



## Letter to the Editor

## Sorption study of an acid dye from an aqueous solution on modified Mg–Al layered double hydroxides

## ARTICLE INFO

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## ABSTRACT

In this paper, experimental methods and results are reported on the removal of the dye Green Bezanyl-F2B (an acid dye) from  $MgAlCO_3$  (HT) and from intercalated anionic surfactant, "sodium dodecylsulfate (SDS)", into the Mg–Al layered double hydroxides by the calcination–rehydration reaction using Mg–Al oxide precursors calcined at 773 K. Dodecylsulfate hydrotalcite was prepared by the calcination–rehydration method. The surfactant intercalation in the interlayer space of hydrotalcite was investigated by XRD and FT-IR spectroscopy where the resulting materials were found to be similar to those reported in the literature and were used to remove an acid dye from an aqueous solution. Equilibrium time and rate-determining step of the dye Green Bezanyl-F2B sorption were determined. Two simplified kinetic models were tested to investigate the sorption. The adsorption capacity data were also fitted to Langmuir and Freundlich equation as well. The sorption data fitted to the Langmuir model gave good values of the determination coefficient.

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## 1. Introduction

Layered double hydroxides (LDH), also referred to as hydrotalcite like compounds (HT) or anionic clays are an important class of ionic lamellar solids [1]. They have received considerable attention in recent years because of their potential applications such as ion-exchangers, catalysts or catalyst supports, sorbents and antacids [2–5]. The structure of LDH consists of positively charged mixed metal hydroxide layers separated by charge-balancing anions and water molecules.

The general chemical formula of these lamellar solids can be written as  $[M_{1-x}^{2+}M_x^{3+}(OH)_2](A^{n-})_{x/n} \cdot mH_2O$ , where  $M^{2+}$  and  $M^{3+}$  are divalent and trivalent cations respectively, and  $A^{n-}$  is an interlayer anion. Such  $A^{n-}$  anions may be polymers, organic dyes [6,7], surfactants or organic acids [8]. The interesting properties of these materials result from their structural brucite-like sheets. They consist of metal cations octahedrally coordinated to  $OH^-$  and interlayer region containing water and anions which compensate the positive charge of the brucite-like sheets. The synthesis of surfactant intercalated layered double hydroxides reported in the literature led us to consider this type of materials as sorbent for organic solutes and particularly the negatively charged species. The organophilic nature of surfactant in the LDH interlayer would permit the sorption of many types of organic molecules [9,10].

Textile industry wastewater contains a wide variety of dyes which are toxic and have a great influence on photosynthetic activity in aquatic biota. Due to their low biodegradability, dyes are generally removed from aqueous solutions by a sorption process using activated carbon [11]. The cost of this process led to numerous studies on alternative removal methods by use of less expensive natural materials and waste by-products such as sludge, perlite, rice husk, sawdust, bentonite, organophilic bentonites and layered double hydroxides [12–16].

In the present work, we describe (1) the preparation of dodecylsulfate intercalated hydrotalcite by the calcination–rehydration method, using calcined Mg–Al hydrotalcite precursors, (2) the study of the acid dye (Green Bezanyl-F2B) removal process.

The present work compares two different materials (HT and HT-SDS) for treating effluents containing acid dye Green Bezanyl-F2B. The latter is an industrial dye used in the tinting of chemical fibres.

## 2. Experimental

## 2.1. Starting materials

## 2.1.1. Hydrotalcite and calcined hydrotalcite

The layered double hydroxide  $MgAlCO_3$  (HT), with a Mg/Al molar ratio = 2:1 was synthesized by a co-precipitation method at a constant pH of 10, according to the method described by Reichle [17]. A mixed solution of 0.1 mol of  $MgCl_2$  and 0.05 mol of  $Al(NO_3)_3$  in 80 ml of distilled water was added dropwise under vigorous stirring to 100 ml of an aqueous solution containing 0.35 mol of NaOH and 0.09 mol of  $Na_2CO_3$ . During the co-precipitation process, the pH was maintained at a constant value (10) by addition of 1N  $HNO_3$  solution. The obtained gel was stirred during 20 h at 338 K until crystallization. The solid was filtered and washed with distilled water until obtaining a  $Cl^-$  free HT- $CO_3$  ( $AgNO_3$  test). This material was dried at 378 K during 18 h, ground and finally, 0.250 mm-sieved. A fraction of the resulting material was calcined at 773 K during 4 h. The solid obtained is noted HT-C773.

