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SUGARCANE BAGASSE BIOMASS APPLIED TO THE ADSORPTION OF REACTIVE BLUE BB DYE

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Abstract

The textile industry has been contributing to the generation of wastewater containing dyes during the dyeing process. In this sector, Reactive Blue BB is a dye used on a large scale. One method widely applied to dyes wastewater treatment is adsorption. This technique is inexpensive since it can apply industrial by-products as adsorbents. In this work, the adsorption of Reactive Blue BB dye by the sugarcane bagasse, a by-product from the sugar and alcohol industry, was evaluated in batch systems. The adsorption kinetics presented an equilibrium time of 480 minutes, obtaining the best fit to the experimental data with the pseudo-second order model ($R^2 = 0.97$). The Langmuir isotherm adequately represented the equilibrium data ($R^2 = 0.97$), obtaining a maximum adsorption capacity of 4.21 mg g^{-1}. This study demonstrated that the sugarcane bagasse presents a capacity of adsorption for the evaluated wastewater, and it can be applied as an alternative adsorbent.

Keywords: adsorption, reactive blue BB, sugarcane bagasse, textile dyes

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1. Introduction

Since water is an essential resource for life on Earth, its contamination is a global concern (Siddiqui et al., 2019). Unfortunately, a variety of industries (textiles, dyes, paper, and plastic) has been generating a significant amount of organic and inorganic residues, which cause pollution and contamination when improperly released to the environment. These industrial wastewaters can contain artificial chemicals that have an impact on human health and ecological security, causing a public concern (Zhou et al., 2019a).

Dye-containing wastewater is released into the environment in significant amounts, worldwide the industries use approximately 10,000 different dyes, and annually 0.7 million tons of dyes are produced (Zhou et al., 2019b). The textile industry stands out in the environmental scenario due to a large amount of effluent generated, since approximately 10-15% of the total used dyes end up as wastewater (Li et al., 2018; Zhou et al., 2018). Consequently, approximately 50,000 tons of dyes are released without adequate treatment into water bodies annually (Siddiqui and Chaudhry, 2018).

The presence of these highly colored residues into water bodies affects the quality parameters of water, such as dissolved oxygen, biochemical oxygen demand (BOD) and chemical oxygen demand (COD) (Siddiqui and Chaudhry, 2019). Besides, the discharge of some dyes into the environment is dangerous for human and aquatic life due to carcinogens, hazardous and toxic compounds they can generate (Siddiqui and Chaudhry, 2018). Dye-containing wastewater also can contain harmful components that cause oxygen

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deficiency and have mutagenic properties (Valley et al., 2019). Moreover, the effluent color can interfere in the light transmission into the water, impairing the photosynthetic activity of the plants in the ecosystem (Juchen et al., 2018). Due to these problems, the discharge of this wastewater without adequate treatment to the environment is unacceptable.

Some techniques used to treat dyeing wastewater are not effective because dyes are resistant to degradation (Zhou et al., 2019b). Adsorption is widely used for wastewater remediation because is a technique that does not need expensive costs, does not require long procedures and sophisticated instrumentation (Chaudhry et al., 2016a; Zhou et al., 2018). The principle of the adsorption is based on the contact of a solid surface with a fluid. On this surface, the molecules of the solute are accumulated due to the unbalanced surface forces (Campos et al., 2012). For this technology to be sustainable, it is interesting that the adsorbent is easily prepared, has low cost, can be applied for various pollutants, is non-toxic, has resistance towards oxidizing and reducing agents, and has fast adsorption kinetics (Chaudhry et al., 2016b; Siddiqui and Chaudhry, 2018). In addition, it is important to evaluate the adsorption conditions, since this process is dependent on the temperature, adsorbate concentration, and adsorbent aggregation, the nature of the adsorbent and adsorbate, and the interaction between them. During the past decades, researchers have been exploring agricultural solid wastes, along with industrial by-products as adsorbents, due to the low cost and high availability (Zhou et al., 2019b). A great variety of low-cost materials from industrial or agricultural waste can be applied as adsorbents; their application is advantageous because its use as an adsorbent can aggregate value to a material not valuable to the industry. For instance, the adsorption potential of sawdust, bark, cotton fiber, rice husk, fly ash and red mud has already been studied (Zhou et al., 2019b).

