solution was controlled at 10 using NaOH 2M. The solution was stirred at 65°C for 24 hours. Then, the precipitate was filtered and washed using distilled water to reduce the impurities. Finally, the precipitate was oven-dried at 100°C overnight.

2.3 Synthesis of Ca/Al-Graphite

Ca/Al-Graphite composite was synthesized using the co-precipitation method under alkaline conditions (pH 10). A solution of 15 mL $Ca(NO_3)_2.4H_2O$ 0.75 M and 15 mL $Al(NO_3)_3.9H_2O$ 0.25 M were homogenized, and then 3 g of graphite was added. The pH solution was kept at 10 using NaOH 2 M and stirred at 65°C for 24 hours. The precipitate was filtered and washed using distilled water. Then it was oven-dried at 100°C overnight.

2.4 Adsorption Study

The adsorption study was carried out with pH, contact time, initial concentration, and temperature parameters. The effect of pH was carried out by controlling the pH of 25 mL DO 50 mg/L solution in a range of 2-11 using HCl 0.1 M and NaOH 0.1 M, followed by an adsorption process using 0.05 g adsorbent for 120 minutes stirring. The effect of contact time was examined by preparing 0.05 g adsorbent in 25 mL direct orange 50 mg/L solution and varying adsorption time in 0-200 minutes. The effect of initial concentration and temperatures was carried out by varying the dye concentration to 50, 75, 100, 125, and 150 mg/L and varying the temperatures in a range of 30-60°C. Then the adsorption reached in a system of 0.05 g adsorbent in 20 mL DO solution.

2.5 Regeneration Study

An adsorption-desorption mechanism examined the regeneration. The adsorption process using 20 mL DO 100 mg/L, repeatedly. Then, the desorption process using 20 mL of different solutions (distilled water, NaOH 1 M, HCl 1 M, acetone 1 M, and ethanol 1 M solution) under 120 minutes of stirring. The concentration excess of dye solution for every cycle was measured using a spectrophotometer UV-Vis.

3. RESULTS AND DISCUSSION

Ca/Al-graphite composite as functionalized material was characterized using XRD, FT-IR, BET, and TGA to observe the crystal structure changes, structure bonding that formed, surface area improvement, and structure transformation according to the different temperatures (Botan and de Bona Sartor, 2020; Cardinale et al., 2022; Shabanian et al., 2020; Walton and Snurr, 2007). Figure 1 shows the XRD pattern of the Ca/Al-graphite composite, which figures out the unique peak in every precursor of LDH and graphite. The peak of Ca/Al (LDH) is shown in 2θ of 10.205°(003), 18.083°(012), and 20.45°(004), while the composited graphite's peak is shown in 2θ of 26.532°(002), 44.52°(101), 54.52°(004), and 77.38°(006) (Siregar et al., 2022; Vinsiah et al., 2020; Wei et al., 2020). The peak at 10.205° is the basal spacing of Ca/Al (LDH). The low peak intensity of graphite was noticed as an amorphous structure (Vinsiah et al., 2020).

Figure 2 figured out the FT-IR analyses of the Ca/Al-graphite composite and shows several specific peaks owned by the LDH and graphite structure that united. The wide band detected

