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Removal of Remazol Brilliant Blue R From Aqueous Solution by Adsorption Using a Calcined Layered Double Hydroxides [Zn_2-Al-CO_3]

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- ✓ Isotherms;
- ✓ Reconstruction
- ✓ Intercalation

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ABSTRACT

In this work, we studied adsorption of a textile dye namely Remazol Brilliant Blue R (RBBR) by calcined layered double hydroxide (CLDH). This material was synthesized by the coprecipitation method of constant pH 10 with a metal ratio Zn/Al = 2 and it is calcined at a temperature of 500°C for 5 h. These materials have great potential for removing effluent pollutants. It was aimed to study the performance of a calcined [Zn_2-Al-CO_3] for the removal of RBBR and optimizes experimental parameters such as contact time, pH and mass ratio RBBR/CLDH. The retention kinetics follows the pseudo-second order model which involves chemisorption mechanism. The adsorption isotherms are of type H and are in good agreement with the Langmuir model. The results of XRD and IR indicate that the retention is governed by adsorption of the textile dye to the available sites on the surface and by intercalation between the LDH layers reconstructed. The removal of RBBR in anionic form by CLDH gave satisfactory results, the retention is total (100%) when the mass ratio RBBR/CLDH is between 0.20 and 0.40 and the retention capacity reached 1000 mg/g.

1. Introduction

In recent years, the work focuses on environmental toxicology because of the misuse of chemicals and serious consequences may beings to water resources [1]. For this, we must have techniques and effective ways to fight against this scourge and protect the quality of water resources [2]. The textile industry consumes considerable amounts of water during dyeing and finishing operations, containing high concentrations of dyes [3]. A lot of synthetic dyes are used for many purposes such as textile, paper, food, and pharmacy... There are more than 100.000 dyes produced in the world. Approximately, 10-15 % of synthetic dyes are directly released to environment as effluent [4]. The water pollution is the result of the discharge of wastewater without treatment or with an insufficient level of treatment [5]; this causes degradation of the ecosystem [6]. The problem is even more serious in the case of industrial effluents which have a toxic character much more pronounced. The presence of dyes in wastewater poses severe problems to the environment [7,8], due to their synthetic nature, poor degradability and difficulty to treat by conventional methods. Dyes are stable to light and heat and some are proven to be toxic to microorganisms [9], having a negative impact upon photosynthetic activity in aquatic systems [10]. Treatments aimed to eliminate dyes from a wastewater such as textile dye Remazol Brilliant Blue R (RBBR) which is one of the most important dye used in textile industries and also toxic, carcinogenic and recalcitrant [11-13].

Current research is then directed towards low-cost treatment processes using materials such as clays, bentonite [14-16], activated carbon, phosphate materials and other adsorbent materials such as layered double hydroxides(anionic clay) [17]. The problem of wastewater treatment is mainly the management of waste generated in wastewater treatment plants or sewage treatment plants [18]. For this, our choice fell on a layered double hydroxide (LDH) because it does not generate sludge [19], low cost synthesis, recyclable and non-toxic

[20,21]. LDH are considered among the most promising inorganic materials [22], their general formula can be normally expressed as $[M^{II}_{1-x}M^{III}_{x}(OH)_2]^{x+}[(A^{n-}_{x/n}), mH_2O]$, where M^{II} and M^{III} are divalent and trivalent metal cations that occupy octahedral sites in the hydroxide layers, A^{n-} is an exchangeable anion located in the interlayer space between two hydroxide layers (Fig. 1) [23].

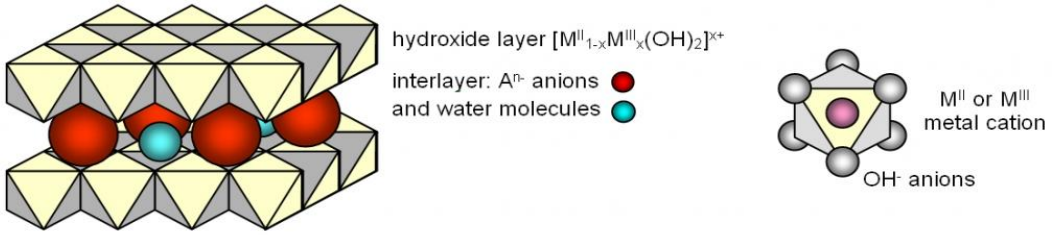

Figure 1. Schematic representation of LDH phase.

