Color removal from textile effluent using *Azadirachta indica* leaf powder as an adsorbent

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Abstract

The removal of dyes present in industrial effluent has received great attention in the past few years. This is partly due to increasing environmental awareness and the implementation of ever stricter environmental rules. Adsorption is one of the techniques used for the effective removal of dyes. However, the efficiency of the adsorption process depends on the choice of a suitable adsorbent. Because of the high cost of some conventional adsorbents, researches have been seeking alternatives, such as Neem tree leaves. The Neem tree (*Azadirachta indica)* is native to India and its importance has been recognized by the US National Academy of Sciences, which published a report in 1992 entitled “Neem – a tree for solving global problems”. The aim of this study was to investigate the use of Neem leaves as an alternative adsorbent for the removal of dyes present in textile effluents. The dyestuff studied was the reactive dye Remazol Blue RR. In order to obtain the best removal conditions for this dye, the influence of the following parameters was investigated: aqueous solution pH, agitation, addition of sodium sulfate and temperature of mixture. Spectrophotometry was used to measure the concentration of the dye remaining in the fluid phase after the adsorption process using Neem tree leaves. The Langmuir model was applied to adjust the experimental data on changes in the concentration of the solute adsorbed by the solid phase (adsorbent) according to the equilibrium concentration of the fluid phase and showed good correlation. According to the experimental data, the removal efficiency of Remazol Blue RR was 90%. Therefore, the removal of dyes present in textile effluent using Neem tree leaves showed good results, and this adsorbent could be used in industries in the treatment of textile effluent.

Keyword: Reactive dye, *Azadirachta indica,* Remazol Blue RR, Dye removal.

1. Introduction

In the textile industry, the activities involving dyeing generate problems due to the discharge of toxic effluents, originating from the byproducts generated. If not treated properly before being discharged into natural water bodies, the effluent from this industry may reach potable water resources, causing serious ecological concern. Therefore, the development of new technologies for the removal of color from industrial effluents has received a lot attention over the past few years, partly driven by an increasing environmental awareness and the implementation of ever stricter environmental rules (HOLME, 1984; MOTSCHI, 1994). The techniques available in the literature for the discoloration of wastewater include adsorption, precipitation, chemical and electrochemical degradation and biodegradation processes (GUARATINI & ZANONI, 1999).

Adsorption has been used successfully in the removal of color from effluents. Activated carbon is the most used adsorbent. Due to its high cost and considering the enormous quantity of effluent produced by textile industries, researches are turning toward the use of alternative adsorbents, also called non-conventional low-cost adsorbents.

An example of this group of adsorbents is the Neem tree leaf. The Neem tree (*Azadirachta indica*), of the family Meliaceae, is native to India and was adapted for its growth in Brazil a few years ago. The leaf has polar groups on the surface, which gives it a high cationic exchange capacity (PARROTA & CHATURVEDI, 1994; DOHAREY & SINGH; 1989).

The aim of this study is to investigate the efficiency of Neem tree leaves in the removal of dyes present in textile effluents, as well as to verify the influence of different parameters on the color removal process.

2. Materials and methods

The dye used in this study was reactive dye Remazol Blue RR. The molecular mass of this dye is 950.0 g/mol.

The adsorbent used during the color removal experiments was Neem tree leaf powder (*Azadirachta indica*). The leaves were kindly given by EMBRAPA MILHO E SORGO – MG.

The reagent sodium sulfate ($Na_2SO_4.10H_2O$) was added to the dye solution in order to simulate real effluent and dyeing bath conditions. To adjust the pH, 0.1 M sodium hydroxide (NaOH) was added to the aqueous solutions.

2.1. Experimental procedure

The adsorption experiments were carried out in batch processes. In each experiment 20 mL of the dye solution was mixed with 1 g of Neem leaf powder (NLP) in a glass tub and left to stand. After a predetermined time interval, previously defined by the kinetics experiments, the mixture was filtered and the quantity of dye not adsorbed, i.e. that remaining in solution, was measured using a Shimadzu UV mini 1240 spectrophotometer.

The same experiment was repeated using different parameters: agitation, pH (8, 10 12), addition of sodium sulfate (0%, 2%, 4% and 6% of the solution mass) and temperature (25, 40 and 60°C).

2.2. Equilibrium isotherms

The adsorption isotherms were determined by batch processes. The procedure used is described at Section 2.1.

Samples containing different predetermined concentrations of the adsorbate (50-2000 mg/L) were volumetrically transferred to the adsorption containers, which contained a constant mass of the adsorbent. After mixing, the containers were left to stand for a time interval predefined by the kinetics experiments. After the equilibrium was reached, samples of each container were collected and quantified through absorbance readings using a spectrophotometer.