## 2.1.2. Surfactant modified hydrotalcite

The surfactant (sodium dodecylsulfate) was intercalated using the calcined hydrotalcite rehydration method which was used in the literature for the insertion of several types of organic molecules [18–19].

An aqueous solution of 1.44 g of sodium dodecylsulfate (SDS) in 100 ml of distilled water was refluxed during 1 h at 353 K and then cooled under nitrogen atmosphere. A suspension of 1 g of HT-C773 in the prepared sodium dodecylsulfate solution was vigorously stirred during 24 h under nitrogen and centrifuged. The solid was washed three times with hot distilled water and dried at 338 K during 24 h. The material noted HT-SDS for dodecylsulfate was ground and passed through a 0.250 mm sieve.

### 2.1.3. Sorbate

Green Bezanyl-F2B was provided by the Ciba Society (Zurich, Switzerland) and was used as received. Synthetic test dye solution was prepared by dissolving accurately a known amount of dye (1 g/L) in distilled water and subsequently diluting to required concentrations.

### 2.1.4. Characterization of the prepared materials

X-ray powder diffraction data of the hydrotalcite (HT) and SDS intercalated hydrotalcite (HT-SDS) were collected with monochromatic Cu K $\alpha$  radiation using a Phillips PW3710 diffractometer. The FT-IR spectra of the samples in the 4000–400 cm $^{-1}$  were obtained with a FOURIER MATTSON genesis series FT-IR spectrophotometer.

## 2.2. Study of green Bezanyl-F2B removal with HT and HT-SDS

### 2.2.1. Kinetic study on the removal of Green Bezanyl-F2B

Kinetic studies were conducted to find out the equilibrium time for Green Bezanyl-F2B uptake by HT and HT-SDS. In this study, initial Green Bezanyl-F2B concentration was fixed at 50 mg/L (HT) or 100 mg/L (HT-SDS) and the adsorbent dose at 0.5 g/L. The pH of the solution was maintained at 5. Suspensions were stirred for different time intervals (10–360 min) at room temperature, and then centrifuged. The dye concentration in the supernatants was analysed colorimetrically using a spectrophotometer (HACH DR/4000 U) by measuring absorbance at  $\lambda_{\text{max}} = 644$  nm. A six-point calibration curve (5–60 mg/L) was run before each analysis. In this concentration range, the absorbance/concentration curve was linear ( $R^2 > 0.99$ ). The linear Beer-Lambert relationship was used in the concentration analysis.

### 2.2.2. Sorption isotherms

The sorption isotherms were established using HT and HT-SDS suspensions in Green Bezanyl-F2B solution (solid/solution ratio = 0.5 g/L) in a range from 15 to 70 mg/L for HT and from 50 to 160 mg/L for HT-SDS. The pH of the solution was adjusted at 5. The suspensions were stirred during the equilibrium time at room temperature, and then centrifuged. The dye concentration was determined as above.

### 2.2.3. Effect of initial pH on dye removal

This effect was studied on suspensions of HT and HT-SDS in 50 or 100 mg/L of Green Bezanyl-F2B solution (solid/solution ratio = 0.5 g/L). The pH of the suspensions was adjusted to values in the range from 5 to 9. The suspensions were stirred during the equilibrium time at room temperature, and then centrifuged. The dye concentration was determined as above.

## 3. Results and discussion

### 3.1. Characterization of materials

The X-ray powder diffraction pattern of HT (Fig. 1) shows peaks at 7.58 Å ( $d_{003}$ ), 3.78 Å ( $d_{006}$ ), 2.58 Å ( $d_{012}$ ), 2.31 Å ( $d_{015}$ ), 1.95 Å ( $d_{018}$ ), 1.52 Å ( $d_{110}$ ), 1.49 Å ( $d_{113}$ ) and 1.41 Å ( $d_{116}$ ), which are similar to those reported by several authors [20–22]. However,

