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Research Article

Batch Adsorption of Maxilon Red GRL from Aqueous Solution by Natural Sugarcane Stalks Powder

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Sugarcane stalks powder was tested for its efficiency of removing a textile dye Maxilon Red GRL from aqueous solution. Different parameters affecting dye removal efficiency were studied. These parameters include contact time, initial dye concentration, adsorbent dose, ionic strength, pH, and temperature. Langmuir and Freundlich isotherm models were applied to the equilibrium data. The data fitted well with the Langmuir isotherm ($R^2 > 0.99$). The maximum monolayer adsorption capacity (Q_0) was found to be 20.96 mg/g at an initial pH of 7.2. The temperature variation study showed that dye adsorption is exothermic and spontaneous with increased randomness at the solid solution interface. The results indicated that sugarcane stalks could be an alternative for more costly adsorbents used for dye removal. The kinetic of the adsorption process followed the pseudo second-order kinetics model.

1. Introduction

The textile industry is among important sources of contamination responsible for the continuous pollution of the environment. Production of textiles and volume of wastewater containing processed textile dyes steadily increase [1]. Large amounts of the dyestuff are lost directly into wastewater and consequently have a detrimental effect on flora and fauna [2]. There are more than 100,000 dyes available commercially; most of which are difficult to decolorize due to their complex structure and synthetic origin as they are designed to resist fading upon exposure to different factors as light, water, and oxidizing agents and as such are very stable and difficult to degrade [3, 4]. The polluting effects of dyes against aquatic environment can also be the result of toxic effects due to their long time presence in environment (i.e., half-life time of several years), accumulation in sediments but especially in fishes or other aquatic life forms, and decomposition of pollutants in carcinogenic or mutagenic compounds but also low aerobic biodegradability [5–7].

Various techniques have been employed for the removal of dyes from wastewaters including, physical, physico-chemical, and chemical processes [8–12]. All these methods have different color removal capabilities, costs, and operating rates. One of the most effective and proven treatments with potential application in textile wastewater treatment is adsorption. This process has been found to be superior compared to other techniques for wastewater treatment in terms of its capability for efficiently adsorbing a broad range of adsorbates and its simplicity [13, 14]. Adsorption process consists in the transfer of soluble organic dyes (solutes) from wastewater to the surface of solid, highly porous, particles (the adsorbent). The adsorbent has a finite capacity for each compound to be removed, and when is "spent" must be replaced by fresh material (it must be either regenerated or incinerated). The most important factors influencing dye adsorption efficiency are: dye/adsorbent interaction, adsorbent surface area, particle size, temperature, pH, and contact time [15–17]. The most used adsorbent is activated carbon beside some other commercial inorganic adsorbents as bentonite, perlite, bauxite, and clays [18–21]. Some low cost adsorbents of agricultural wastes as peat, rice husk, neem leaf powder, tree barks, bagasse pith, wood chips, ground nut shell powder, rice hulls, wood sawdust, and grounded sunflower seed shells are also used for the removal of dyes from textile effluents [22–24]. The use of vegetarian natural materials is advantageous mainly due to their widespread availability and cheapness. Sometimes the regeneration is not necessary, and the "spent" material is conventionally burnt although there is potential for solid state fermentation (SSF) for protein enrichment.

In the present study, sugarcane stalks powder was used as a new low-cost adsorbent for the removal of a textile dye Maxilon Red GRL from aqueous solutions. Few studies were conducted for the removal of Maxilon Red GRL dye by adsorption onto mineral adsorbents as bentonite [25], kaolinite [26], and silica [27]. No study has been reported for the adsorption of the dye on agricultural wastes. The effect of different parameters such as pH, temperature, contact time, initial adsorbent dose, and initial dye concentration were investigated. Finally, the isotherm and thermodynamic parameters for the adsorption of Maxilon Red GRL dye onto sugarcane stalks powder were evaluated.