Through the absorbance measured and the calibration curve drawn, it is possible to determine the final concentration of the dye present in the fluid phase. Therefore, the concentration of the solute adsorbed on the solid phase can be determined by a mass balance of the adsorbate.

The experimental data for changes in the concentration of the solute adsorbed on the solid phase (adsorbent) according to the equilibrium concentration of the fluid phase were adjusted using the Langmuir model, as shown in the following equation:

$$q_e = \frac{q_m.K_L.C_e}{1+K_L C_e} \tag{1}$$

where C_e is the equilibrium concentration of the solute in the fluid phase (mg/L), q_e is the quantity of solute adsorbed on the solid phase (mg/g), q_m is the maximum quantity of the solute adsorbed for a complete monolayer (mg/g) and K_L is the adsorption equilibrium constant (L/mg) or Langmuir constant.

Langmuir isotherm parameters, q_m and K_L, were determined on the basis of the linear form of the Langmuir equation.

3. Results and discussion

In order to remove the dye Remazol Blue RR, the influence of different parameters were verified, such as pH of the aqueous solution, batch agitation, salt addition and temperature. In this way, the conditions to obtain the best efficiency of the color removal process were determined.

3.1. Remazol Blue RR dye removal

3.1.1. Influence of pH

The pH is an important factor controlling the adsorption process. In order to find the best pH for Remazol Blue RR removal, experiments were carried out varying the pH from 8 to 12.

Langmuir adsorption isotherms (C_e versus q_e) for different values of pH (8, 10 and 12), are shown in Figure 1 and the Langmuir parameters are given in Table 1.

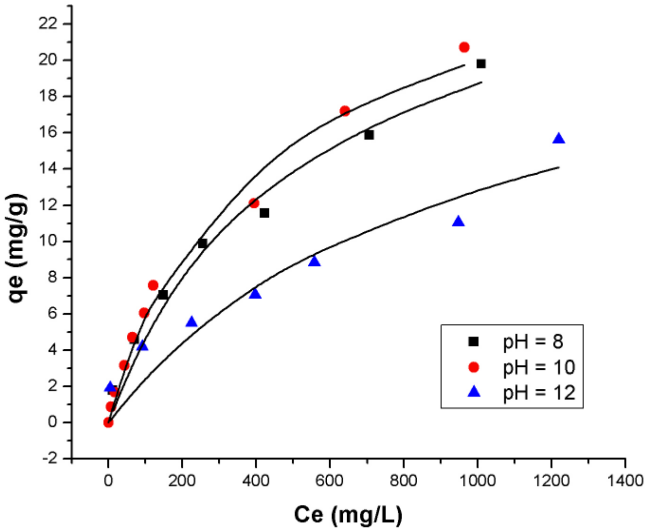

Figure 1: Langmuir isotherms for different pH values.

pH	q_m (mg/g)	R^2
8	18.656	0.981
10	22.573	0.983
12	15.360	0.930

Table 1: Langmuir isotherm parameters for different pH values.

The influence of pH on the adsorption equilibrium can be observed in Figure 1. It is possible to verify that the pH of the medium influences strongly the adsorption process.

3.1.2. Influence of agitation

In order to optimize the adsorption phenomenon, the influence of agitation was studied. Some samples of the concentrated solutions of the studied dye were kept under constant agitation and other were left to stand.

Through the results obtained from several experiments carried out with different concentrations of the solution, but the same adsorbent mass, it was possible to draw

Langmuir adsorption isotherms, as shown in Figure 2. The Langmuir parameters are given in Table 2.

Agitaton	q_m (mg/g)	R^2
With agitation	16.207	0.965
Without agitation	22.883	0.985

Table 2: Langmuir isotherm parameters for experiments with and without agitation

On analyzing the Langmuir isotherm parameters given in Table 2, as well as in Figure 2, it can be verified that the q_m (maximum adsorption capacity) values for the two cases are close, and as the utilization of the process without agitation allows an energy saving, the subsequent experiments were carried out without agitation.

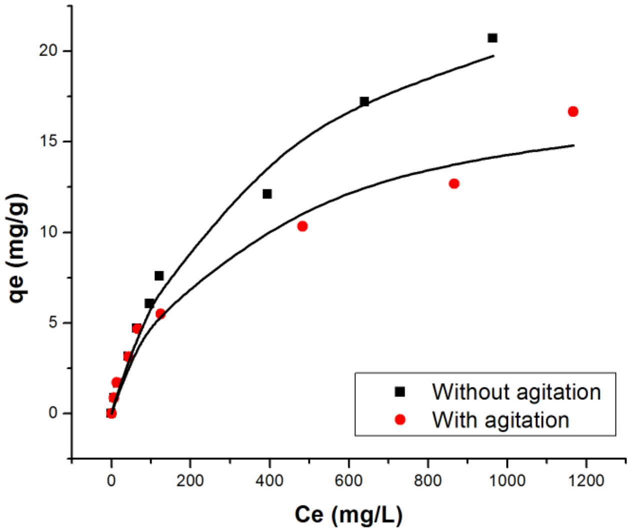

Figure 2: Langmuir isotherms for experiments with and without agitation.