The aim of this work is to optimize the parameters influencing the adsorption of the RBBR dye by a calcined matrix of $[Zn_2\text{-}Al\text{-}CO_3]$ (CLDH), such as pH, contact time, initial concentration of RBBR and mass ratio (RBBR/ CLDH).

2. Materials and methods

2.1.Adsorbent preparation and characterization by XRD and IR

Concerning the synthesis of the precursor, we used the pH constant at said coprecipitation method [24]. About 250 ml of distilled water are placed in a reactor and maintained under magnetic stirring at 25 °C. The optimization of various factors influencing the coprecipitation was carried out. Phase $[Zn_2\text{-}Al\text{-}CO_3]$ having relatively better crystallinity was prepared by coprecipitation at pH 10 from a mixture of metal salts ($ZnSO_4$, $7H_2O$) 0.5M (Sigma-Aldrich) and ($Al(NO_3)_3$, $9H_2O$) 0.5M (Sigma-Aldrich). The pH was kept constant by adding a solution containing 0.75M of Na_2CO_3 and 0.25M of NaOH, as a metal ratio Zn/Al = 2 and a ripening time of 72 hours under moderate agitation, then this material is calcined at 500 °C during 5 h.

Characterization of the solid obtained by XRD (Fig. 2) showed that the phase corresponds to a pure LDH. The solid consists of a well-crystallized single phase with large constituting crystallites. The lattice parameters refined on the hexagonal setting with a Rhombohedral symmetry (space group: R-3m). The cell parameters, obtained by refinement according to the least square method, are: **a** = 0.306 nm; **c** = 2.292 nm and **d** = **c** / 3 = 0.764 nm.

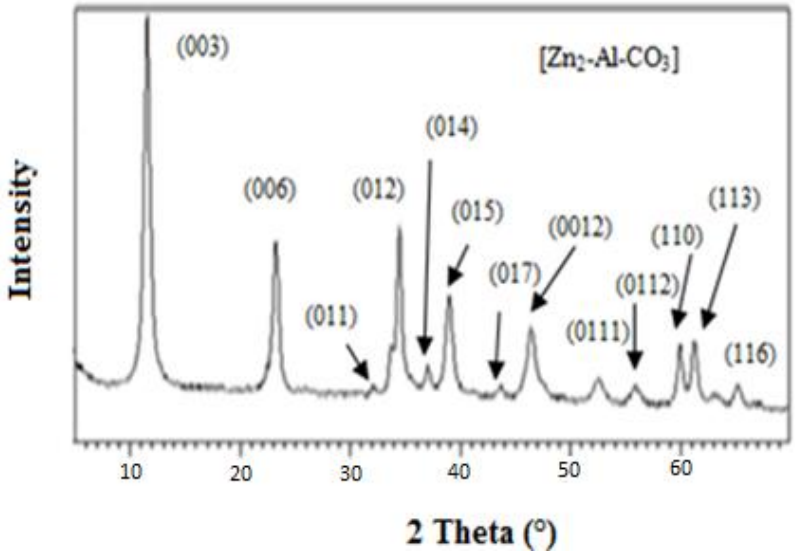

Figure 2. XRD pattern of $[Zn_2\text{-}Al\text{-}CO_3]$ precursor.

The IR spectrum of the interlcalated phase by carbonate anions (Fig. 3) shows bands at 3470 and 1630 cm^{-1} corresponding respectively to the valence vibrations of hdroxyl groups of the brucite layers (ν_{OH}) and vibration deformation molecules of interlayer water (δ_{H2O}). The carbonate anion free said belongs to D_{3h} symmetry group and only ν_2 vibration, ν_3 and ν_4 respectively to 879, 1429 to 1492 and 706 cm^{-1} are active in the infrared in a

reference compound such as $CaCO_3$ [25]. In [Zn_2-Al-CO_3], the shoulder at 1550 cm^{-1} and the appearance in the vicinity of 1150 cm^{-1} band of the ν_1 show that some carbonate anions may exist in a different symmetry, suggesting that they are not inserted between the sheets but rather adsorbed on the surface of monocrystallites [26].