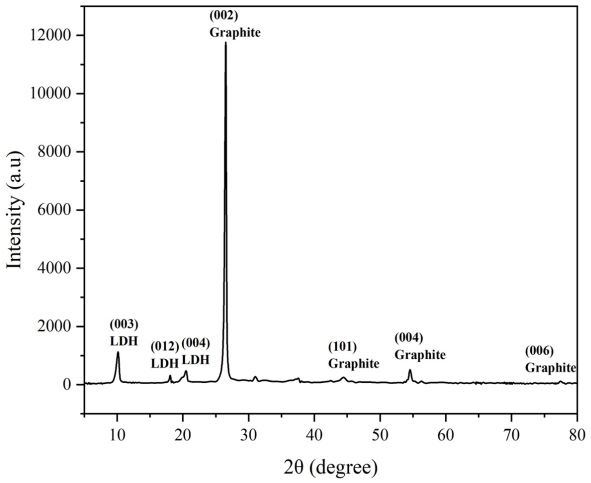

Figure 1. XRD Pattern of Ca/Al-Graphite Composite

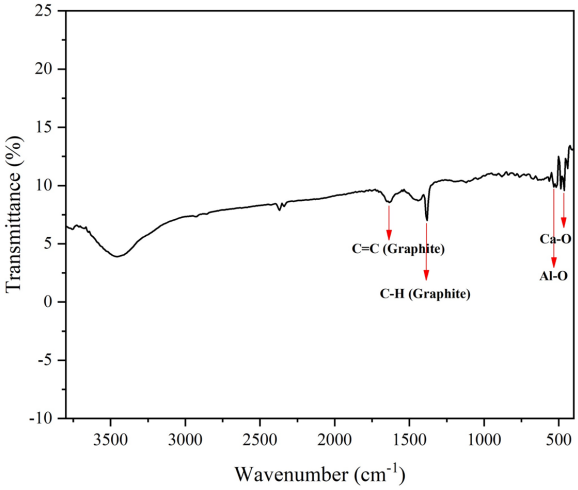

Figure 2. FT-IR Spectrum Analysis of Ca/Al-Graphite Composite

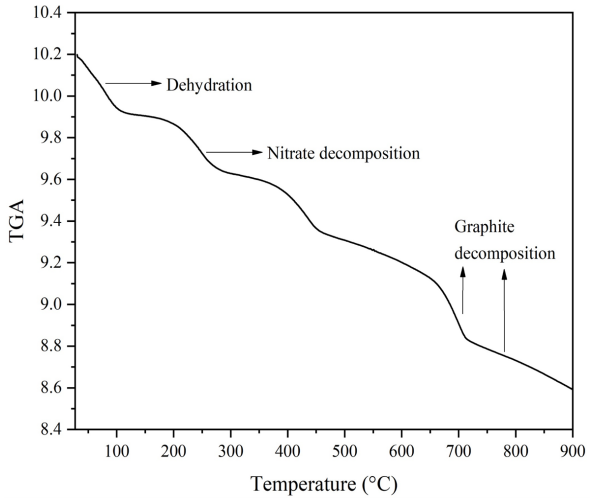

Figure 3. TGA Pattern on Ca/Al-Graphite Composite Analysis

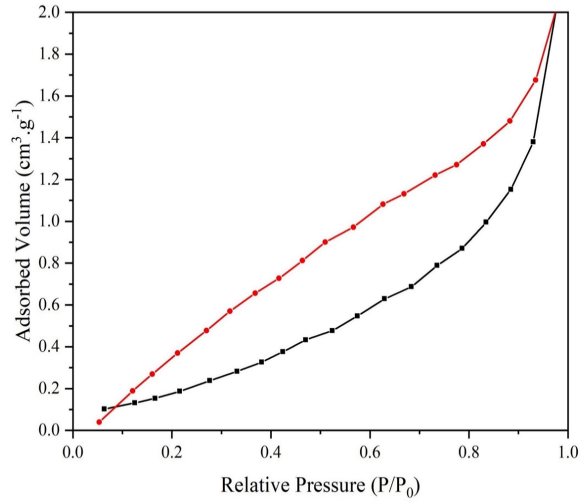

Figure 4. BET Model Profile of Ca/Al-Graphite Composite

at 3456 cm^{-1} as O-H vibration from water molecules located between the interlayer of LDH (Shabanian et al., 2020). Then, the duplet band located at 516 and 640 cm^{-1} was detected as metal-oxygen binding from the LDH structure (Ca-O and Al-O, respectively) (Siregar et al., 2022). Moreover, the double band at 1635 and 1381 cm^{-1} was detected as C=C and C=H binding from graphite web structure (Vinsiah et al., 2020).

The thermal behavior of the Ca/Al-graphite composite was carried out using a TGA instrument with nitrogen media, and the result was figured out in Figure 3. The dehydration process as water molecules decomposition from the composite structure was detected at 100°C (Kanungo and Mishra, 1996). Nitrate molecules loss by thermal decomposition was detected at 270°C (Zhao et al., 2022). Furthermore, double decreasing area at 700 and 760°C detected as graphite structure decomposition and transforming to oxide form (Zhang et al., 2018).

