

REMOVAL OF REACTIVE BRILLIANT ORANGE X-GN FROM AQUEOUS SOLUTIONS BY Mg-Al LAYERED DOUBLE HYDROXIDES

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Abstract—Among the many techniques used to remove toxic dyes from the environment, layered double hydroxides (LDH) are considered to be especially environmentally friendly, but, this quality may be altered by variations in the octahedral Mg/Al molar ratios in the LDH structure. The aim of the present study was to synthesize environmentally sound LDH for use as an economically viable sorbent for the adsorption of reactive brilliant orange X-GN. Layered double hydroxides with Mg/Al molar ratios of 2:1 and 4:1 were prepared by co-precipitation. The materials obtained were characterized by powder X-ray diffraction (XRD), Fourier-transform infrared (FTIR) spectroscopy, X-ray fluorescence spectroscopy (XRF), and surface-area analysis. Batch experiments were carried out to investigate the effects of contact time, pH, adsorbent dosage, and initial dye concentration on the adsorption behavior of the reactive brilliant orange X-GN by Mg-Al LDH. The results showed that the optimum pH value for dye adsorption was 3.0, at which the adsorption capacities of the reactive brilliant orange X-GN by the 2:1 LDH and the 4:1 LDH at 298 K were 79.370 mg/g and 83.343 mg/g, respectively. Further analysis of the dye-adsorption kinetics show that they fit the pseudo second-order model well. The adsorption equilibrium data showed that the Langmuir model provided better correlation of the equilibrium data than the Freundlich model. This result indicates that LDH provide specific homogeneous sites where monolayer dye adsorption occurs. The results of XRD and FTIR analyses of LDH before and after the dye adsorption demonstrated that the adsorption mechanisms were ion exchange and coulombic attraction.

Key Words—Dye Adsorption, Layered Double Hydroxides, Reactive Brilliant Orange X-GN.

INTRODUCTION

Reactive dyes are used extensively in the textile industry for their bright color, high solubility, ease of application, and low energy consumption (Aksu and Dönmez, 2003). The exact amount of reactive dyes produced worldwide and the quantity discharged into the environment are unknown, but Forgacs *et al.* (2004) estimated that ~40 tons of reactive dyes per year are lost in their production and use. The presence of dyes causes visual pollution and can be carcinogenic even at low concentrations. Moreover, many dyes and their breakdown products in wastewater are toxic to aquatic life forms (Weisburger, 2002).

A wide range of conventional treatment techniques such as oxidative processes, Fenton's reagent, ozonation, electrochemical destruction, activated carbon sorption, and membrane filtration have been applied to remove dyes from wastewater (Robinson *et al.*, 2001). Each technique has technical and economic limitations (Pearce *et al.*, 2003).

Layered double hydroxides (LDH), also known as hydrotalcite-like compounds or anionic clays, are a class of two-dimensional nanostructured anionic clays. They exist in nature and are also relatively simple and economical to synthesize (Goh *et al.*, 2008). The structure of the LDH consists of octahedral double hydroxyl layers with exposed positive surface charges and anions in the hydrated interlayer regions which compensate the positive charges of the brucite-like sheets. The surface positive charges of the LDH can be attributed to the isomorphous replacement between the divalent and trivalent cations located at the center of the octahedron (Yong *et al.*, 2000). In recent decades, LDH have been used widely as adsorbents to remove arsenic (Goh *et al.*, 2009), mercury (Nakayama *et al.*, 2007), phosphate (Cheng *et al.*, 2009), 2-chlorophenol (Chuang *et al.*, 2008), phenol, and 4-nitrophenol (Chen *et al.*, 2009) from aqueous solutions due to their large surface areas and ion-exchange capacities. Layered double hydroxides are considered to be environmentally friendly materials (Goh *et al.*, 2008; Reinholdt *et al.*, 2009) and could potentially be used as adsorbents for removing active dyes.

The purpose of the present study was to synthesize LDH with Mg/Al molar ratios of 2:1 or 4:1, then to use them as adsorbents for reactive brilliant orange X-GN in order to understand the adsorption interactions between

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LDH and anionic dyes, including reaction kinetics and changes in dye and LDH configurations.