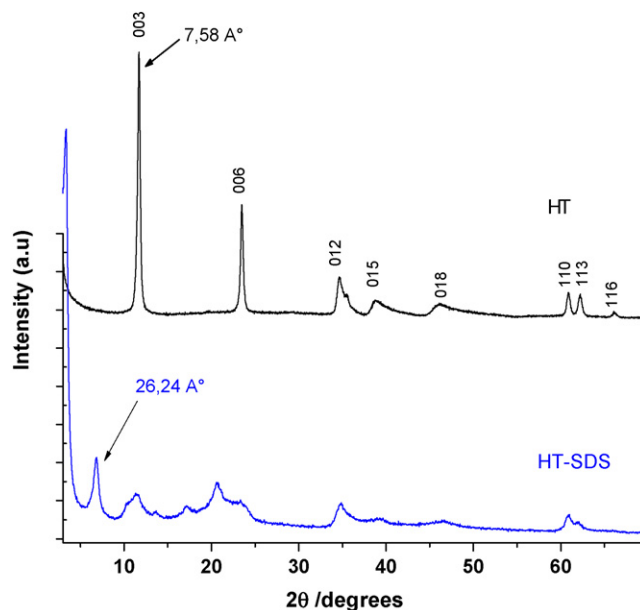


Fig. 1. X-ray powder diffraction patterns of Mg–Al hydrotalcite (HT), dodecylsulfate intercalated hydrotalcite (HT-SDS).

the HT-SDS diffraction patterns show a shift to 26.24 Å, indicating the intercalation of the surfactant ions in the hydrotalcite interlayer. The same order of magnitude for these  $d_{003}$  values has been reported in the literature [23–25].

The infrared spectrum of HT (Fig. 2) shows the characteristic absorption bands of a hydrotalcite, particularly a broad band at 3421 cm $^{-1}$  (due to the interlayer water molecules) with a shoulder near 3200 cm $^{-1}$  (due to the H-bonded stretching vibration). The weak peak at 1637 cm $^{-1}$  can be assigned to the H $_2$ O bending vibration of interlayer water. The strong peak at 1359 cm $^{-1}$  can be assigned to the vibration of carbonate species. The bands in the range of 500–700 cm $^{-1}$  are attributed to metal–oxygen–metal stretching.

The postulated surfactant (SDS) intercalation in the hydrotalcite interlayer is confirmed by the infrared spectra in Fig. 2. The presence of dodecylsulfate ion in the sample is evidenced by the C–H stretching vibration bands 2955, 2919 and 2851 cm $^{-1}$  and a C–H bending vibration band at 1467 cm $^{-1}$ . The sulfate S=O stretching vibration bands at 1215 and 1065 cm $^{-1}$  were also observed. Besides

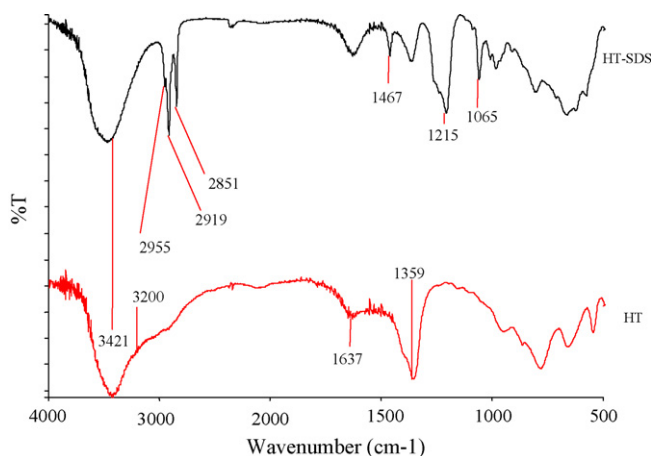


Fig. 2. FT-IR spectra of Mg–Al hydrotalcite (HT), dodecylsulfate-intercalated hydrotalcite (HT-SDS).

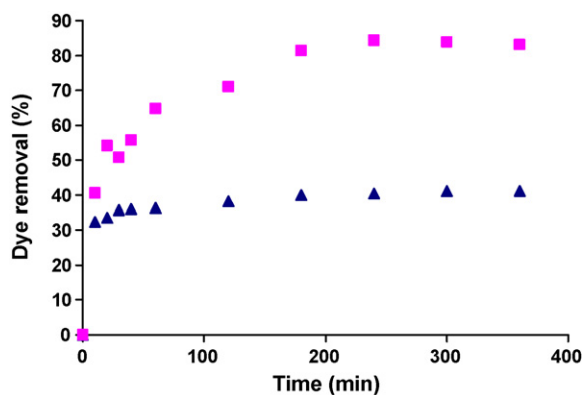


Fig. 3. Effect of contact time on percentage of dye removal by HT (▲) and HT-SDS (■).

the hydroxyl vibration bands of hydrotalcite and the C–H vibration bands were observed in the spectrum of HT-SDS. The two spectra of Fig. 2 are similar to those reported in the literature for the same materials [26,27].