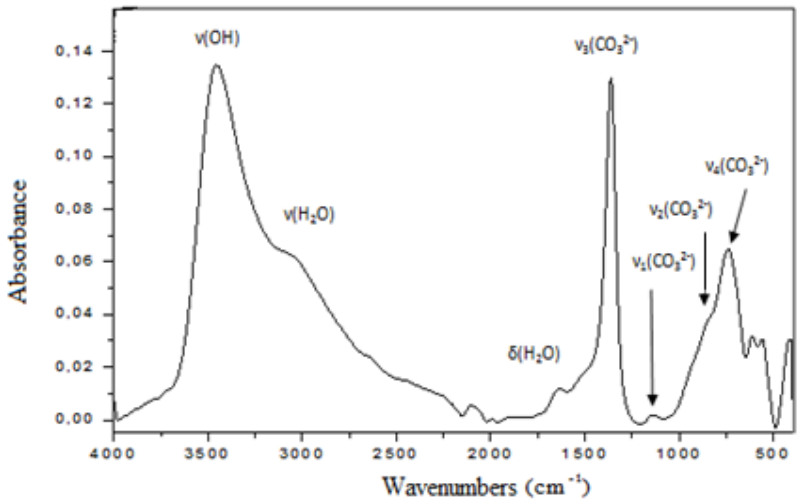

Figure 3. IR spectrum of [Zn_2-Al-CO_3] precursor.

2.2. Retention experiments

Amounts of 30, 50 or 60 mg of [Zn_2-Al-CO_3] calcined at 500 °C were suspended with 100 ml of RBBR solutions at variable initial concentrations between 0.1 and 0.8 g /L. After filtration, the solids obtained were dried at room temperature for 48h and then analyzed by XRD and IR spectroscopy. The supernatants were recovered and the residual dye concentration was determined by UV-Vis spectroscopy. The absorbance was measured at 594 nm on a Jenway 6300 spectrophotometer.

The RBBR quantity retained by a calcined [Zn_2-Al-CO_3], Q, was calculated as the difference between initial and equilibrium (final) concentrations of the dye in solution (C_i and C_e, respectively) by mass of the adsorbent (m) in the volume of solution (V) using the following equation:

$$Q = (C_i - C_e).V/m$$

2.3. Analytical techniques

The XRD equipment used was a Siemens D 501 diffractometer. Samples of unoriented powder were exposed to copper K_α radiation (λ= 0.15415 nm). Measurement conditions were 2h, range 5-70°, step size: 0.08-2h, and step counting time: 4s. Data acquisition was effective on a DACO-MP microcomputer. Unit cell constants were calculated using a least squares refinement.

Absorbance IR spectra were recorded on a JASCO6300 PC spectrophotometer, at a resolution of 2 cm^{-1} and averaging over 100 scans, in the range 400–4000 cm^{-1}.

3. Resulats and Discussion

3.1. Adsorption equilibrium

Preliminary adsorption experiments were conducted to determine the optimal conditions for the retention of RBBR on CLDH regarding the pH value, contact time (t_c), initial concentration (C_i) of adsorbate and the mass ratio RBBR/CLDH.

3.1.1. Effect of pH

The influence of pH on the reconstruction phenomenon was examined using 50 mg of CLDH and an initial concentration of RBBR solution equal to 200 mg/L. Then, the suspensions were adjusted to pH values between 4 and 10. The results of the retention dye RBBR by CLDH in function of pH of the solutions are shown in Figure 4. It may be noted that there is a maximum retention for pH values between 5 and 9 with a slightly higher value for pH 5. For pH < 5, the retention of RBBR decreased can be attributed to a partial dissolution of the matrix by hydrolysis [27]. At pH > 9, the low value of the CLDH adsorption capacity could be the result of

competition with the carbonate anions (CO_3^{2-}) to the adsorption sites; this suggests that the carbonate ions exhibit high affinity for the LDH matrix [28]. The choice of a pH 5 will be applied thereafter in this work.