3.1.3. Influence of sodium sulfate addition

In order to simulate real conditions of dyeing bath effluents, different quantities of sodium sulfate ($Na_2SO_4.10H_2O$) were added to the solution. The quantities studied were: 0, 2, 4 and 6% of the aqueous solution mass.

The Langmuir isotherms for the different quantities of sodium sulfate are shown in Figure 3, and the respective parameters are given in Table 3.

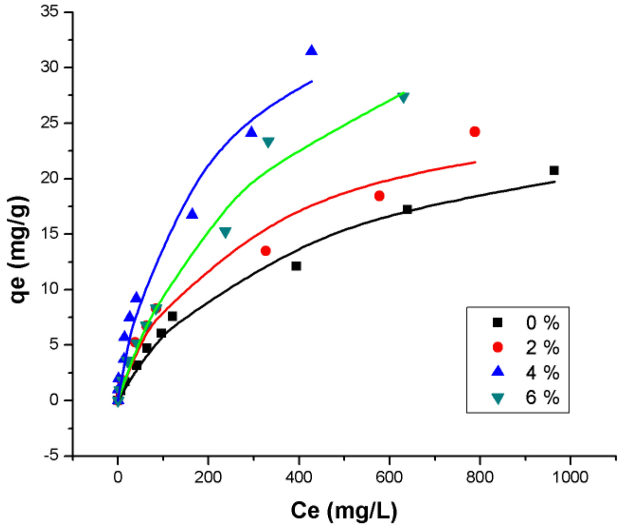

Figure 3: Langmuir isotherms for different quantities of sodium sulfate.

Sodium Sulfate(%)	q_m (mg/g)	R^2
0	22.883	0.985
2	24.630	0.962
4	32.467	0.965
6	28.571	0.984

Table 3: Langmuir isotherm parameters for solutions with 0, 2, 4 and 6% of salt.

According to the Langmuir parameters given in Table 3, it can be observed that the maximum adsorption capacity, q_m, increases from 22.883 to 32.467 mg/g with the increase from 0% to 4% sodium sulfate. However, it decreases to 28.571 mg/g with the continued increase to 6% sodium sulfate. Therefore, it is concluded that 4% of sodium sulfate is the best value for the color removal process.

Considering that in textile industries sodium sulfate is used to increase the color fixation on the cellulosic fiber of the fabric, by analogy, it can be assumed that sodium sulfate also influences positively the fixation of dyes on NLP.

3.1.4. Influence of temperature

Adsorption is an exothermic phenomenon and its efficiency should, at constant pressure, decrease with increasing temperature.

Langmuir adsorption isotherms for different temperatures (25, 40 and 60°C) are shown in Figure 4. The respective Langmuir parameters are given in Table 4.

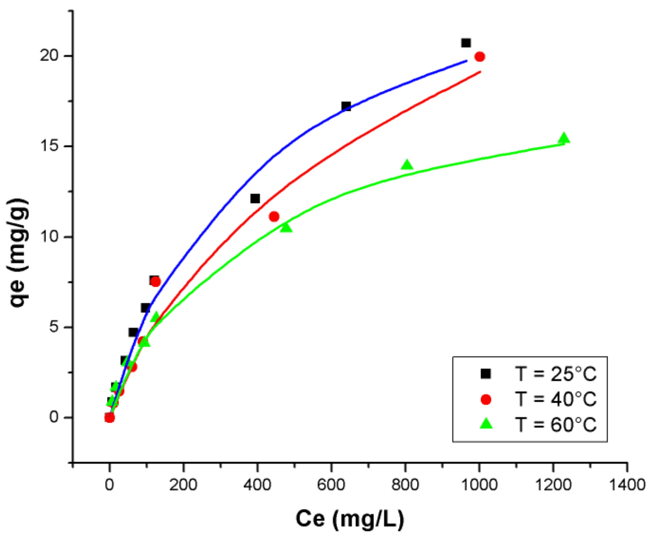

Figure 4: Langmuir isotherms for different temperatures, 25, 40 and 60°C.

