

## Layered Double Hydroxide/Chitosan Composite (Mg-Al/CT) as a Selective Adsorbent in Congo Red Adsorption from Aqueous Solution

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### ABSTRACT

Layered double hydroxide (LDH) can be used as an adsorbent to remove pollutants from aqueous solutions, but it drawbacks where the structure is easily damaged so that it cannot be reused in the adsorption process and has a low adsorption capacity. This can be overcome through the development of layered double hydroxide material composited with chitosan support material. In addition to utilizing waste, chitosan is selected as supporting material in the layered double hydroxide modification process, because it is cheap, has high selectivity, and is biodegradable. In this study, the adsorbent was applied in the process of removing Congo Red (CR). The LDH modification process using chitosan was successfully carried out, as seen from XRD analysis which resembled the base material (Mg-Al) and support (CT), the BET analysis which showed an increase in surface area, as well as from the large adsorption capacity value and the regeneration process which tends to be stable after compositing is done.

**Keywords:** adsorption, Congo Red, modified of layered double hydroxide, regeneration.

### INTRODUCTION

Color is the first recognized contaminant in wastewater. The waste generated from the use of these dyes can have a serious impact on humans and other living things (Elmoubarkia et al., 2017). Congo Red (CR) is a type of red azo dye that is commonly found in the textile industry (Zhang et al., 2018). Congo Red has the chemical formula  $C_{32}H_{22}N_6Na_2O_6S_2$  with the structure sodium ben-zidindiazo-bis-1-naphthylamine-4-sulfonate in it. Congo Red has a molecular weight of 696.665 g/mol (Vinsiah et al., 2017) (Fig. 1).

Several methods can be used to remove dyes, such as biological treatment, coagulation (Banerjee & Chattopadhyaya, 2017), photocatalysis (Kaur et al., 2021), and adsorption (Wang et al., 2020). The adsorption method is a reliable alternative because it is easy to operate (Ali et al., 2020), simple (Noreena et al., 2020), available for various adsorbents, and has high efficiency (Zhang et al., 2019). Several factors can affect the adsorption process, such as adsorbate concentration, effect of temperature, contact time, surface area from material, and particle size (Sonia et al., 2020).

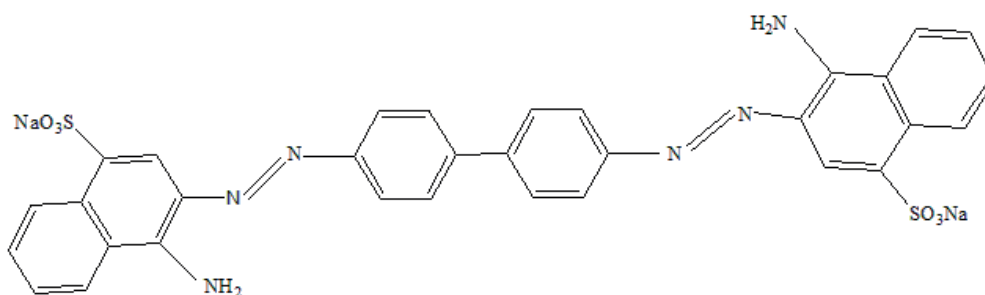


Figure 1. Congo Red

Layered double hydroxide (LDH) can be used to adsorb anionic dyes but cannot be applied to cationic dyes, so it is necessary to modify the layered double hydroxide so that it can absorb various types of dyes. According to Wu et al., (2017) layered double hydroxide has limitations as an adsorbent to adsorb dyes and its repeated use is not effective. Layered double hydroxide can be modified with composites. Supporting materials that can be used in the layered double hydroxide modification process include activated carbon, graphite, biochar, hydrochar, and chitosan (Pietrelli et al., 2014).

Chitosan (CT) is environmentally friendly, inexpensive, has good stability, and abundant availability, which makes it widely used in the layered double hydroxide modification process (Barkhordari & Alizadeh, 2021). Chitosan is used in the adsorption process of pollutants in aqueous solution (Zhang et al., 2016). Chitosan is an organic compound containing  $\text{NH}_2$  and  $-\text{OH}$  groups in its molecular structure so it can be used as an adsorbent in the adsorption process (Dey et al., 2016; Muinde et al., 2020). Chitosan can be extracted from the shells of shrimp, crab, clams, and lobster (Mulyani et al., 2019).