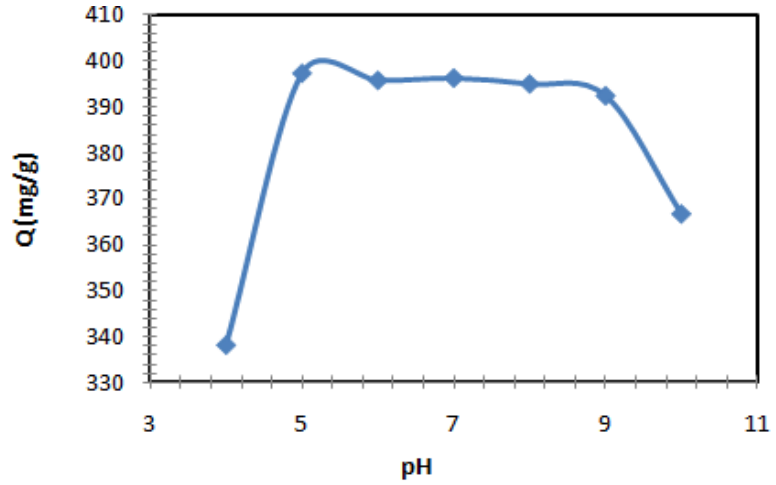

Figure 4. Amount of RBBR retained by a calcined [Zn_2-Al-CO_3] at different pH values.

3.1.2. Effect of contact time

The variations in quantities of RBBR retained by 50 mg of CLDH, depending on the contact time for three initial concentrations of the dye are shown in Figure 5.

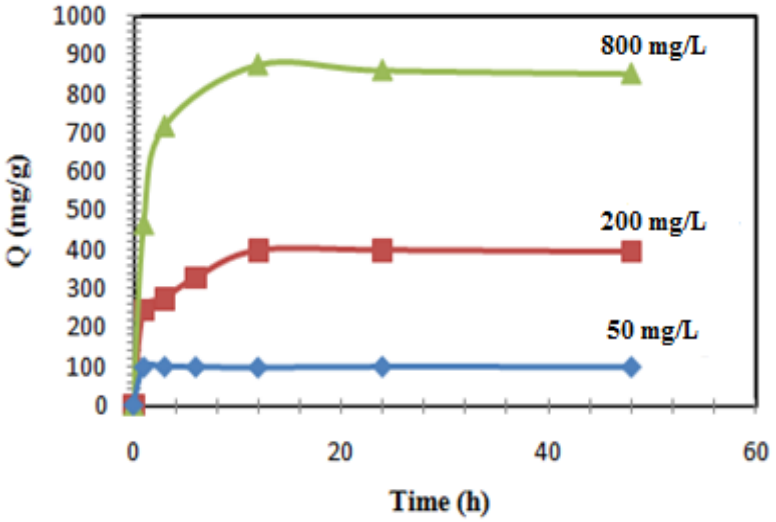

Figure 5. Amount of RBBR sorbed by 50 mg of a calcined [Zn_2-Al-CO_3] versus contact time at three different RBBR initial concentrations.

From Figure 5 it is noted that the adsorption rate is fast at the beginning of the process and becomes slower during the stirring time and equilibrium is reached after 12 h for both large concentrations and 3 h for C_0 = 50 mg/L. To be sure that the equilibrium state is reached, 24 hours was applied.

3.2.Adsorption Kinetics **(***Pseudo-first and pseudo-second order models)*

Two models in Figure 6 were attempted to test the kinetics of interactions with CLDH and RBBR anions are expressed as follows:

$$\ln (Q_e - Q_t) = \ln Q_e - k_1 t \quad : \quad \text{pseudo-first order} \quad [29]$$

$$t/Q_t = 1/(k_2 Q_e^2) + (1/Q_e)\, t \quad : \quad \text{pseudo-second order} \quad [30]$$

With Q_e the amount retained at equilibrium (mg/g), Q_t an amount retained at a given time (mg/g) and k_1 is a kinetic constant of pseudo-first order (h^{-1}) and k_2 is a kinetic constant of the pseudo-second order (g/mg/h).