MATERIALS AND METHODS

Preparation and structural characterization of Mg-Al LDH

The Mg-Al LDH used in this work were synthesized in the authors' laboratory using a procedure adapted from the co-precipitation method (Dadwhal *et al.*, 2008). A solution (140 mL) containing 0.7 moles of NaOH and 0.18 moles of Na₂CO₃ was added dropwise to a second solution (180 mL) containing 0.04 moles of Al(NO₃)₃·6H₂O and 0.08 or 0.16 moles of Mg(NO₃)₂·6H₂O under magnetic stirring at 50°C. The resultant slurry was aged without stirring for 24 h at 60°C. The solid phase was then separated by centrifugation at 4000 × g for 10 min. The resultant cake was washed eight times with deionized water so as to completely remove Na salts from the LDH. Afterward, the sample obtained was dried at 80°C, crushed using a ceramic mortar, sieved through a 200 mesh screen, and denoted as 2:1 LDH and 4:1 LDH, respectively.

The XRF analysis of the synthesized material indicated that the Mg/Al molar ratios of the LDH prepared were ~2.1 and ~3.8, respectively. The surface areas were measured using the ASAP 2020 volumetric adsorption analyzer (Micromeritics Instrument Corporation, USA). The samples were degassed at 110°C for 12 h. The XRD patterns were obtained using a Rigaku D/max-III diffractometer equipped with CuKα radiation (λ = 0.154 nm) operating at 40 kV and 40 mA. The data were collected at room temperature (25°C) in the range 2–70°2θ with a scanning speed of 4°2θ/min. The FTIR spectrum of each sample was recorded over the range 4000 to 400 cm⁻¹ using the KBr pellet technique on a Perkin Elmer 1725X FTIR spectrometer. The concentration of the sample in KBr was 0.5%.

Adsorption of reactive brilliant orange X-GN

The reactive brilliant orange X-GN was obtained from a local business, Advanced Technology Industry, Ltd. (Guangdong, P.R. China), and used as a target dye in all trials without further purification. The chemical structure (Figure 1) of the dye showed that the main functional group was SO₃²⁻.

The reactive brilliant orange X-GN adsorption experiments were conducted in 150 mL stoppered conical flasks by mixing a 50.00 mL reactive dye

solution of the selected concentration with 0.05 g of LDH as adsorbent, followed by immediate shaking of the suspension in a constant-temperature water bath for 60 min. The pH of the dye solution was adjusted to 3.0 by addition of either 0.1 mol/L HCl or 0.1 mol/L NaOH. The mixtures were filtered through a 0.45 μm cellulose acetate membrane. The concentrations of the reactive dye were determined using a Shimadzu 2501 PC UV-vis spectrophotometer. The adsorption capacity, *q_e*, was given by the following equation:

$$q_e = (C_0 - C_e)V/m \quad (1)$$

where *q_e* (mg/g) is the equilibrium adsorption capacity, *C₀* (mg/L) is the initial concentration of the reactive brilliant orange X-GN, *C_e* (mg/L) is the equilibrium concentration of the reactive brilliant orange X-GN in solution, *V* (L) is the suspension volume, and *m* (g) is the dry weight of adsorbent.

RESULTS AND DISCUSSION

XRD analysis

The XRD patterns (Figure 2) show the 2:1 LDH and 4:1 LDH before and after adsorption of the reactive dye. The sharp and symmetric peaks at lower 2θ values suggested that the two LDH were crystallized with well ordered layered structures (Wang *et al.*, 2009). The average metal–metal distances inside the 2:1 LDH and 4:1 LDH (Table 1) were 3.05 Å and 3.08 Å, respectively. The interlayer distance between the brucite-like sheets increased from 22.94 Å to 23.85 Å as the Mg/Al molar ratio increased from 2.1:1 to 3.8:1. This could be interpreted as a result of fewer Mg atoms being substituted isomorphously by Al within the structure of the LDH and the electrostatic attraction between the positively charged layers being reduced as a result (You *et al.*, 2001). With the Mg/Al molar ratios of 2 and 4, the original *d* spacing values (*d*₀₀₃) of the 2:1 LDH and of the 4:1 LDH were 7.65 Å and 7.95 Å, respectively. After interaction with the reactive dye, the *d*₀₀₃ spacing of the 4:1 LDH was increased to 7.99 Å. The changes in the *d*₀₀₃ spacings of 4:1 LDH proved partial replacement of interlayer carbonate ions by the dye. The change in the basal spacing also indicated that the 4:1 LDH adsorbed more of the dye than the 2:1 LDH.