The adsorption capacity of 2,4-dichlorophenol from 1.33 mg/g to 1.96 mg/g after modification of layered double hydroxide (Co-Fe) using chitosan (Yang et al., 2018). Layered double hydroxide Mn-Fe material composited with chitosan could be used repeatedly to remove indigo carmine dye, as seen from the regeneration process which did not experience a significant decrease in four consecutive cycles of 98.80%; 91.07%; 81.29% and 64.37% (Khalili et al., 2021).

In this work, Mg-Al was modified using chitosan as supporting material. The Mg-Al/CT composite was applied for CR dye removal. The synthesized material was then analyzed using XRD, FTIR, and BET. In this research, the selectivity process for mixed dyestuffs was carried out first, followed by kinetics, thermodynamics, and regeneration studies.

## EXPERIMENT

### Chemicals and instrumentations

In this research, materials include  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (256.41 g/mol),  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (375.13 g/mol), chitosan extracted from shrimp shells through demineralization and deproteinization process, distilled water from PT. Bratachem Indonesia, NaOH (40 g/mol), Congo Red (CR), Methylene Blue (MB), Malachite Green (MG), and Rhodamine-B (Rh-B). The materials were characterized using XRD analysis from Rigaku Miniflex-6000, FTIR from Shimadzu Prestige-21. UV-Visible Bio-Base spectrophotometer BK-UV1800 was used to measure dye concentration.

### Preparation of Mg-Al

Mg-Al can be synthesized using the coprecipitation method;  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  solution (19.23 g, 100 mL) and  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  solution (9.378 g, 100 mL) were mixed (Badri et al., 2021). The Mg-Al mixture was added with NaOH (2 M) until the pH of the solution was 10 and stirred at  $80^\circ\text{C}$  for 24 hours. After stirring is complete, the precipitate is filtered and rinsed using distilled water. The precipitate was dried and the resulting solid was characterized using XRD, FTIR, and BET analysis.

### Preparation of chitosan

Extraction of chitosan from shrimp shells can be carried out by demineralization and deproteinization processes (Mohadi et al., 2021). The demineralization process was carried out by smoothing the shrimp shells and putting 100 g of the finely chopped shrimp shells were placed into beaker, then HCl was added a ratio of 1:10 (w/v). The mixture was stirred at  $60^\circ\text{C}$  for 3 hours. After completion, the mixture was filtered and the residue dried at  $70^\circ\text{C}$ . The demineralized residue was put into

beaker and NaOH was added with a similar ratio. The mixture was stirred for 1 hour at 60 °C. The mixture was then filtered and the precipitate was dried at 70°C. The chitosan obtained was characterized using XRD, FTIR, and BET analysis.

### Preparation of Mg-Al/CT

The Mg-Al/CT preparations were made by the coprecipitation method (Elanchezhian & Meenakshi, 2017). Total of 15 mL (0.75 M) Mg solution and 15 mL (0.25 M Al solution). The solution was mixed and adjusted the pH to 10 using 2 M NaOH then the mixture was stirred for 1 hour. After the stirring process is complete, 3 g of chitosan is added and then stirred for 72 hours at 80°C. The precipitate was dried and characterized using XRD, FTIR, and BET analysis.

### Selectivity of dyes

20 mg/L (CR, Rh-B, MG, and MB) as much as 10 mL, added 0.02 g of adsorbent, then stirred with variations of time (0, 15, 30, 60, and 120 minutes) and the adsorbed was measured using a UV-Vis spectrophotometer.

### Kinetic study

20 mL of CR solution (200 mg/L, 0.02 g adsorbent), then stirred with various contact times (0, 10, 20, 30, 60, 90, 120, 150, 180, 200, and 250 minutes). After the stirring is complete, the Congo Red solution and the adsorbent were separated using centrifugation. The filtrate was measured at the maximum wavelength of Congo Red (466 nm) using a UV-Visible spectrophotometer.

### Adsorption process

The adsorption process was carried out using variations in CR concentration (60–100 mg/L) and adsorption temperature (30–60°C). Then the filtrate was measured at the maximum wavelength of CR (466 nm) using a UV-Visible spectrophotometer.

### Regeneration

The regeneration process was carried out by adsorption of 25 mL CR dye (100 mg/L, 1 g adsorbent). After the adsorption process was completed, measurements were conducted using a UV-Vis spectrophotometer. Then the adsorbent containing

the dye was desorbed using an ultrasonic device. After completion, the adsorbent was dried using an oven and continued with the adsorption process with the same procedure for seven cycles.