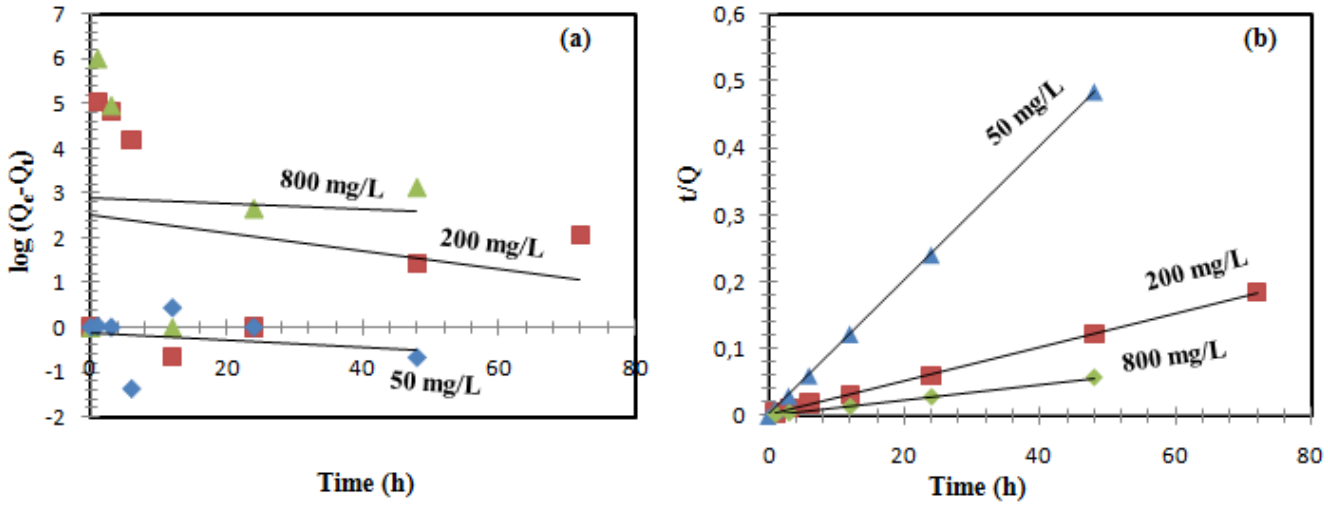

Figure 6. Pseudo-first order model (a) and pseudo-second order model (b) for the kinetic of RBBR adsorption onto CLDH.

The results linearization both models are summarized in Table 1. It can be said that the RBBR retention kinetics by CLDH is in good agreement with the model of the pseudo-second order which involves chemisorption mechanism.

Table 1. Parameters of kinetic models of pseudo-first and pseudo-second order and the maximum adsorption amounts of RBBR.

	Pseudo-first order					**Pseudo-second order**				
C_0 (mg/L)	**Equation**	**k_1** (h^{-1})	**$Q_{e_{th}}$** (mg/g)	**$Q_{e_{exp}}$** (mg/g)	**R^2**	**Equation**	**k_2**(g/mg/h)	**$Q_{e_{th}}$**(mg/g)	**$Q_{e_{exp}}$** (mg/g)	**R^2**
50	$Log(q_e-q_t)$ = -0,0076.t -0,1221	0,0076	0,885	99,74	0,121	t/q_t = 0,0101.t +0,005	0,02	99,01	99,74	1
200	$Log (q_e-q_t)$ = -0,02.t +2,5211	0,02	12	397,18	0,051	t/q_t = 0,0025.t +0,0012	0,003	400	397,18	0,999
800	$Log (q_e-q_t)$ = -0,006.t +2,8877	0,006	17,95	860,51	0,002	t/q_t = 0,0012.t +0,0005	0,002	833,33	860,51	0,999

3.3. Study of adsorption isotherms

The study of adsorption isotherms aims to determine the retention capacity of the RBBR dye by [Zn_2-Al-CO_3] calcined and uncalcined and the type of adsorption mechanism. The study that we conducted was carried out with different initial concentrations of RBBR and different LDH masses equal to 30, 50 and 60 mg, calcined at 500 ° C and a mass of 50 mg of uncalcined LDH. The results are represented in Figure 7.

