

REMOVAL OF THE REACTIVE REMAZOL BLACK B AND REMAZOL RED FROM AQUEOUS SOLUTIONS BY ADSORPTION ONTO TREATED RESIDUE FROM THE ALUMINUM INDUSTRY

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Abstract— In this paper, the use of a clay residue as an adsorbent of the Remazol Black B (RB) and Remazol Red RB133 (RR) is studied. The residue is generated in a large amount on the Aluminum industry, especially the Arconic Alumínio S/A, Itapissuma, Pernambuco State. The thermally treated residue behavior was studied by performing a kinetic and equilibrium experiments. The variables evaluated in all experiments were the adsorption capacity (q) and adsorption efficiency (%). Adsorption experiments were performed at environmental conditions, with 50 mL of dye solution, with controlled stirring speed, solution pH and adsorbent amount. In the kinetic studies was verified that for RB and RR, the system achieves the equilibrium state after 5 min. In the equilibrium, both reactive dyes obtained their maximum adsorption capacity (q_m) with 1.0g of adsorbent and pH 4.0 and 6.5; with q_m 0.98 mg·g⁻¹ and 6.88 mg·g⁻¹ for the RB and RR, respectively.

Keywords— Adsorption, Residue, Treatment, Dye, Remazol.

I. INTRODUCTION

On the textile industry, the process of washing and rinsing are the ones which consume the most of the water and which generate the major part of effluents. On the environmental point, the textile industry has been considered as one of great impact, due its discharges of huge volumes of rejects with high contents of organic compounds and strongly colored effluents (McKay, 1981; Alinsafi *et al.*, 2006). Dyes can be classified as a function of the charge in anionic, cationic and non-ionic compounds and are released into effluents from textile, leather, shoes polish, wood stain, paper, food and cosmetic industries, among others (Moreira *et al.* 2017). Among them, the anionic and reactive dyes are widely used as colorant in the textile industry in wool, silk, acrylic, leather or nylon (Aguiar *et al.*, 2017).

Reducing the amount of water consumed in the textile industry presents a major challenge; nevertheless recirculating treated process water is one possible option. In order that process water can be safely reused, however, it must first be treated to lessen its concentration of suspended and dissolved matter; in this issue, previous researchers have developed and studied new types of adsorbents (Cestari *et al.*, 2008). The reuse practice has been applying for different industrial sectors in the world due to its low environmental impact and its return in investment costs. Furthermore, the application of clay minerals for decolorizing water represents economic viable and locally available materials that can be used

substantially for pollution control and management (Cardoso *et al.*, 2011). Using a residue to treat another residue is a new philosophy, which can bring many benefits to those who adopt it: saving of money on conventional residue treatments and decrease on the cost of traditional methods to remove toxic substances from liquid effluents. The objective of this work is purpose a new use to the residue of the aluminum industry as an adsorbent of two reactive dyes widely used in the textile industry – the Remazol Black B and Remazol Red RB133 (denominated in this work by RB and RR, respectively).

II. METHODS

A. Reactive dyes

Textile laundries located at the west region of Pernambuco State generates large volume of textile effluents, majorly with low concentrations of dyes (5.0 – 20.0 mg·L⁻¹). The dyes used in the experiments were the Remazol Black B (RB) and Remazol Red RB133 (RR), due to their large use in the dyeing processes in the textile industries and for belonging to a reactive dye class. All the dyes were supplied by Dystar do Brasil S/A. The Fig. 1 presents the molecular structures for the RB (a) and RR (b) respectively.

Generally, the reactive dyes present some functional chromophore groups, such as azo, nitro, anthraquinone, etc. and auxiliary groups which are responsible for fixing the color on the fibers. However, the most representative and widely employed belong to the azo dyes' family. They are characterized for present one or more N = N bounded to an aromatic group. The reactive dyes also present the *solubilizing groups* in their molecular structures, represented by the sulfonic and vinyl groups, they are responsible for their solubility, their migration degree and washability; and the *reactive groups*, responsible for their reactivity with the fibers, especially the celluloses (Horrocks and Anand, 2000).