FTIR analysis

Additional evidence for the intercalation of the reactive dye was provided by FTIR spectroscopy in the

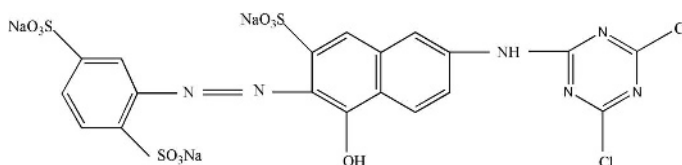


Figure 1. Chemical structure of the reactive brilliant orange X-GN.

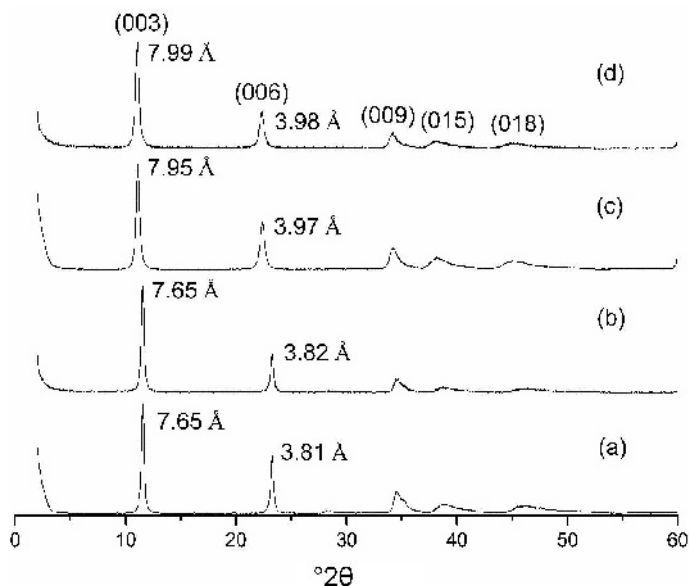


Figure 2. XRD patterns of LDH before and after dye adsorption: (a) 2:1 LDH, (b) 2:1 LDH-dye, (c) 4:1 LDH, (d) 4:1 LDH-dye.

frequency range $4000\text{--}400\text{ cm}^{-1}$ (Figures 3, 4). The broad adsorption peak between 3600 and 3300 cm^{-1} was due to the vibration of structural OH^- groups from the brucite-like sheets as well as from the interlayer water molecules. The peaks at $\sim 1650\text{ cm}^{-1}$, 1370 cm^{-1} , and 1384 cm^{-1} were attributed to the vibration of CO_3^{2-} (Yang *et al.*, 2005; Beaudot *et al.*, 2004). The peaks at 1198 cm^{-1} and 1050 cm^{-1} correspond to the vibration of SO_3^{2-} . Two new peaks at $\sim 1050\text{ cm}^{-1}$ and 1224 cm^{-1} appeared in the spectra for both LDH after adsorption of the dye. The peak at 1050 cm^{-1} corresponds to the intercalation of the SO_3^{2-} in the brucite-like sheets. The peaks at 1238 cm^{-1} and 1198 cm^{-1} were shifted to $\sim 1224\text{ cm}^{-1}$, suggesting coulombic attraction of the SO_3^{2-} by the positively charged surface of the LDH. The peak at 1650 cm^{-1} was shifted to a smaller wavenumber and the intensity of the peaks at 1370 cm^{-1} and 1384 cm^{-1} was reduced after adsorption of the dye, demonstrating ion exchange between CO_3^{2-} and SO_3^{2-} .