### 3.2. Study of Green Bezanyl-F2B removal with HT and HT-SDS

#### 3.2.1. Contact time effect on dye sorption

Plots of Fig. 3 show the percentage dye removal with hydrotalcite (HT), dodecylsulfate intercalated hydrotalcite (HT-SDS) versus contact time. Sorption equilibrium was reached after two hours with HT and after more than three hours with HT-SDS. Moreover the percentage dye removal by HT-SDS and HT were respectively 84 and 40%.

The percentage removal ( $R\%$ ) of dye was calculated using the following relationship:

$$R\% = \frac{C_i - C_t}{C_i} \times 100 \quad (1)$$

where  $C_i$  (mg/L) is the initial concentration of the dye solution,  $C_t$  (mg/L) is the concentration of the dye solution at the adsorption equilibrium at time  $t$ . These preliminary results led us to deduce that the intercalation of SDS in the hydrotalcite results in a better acid dye sorption capacity. This investigation is only devoted to the HT-SDS which is more efficient than HT in the acid dye removal process.

#### 3.2.2. Kinetic modeling

The sorption kinetics is an important aspect of pollutants removal process control. The Lagergren's first order kinetic model [28] and the Ho's pseudo second order model [29] are the most frequently used in the literature to describe the sorption process.

The Lagergren's first order model [28] is expressed by the equation:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} \times t \quad (2)$$

where  $q_t$  and  $q_e$  (mg/g) are the amounts of dye adsorbed per unit mass of the adsorbent at time  $t$  and at equilibrium, respectively and  $k_1$  ( $\text{min}^{-1}$ ) is the first order rate constant of adsorption.

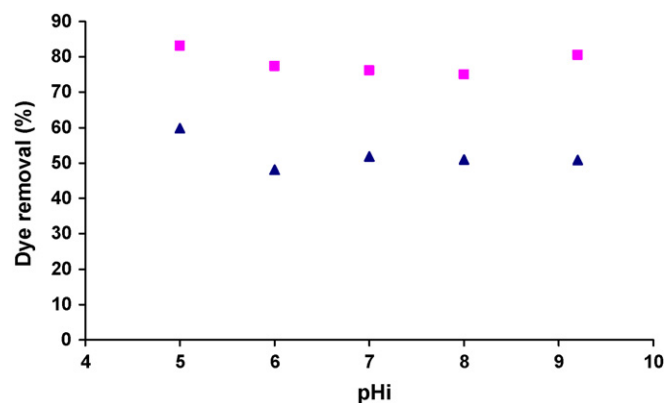


Fig. 4. Effect of initial solution pH on dye removal from aqueous solutions by HT (▲) and HT-SDS (■).

Eq. (2) shows a linear relationship between  $\log(q_e - q_t)$  and  $t$ . Linear regression calculations allow to obtain the values of the rate constant  $k_1$  and the equilibrium removal capacity  $q_e$ . These parameters ( $k_1$ ,  $q_e$ ) were calculated for the sorption of Green Bezanyl-F2B on HT and HT-SDS and are reported in Table 1.

The calculated values from the first order kinetic model show that the experimental results fit this model with determination coefficient values of 0.964 and 0.946 for HT and HT-SDS respectively. Moreover large differences between experimental and calculated values of the equilibrium sorption capacities are observed (Table 1). We conclude that the first order kinetic model is not adequate to describe the sorption process. Due to its good correlation with the experimental results, the more recent pseudo second order model has been extensively used by several researchers in the same field [29]. This model is expressed by equation (3):

$$\frac{t}{q_t} = \frac{1}{q_e} t + \frac{1}{k_2 q_e^2} \quad (3)$$

where  $k_2$  is the second order rate constant ( $\text{g mg}^{-1} \text{min}^{-1}$ ).

The plots of  $t/q_t$  versus  $t$  should give straight lines where slopes and intercepts are respectively  $1/q_e$  and  $1/(k_2 q_e^2)$ . The values of the rate constant  $k_2$  and of the equilibrium sorption capacity  $q_e$  are calculated from these parameters. The calculated  $k_2$  and  $q_e$  values and the corresponding linear regression determination coefficient  $R^2$  values for 50 mg/L (HT) and 100 mg/L (HT-SDS) initial concentrations are also reported in Table 1. Good correlation is observed between experimental data and the pseudo second order kinetic model with determination coefficient values higher than 0.99. Values of the rate constant  $k_2$  are of the same order of magnitude as those observed in the literature [30].