B. Residue Treatment

The fresh residue was provided by Arconic Alumínio S/A. It is composed of a mixture of two types of earth: the Celite 503, a diatomite earth usually found in Mexico and widely applied on oil filters; and the Tonsil Op-

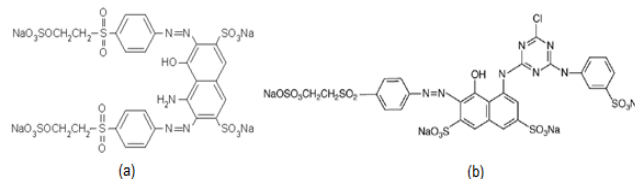


Fig. 1 – molecular structures of the dyes: a) Remazol Black B; b) Remazol Red RB133.

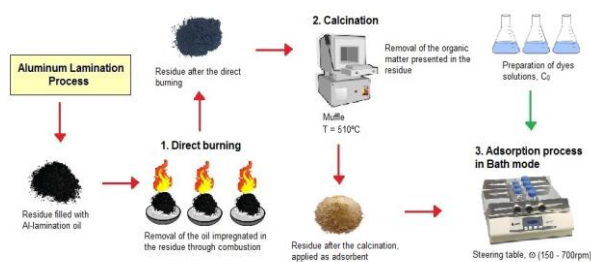


Fig. 2. Graphical scheme of the residue treatment process and its use as adsorbent of reactive dyes in a bath process.

timum, an acidified Fuller's earth, which has bleaching properties. After its use on the lamination process in the aluminum industry, the mix of filtering earths becomes steeped in oil, soaps and aluminum filings. The residue samples were submitted to a direct combustion aiming to eliminate the excess of oil. The residue was then calcined in a muffle at 510°C for 4h until the organic matter impregnated on the filtering earth was completely removed. The residue treatment process, as well as the experimental chain are presented in the Fig. 2.

C. Characterization methods

The characterization of the adsorbent after the direct burning and the calcination process, and also of its raw materials (Celite 503 and Tonsil Optimum), was made through the analysis of Specific Superficial Area through the Brunauer, Emmet and Teller (BET) Method, realized by a Specific superficial area analyzer model Quantachrome Nova 100e (USA). The specific surface area (S_{BET}) and the total pore's volume (V_P) of the samples was obtained by assuming the section area of nitrogen molecule to be 0.098 nm². Microscopy analysis of the adsorbent and the raw materials has been made by using a scanning electron microscope with DES model Shimadzu SS-550 (Japan) to observe possible changes on its morphological structure after the thermic treatment. The mineralogical compositions of the raw earth and the thermally treated residue samples were determined from X-ray patterns. X-ray Diffraction (XRD) analyses were realized by using an X-ray diffractometer Rigaku Ultima (Japan).

The average particles size was measured through sifting process and it was found 44 μm (< 325 Mesh).

D. Batch Sorption Experiments

The dyes' initial concentration was established in 10 mg·L⁻¹. A volume of 1000 mL of 10 mg·L⁻¹ dye solution was prepared for RB and RR and aliquots of 50 mL of each dye solution were transferred to erlenmeyers of 125 mL. Solution pH was established at 6.5. An aliquot of 1.0 g of adsorbent were put in contact with the dye solution for 10 min under 350 rpm steering. After the contact time, the samples were then centrifuged to separate the adsorbent from the solution. The solution concentrations were measured in the UV-Vis spectrophotometer AquamatTM. The values of λ_m were obtained through a previous UV scanning of the samples of RB and RR solution, with initial concentration of 100 mg·L⁻¹. A calibration curve was built for each dye absorbance vs concentration at the range 0.11–100 mg·L⁻¹.

Afterwards, the dye removal efficiency and the adsorption capacity were calculated using a data treatment software. The percentage of dye removal (dye removal efficiency, %) and the adsorption capacity (q_A in mg·g⁻¹) were calculated using the follow relationships (Eq. 1 and 2):

Percentage of removal:

$$(\%) = \frac{C_A^0 - C_A}{C_A^0} \times 100 \quad (1)$$

Adsorption capacity:

$$q_A = \frac{(C_A^0 - C_A)V}{m} \quad (2)$$

where C_A^0 is the initial concentration (mg·L⁻¹), C_A is the equilibrium state or final concentration (mg·L⁻¹), V is the volume of the solution (L) and m is the mass of the adsorbent (g).