The surface area analysis was carried out using the BET model isotherm, and the result is shown in Figure 4. The adsorption-desorption plot shape fitted to type IV, which is indicated by the formation of a hysteresis loop (Mohadi et al., 2023). In detail, this explains that the gas capillary condensation occurred in the mesopore at relatively high pressure. The loop hysteresis shows a narrow edge and wide body classified as H2-type (Cychosz and Thommes, 2018). The analysis calculated the surface area as 16.795 m^2/g with pore volume and diameter of 0.040 cm^3/g and 2.897 nm, respectively. The composite mechanism has repaired the particle size and pore condition (Monakhova et al., 2021).

The adsorption study was initiated by screening the optimum pH in the adsorption system at 2-11. The optimum pH adsorption

Table 1. Kinetic Parameter for DO Adsorption

Dye	Initial Concentration (mg/L)	Qexperiment (mg/g)	PFO			PSO		
			Qe-calc (mg/g)	R^2	K_1	Qe-calc (mg/g)	R^2	K_2
DO	50	48.023	284.577	0.878	0.033	55.248	0.971	0.0007

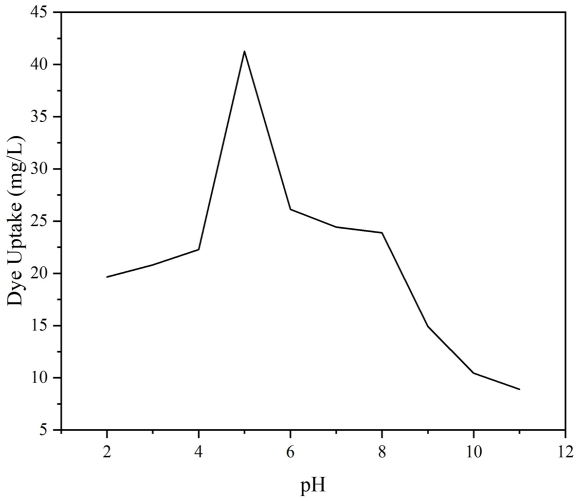

Figure 5. Plotting of pH Optimum Adsorption of DO onto Ca/Al-Graphite Composite

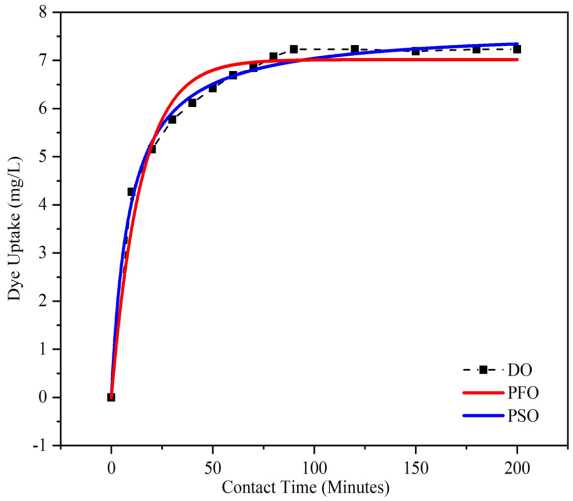

Figure 6. The Effect of Contact Time Adsorption of DO on Ca/Al-Graphite and the Fitting Toward Kinetic Model Adsorption

of DO using Ca/Al-graphite composite was achieved at 5, and the result was plotted in Figure 5. The acid condition from the optimum pH triggers protonation at composite surfaces and promotes the adsorption process by providing a binding site for