Effect of contact time and adsorption kinetics

The equilibrium time and adsorption rate of the dye onto the 2:1 and 4:1 LDH can be determined by means of the kinetic curves (Figure 5) which showed that the adsorption processes were rapid over the initial 5 min, and reached equilibrium within ~ 30 min. This could be interpreted as follows: in the first stage (within 5 min), the concentration of the reactive dye was relatively high and more adsorption sites were on the surface of the LDH. The dye reacted easily with the adsorption sites. In the later stage, the sites were almost all taken up and the dye concentration became lower, resulting in the slower rate of adsorption.

In order to evaluate the kinetic mechanisms that control the adsorption process and potential rate-controlling steps, the pseudo first-order, pseudo second-order, and Elovich equation models (Anirudhan and Radhakrishnan, 2009; Özacar and Sengil, 2005) were used to interpret the experimental data at an initial

Table 1. Textural characteristics of the LDH.

	Chemical composition Mg/Al	BET surface area S (m^2/g)	Pore volume V (m^3/g)	Lattice parameter		
				a^a (Å)	c^b (Å)	c_1^c (Å)
2:1 LDH	2.1	51.6	0.35	3.05	22.94	22.96
4:1 LDH	3.8	59.2	0.38	3.08	23.85	23.98

^a a = average metal–metal distance inside the LDH = $2 \times d_{110}$.

^b c = interlayer distance regulated by the size and charge of the anion placed between the brucite-like sheets = $3 \times d_{003}$.

^c c_1 = interlayer distance regulated by the size and charge of the anion placed between the brucite-like sheets after the adsorption of reactive orange, X-GN.

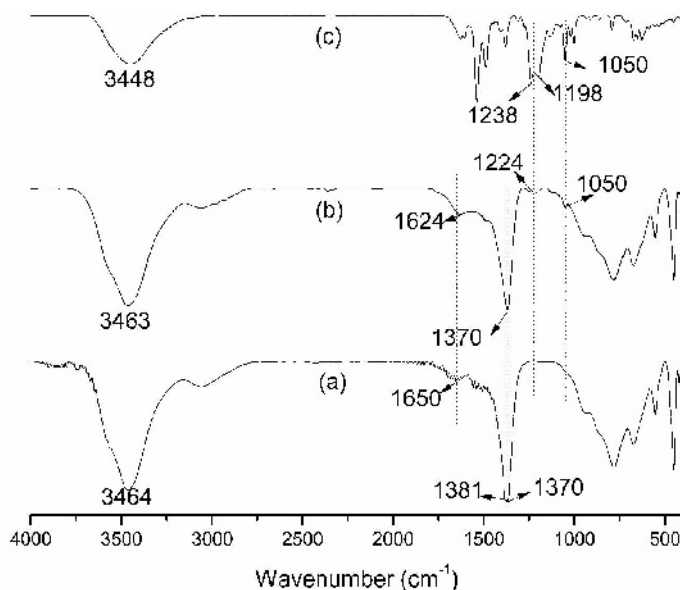


Figure 3. FTIR spectra of (a) 2:1 LDH; (b) 2:1 LDH-dye; (c) reactive brilliant orange X-GN.

dye concentration of 100 mg/L, an initial temperature of 25°C, and at initial pH 3.0. On comparison, only the pseudo second-order equation was applicable; this could be expressed by

$$d_q/dt = K(q_e - q_t)^2 \quad (2)$$

where q_e (mg/g) and q_t (mg/g) were the amounts of the reactive dye adsorbed on the LDH at equilibrium and at any time, t (min), respectively; and K (g/mg/min) was

the adsorption constant of the pseudo second-order model.

Integrating equation 2 for the boundary conditions $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_t$, the equation becomes

$$t/q_t = 1/Kq_e^2 + t/q_e \quad (3)$$

The kinetic parameters (Table 2) for adsorption of the reactive dye on the 2:1 and 4:1 LDH fit the pseudo