E. pH influence and the zero point of charge

The zero point of charge (pH_{ZPC}) was obtained to the treated residue in order to estimate the ranges where the surface is charged positively – below ZPC – and negatively – above ZPC. The point of zero charge (pH_{pzc}) of the adsorbent was determined by adding 2.5 g of the adsorbent to several Erlenmeyer flasks. The total volume of solution with the adsorbent in each flask was established at 50 mL. A range of initial pH values ($pH_{initial}$) of the solutions were adjusted from 2.0 to 12.0 by adding solutions of 0.1 mol·L⁻¹ of HCl and 0.1 mol·L⁻¹ of NaOH, which were also used to adjust the adsorbate pH. The influence of the pH rates was evaluated concomitantly with the equilibrium studies. It was obtained the equilibrium curves for each initial solution's pH value and was observed the results for the maximum adsorption capacity (q_m) and its variation according to the solution pH changes.

F. Adsorption kinetics

The studies of adsorption kinetics were applied with the objective to discover the moment which the system achieves the equilibrium state. To proceed with the experiments, the time varied between 1 to 30 min and the same volume of dye solution was used. All the experiments were conducted at environmental temperature (25°C), and using the dye solutions at their natural pH values – 6.5 for RB and RR. The adsorbent mass influence was observed in the same study through the variation of the adsorbent amount between 0.5g and 1.0 g.

G. Adsorption Isotherms

The equilibrium studies were realized by varying the dyes' initial concentration: 2–50 mg·L⁻¹, afterwards it was possible to determine the adsorption isotherm models which better describe the adsorption mechanisms. The classical adsorption isotherms which are most applied are the Langmuir's and the Freundlich's. After the experiments, all data were treated and the curves were plotted, then the experimental data were linearized to obtain the isotherm which best represent the adsorption process; the process parameters, such as the equilibrium constant (K_A) and the maximum adsorption capacity (q_m) were obtained. The adsorbent saturation is affected by the dye concentration, and at identical values of initial concentrations, the dye uptake is lower for the

coarsest particles, according to the results obtained with silica. For finer particles, on the other hand, the values did not differ much (McKay *et al.*, 1981).

The model used to describe the adsorption process studied in this paper was the Langmuir's, whose adsorption isotherm has been applied to many other real sorption processes and it has been used to explain the sorption of dyes onto various adsorbents (Ruthven, 1984). The experimental results have been fitted to the Langmuir model. The Langmuir adsorption isotherm follows the expression in the Eq. 3:

$$\frac{q_A}{q_m} = \frac{K_A C_A}{1 + K_A C_A} \quad (3)$$

where q_m is the maximum adsorption capacity or the adsorption capacity when the system achieves the equilibrium, K_A is the equilibrium constant, C_A the adsorbate concentration at equilibrium state and q_A the adsorption capacity for each concentration.

III. RESULTS AND DISCUSSION

A. Scanning Electron Microscopy with DES

The Fig. 3 shows the microphotographs of the raw material, Celite 503 and Tonsil Optimum respectively. The Fig. 3 - c) shows the residue after direct burning and after calcination in 500°C. The right images show the material after the adsorption process. It can be observed on the Fig. 3 a) to d) that after the burning, the material's structure remains the same. Furthermore, after calcination (Fig. 3-d), there was a "smoothing" on the thermally treated residue surface area.