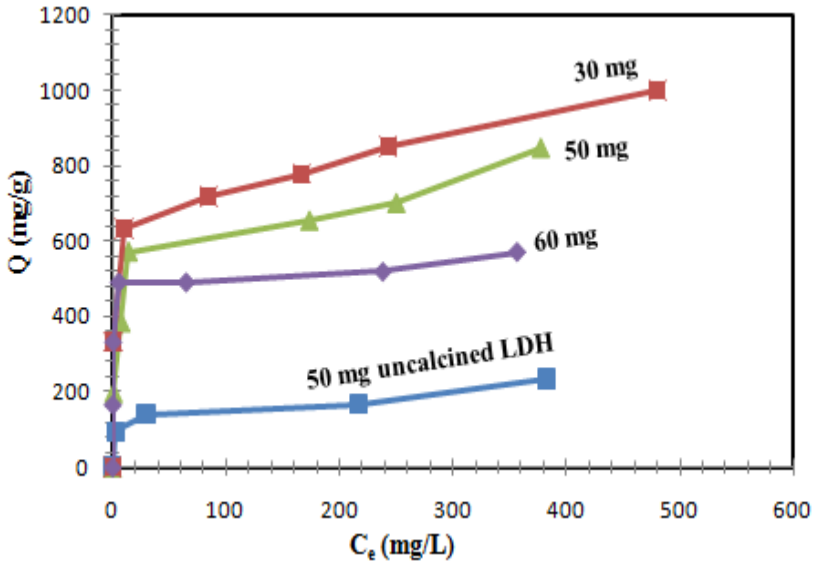

Figure 7. Adsorption isotherms of RBBR onto LDH and CLDH at different adsorbent doses.

The transformed linear Freundlich and Langmuir adsorption isotherms models are shown in Figure 8.

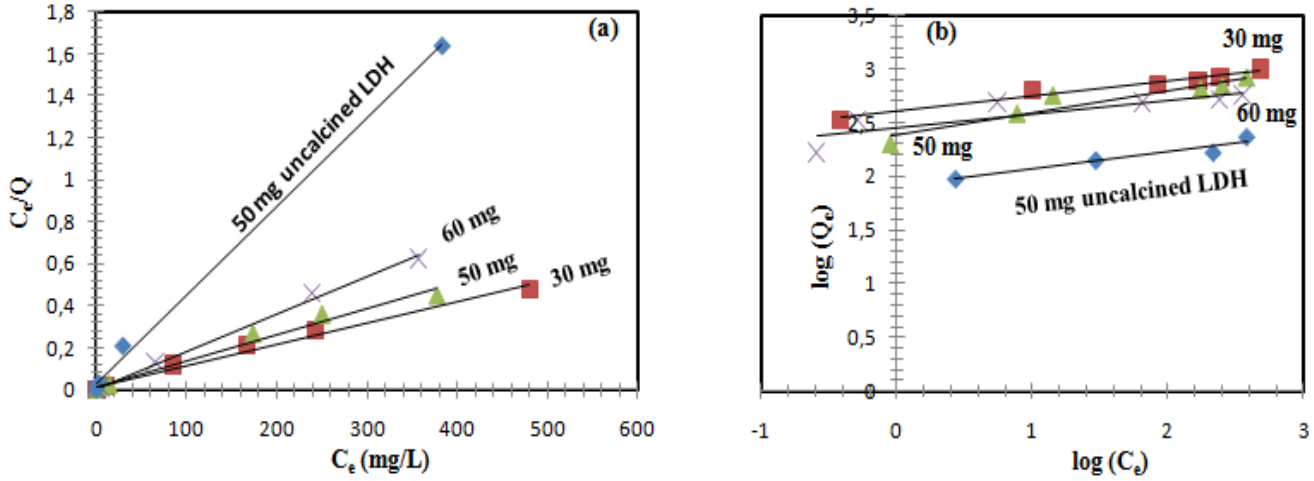

Figure 8. Linearized Langmuir (a) and Freundlich (b) isotherms plots of RBBR adsorption by a calcined and uncalcined LDH.

From the results of table 3, it can be said that the four isotherms are in good agreement with the Langmuir model.

Table 2. Langmuir and Freundlich isotherms model constants and correlation coefficients for retention of RBBR onto calcined and uncalcined LDH (un).

	Langmuir isotherm			**Freundlich isotherm**		
m_{CLDH} (mg)	**Q_m** (mg/g)	**K** (L/mg)	**R^2**	**K_f** (mg/g)	**n**	**R^2**
30	1000,00	0,061	0,986	13,492	7,067	0,961
50	833,33	0,093	0,982	10,833	4,764	0,898
60	555,56	0,346	0,997	11,711	7,924	0,715
50 (un)	238,09	0,129	0,998	6,715	6,192	0,927