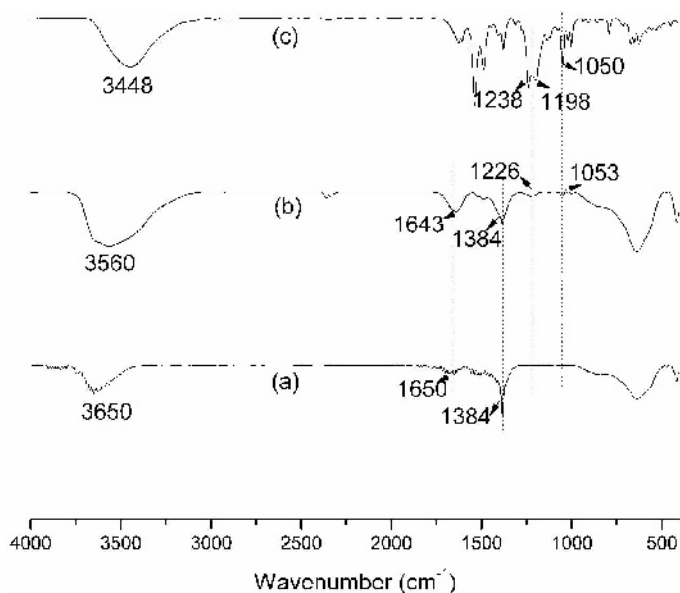


Figure 4. FTIR spectra of (a) 4:1 LDH; (b) 4:1 LDH-dye; (c) reactive brilliant orange X-GN.

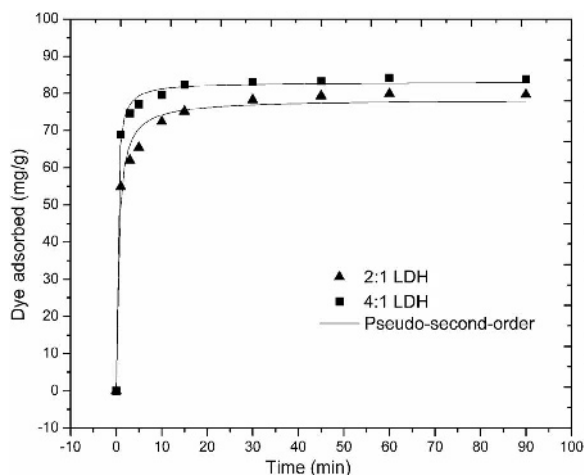


Figure 5. Pseudo second-order kinetic model of the adsorption of reactive brilliant orange X-GN by 2:1 LDH and 4:1 LDH. Initial reactive dye concentration: 100 mg/L; temperature: 25°C; pH: 3.0; amount of LDH: 50 mg/50 mL; stirring rate: 200 rpm.

second-order model well in the present study. The values of theoretical q_e (Table 2) for both LDH were in good agreement with the experimental values. The correlation coefficients obtained were >0.98 , indicating that the rate-limiting step of the adsorption mechanism was chemical sorption involving force through sharing or exchange of electrons between the dye and LDH, but not involving a mass transfer in solution.

Effect of initial pH

The adsorption of the reactive dye by the 2:1 and 4:1 LDH at different initial pH values ranging from 3.0 to 10.0 showed that the optimum pH value for adsorption of the dye was 3.0 (Figure 6). The adsorption capacity of the reactive dye decreased with increasing pH with a plateau region from ~ 5.0 to 9.0; similar behavior was reported by Netpradit *et al.* (2004) and Al-Degs *et al.* (2008). The decrease in adsorption at higher pH may have been due to the abundance of OH^- ions.

Effect of adsorbent dosage

Optimum dosages of adsorbents for removal of reactive dye in a batch adsorption system are crucial for their cost-effective application. The adsorption

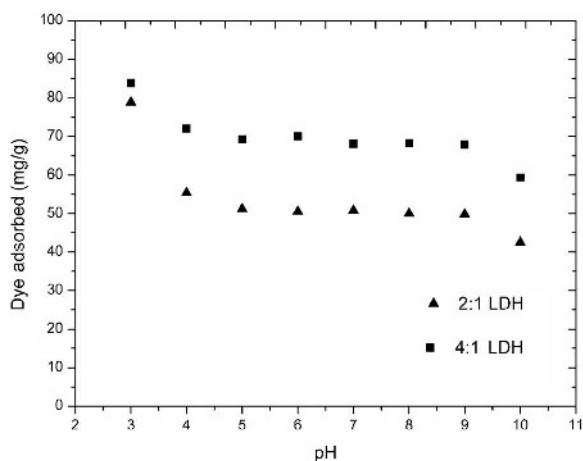


Figure 6. Effect of initial solution pH on the adsorption of the reactive brilliant orange X-GN by 2:1 LDH and 4:1 LDH. Initial reactive dye concentration: 100 mg/L; temperature: 25°C; contact time: 60 min; amount of LDH: 50 mg/50 mL; stirring rate: 200 rpm.