The sugarcane bagasse is a by-product of the alcohol and sugar industry, it is generated after the sugarcane milling process, and lignin and cellulose have significant amounts in its composition (Gusmão et al., 2013). The use of sugarcane bagasse as an adsorbent material is an interesting alternative since this biomass is produced in high amounts. In this context, the present study aims to evaluate the adsorption of the Reactive Blue BB dye by sugarcane bagasse.

2. Material and methods

2.1. Preparation of the adsorbent

The adsorbent utilized in the experiments was the sugarcane bagasse, a fibrous by-product resulted from the sugarcane milling process. It was provided by an alcohol and sugar industry from the northwest of Paraná, Brazil. In order to prepare the adsorbent, it was rinsed with water to remove the impurities, followed by a rinsing step with distilled water, and after it was dried at 80ºC. The dried biomass was separated in a sieve, and the particle size of 2.8 mm (7 mesh) was used in the adsorption experiments.

2.2. Dye solution preparation

The Reactive Blue BB (Remazol Blue BB, Reactive Blue 220, CAS:128416-19-3) commercial dye was provided by a dyeing industry, this dye has the molecular formula $C_{22}H_{15}CuN_4Na_2O_{12}S_3$ (Boduroğlu et al., 2014). The solutions were prepared by diluting the dye in distilled water, using a stock solution of 500 mg L^{-1}. The pH was adjusted using a solution of hydrochloric acid (0.5 mol L^{-1}).

2.3. Molecular absorption spectra and analytical curve

To obtain the wavelength of maximum absorbance of the dye, molecular absorption spectra with a range from 350 nm to 800 nm were evaluated (Shimadzu UV-1601). A stock solution of the dye (1000 mg L^{-1}) was prepared, and the pH was adjusted from 1 to 10 with solutions of NaOH and HCl. The measurements were performed in a quartz cuvette using an aliquot of 4 mL of the dye solution.

To determine the analytical curve, dye solutions were prepared with concentrations varying from 10 to 200 mg L^{-1}, and their absorbances were measured. From the data linearization, the concentration limit in which the Lambert-Beer law was still valid was determined.

2.4. Adsorption experiments

For the adsorption experiments, the experimental conditions were: particle size of 2.8 mm, the temperature of 30ºC, solution volume of 0.05 L, and stirring speed of 120 rpm.

The Erlenmeyer flasks were shaken in a mechanical incubator (Tecnal, TE-424). After the adsorption, the adsorbent was separated from the dye solution using a domestic sieve. Then the solution was centrifuged to remove the fine particles.

The dye concentration was determined by a spectrophotometer (Shimadzu, UV-1601) at a wavelength of 610 nm. The amount of dye adsorbed per gram of sugarcane bagasse at equilibrium (q_e, mg g^{-1}) was determined using the mass balance equation (Eq. 1), and the percentage of removal using the Eq. (2).

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

$$Removal\ (\%) = [(C_0 - C_e)/C_0] * 100 \quad (2)$$

where: C_e is the dye solution concentration at equilibrium (mg L^{-1}); C_o is the initial dye solution concentration (mg L^{-1}); V is the solution volume (L); m is the mass of adsorbent on a dry basis (g).

2.5. Influence of the adsorbent dosage and solution pH

The experiments related to the adsorbent dosage and solution pH applied an initial concentration of dye of 80 mg L^{-1}. In dosage experiments, the pH used was 2, and the biomass concentrations varied from 2 to 20 g L^{-1}. Based on the best biomass dosage result, the influence of the solution pH was evaluated analyzing the values of 2.3, 2.7, 3.0, 4.2, and 8.6. Eq. (1) and Eq. (2) were applied in order to determine the dye removed by the sugarcane bagasse.

2.6. Adsorption kinetics

The adsorption kinetics was evaluated applying the best adsorbent dosage and pH previously determined. This experiment is applied to obtain the contact time necessary to the system adsorbent/dye to reach the equilibrium. The experiment was carried out using an initial dye solution of 110.5 mg L^{-1}, and aliquots were collected in determined time intervals (5-960 minutes).

The amount of dye adsorbed per gram of sugarcane bagasse (q_t, mg g^{-1}) for each evaluated time was determined according to Eq. (3).

$$q_t = [(C_0 - C_t) * V]/m \quad (3)$$

where: C_t is the concentration of the dye solution (mg L^{-1}) at a specific time.