Table 2. Isotherm Parameters for DO Adsorption

Temperature (°C)	Model of Isotherm Adsorption					
	Langmuir			Freundlich		
	Qmax	KL	R^2	n	KF	R^2
30	10.010	0.089	0.980	0.425	1.731	0.928
40	105.263	0.114	0.988	0.498	5.176	0.898
50	111.111	0.139	0.993	0.532	2.251	0.904
60	112.36	0.214	0.998	0.909	1.527	0.996

the DO dye as an anionic dye (Normah et al., 2021a). The pH 5 gives appropriate composition in providing an active site and functional group that optimizes the adsorption system of the adsorbent. A considerable amount of dye particles adsorbed in this pH range suggests involvement in chemical interaction, such are hydrogen bonding, Van der Waals force, and hydrophobic interaction (Mahmoud et al., 2021). Furthermore, the increasing pH value triggers decreasing adsorption capacity due to the competitive effect between -OH from the solution's alkaline condition and the dyes' anion charge (Yadav and Dasgupta, 2022).

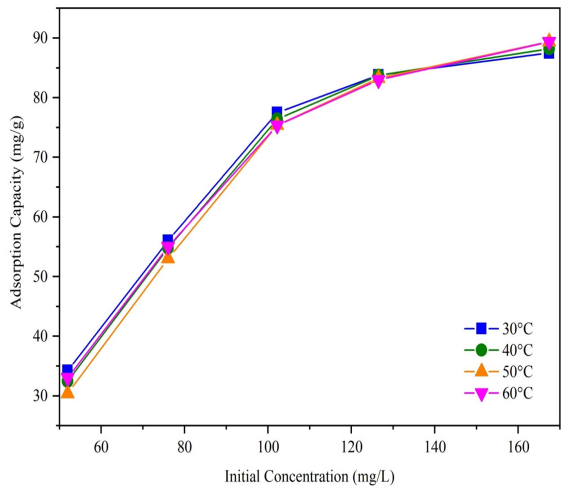

Figure 7. The Effect of Temperature and Concentration of DO Adsorption on Ca/Al-Graphite Composite

The effect of contact time in DO adsorption onto Ca/Al-graphite composite was calculated using the kinetic adsorption model of the pseudo-first-order (PFO) and pseudo-second-order (PSO) equations. The analysis result is shown in Figure 6 (Mo-

Table 3. Thermodynamic Parameters for DO Adsorption

Concentration (mg/L)	T (°C)	Qe (mg/g)	ΔH (kJ/mol)	ΔS (kJ/mol.K)	ΔG (kJ/mol)
50	30	34.231	27.528	0.096	-1.578
	40	37.462			-2.538
	50	40.385			-3.499
	60	43.615			-4.459
75	30	56.023	27.256	0.098	-2.483
	40	59.792			-3.464
	50	63.023			-4.446
	60	67.177			-5.425
100	30	77.485	21.306	0.080	-2.822
	40	81.408			-3.619
	50	85.408			-4.415
	60	88.946			-5.211
125	30	83.792	14.595	0.054	-1.692
	40	88.715			-2.230
	50	93.254			-2.767
	60	97.1			-3.305
150	30	87.531	11.410	0.038	-0.226
	40	93.223			-0.610
	50	99.377			-0.994
	60	103.992			-1.378