## RESULTS AND DISCUSSION

The sharp peaks of Mg-Al shown in Figure 2(a) angles  $2\theta = 11.47^\circ(003)$ ,  $22.86^\circ(006)$ ,  $34.69^\circ(009)$ , and  $61.62^\circ(110)$  indicating Mg-Al has a high crystal structure similar to the research conducted by (Bolbol et al., 2019). The widened peaks defined as amorphous peaks of chitosan at angles of  $2\theta = 7.93^\circ$  and  $19.35^\circ$  with plane reflections (002) and (006) are shown in

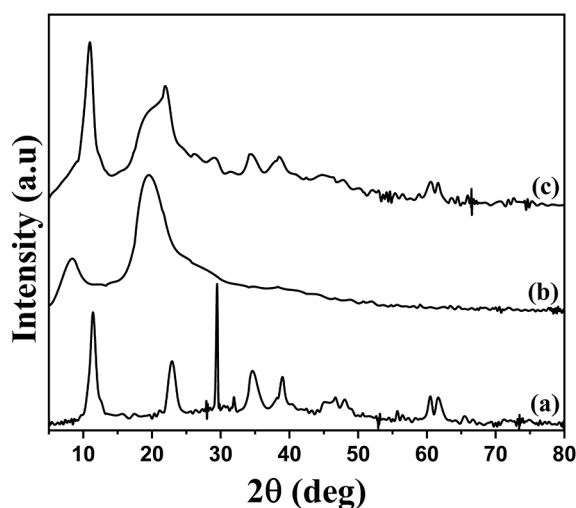


Figure 2. XRD analysis of Mg-Al (a), CT (b), Mg-Al/CT (c)

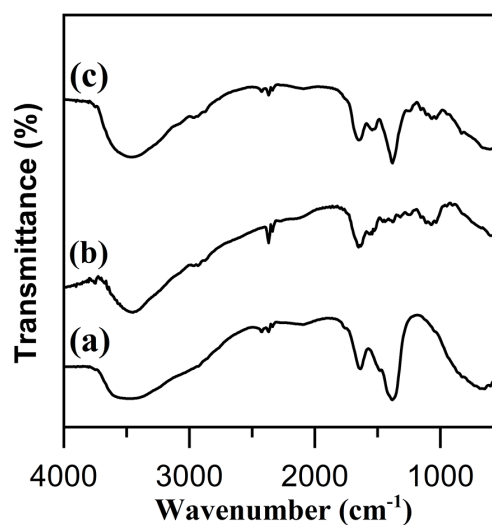


Figure 3. FTIR analysis of Mg-Al (a), CT (b), Mg-Al/CT (c)

Figure 2(b). The diffraction pattern of Mg-Al/CT is shown in Figure 2(c). Figure 2(c) shows the same peaks as Mg-Al and chitosan diffraction, namely  $10.83^\circ(003)$ ,  $19.52^\circ(006)$ , and  $60.6^\circ(110)$  (Li et al., 2006).

Figure 3 shows the FTIR spectrum of each material. Vibration peaks associated with stretching  $\text{NH}_2$  and OH groups appear at wavenumber  $3447\text{ cm}^{-1}$ . The peak at wavenumber  $1657\text{ cm}^{-1}$  was associated with the  $\text{CONH}_2$  group of chitosan (Kumar & Koh, 2012). The peak that appears at wavenumber  $1381\text{ cm}^{-1}$  indicates the presence of  $\text{NO}_3^-$  anion vibrations from LDH (Shabanian et al., 2020). The FTIR spectrum synthesized is similar to that reported by Chen et al., (2017) with different absorption intensities.

The BET analysis of each material is shown in Figure 4. It confirms that each material follows a type IV isotherm with hysteresis loop. The type IV isotherm indicates that the adsorption and desorption patterns of the material do not overlap. The hysteresis loop indicates that the material has mesopores (Siregar et al., 2021). Layered double hydroxide (Mg-Al) has a surface area of  $8.963\text{ m}^2/\text{g}$ , chitosan (CT) has a surface area of  $3.288\text{ m}^2/\text{g}$ ,