For most adsorbent/adsorbate systems, it is common to describe the adsorption kinetics using the pseudo-first order model (Eq. 4, Lagergren (1898)) and pseudo-second order model (Eq. 5, Ho et al. (1996)).

$$q_t = q_e * [1 - exp(-k_1 * t)] \quad (4)$$

$$q_t = q_e * [(q_e * k_2 * t)/(1 + q_e * k_2 * t)] \quad (5)$$

where: q_e is the amount of dye adsorbed at equilibrium (mg g^{-1}); k_1 is the kinetic constant of the pseudo-first order model (min^{-1}); t is the time (min); k_2 is the kinetic constant of the pseudo-second order model (g mg^{-1} min^{-1}).

The experimental data were fitted to the models using the Statistica software and the nonlinear Simplex method.

2.7. Equilibrium study

The equilibrium isotherm was obtained applying the same experimental conditions previously described. It was varied the initial dye concentration (10 to 500 mg L^{-1}), and it was applied a contact time enough to reach the system equilibrium (obtained from the kinetic study). The amount of dye adsorbed per gram of sugarcane bagasse at equilibrium (q_e, mg g^{-1}) was calculated using Eq. (1).

The adsorption isotherms represent the equilibrium between the adsorbent and adsorbate. They can be described by models such as Langmuir (Eq. 6, Faust and Aly (1987)) and Freundlich (Eq. 7, Mortimer (1993)).

$$q_e = (q_{max} * k_L * C_e)/(1 + k_L * C_e) \quad (6)$$

$$q_e = k_F * C_e\text{^}(1/n) \quad (7)$$

where: q_{max} is the maximum adsorption capacity related to a single layer of adsorbate (mg g^{-1}); k_L is the affinity constant (L g^{-1}); C_e is the liquid phase concentration at equilibrium (mg L^{-1}); k_F and n are the Freundlich constants.

The experimental data were fitted to the models using the Statistica software and nonlinear Simplex method.

3. Results and discussion

Fig. 1 represents the molecular absorption spectra (350 – 800 nm) obtained for the Reactive Blue BB reactive dye according to different solution pH values (1 to 10).

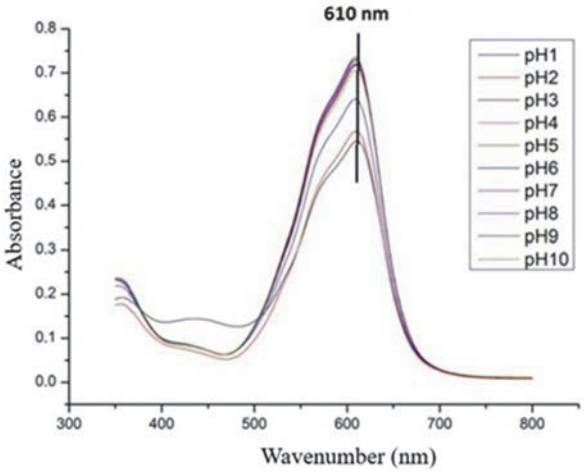

Fig. 1. Molecular absorption spectra to different pH values of the Reactive Blue BB dye solution

The reactive dye did not show any significant changes in the band position for the different pH values evaluated (Fig. 1). However, a reduction of the intensity of the band occurred with a pH of 1, 2, and 3, indicating minor changes in the energy levels of the molecular orbitals of the dye.

The maximum molecular absorption occurred at 610 nm. According to Niebisch (2009), this peak corresponds to the presence of azo bonds in the dye structure. Since there was a minor variation among the wavelengths obtained, the wavelength of 610 nm was employed to determine the absorbance in the adsorption experiments. The study of Aksu and Donmez (2003) used 600 nm to read the absorbance of Remazol Blue dye.

The analytical curve for the Reactive Blue BB dye (pH of 2.3) presented linearity between absorbance and concentration, obtaining a coefficient of determination (R^2) of 0.999.

Fig. 2 shows the dye removal according to different dosages of the sugarcane bagasse adsorbent.