2. Experimental

2.1. Materials. All chemicals used were of analytical grade and were used without further purifications. The dye was obtained from commercial market. Stock solution of Maxilon Red GRL dye contains 200 mg dye/L and was prepared by dissolving the dye in double distilled water. The working solutions were obtained by diluting the stock solution with twice distilled water. The chemical structure of Maxilon Red GRL dye is 1,2-dimethyl-3-((4-(methyl(phenylmethyl) amino)phenyl)azo)-1,2,4-tetrazolium bromide (Figure 1).

2.2. Preparation of Sugarcane Stalks. Sugarcane stalks residues were collected from the cultivated area around Benha city and then washed several times with distilled water to remove dust. The samples were sun dried for 48 hrs and dried again in a drying oven overnight at 80°C. The dried stalks samples were ground sieved through a 1 mm sieve and kept dry in a closed container till use.

2.3. Determination of pH of Point of Zero Charge (pH_{pzc}) of the Adsorbent. In order to determine the pH of point of zero charge, 0.15 g of sugarcane stalks was taken to different 100 mL conical flasks containing 50 mL of 0.01 M NaCl; the pH of which was adjusted from 2 to 10 by addition of 0.01 M HCl or NaOH for each flask. The conical flasks were sealed and placed in a shaker for 24 hours. The content of the flasks was filtered, and pH was then measured by pH meter.

2.4. Batch Adsorption Studies. Batch adsorption experiments were carried out at room temperature (25°C ± 1). Exactly 25 mL of dye solution of known initial concentration (50 mg/L) was stirred at a constant stirring rate (400 rpm) with a required dose of adsorbents (0.05–0.2 g) for a specific period of contact time (15–120 min) using a magnetic stirrer, after noting down the initial pH of the solution to the optimum pH. The pH of the solutions was adjusted to the required value by adding either 1 M HCl or 1 M NaOH solution. After equilibrium, the final concentration (C_e) was measured using absorbance values with a JascoV-530

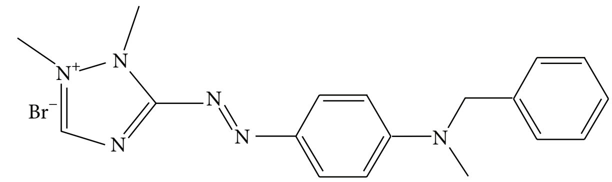

Figure 1: Chemical structure of Maxilon Red GRL dye.

(UV-Vis) spectrophotometer (Japan) and compared with the absorbance value of the initial solutions (C_0). The absorbance was measured at 530 nm. Experiments were carried out in duplicate, and mean values are presented. The percentage removal of dye was calculated using the following equation:

$$\%\ \text{Dye Removal} = \frac{C_0 - C_e}{C_0}, \quad (1)$$

where C_0 and C_e are the initial and final (equilibrium) concentrations of dye (mg/L), respectively. Blanks containing second distilled water were used for each series of experiments as controls. The amount of dye adsorption per unit mass of sugarcane stalks powder at equilibrium q_e (mg/g) was calculated by the following equation:

$$q_e = \frac{(C_0 - C_e)\,V}{W}, \quad (2)$$

where V is the volume of the dye solution (mL) and W is the weight of adsorbent (g) added to volume V.

3. Results and Discussion

3.1. Point of Zero Charge (pH_{pzc}) of the Adsorbent. pH_{pzc} of an adsorbent is important because it indicates the net surface charge of the adsorbent in solution. The pH_{pzc} is the point where the curve of pH_{final} versus $pH_{initial}$ intersects the line $pH_{initial} = pH_{final}$ (Figure 2). The point of zero charge of sugarcane stalks was determined as 4.8.