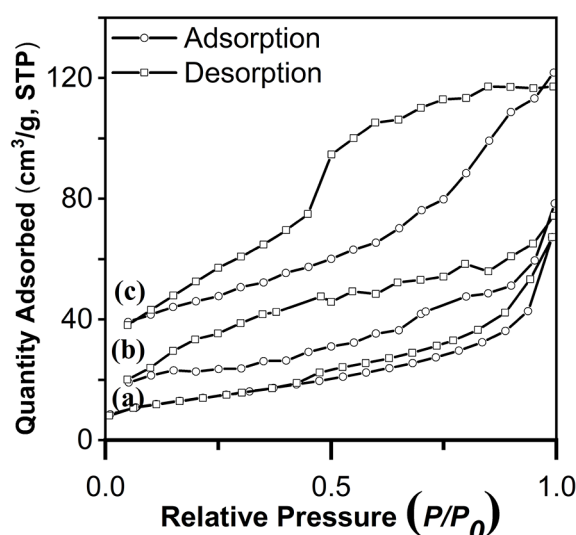


Figure 4. BET analysis of Mg-Al (a), CT (b), and Mg-Al/CT (c)

Table 1. BET analysis of materials

Materials	Surface area ( $\text{m}^2/\text{g}$ )	Pore volume ( $\text{cm}^3/\text{g}$ ) BJH	Pore size (nm), BJH
Mg-Al	8.963	0.027	3.169
CT	3.288	0.006	6.092
Mg-Al/CT	24.556	0.027	3.929