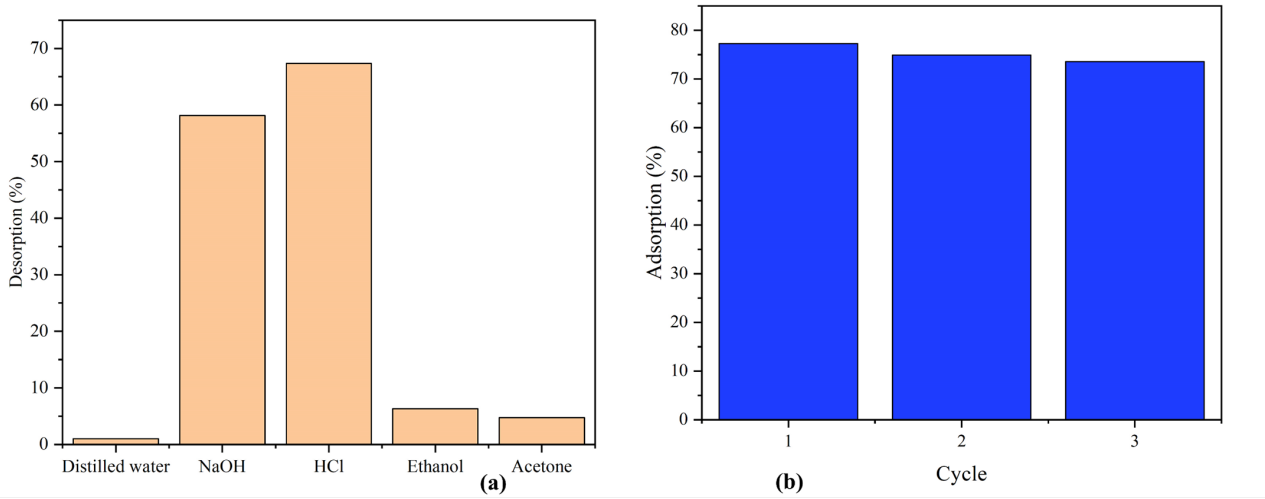

Figure 8. Effectiveness of Different Solutions on DO Desorption (a) and Degree of Regeneration Adsorbent (b)

hadi et al., 2018). According to the fit-test, the trend of kinetic adsorption follows the PSO of the kinetic adsorption model. According to Hakim et al. (2023), the PSO model explains that the interaction in the adsorption of the adsorbate occurs through the chemisorption mechanism. The kinetic parameter is shown in Table 1.

The effect of different temperatures and initial concentration adsorption was observed using the isotherm and thermodynamic adsorption model. The isotherm adsorption analysis was calculated by Langmuir and Freundlich's model equation (Freundlich, 1907; Langmuir, 1916). According to Chung et al. (2015), the differences between Langmuir and Freundlich models are assuming the adsorption scheme occurs by monolayer (directly adsorbate to adsorbent) and multilayer (growth in heterogeneous places), respectively. According to Figure 7, the result of isotherm adsorption analysis follows the Langmuir adsorption model, meaning the adsorption occurs in a monolayer (chemisorption) (Taher et al., 2018; Taher et al., 2019). This statement was supported by Table 2 data showing the higher R^2 value in the Langmuir adsorption model. The maximum adsorption capacity of Ca/Al-graphite composite toward DO dye is 112.36 mg/g.

The thermodynamic adsorption analysis was achieved by calculating the enthalpy (ΔH), entropy (ΔS), and Gibbs free energy (ΔG) equation (Motomura, 1978). According to Table 3, the thermodynamic parameters show several essential points, including the positive value of ΔH that indicates the adsorption optimal occurs by endothermic, the positive value of ΔS that indicates the DO adsorption reaction feasible in the increasing degree of freedom, and the negative value of ΔG that means the adsorption occurs spontaneously (Hakim et al., 2023; Siregar et al., 2022; Sahmoune, 2019). The adsorption capacity tends to increase aligned with increasing temperature because the high temperature can overcome the repulsive forces in the adsorption system. This statement supports by the ΔG value (Malima et al., 2021).

The regeneration study includes the adsorption and desorption schemes. The desorption scheme was initiated by screening the washer solution. According to Figure 8a, the HCl solution provides the highest desorption ability, meaning it has the highest ability to cut the DO binding to the adsorbent (Moyo, 2013; Pan et al., 2022). Furthermore, in Figure 8b, Ca/Al-graphite composite regeneration shows considerable ability due to the high adsorption percentage after three cycles, with 73.559% removal at the third cycle.

4. CONCLUSIONS

The collaborative material of the Ca/Al-graphite composite was successfully formed using the co-precipitation method. The XRD analysis confirmed the compositing result by figuring out the Ca/Al (LDH) and graphite peaks in an XRD graph. Adsorption features of surface area from the Ca/Al-graphite composite were achieved at 16.795 m^2/g. According to the adsorption study, the optimum removal of the DO occurred at a pH of 5 for 120 minutes of adsorption. The adsorption capacity of the Ca/Al-graphite composite is 112.360 mg/g. The Ca/Al-graphite composite's regeneration work showed proper desorption using an HCl solution and achieved a stable DO removal for three cycles with 73.559% removal at the third cycle.

5. ACKNOWLEDGEMENT

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