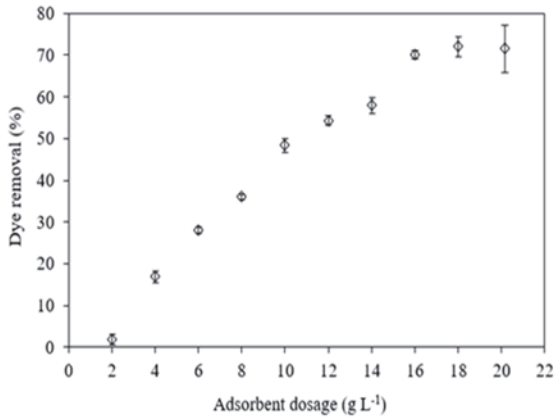

Fig. 2. Dye removal for different biomass dosages (C_o = 80 mg L^{-1}, T = 30 ºC, t = 1410 minutes, pH = 2.3, 120 rpm)

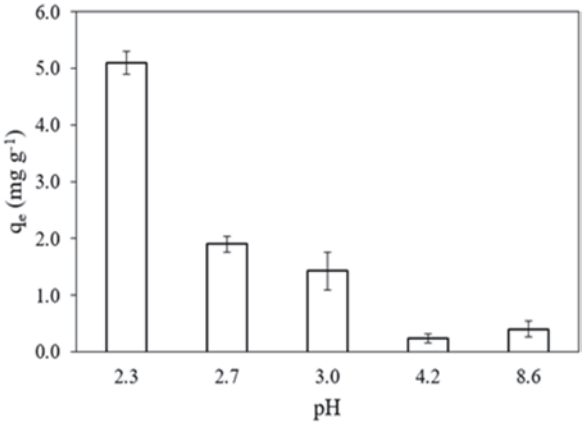

Fig. 3. Influence of pH on dye adsorption by sugarcane bagasse (C_o = 80 mg L^{-1}, V = 0.05 L, T = 30 ºC, 120 rpm, t = 1410 minutes, dosage = 16 g L^{-1})

It can be observed (Fig. 2) that the dye removal increased with the increment of the biomass dosage. In the curve begging the adsorption efficiency increased as the adsorbent dosage increased, due to more available adsorption sites. However, in higher absorbent dosages, the removal efficiency tended to a constant value. The same effect was observed in other industrial by-products applied as adsorbents (Garg et al., 2003; Gong et al., 2005; Juchen et al., 2018; Ratnamala et al., 2015).

Applying the dosage of 16 g L^{-1}, the percentage of removal reaches a value of around 70%, and the dosage increase to 18 and 20 g L^{-1} did not increase the dye removal significantly. The removal tends to stagnate, and the increase of the adsorbent amount is no longer advantageous, since the application of a higher amount of adsorbent is no effective to increase the dye removal, being not a practical application from an economic perspective (Zhou et al., 2019b). Based on this result, the dosage of 16 g L^{-1} was chosen for the following adsorption experiments, as a further increase in dosage did not improve much the dye adsorption.

Fig. 3 shows the results obtained from the study of the pH influence on the amount of dye removed. The pH condition can influence the surface charge on the active sites of adsorbent (Huang et al., 2018). The result showed that the adsorption of the reactive dye was enhanced by acidic pH (Fig. 3). This effect occurs because of the electrostatic attraction effect. At low pH the adsorbent surface is positively charged, resulting in a strong attraction between the biomass surface and the dye anionic molecules, which have a negative charge (Garg et al., 2003; Juchen et al., 2018). In other words, at low pH, the process supports the removal of anionic dyes and disfavor the adsorption of cationic dyes (Zhou et al., 2019b).

As shown in Fig. 3, the highest dye removal was reached with a pH of 2.3 (5.09 mg g^{-1}), followed by a pH of 2.7 with the removal of 1.90 mg g^{-1}. This higher removal of dye with a lower pH value requires a significant amount of acid added to adjust the solution pH to 2.3, which makes this procedure unfeasible. Similar results were obtained by Honorio et al. (2016) in the adsorption of Reactive Blue BF-5G dye by soybean hulls. These authors chose to follow the experiments using a pH of 2 due to the less costly process to correct the pH. Based on the pH effect results and aiming for an economic process, the pH value of 2.7 was chosen for the following adsorption experiments.

Aksu and Donmez (2003) studied the biosorption by yeast of a similar dye, the Remazol Blue dye, in the study the pH of 2 also showed the maximum biosorption. According to the authors, since the reactive dyes release anions to the solution, the electrostatic attraction happens between the adsorbent positively charged surface and the negatively charged dye anions. Merci et al. (2019) also obtained the best adsorption at pH 2 in the adsorption of Remazol Brilliant Blue BB applying as adsorbents sugarcane and green coconut fibers.