Temperature (°C)	q_m (mg/g)	R^2
25	22.883	0.985
40	19.646	0.973
60	16.528	0.989

Table 4: Langmuir isotherm parameters for different temperatures.

On comparing the Langmuir parameters given in Table 4, it can be seen that the maximum adsorption capacity, q_m, decreases with increasing temperature, confirming the exothermic behavior of adsorption processes.

After verifying the influence of different parameters on the removal of Remazol Blue RR, the best conditions for this dye removal were obtained, as given in Table 5.

Parameters	Best condition
Agitation	Without agitation
pH	10
Sodium sulfate	4%
Temperature	60°C

Table 5: Best conditions for Remazol Blue RR dye removal.

The results obtained using all of the best conditions given in Table 5 in one experiment for color removal are shown in Figure 5.

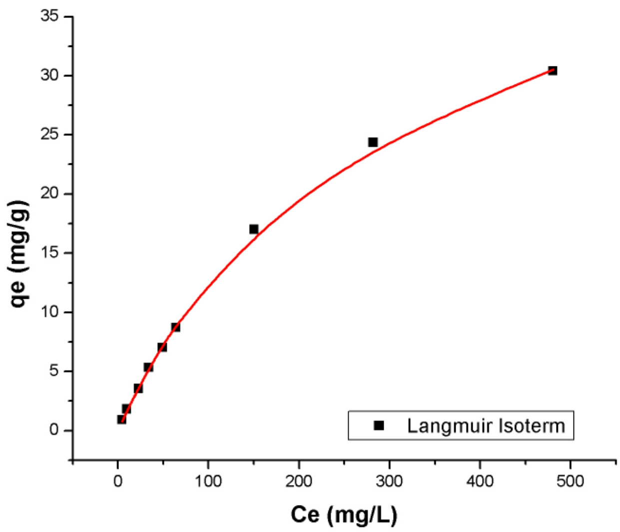

Figure 5: Langmuir adsorption isotherm for Remazol Blue RR dye – best conditions.

Isotherm	q_m (mg/g)	R^2
Langmuir	33.898	0.999

Table 6: Langmuir parameters for Remazol Blue RR dye – best conditions.

Carrying out the experiments under the best conditions of operation, it was possible to obtain a removal of 90% of the dye Remazol Blue RR from aqueous solution.

4. Conclusions

A study was carried out to investigate dye removal from aqueous effluents, through adsorption processes using NLP as the adsorbent. For Remazol Blue RR color removal, the influence of different parameters, such as aqueous solution pH, bath agitation, salt addition and temperature, was verified.

The equilibrium isotherms indicate that a basic medium favors the dye adsorption, and pH 10 showed a better efficiency in the adsorption process. No significant difference was found between systems with and without agitation in terms of color removal and, thus, systems without agitation were used with the aim of achieving a more economic process. The presence of sodium sulfate in the dye solution had a significant influence on the dye fixation on NLP. The amount of sodium sulfate which gave the best results in this study was 4% of the dye solution mass. An increase in temperature did not favor the dye transference process from the fluid phase to the solid adsorbent phase, confirming the exothermic behavior of the process. However, an increase in temperature together with sodium sulfate addition (4%) favored greatly the dye adsorption process. According to the experimental data, Remazol Blue RR removal was 90%. On the basis of the results obtained, it is possible to observe that the use of NLP as an adsorbent showed good efficiency in the removal of the textile dye studied. Thus, the use of NLP as an adsorbent could be applied as one of the stages in textile effluent treatment processes.

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References

DOHAREY, K. L.; SINGH, R. P. Evaluation of Neem (*Azadirachta indica A. Juss*) seed kernel extracts against chafer beetles. **Indian Journal of Entomology.** v. 51, p. 217-220, 1989.

GUARATINI, C. C. I.; ZANONI, M. V. B. Corantes Têxteis. **Revista Química Nova,** São Paulo, v. 23, p. 71-78, 1999.

HOLME, J. **Developments in the Chemistry and Technology of Organic Dyes.** J. Griffiths Ed., Blackwell Scent. Publ., Oxford, 1984.

KIMURA, I. Y. *et al*. Efeito do pH e do tempo de contato na adsorção de corantes reativos por microesferas de quitosana. **Polímeros: Ciência e Tecnologia.** v. 9, n. 3, p. 51-57, jul/set 1999

MOTSCHI, H.; **Chemical Safety.** M. Richardson Ed.; V. C. H. Publ., 1994. 329 p.

PARROTA, J. A.; CHATURVEDI, A. N. ***Azadirachta indica A. Juss.*** **Neem, margosa**. SO-ITF-SM-70. New Orleans, LA: U.S. Department of Agriculture, Forest Service, Southern Forest Experiment Station. 1994, 8p.