The DES analysis showed the elements composition of the diatomaceous clay Celite 503 presented in the residue is: O (70.07%), Si (29.53%), basically due to the presence of silicon dioxide (SiO_2), which is the main component in the diatomaceous earth (Al-Ghouti *et al.*, 2009), followed by Na (0.36%) and K (0.05%); the elements which compounds the Fuller's earth Tonsil are: mostly O (68.66%), Si (21.32%), and in a less rate, Al (4.46%), C (3.23%) and Fe (1.17%). The fresh Bentonite based Fuller's earth is majorly compounded by SiO_2 (71.27%), followed by Al_2O_3 (12.56%) and Fe_2O_3 (5.40%) (Oubagaranadin *et al.*, 2007), data in accordance to the atomic wt.% summary presented at the Table 1. The results point to an increase in the oxygen content

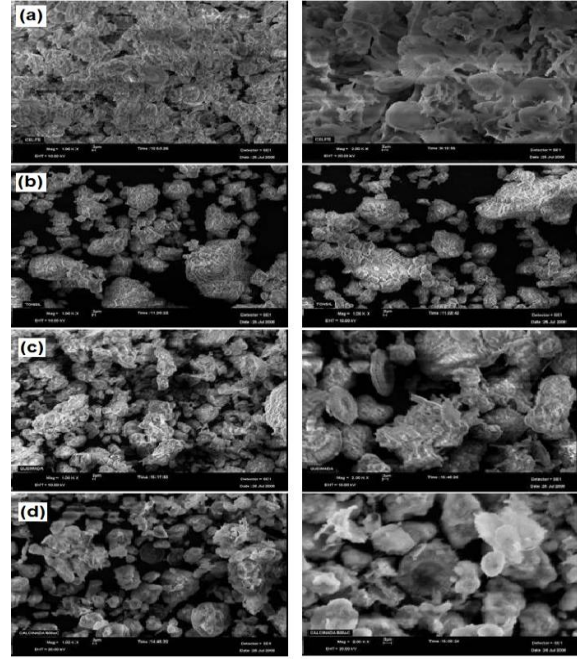


Fig. 3. a) diatomaceous earth – Celite 503; b) Fuller's earth – Tonsil Optimum; c) filtration residue after being submitted to direct burning; d) filtration residue after calcination process.

for the diatomaceous clay from 36.12% to 70.07%, dues to the oxides formed in the direct burning process and in the calcination in 500°C. The metallic elements which compound the raw materials remain practically unchanged after the direct burning and calcination process (Si, Al, Ca, K). It's possible to verify the output of the magnesium after the calcination of the Fuller's earth – Tonsil.

B. BET analysis

In the BET analysis, a significant reduction of pore's area and volume was observed after calcination in comparison with the fuller's earth Tonsil as presented on Table 2. The calcined residue presented high specific surface area in comparison to the burned residue due to an elimination of the excess of the organic material impregnated on its surface. The morphological parameters of the treated residue were: specific surface area $S_{BET} = 55.39 \text{ m}^2 \cdot \text{g}^{-1}$ and total pore volume $V_P = 2.34 \cdot 10^{-2} \text{ cm}^3 \cdot \text{g}^{-1}$. The surface area is considerable low in comparison to

Table 1. DES analysis result for the raw material, the burned and the calcinated residue (in 510°C).

VIRGIN			BURNED			CALCINED AT 510°C		
Element	Spect. Type	Atomic (%)	Element	Spect. Type	Atomic (%)	Element	Spect. Type	Atomic (%)
CELITE			CELITE			CELITE		
O	ED	36.12	O	ED	54.65	O	ED	70.07
Na	ED	0.66	Na	ED	0.26	Na	ED	0.36
Si	ED	62.76	Si	ED	45.09	Si	ED	29.53
Ca	ED	0.47				K	ED	0.05
Total		100.00	Total		100.00			100.00
TONSIL			TONSIL			TONSIL		
C	ED	5.39	C	ED	2.47	C	ED	3.23
O	ED	65.64	O	ED	51.57	O	ED	68.66
Mg	ED	1.67	Mg	ED	1.25			
Al	ED	4.45	Al	ED	7.99	Al	ED	4.46
Si	ED	19.98	Si	ED	31.76	Si	ED	21.32
K	ED	1.21	K	ED	2.03	K	ED	1.03
Ca	ED	0.29	Ca	ED	0.37	Ca	ED	0.12
Fe	ED	1.36	Fe	ED	2.57	Fe	ED	1.17
Total		100.00	Total		100.00	Total		100.00

Table 2. BET analysis results for the raw material and the treated the raw material and the treated residue.