3.4. Characterization of the reconstructed phase

3.4.1. Study by X-ray diffraction

XRD pattern for [Zn_2-Al-RBBR] reconstructed with a mass ratio RBBR/CLDH equal to 0,8 are shown in Figure 9. The XRD pattern of CLDH (Fig. 9b) shows that layered structure of the original LDH was completely destroyed and indicates only mixed oxides peaks [31,32], suggesting an almost total decomposition of the original LDH and elimination of most interlayer carbonate anions and water. After RBBR adsorption by CLDH, the layered structure of the recovered products was reconstructed (Fig. 9c) because of the "memory effect" [33]. The diffractogram X obtained after retention of dye indicate that this phase correspond to hydrotalcite-like materials. However, the crystallinity of reconstructed phase is lowered; this is detected by peak broadening and the decrease of their intensities. The phase obtained has a slight amorphousness [34-36]. The presence of the lines at 61° in 2θ (line (110)) confirms the reconstruction of a LDH phase (hydrotalcite). Also the presence of the line (003) and its displacement towards the low values 2θ giving interlamellar distance on the order of 1 nm (**d**=1 nm), confirms the existence of such LDH phase intercalated by the textile dye RBBR.This value of the interlamellar distance allows us to confirm that the RBBR anion intercalated between LDH sheets adopt a parallel arrangement to these latter.

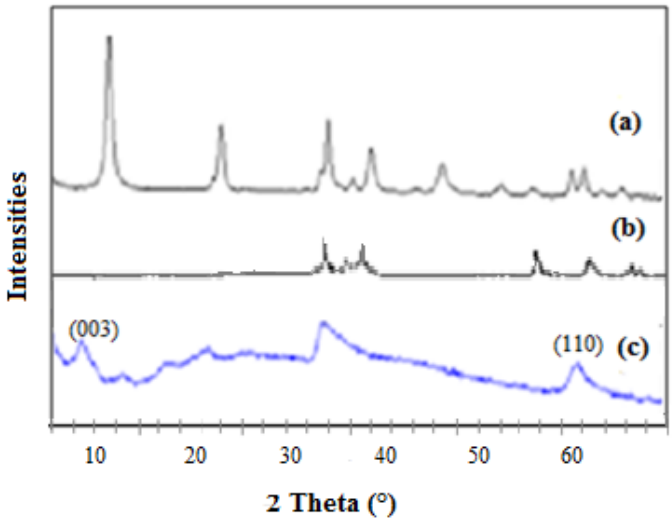

Figure 9. XRD of the matrix LDH (a), the matrix LDH calcined at 500°C (b) and the phase obtained after retention (reconstruction) (c) with a mass ratio RBBR/CLDH equal to 0,8.

3.4.2. Study by Infrared spectroscopy

This technique allows demonstrating the presence of the RBBR anion and interactions which may exist with the matrix (Fig. 10). Broadband and intense to 3400 cm^{-1} corresponds to (ν_{OH}) stretching vibrations. The characteristic vibrations of RBBR located around 1600 cm^{-1} correspond to the bonds of C = C vibrations of the benzene ring. In the region between 1100 and 1260 cm^{-1}, symmetric and asymmetric vibrations of sulfonate groups are observed, proving that the textile dye RBBR was well adsorbed on the reconstructed sheets (LDH phase).

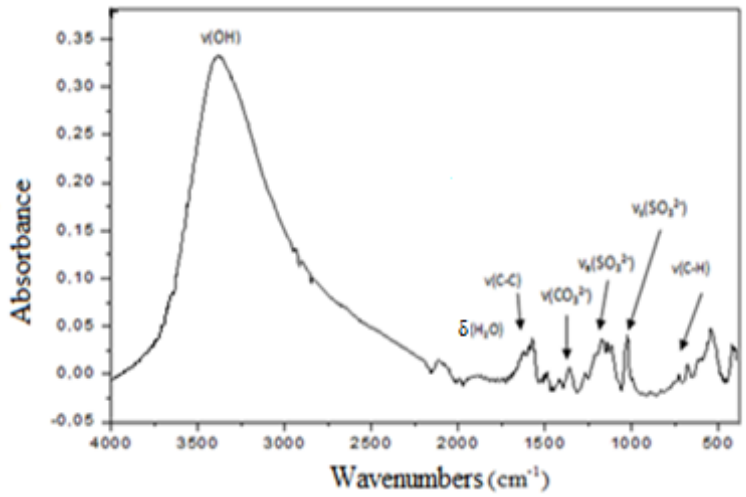

Figure 10. IR spectrum of [Zn_2-Al-RBBR] obtained after retention of RBBR by CLDH.