3.2. Effect of Contact Time and Initial Dye Concentration. The effect of contact time on the percentage color removal of the dye was investigated at different initial dye concentrations 25, 50, 75, and 100 mg/L as shown in Figure 3. The percentage removal of dye by sugarcane stalks powder was rapid in the beginning due to larger surface area available of adsorbent, but it gradually decreased with time until it reached equilibrium. The plot reveals maximum percent removal of the dye after about 60 min of continuous stirring. From Figure 3, it is clear that the removal of dye was dependent on the concentration of the dye, and the process was faster at higher concentrations. While the percent of color removal decreases with increasing initial dye concentration (C_0) from 91.0% at 25 mg/L to 26.4% at 100 mg/L, the adsorption capacity (q_e) increased from 11.37 mg/g to 13.20 mg/g, respectively. The shapes of the curves are similar and approximately independent on the initial dye concentration which indicates a monolayer formation of the dye on the external surface [28]. A similar trend was reported for the adsorption of dyes onto different biosorbents [29–32].

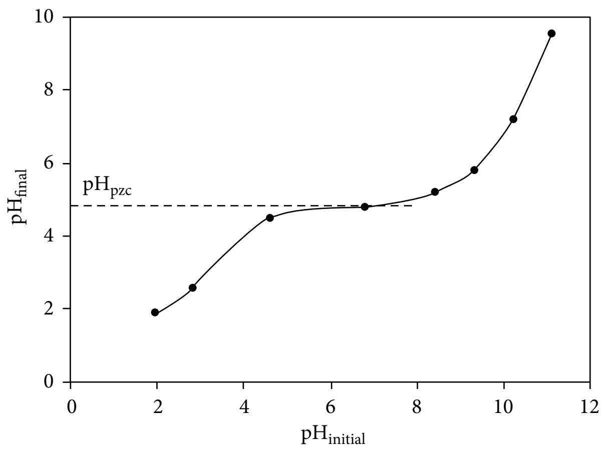

Figure 2: Determination of point of zero charge of sugarcane stalks powder.

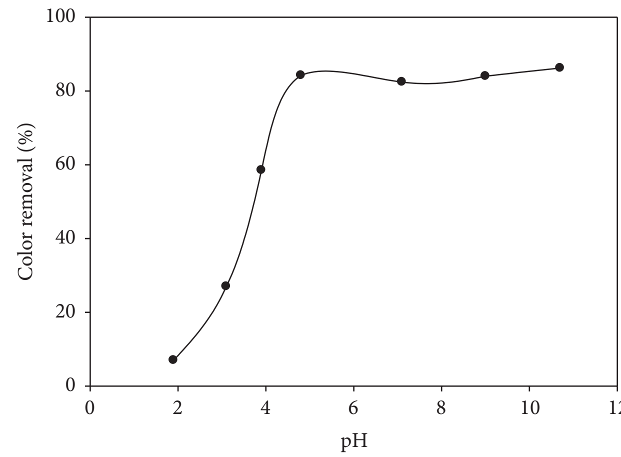

Figure 4: Effect of solution pH on the dye color removal on sugarcane stalks powder.

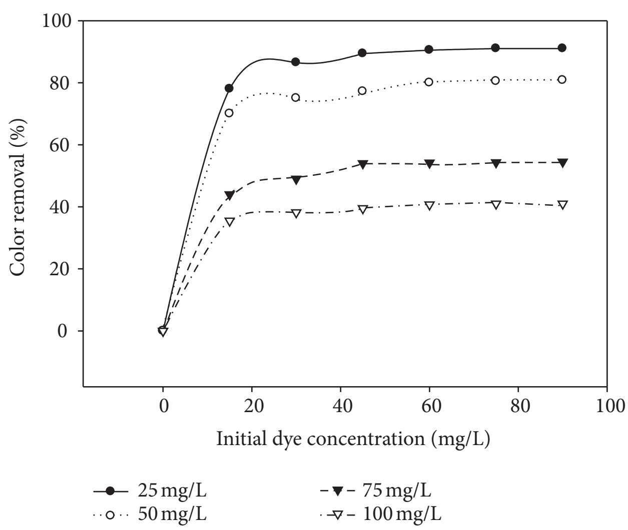

Figure 3: Relation between contact time and dye color removal at different Maxilon Red GRL concentrations.