Sample	Surface area, $\text{m}^2\cdot\text{g}^{-1}$	Pores' volume, $10^{-2}\text{ cm}^3\cdot\text{g}^{-1}$
Diatomaceous clay – Celite	2.97	0.087
Fuller's Earth – Tonsil	136.80	5.962
Burned residue	46.03	1.414
Calcined residue (in 510°C)	55.39	2.341

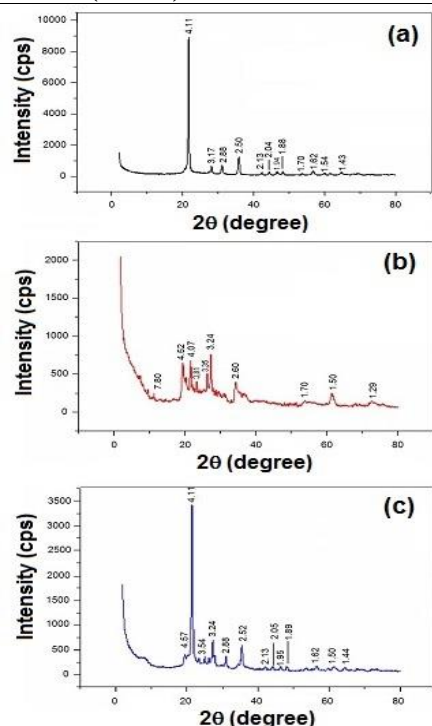
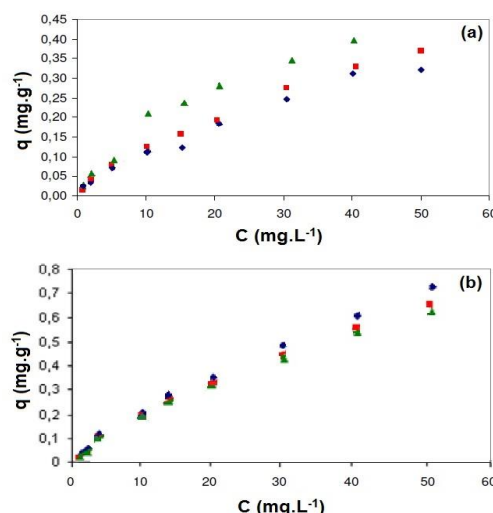


Fig. 4. X-ray diffraction graphics for: a) Celite 503 clay; b) Fuller's earth Tonsil Optimum; c) calcined residue at 500°C, used as adsorbent material.

classical adsorbents, such as activated carbon, which presents an elevated surface area, $S_{\text{BET}} = 1250 \text{ m}^2\cdot\text{g}^{-1} \sim 2400 \text{ m}^2\cdot\text{g}^{-1}$. The total volume of pores is also considerably low in comparison to the activated carbon: $V_P = 0.57 \text{ cm}^3\cdot\text{g}^{-1}$ (Oubagaranadin *et al.*, 2007). The BET specific surface area obtained by Tlili *et al.* (2012) for the Spanish virgin diatomaceous clay, showed $S_{\text{BET}} = 3.07 \text{ m}^2\cdot\text{g}^{-1}$, quite close to the Celite clay result, which compounds one of the raw materials of the residue. According to the author, the calcination process, which included heating the samples at 400 °C and 800 °C for 3h, and at 1000 °C for 1.5h, 3h and 6h, showed that the BET surface values decrease with increasing temperature of thermal treatment – this behavior was observed comparing the results for the Fuller's earth Tonsil to the burned and calcined residue samples, which decreased from $136.80 \text{ m}^2\cdot\text{g}^{-1}$ to $46.03 \text{ m}^2\cdot\text{g}^{-1}$ and $55.39 \text{ m}^2\cdot\text{g}^{-1}$, respectively.