For both materials, the differences between the experimental and calculated values of the equilibrium removal capacity were lower than 3%.

#### 3.2.3. pH effect on dye removal

Fig. 4 shows the plots of HT and HT-SDS sorption capacity versus pH ranging from 5 to 9. It was observed that the sorption capacity is practically constant in this pH range. Similar observation was reported in the literature with other sorbents [31,32]. This indi-

Table 1

Comparison of experimental and calculated values for the first and the second order sorption rate constant

Sorbents	$q_{e,\text{exp}}$ (mg/g)	$k_1$ ( $\text{min}^{-1}$ )	$q_{e,\text{cal}}$ (mg/g)	$R^2$	$k_2$ ( $\text{g mg}^{-1} \text{min}^{-1}$ )	$q_{e,\text{cal}}$ (mg/g)	$R^2$
HT	40.48	0.0106	8.81	0.964	0.0055	41.66	0.999
HT-SDS	168.76	0.0143	98.80	0.946	0.0003	172.41	0.991

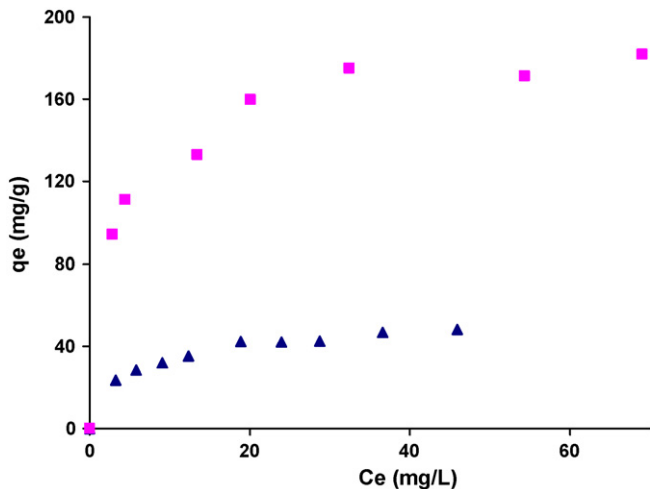


Fig. 5. Adsorption isotherms of dye removal on different sorbents HT (▲) and HT-SDS (■).

cates that HT and HT-SDS remove the anionic dye with the same efficiency in this range of pH values.

### 3.2.4. Sorption isotherms

The equilibrium sorption experimental data obtained in this study were analyzed using the commonly used Freundlich and Langmuir isotherm models.

The empirical Freundlich model which is known to be satisfactory for low concentrations is expressed by the equation:

$$q_e = k_F \times C_e^{1/n} \quad (4)$$

where  $q_e$  is the equilibrium sorption concentration of solute per gram of adsorbent (mg/g),  $C_e$  is the equilibrium aqueous concentration of the solute (mg/L);  $k_F$  and  $n$  are Freundlich constants which are related to adsorption capacity and intensity of adsorption.

Eq. (4) can be linearized in its logarithmic form which enables the determination of Freundlich constants as below:

$$\log q_e = \frac{1}{n} \times \log C_e + \log k_F \quad (5)$$

The Langmuir isotherms model is described by the following equation:

$$q_e = \frac{Q_{\max} b C_e}{1 + b C_e} \quad (6)$$

where  $q_e$  (mg/g) is the amount of dye removed per gram of sorbent,  $Q_{\max}$  (mg/g) is the maximum sorption capacity,  $C_e$  (mg/L) is the dye concentration in the equilibrium solution, and  $b$  (L/mg) is the Langmuir constant related to the adsorption energy. For convenience of plotting and determining the Langmuir constants the Langmuir equation can be rearranged to linear form as below:

$$\frac{C_e}{q_e} = \frac{C_e}{Q_{\max}} + \frac{1}{Q_{\max} b} \quad (7)$$

Sorption isotherms of Green Bezanyl-F2B retention by HT and HT-SDS are shown in Fig. 5. It is evident that sorption capacities of HT-SDS are much larger than those of HT. The adsorption capacity of HT is less than that HT-SDS because the latter is more hydrophobic. These results suggest that HT-SDS may be used to remove anionic dyes of relatively high concentrations, whereas HT can only be used to remove anionic dyes of low concentrations [32]. Figs. 6 and 7 show Freundlich and Langmuir isotherms for the Green Bezanyl-F2B sorption on HT and HT-SDS. A correct fit of the Freundlich model is observed but the Langmuir model fits better