capacities were decreased with increasing amounts of adsorbents as the removal rate was increased (Figure 7). Equilibrium removal rates of the reactive dye were 96% by 2:1 LDH and 98% by 4:1 LDH at adsorbent dosages of 0.1 g. Considering both the adsorbent dosage and the rate of removal of the dye, the optimum adsorbent dosage was 0.05 g/50 mL for both materials.

Effect of initial concentration and adsorption isotherms

The adsorption isotherms (Figure 8) of the reactive brilliant orange X-GN onto the LDH were determined at initial concentrations from 20 to 120 mg/L. The adsorption capacities increased notably with increase in the lower initial concentrations until the plateau was reached. With different Mg/Al ratios, the LDH exhibited different dye-adsorption characteristics. The maximum adsorption capacity of the 4:1 LDH was higher than that of the 2:1 LDH. The greater dye-adsorption capacity of the 4:1 LDH could be attributed to its larger interlayer basal spacing, and to its greater surface area and pore volume (Table 1).

In the present study, the equilibrium adsorption data were analyzed using the Langmuir, Freundlich, and Redlich-Peterson models (Noeline *et al.*, 2005; Justi *et al.*, 2005), the linear forms of which are as follows:

Table 2. Pseudo second-order kinetic parameters for the adsorption of reactive brilliant orange X-GN by 2:1 and 4:1 LDH.

	R^2 ^a	K (g/mg/min)	q_e (mg/g)	q_e (experimental) (mg/g)
2:1 LDH	0.983	43.451	78.372	79.370
4:1 LDH	0.996	19.529	83.081	83.343

^a correlation coefficient

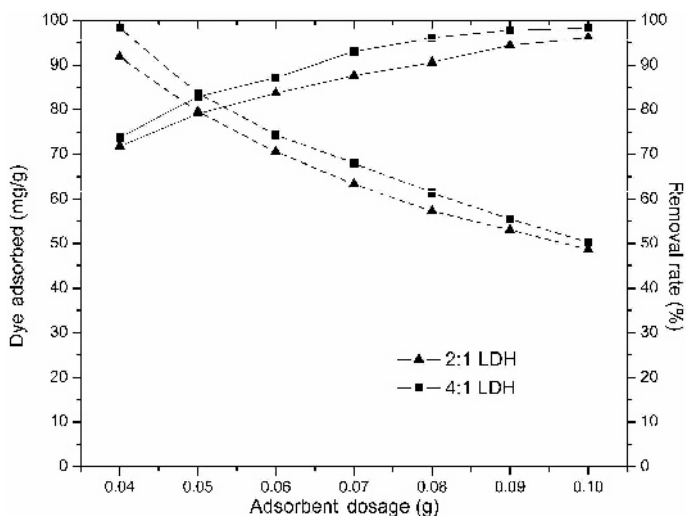


Figure 7. Effect of the dosage of 2:1 LDH and 4:1 LDH on the amount of dye adsorbed and on the rate of removal. Initial reactive dye concentration: 100 mg/L; temperature: 25°C; pH: 3.0; contact time: 60 min; stirring rate: 200 rpm.

$$q_e = bq_m C_e / (1 + bC_e) \tag{4}$$

$$q_e = K_f C_e^{1/n} \tag{5}$$

$$q_e = K_{RP} C_e / (1 + \alpha C_e^\beta) \tag{6}$$

where q_e (mg/g) is the equilibrium adsorption capacity; C_e (mg/L) is the equilibrium concentration of the reactive orange in the solution; q_m (mg/g) is the maximum adsorption capacity on the adsorbent; $1/n$ indicates the adsorption intensity; K_f is a constant related to the sorption capacity; K_{RP} and α are adsorption constants; and β is a heterogeneity index of between 0 and 1.