C. X-Ray diffraction

In the XRD analysis, a peak on $2\theta = 2.17^\circ$ was observed, which characterizes the Tonsil earth, an activated diatomaceous clay, as presented in the Fig. 4 – a) and c). It can be observed on the picture 4 – c) that the residue has characteristic peaks of Celite 503 and the Tonsil Optimum, confirming the mixing of both raw materials which compound the adsorbent. It is also possible to verify that the calcination process has not caused great

Fig. 5. Equilibrium curves for a) RB and b) RR. Experiments proceeded with 1.0 g of adsorbent, adsorbate volume of 50 mL, $T = 25^\circ\text{C}$, steering velocity of 350 rpm, initial dye's concentration range: from $0.5 \text{ mg}\cdot\text{L}^{-1}$ to $50 \text{ mg}\cdot\text{L}^{-1}$. Experiments proceeded: (●) at pH 6.5; (♦) at pH 8.0; and (▲) at pH 4.0.

changes on the clay material composition, only the increase of pick's intensity in 2θ from 23° , which is due to the oxides formed on calcination, which aggregated on the material's surface, giving to the material a smooth aspect, as observed on the Electronic microscopy's photographs. Benkancem *et al.* (2016) analyzed the diatomaceous clay before and after acid treatment, it was observed essentially an amorphous silica phase, revealed by a large peak between 16° and 26° (2θ), but also presents characteristic peaks of quartz and carbonates, such calcite and ankerite.

D. Dye characterization

UV-vis scanning presented a single peak, with the following values for λ_m : 597 nm and 514 nm, for RB and RR, respectively. These data were used to build the calibration curve for each dye concentration; moreover, single peaks in UV-vis spectra point to the presence of dye monomers in the sample (Macedo *et al.* 2011), and no formation of dimmers or agglomerates was observed. DB and DR solution natural pH measured prior adsorption experiments was 6.5 ± 0.2 .

E. Effect of pH

The equilibrium curves for the three different pH rates are presented in the Fig. 5 – a) and 5 – b) for the experiments performed to the RB and RR, respectively. The maximum adsorption capacity (q_m) obtained after applying the Langmuir isotherm for the adsorption of RB were: $0.98 \text{ mg}\cdot\text{g}^{-1}$ (pH 4.0); $0.26 \text{ mg}\cdot\text{g}^{-1}$ (pH 6.5); $0.63 \text{ mg}\cdot\text{g}^{-1}$ (pH 8.0). In the same way, were obtained the following values of q_m for the RR: $6.88 \text{ mg}\cdot\text{g}^{-1}$ (pH 4.0); $0.96 \text{ mg}\cdot\text{g}^{-1}$ (pH 6.5); $2.37 \text{ mg}\cdot\text{g}^{-1}$ (pH 8.0). For the RB and RR, the values of adsorption capacity (q) were higher at pH 4.0; at high pH ranges ($\text{pH} > 6.0$) the values were not satisfactory, indicating that the adsorption is favorable when $\text{pH} < \text{pH}_{\text{ZPC}}$ (Fig. 6). It was also observed that pH changes had not interfered at the dyes solution wavelength (λ_m).

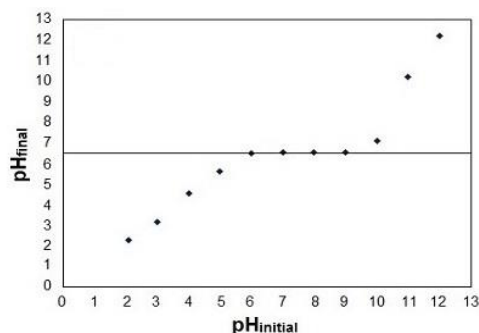
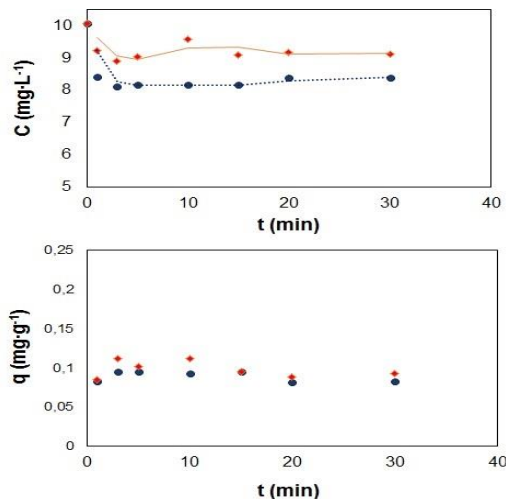
Fig. 6. Point zero of charge (pH_{ZPC}) of the thermally treated residue.