It is noted that the material characteristics LDH vibration (around 430 cm^{-1} and 638 cm^{-1}) were observed with apparent intensities. Bands between 700 and 800 cm^{-1} correspond to vibration (C-H) out of deformation benzene rings. The two bands at 1280 cm^{-1} and 1060 cm^{-1} correspond respectively to the antisymmetric and symmetric vibrations (S-O). According to analysis by infrared spectroscopy can be confirmed the presence of characteristic vibration bands of different active groups of RBBR [37-40].

3.5. Effect of mass ratio RBBR/CLDH

There will be the total elimination when the mass ratio RBBR/CLDH between 0.2 and 0.4. The retention capacity reached 1000 mg/g, and a removal rate of RBBR reached 100% for optimal mass ratio RBBR/CLDH equal to 0.4.These results show the efficiency of our material to the total elimination of such types of pollutants compared to other materials such as a biomaterial based on eucalyptus which reached 99.46% but the maximum retained amount of Methylene Blue dye does not exceed 137 mg/g [41]. Also other materials have been used recently but their retained quantities does not exceed 50 mg/g as hydroxyapatite for the elimination of Reactive Yellow 4 [42] or natural untreated clay for adsorption of Reactive Red 120 [43].

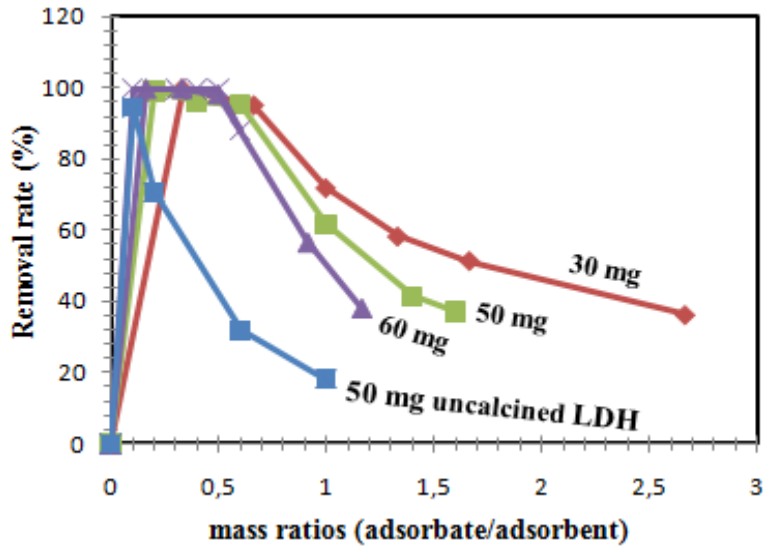

Figure 12. Curves of different mass ratio (RBBR/CLDH) for adsorption of RBBR by different masses of a calcined and uncalcined [Zn_2-Al-CO_3].

Conclusion

The aim was to study the possibility of elimination of the textile dye RBBR by a calcined [Zn_2-Al-CO_3].
So according to the results obtained we can conclude that:

- The isotherms are in good agreement with the Langmuir model, class H, which means that there is a very high affinity between the adsorbate (RBBR) and the adsorbent (CLDH).
- The values of constants K isotherms are relatively comparable, which reflects the type of interaction is the same whatever the mass of adsorbent.
- Retention is inversely proportional to the mass of the CLDH, the amount of retention increases for low masses by a calcined LDH.
- The maximum quantities adsorbed Q_m obtained by the Langmuir model are very close to those determinated experimentally.
- The amount of maximum retention reached 1000 mg/g.
- The elimination is total for optimal mass ratio RBBR/CLDH equal to 0.4. By against the dye retention by the uncalcined LDH is always less than 100% whatever the mass ratio used.
- Analysis by XRD showed that the retention of RBBR was performed by a reconstruction of a material LDH phase intercalated by the textile dye RBBR.
- The additional analysis by IR spectroscopy confirmed this result by the appearance of different characteristic vibration bands of RBBR dye.

Finally, we can move forward from these results that the material used has a relatively low cost synthetic and proved effective for the total elimination of such pollutants (dyes). It may also be recyclable by a series of anionic exchange reactions of the dye by the carbonate anion. This allows us the recovery of the pollutant and the washing waters for further reuse.

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