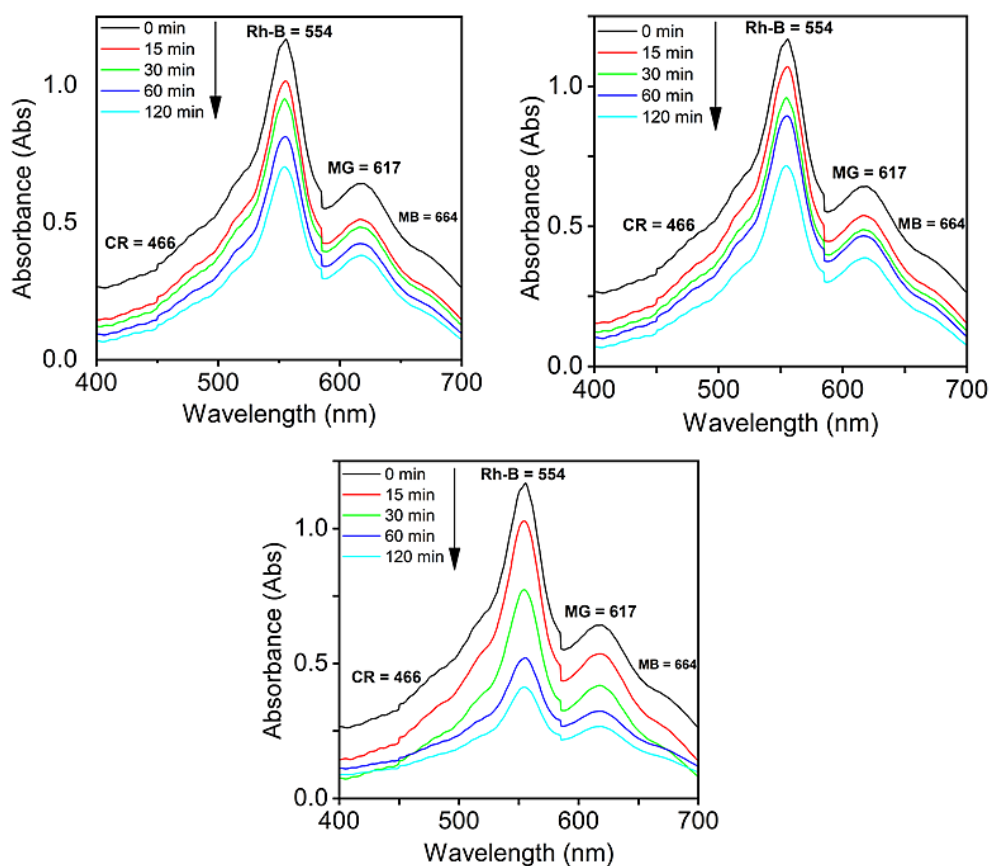


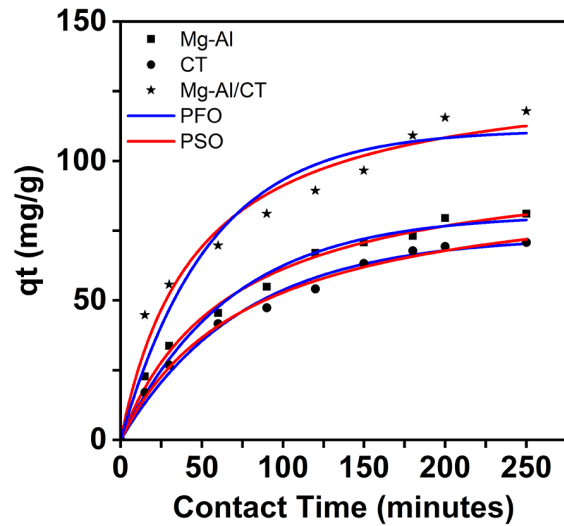
Figure 5. Selectivity of dyes mixture CR, Rh-B, MG and MB with variation contact time onto Mg-Al (a), CT (b), and Mg-Al/CT (c)

**Table 2.** Kinetic parameter

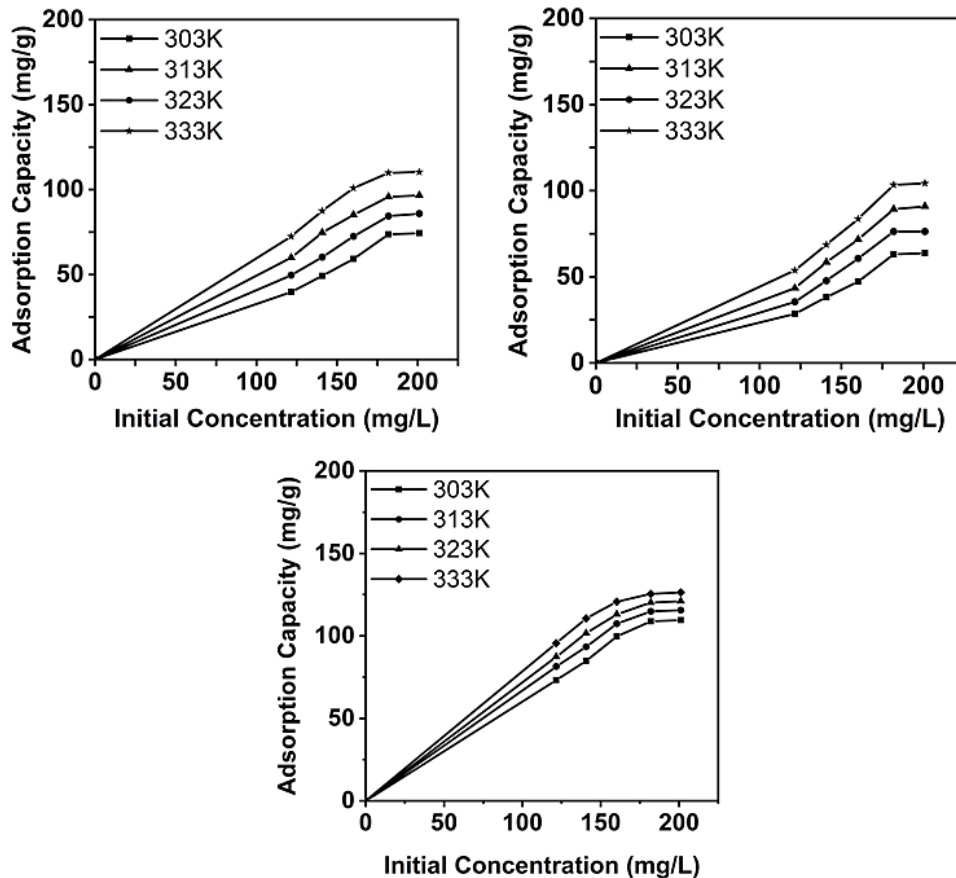
Adsorbent	$Q_{e\_exp}$ (mg/g)	PFO			PSO		
		$Q_{e\_Calc}$ (mg/g)	$R^2$	$k_1$	$Q_{e\_Calc}$ (mg/g)	$R^2$	$k_2$
Mg-Al	81.061	87.720	0.907	0.016	101.010	0.991	0.0002
CT	70.833	83.869	0.965	0.018	93.458	0.991	0.0001
Mg-Al/CT	117.803	120.837	0.876	0.015	128.205	0.951	0.0001

$m^2/g$ , and Mg-Al/CT composites have a surface area of  $24.556 m^2/g$  shown in Table 1. The Mg-Al/CT composites showed the largest surface area compared to their constituent materials (Mg-Al and CT). This confirmed that the Mg-Al modification process using chitosan was successfully carried out.