Fig. 7. Adsorption kinetics for RB: (●) experiments proceeded with 1.0 g of adsorbent; (♦) experiments proceeded with 0.5 g of adsorbents.

According to the literature data, it is rather difficult to determine an absolute point of zero charge (PZC) for the clays (Errais *et al.*, 2012). However, the Fig. 6 suggests that at pH 6.5 the residual charges of the treated residue will be zero. The hydroxyl groups present on the surface of the diatomite can gain or lose a proton; at low pH, surface sites are protonated and the surface becomes positively charged and increases the electrostatic interactions with the sulfonate groups presents in the RB (Al-Ghouti *et al.*, 2009).

F. Adsorption kinetics

By the kinetics experiments it was possible discover the time in which the adsorption system achieves the equilibrium state, in the following experimental conditions: initial concentration of $10.0 \text{ mg}\cdot\text{L}^{-1}$, volume of solution 50 mL and temperature 25°C . The Fig. 7 and 8 presents the curves of adsorption capacity (q) $\times$ time and the equilibrium concentration (C_A) $\times$ time obtained after the adsorptive process. From Fig. 7 it is possible verify that the RB achieves the equilibrium state at 5 min with $q = 0.10 \text{ mg}\cdot\text{g}^{-1}$. The RR also achieved the equilibrium at a time of 5 min with $q = 0.20 \text{ mg}\cdot\text{g}^{-1}$ (Fig. 8) The results indicate that the adsorption rate of the two dyes onto the thermally treated residue is very fast at the early stages (0–5 min) and then slows down gradually, similar behavior was observed by Al-Ghouti *et al.* (2009), in which the adsorption of basic Methylene Blue onto Jordanian diatomite reached the equilibrium was after 7 min. In both experiments, it was verified the influence

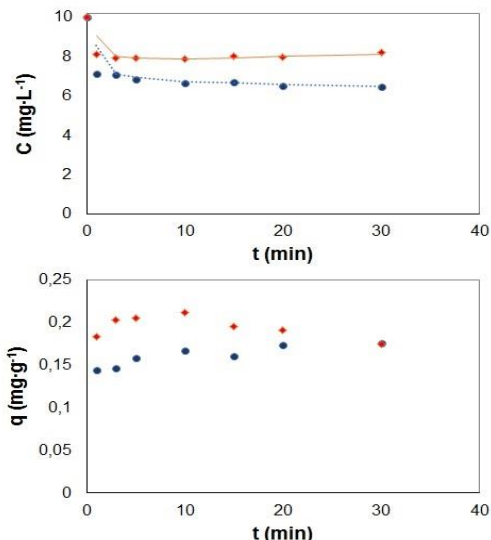


Fig. 8. Adsorption kinetics for RR: (●) experiments proceeded with 1.0 g of adsorbent; (♦) experiments proceeded with 0.5g of adsorbents.

of the adsorbent mass in the results; higher values for the adsorption capacity were achieved with a lower amount of adsorbent— 0.5g. The RR kinetic curve, shows a desorption process after 15 min; this phenomenon occurs after 20 min for the RB. Furthermore, the lower time necessary to the system achieve the equilibrium state (between 1 and 5min) is characteristic of the physisorption process, which the adsorbate sticks to the adsorbent surface through weak intermolecular interactions such as Van der Waals forces, hydrophobicity, hydrogen bonding, polarity, static interactions, dipole-dipole interactions and π - π interactions (Ruthven, 1984).

G. Equilibrium studies

The equilibrium experiments were conducted at environmental conditions of temperature and pressure and also following the operational conditions obtained at the preliminary experiments: adsorbent mass 1.0 g, stir speed 350 rpm, solution's volume of 50 mL, pH 4.0 and time of contact of 10 min. The equilibrium curve $q \times C$ and the linearized isotherm for the RB and RR is presented in the Fig. 9 and 10 respectively.