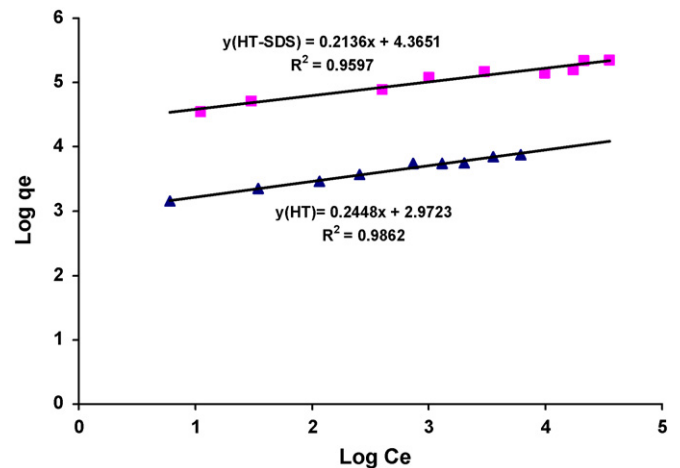


Fig. 6. Freundlich isotherms of dye removal from aqueous solutions with HT (▲) and HT-SDS (■) (solid solution ratio = 0.5 g/L).

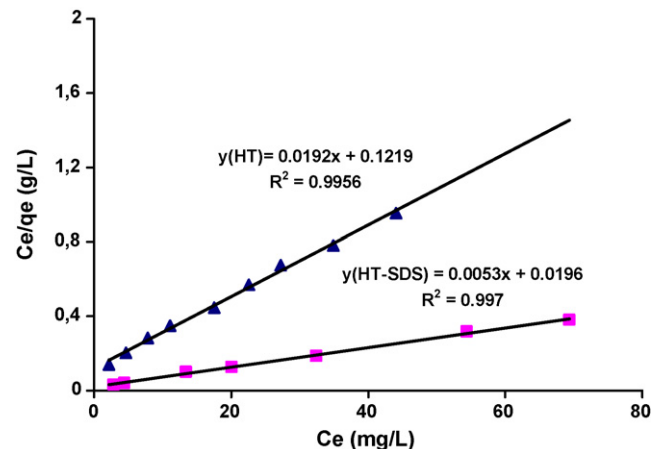


Fig. 7. Langmuir isotherms of dye removal from aqueous solutions with HT (▲) and HT-SDS (■) (solid solution ratio = 0.5 g/L).

Table 2

Langmuir and Freundlich constants for dye sorption on HT and HT-SDS

Sorbents	Langmuir constants			Freundlich constants		
	$Q_{\max}$ (mg/g)	$b$ (L/mg)	$R^2$	$k_F$	$n$	$R^2$
HT	52.08	0.157	0.995	19.83	4.08	0.986
HT-SDS	188.68	0.270	0.997	78.65	4.68	0.959

the experimental results. The determination coefficient values for this model were higher than those of Freundlich model (0.995 and 0.997 for the Langmuir model against 0.986 and 0.959 with HT and HT-SDS respectively). The calculated parameters of Freundlich and Langmuir are reported in Table 2. The maximal sorption capacity  $Q_{\max}$  of HT-SDS is much larger than that of HT (188.68 mg/g against 52.08 mg/g) while the value of the parameter  $b$  related to adsorption energy is higher for HT-SDS than for HT (0.270 L/mg against 0.157 L/mg).

## 4. Conclusion

In this study, a dodecylsulfate ion was intercalated in the interlayer space of a hydrotalcite. HT and HT-SDS were prepared, characterized by X-ray and FT-IR diffraction analysis, with the products evaluated for their ability to adsorb an acid dye (Green



Bezanyl-F2B) from aqueous solutions. The results indicate that HT-SDS are better adsorbents for the removal of anion pollutants than HT. The kinetic and sorption data fitted well the second order kinetic model and the Langmuir model respectively with good values of the determination coefficient. The sorption process was found to be pH independent.

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