The dye mixtures (CR, Rh-B, MG, and MB) were measured at a wavelength of 400–700 nm (Figure 5). The graph of the selectivity of the dye mixture shows that with increasing adsorption time, the absorbance obtained also decreases. At the 120 minutes, there was a significant decrease in absorbance and it was seen that CR had the most significant decrease in absorbance compared to other dyes. Then the CR dye was carried out by adsorption processes such as



**Figure 6.** Adsorption contact time



**Figure 7.** Effect of adsorption on Mg-Al (a), CT (b), Mg-Al/CT (c)

from the influence of contact time, isotherm, thermodynamics, and regeneration.

Figure 6 shows the optimum contact time at 200 minutes, under optimum conditions there was an increase in the maximum absorption of the dye or the equilibrium rate of adsorption so that the addition of time had no significant effect. Table 2 shows the adsorption kinetics of each adsorbent tends to follow Pseudo Second Order (PSO) which is indicated by the value of  $R^2$  which is close to 1 and the value of  $k_2$ , which indicates the adsorption process is faster in Pseudo Second Order. and the difference in theoretical and

experimental  $Q_e$  values in Pseudo Second Order is not significant (Guechi & Hamdaoui, 2016).

The maximum adsorption process occurred at 333 K and at 120 minutes as shown in Figure 7.

The suitable isotherm model can be determined by looking at the data of the coefficient of determination ( $R^2$ ). If the  $R^2$  value is closer to 1, it can be said that there is a greater influence and the relationship between variables stronger. Table 3 shows that the  $R^2$  value tends to follow the Freundlich equation, which means that the absorption of Congo Red occurs by physisorption (physical absorption). Physical adsorption occurs because

**Table 3.** Isotherm adsorption

Adsorbent	Adsorption Isotherm	Adsorption Constant	T (K)			
			303	313	323	333
Mg-Al	Langmuir	Qmax	46.083	84.746	63.694	120.482
		kL	0.006	0.005	0.152	0.008
		$R^2$	0.981	0.951	0.938	0.835
	Freundlich	n	0.464	0.563	1.066	0.582
		kF	3.410	3.991	1.471	1.090
		$R^2$	0.987	0.994	0.998	0.973
CT	Langmuir	Qmax	39.216	53.476	67.568	116.279
		kL	0.005	0.005	0.006	0.005
		$R^2$	0.972	0.985	0.944	0.975
	Freundlich	n	0.330	0.267	0.248	0.277
		kF	3.331	4.861	9.307	7.705
		$R^2$	0.975	0.999	0.984	0.995
Mg-Al/CT	Langmuir	Qmax	192.308	222.222	263.158	344.828
		kL	0.025	0.032	0.014	0.067
		$R^2$	0.798	0.899	0.907	0.956
	Freundlich	n	1.821	2.151	2.606	3.426
		kF	13.005	21.198	18.728	23.361
		$R^2$	0.792	0.838	0.832	0.829

**Table 4.** Thermodynamic adsorption

Adsorbent	T (K)	$Q_e$ (mg/g)	$\Delta H$ (kJ/mol)	$\Delta S$ (J/mol K)	$\Delta G$ (kJ/mol)
Mg-Al	303	74.427	20.058	0.070	-1.151
	313	85.878			-1.851
	323	96.565			-2.551
	333	110.305			-3.251
CT	303	63.740	23.643	0.080	-0.564
	313	76.336			-1.363
	323	90.840			-2.162
	333	104.198			-2.961
Mg-Al/CT	303	129.779	22.875	0.080	-1.503
	313	141.687			-2.308
	323	153.901			-3.112
	333	161.382			-3.917

of physical forces (Siregar et al., 2021). The physically adsorbed molecules are not strongly bound to the surface of the adsorbent, and usually, a fast reversible process occurs, so they are easy to replace with other molecules. The value of ( $n > 1$ ) indicates a physical adsorption process (Rath et al., 2019). The respective adsorption capacities of Congo Red are shown in Table 3. The maximum adsorption capacities obtained using Mg-Al, CT, and Mg-Al/CT were 120.482 mg/g, 116.279 mg/g, 344.828 mg/g, respectively, as shown in Table 3.