The adsorption capacity obtained by the RR was higher than the one obtained by the RB, due to a major electrostatic interaction with the sulfonate groups, presented in higher amount in the RR structure, and which gives an anionic behavior to both dyes. These groups form electrostatic interactions with the oxides and hydroxyl groups on the adsorbent surface; such groups are present in diatomaceous clay's surface, as observed by Al-Ghouti *et al.*, (2009).

Langmuir's isotherm model was applied to the process data and showed a good fit, with higher correlation coefficient (R^2). The adsorption parameters for the RB, obtained after the data linear regression, according to the Langmuir's model, were given as follows: $q_m = 0.98 \text{ mg}\cdot\text{g}^{-1}$, $K_A = 9.29 \cdot 10^{-2} \text{ L}\cdot\text{mg}^{-1}$ and $R^2 = 0.992$. Similar to RB, the adsorption parameters were achieved after the linearization according to the Langmuir model, q_m , K_A and R^2 were, respectively: $6.88 \text{ mg}\cdot\text{g}^{-1}$, $3.96 \cdot 10^{-4} \text{ L}\cdot\text{mg}^{-1}$

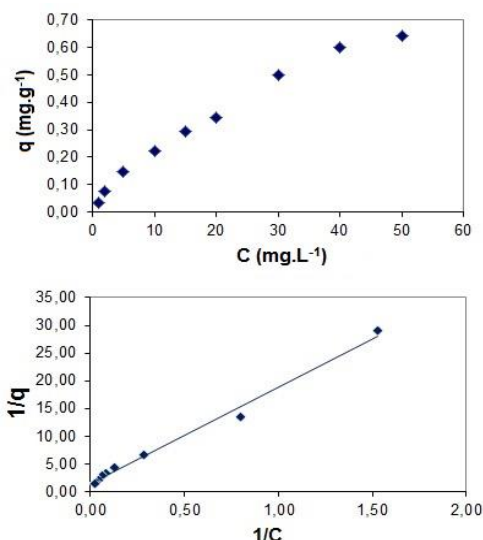


Fig. 9. a) Equilibrium curve ($q \times C$) and b) Langmuir isotherm for RB.

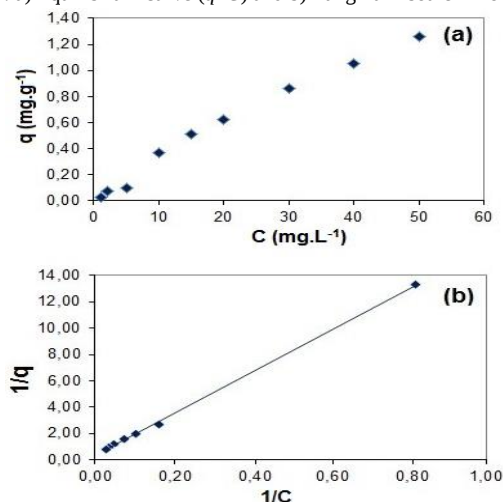


Fig. 10. a) Equilibrium curve ($q \times C$) and b) Langmuir isotherm for RR.

and 0.975. Adsorption studies of Methylene Blue onto raw diatomite showed q_m of $1.72 \text{ mg} \cdot \text{g}^{-1}$; however, this value increased after a treatment with sodium hydroxide (Jian *et al.*, 2013).

IV. CONCLUSIONS

In face of the challenge of developing new species of adsorbents for reactive dyes, a residue of the aluminum lamination was thermally treated and applied to adsorb the reactive Remazol Black B and Remazol Red RB133. Better removal results were obtained at low value of pH 4.0 and adsorbent amount, 1.0 g. In the kinetic studies, was observed that the RB and RR achieved the equilibrium state after 5 min. Values of the maximum adsorption capacity (q_m) presented the same order of magnitude of the reported for the raw diatomaceous clay. Langmuir's isotherm is the one which best describe the adsorption process. After adsorption, the treated residue can be reused in ceramic industry. Moreover, acid or basic activation can be proposed to enhance its q_m in a further study.

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