The experimental kinetic data of the dye adsorption by the sugarcane bagasse and the fitted data according to the pseudo-first and pseudo-second order models are presented in Fig. 4.

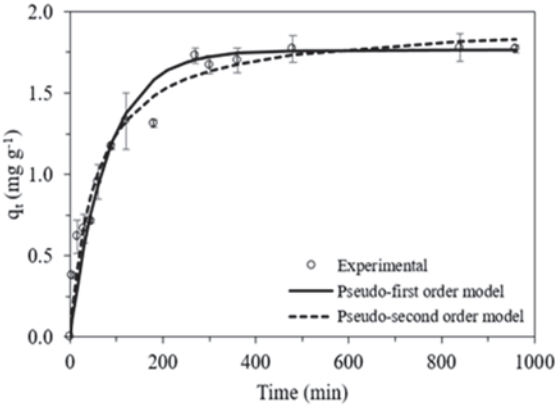

Fig. 4. Experimental and fitted kinetic data of Reactive Blue BB dye adsorption (C_o = 110.5 mg L^{-1}, pH 2.7, T = 30 ºC, 120 rpm, adsorbent dosage = 16 g L^{-1}, 0.05 L of solution)

According to Fig. 4, it can be verified that the equilibrium time was reached in approximately 480 minutes. Similarly, the kinetics of activated carbon from sugarcane bagasse soot to adsorb Methylene Blue dye achieved the equilibrium with 420 minutes (Giusto et al., 2017).

Table 1 presents the parameters estimated by the models and their coefficients of determination. It can be verified from Table 1 that the pseudo-second order model presented the best coefficient of determination (R^2=0.97) when compared to the pseudo-first order model (R^2=0.94). However, the pseudo-first order presented a better prediction regarding the experimental and fitted capacities of adsorption when compared to the pseudo-second order model.

The pseudo-second order model was also the best model to describe the adsorption of other dyes by sugarcane bagasse. For instance, the Methylene Blue adsorption by activated carbon from sugarcane bagasse soot was best described by the pseudo-second-order model (Giusto et al., 2017). In the study of Ferreira et al. (2015), the adsorption kinetics of crystal violet dye by modified sugar bagasse also followed the pseudo-second-order model, and the rate constant increased with increasing temperature.

Aksu and Donmez (2003) studied the biosorption of a similar dye the Remazol Blue Reactive by different types of dried yeasts; the biosorption process also followed the pseudo-second order model. Ratnamala et al. (2015) also obtained that the adsorption kinetics was better represented by the pseudo-second order model in the adsorption of Remazol Brilliant Blue by acid-treated red mud.

Fig. 5 presents the equilibrium isotherms of the Reactive Blue BB dye according to the Langmuir and Freundlich models. The parameters estimated from the models are shown in Table 2.

The lowest value of the objective function and the best coefficient of determination (R^2 = 0.97) were obtained from the Langmuir model when compared to the Freundlich model (R^2 = 0.96). The value of the parameter n from the Freundlich model was 4.12 (1/n=0.24).

Table 1. Parameters obtained from the kinetic fitted data of dye adsorption by sugarcane bagasse according to the pseudo-first and pseudo-second order models

Pseudo-first order	***Parameter value***
k_1 (min^{-1})	0.0126±0.0019
$q_{e,\,mod}$ (mg g^{-1})	1.76±0.06
$q_{e,\,exp}$ (mg g^{-1})	1.77±0.08
Objective function	0.32
R^2	0.94
Pseudo-second order	***Parameter value***
k_2 (g $mg^{-1}min^{-1}$)	0.0095±0.0018
$q_{e,\,mod}$ (mg g^{-1})	1.93±0.07
$q_{e,\,exp}$ (mg g^{-1})	1.77±0.08
Objective function	0.19
R^2	0.97