The values of the isotherm constants (Table 3) were determined by fitting the models above to the experi-

mental data. The fit of the experimental data to the Freundlich model was poorer than either the Langmuir or Redlich–Peterson models. The latter two models gave the largest R^2 values, >0.96 for both LDH. The Langmuir model is built on the assumption of adsorption homogeneity, such as equally available adsorption sites, monolayer surface coverage, and no interaction among adsorbed species. The Langmuir isotherm was more suitable than the Freundlich isotherm, suggesting homogeneous adsorption, which means that the dye adsorption on both LDH was monolayer sorption. The b value of the 4:1 LDH was greater than that of the 2:1 LDH, indicating a higher affinity for the LDH surface. The values of β for both materials were ≈ 1 , suggesting that

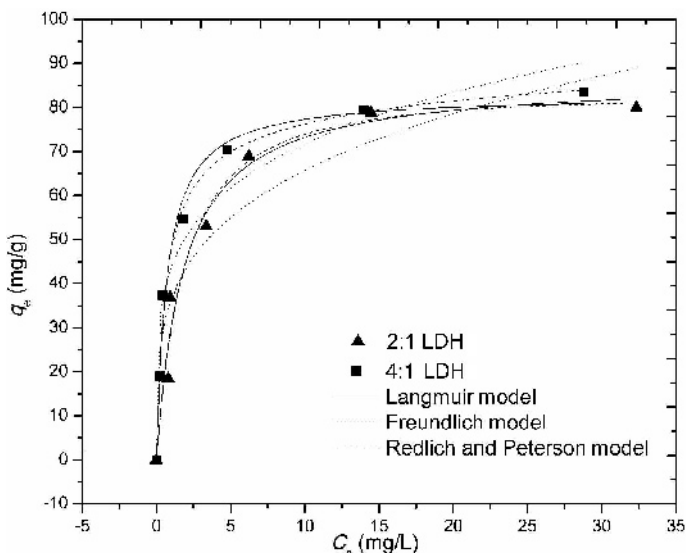


Figure 8. Adsorption isotherms of the adsorption of reactive brilliant orange X-GN by 2:1 LDH and 4:1 LDH. Temperature: 25°C; pH: 3.0; contact time: 60 min; amount of LDH: 50 mg/50 mL; stirring rate: 200 rpm.

Table 3. Parameters for the adsorption of reactive brilliant orange X-GN onto 2:1 and 4:1 LDH by the Langmuir, Freundlich, and Redlich–Peterson models.

	Langmuir parameters			Freundlich parameters			– Redlich–Peterson parameters –			
	R ²	<i>b</i>	<i>q_m</i>	R ²	K _f	<i>n</i>	R ²	K _{RP}	α	β
2:1 LDH	0.971	0.548	86.598	0.896	36.338	3.884	0.965	43.941	0.461	1.030
4:1 LDH	0.986	1.412	82.851	0.934	43.645	4.625	0.989	148.283	2.111	0.941

the adsorption process took place mainly at specific homogeneous sites within the adsorbents.

CONCLUSIONS

The adsorption of reactive brilliant orange X-GN from aqueous solutions by the 2:1 and 4:1 LDH prepared were investigated under different reaction conditions. BET analysis showed that the surface area and pore volume of the 4:1 LDH were larger than those of the 2:1 LDH. The XRD patterns demonstrated that the reactive dye was intercalated into the interlayer space of the LDH as shown by the increase in the *d*₀₀₃ values in the case of the 4:1 LDH. The FTIR spectra revealed that the adsorption of the reactive dye was mainly governed by ion exchange between the dye and carbonate ions and coulombic attraction between the dye molecule and LDH surface.

Adsorption experiments showed that the maximum adsorption capacity of the 4:1 LDH was greater than that of the 2:1 LDH. The kinetics of the dye adsorption are well described by the pseudo second-order rate expression (R²>0.98) with an equilibrium time of 60 min. The equilibrium studies demonstrated that the reactive dye adsorption process was dependent on the solution pH. The adsorption capacity of the reactive dye decreased as the solution pH value increased. The optimal pH value was 3.0. The Langmuir equation provided better correlation, indicating that the adsorption mechanism of the reactive dye was monolayer adsorption.

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