Table 4 shows the thermodynamic data of adsorption using Mg-Al, CT, and Mg-Al/CT. Positive enthalpy value data ( $\Delta H$ ) indicates that the adsorption process is endothermic. The data of entropy or degree of disorder ( $\Delta S$ ) that occurs during adsorption on each adsorbent shows a small value. The greater the concentration of Congo Red, the smaller the degree of adsorption irregularity. Gibbs free energy data ( $\Delta G$ ) shows a negative value which indicates the adsorption process occurs spontaneously for each adsorbent (Siregar et al., 2021).

The Mg-Al/CT regeneration process showed an insignificant decrease for seven different cycles with Mg-Al and CT as shown in Figure 8. This shows that Mg-Al/CT is a good adsorbent in the process of adsorption of CR, this is evidenced by the increased surface area after the composite process, greater adsorption capacity, and better structural stability than Mg-Al and CT.

Table 5 shows the comparison of Congo Red adsorption using various adsorbents. The Congo

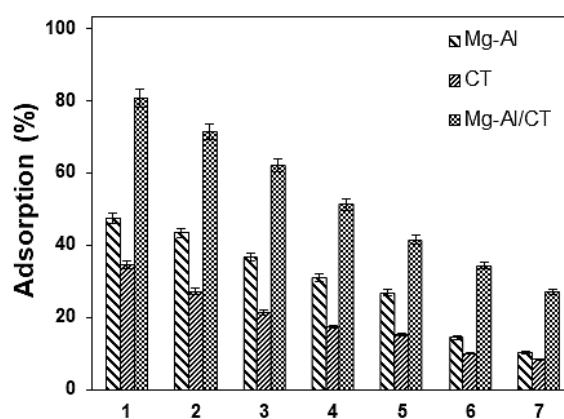


Figure 8. Regeneration cycle of each adsorbent

Red adsorption capacity obtained in this study is greater than the adsorbent in Table 5.

## CONCLUSIONS

The manufacturing of composites based on layered double hydroxide (Mg-Al) and support material of chitosan (CT) were successfully synthesized. The success of material synthesis was proven by the results of XRD, FTIR, and BET analysis. XRD analysis showed the Mg-Al has typical peaks at angles of  $2\theta = 11.47^\circ$  (003),  $22.86^\circ$  (006),  $34.69^\circ$  (009), and  $61.62^\circ$  (110) and peaks of chitosan appear at angles of  $2\theta = 7.93^\circ$  (002) and  $19.35^\circ$  (006). The results of BET analysis showed that the surface area of Mg-Al was  $8.963 \text{ m}^2/\text{g}$  and increased after modifying layered double hydroxide with chitosan to  $24.556 \text{ m}^2/\text{g}$ . Adsorption parameters were studied through

Table 5. Adsorption of CR from aqueous solutions using several adsorbent

Adsorbent	Adsorption capacity (mg/g)	Reference
Metal hydroxides sludge	40	(Attallah et al., 2013)
Diatomaceous Earth	23.2	(Sriram et al., 2020)
Shiitake mushroom	217.86	(Yang et al., 2020)
<i>Aspergillus Niger</i>	3.805	(Hamad & Saied, 2021)
Bread fruit seed shell	16	(Conrad et al., 2016)
TiO <sub>2</sub> /SCEC	18.4	(Widiyowati et al., 2020)
ZnO/chitosan	227.3	(Nguyen et al., 2020)
Magnetic chitosan composite	181.82	(Wang et al., 2013)
Na bentonite	35.84	(Vimonsesa et al., 2009)
Kaolin	5.44	(Vimonsesa et al., 2009)
Zeolit	3.77	(Vimonsesa et al., 2009)
Mg-Al	120.482	This study
CT	116.279	This study
Mg-Al/CT	344.828	This study

variations in contact time, isotherm, and thermodynamics. The kinetic model of each adsorbent tends to follow the pseudo second order (PSO). The isotherm data for each adsorbent shows that the adsorption isotherm model tends to follow the Freundlich isotherm. The adsorption capacity of Mg-Al is 120.482 mg/g, chitosan is 116.279 mg/g, and Mg-Al/CT is 344.828 mg/g. Mg-Al/CT is the most effective adsorbent for adsorption of congo red repeatedly seen from the regeneration cycle which did not experience a significant decrease in adsorption capacity for seven cycles.

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