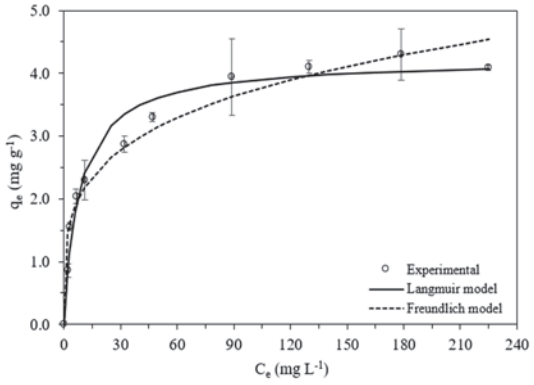

Fig. 5. Reactive Blue BB adsorption isotherms (T= 30 ºC, 120 rpm, adsorbent dosage =16 g L^{-1}, contact time = 480 minutes)

Table 2. Parameters from the equilibrium data of dye adsorption by sugarcane bagasse fitted to the Langmuir and Freundlich models

Langmuir	***Value***
q_{max}(mg g^{-1})	4.21±0.15
q_{exp}(mg g^{-1})	3.94±0.07
k_L(L g^{-1})	0.121±0.022
Objective function	0.61
R^2	0.97
Freundlich	***Value***
k_F	1.22±0.15
n	4.12±0.45
Objective function	0.80
R^2	0.96

According to Freundlich theory, when 1/n is between 0 to 1 it indicates a favorable adsorption process (Gusmão et al., 2013). The Langmuir model best described the data, and this model indicates the chemical adsorption is probably more predominant than physical adsorption, and the adsorption was monolayer and occurred on homogenous adsorption sites (Giusto et al., 2017; Huang et al., 2018).

Langmuir isotherm represents better the experimental data for a similar dye for other adsorbent materials. For instance, in the study of Sathishkumar et al. (2012) which evaluated the adsorption of Remazol Brilliant Blue R by agro-industrial waste Jatropha curcas pods. In the study of Mavinkattimath et al. (2017), the Langmuir isotherm well fitted the equilibrium data for simultaneous Remazol Brilliant Blue and Disperse orange dyes adsorption by red mud.

Furthermore, the results showed a maximum capacity of adsorption of 4.21 mg g^{-1} obtained according to the Langmuir isotherm, a value that is close to the one obtained experimentally (3.94 mg g^{-1}) for the evaluated adsorbent-adsorbate system. Giusto et al. (2017) studied the adsorption potential of Methylene Blue by activated carbon from sugarcane bagasse soot and achieved an adsorptive capacity of 331 mg g^{-1}, much superior value than the present study. Ferreira et al. (2015) also modified the sugarcane bagasse to adsorb crystal violet dye and

obtained a higher maximum adsorption capacity equivalent to 692.1 mg g^{-1}.

Another study showed that chemical modification of another industry by-product, the pine sawdust, could improve the adsorption capacity of dyes (Zhang et al., 2015). In the study of Liu et al. (2019) the modification of straw, an agricultural waste, also showed to increase significantly the adsorption efficiency. The adsorption capacity for Methyl Orange and Methylene Blue dyes by the modified straw was 54 and 3 times higher than the unmodified straw, respectively.

These results demonstrate that the ability to remove several dyes can be enhanced when agricultural waste and industrial by-products are subjected to some biomass modification procedure. However, this biomass modification by chemical and thermal processes/activation adds a cost to the adsorbent material, and consequently to the wastewater treatment process, besides the generation increase of liquid and gaseous waste. Therefore, the use of the industrial byproduct, sugarcane bagasse, as adsorbent material in its natural form it is advantageous since it does not requires any additional operational cost to the process. Generally, the option of using modified or unmodified adsorbent material requires careful technical and economic analysis in terms of building and operation of the treatment system.

4. Conclusions

The present study evaluated the influence of parameters such as pH, adsorbent dosage, and contact time on the adsorption process of the Reactive Blue BB dye by the sugarcane bagasse. Within the visible molecular absorption spectra, the studied dye presented only one peak at the maximum wavelength of 610 nm.

The adsorption process was enhanced in an acidic pH medium, improving the adsorbent/adsorbate interactions.

The kinetics of adsorption presented an equilibrium time of 480 minutes, and the pseudo-second order model best represented the experimental kinetic data (R^2=0.97).

The Langmuir model represented the equilibrium data with a maximum adsorption capacity of 4.21 mg g^{-1}. The sugarcane bagasse, obtained on a large scale in sugar and alcohol industries, presented an adsorptive capacity to remove the Reactive Blue BB textile dye.

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