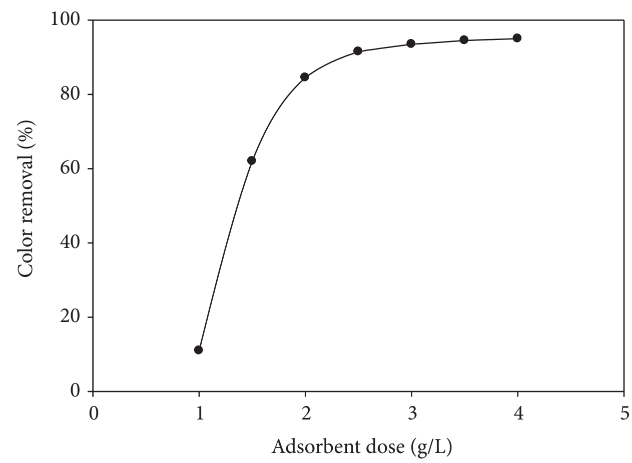

Figure 5: Effect of adsorbing dose of sugarcane stalks powder on the color removal of Maxilon Red GRL.

3.3. Effect of pH. The initial pH of the adsorbate solution plays an important role in the adsorption capacity. The effect of initial pH of dye solution on the percentage removal of dye was studied by varying the initial pH (1.9–10.8) under constant process parameters. The results are shown in Figure 4. The dye adsorbed by sugarcane stalks was lower at lower pH, and its maximum values attained at pH values higher than 4.5, so the optimum pH was attained at slightly acidic or neutral solutions. Maximum removal of the dye (about 84.2%) was achieved at pH 4.8. As the pH was decreased from 4.8 to 1.9, the removal percent decreased to 7.6%. This behavior is expected and can be explained by considering the pH_{zpc} of the adsorbent as well as molecular nature of Maxilon Red GRL (as a cationic molecule). The pH_{zpc} of the adsorbent is 4.8, meaning that the adsorbent's surface is positively charged at solution pHs below 4.8. This causes competition between protons and dye cations for adsorption locations [33]. Above the value of pH_{pzc}, a negative charge is present on the surface of sugarcane stalks causing better dye cations adsorption through the electrostatic attraction.

3.4. Effect of Adsorbent Dose. The influence of adsorbent dose on dye removal by sugarcane stalks powder was performed in a range of 0.5–2 g/L under the conditions specified in Figure 5. The percentage of the dye removal after equilibrium time increased from 11.25% to 95.10% as the adsorbent dose was increased from 1.0 to 4.0 g/L. The increased removal at high dosages is expected because of the increased adsorbent surface area and availability of more adsorption sites [34].

3.5. Effect of Ionic Strength. Generally, various salts and metal ions exist in dye containing wastewater. These salts lead to high ionic strength, which may affect the dye adsorption onto various adsorbents. Figure 6 shows the effect of ionic strength on Maxilon Red GRL uptake from aqueous solution by sugarcane stalks. It was observed that the presence of NaCl decreased the amount of dye adsorbed on sugarcane stalks at equilibrium (q_e). Because the adsorption of basic dyes is likely

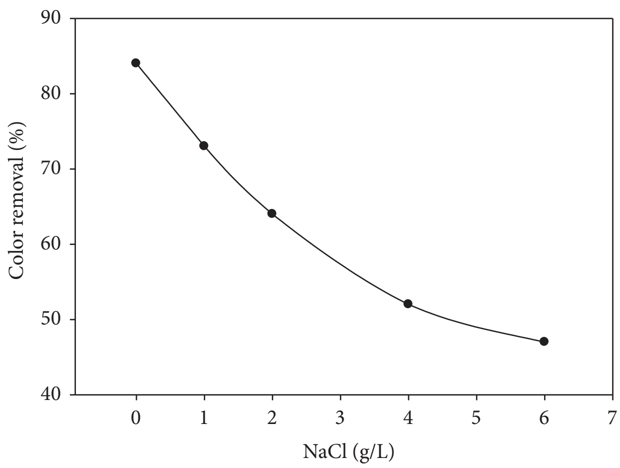

Figure 6: Effect of salinity on Maxilon Red GRL removal onto sugarcane stalks powder.

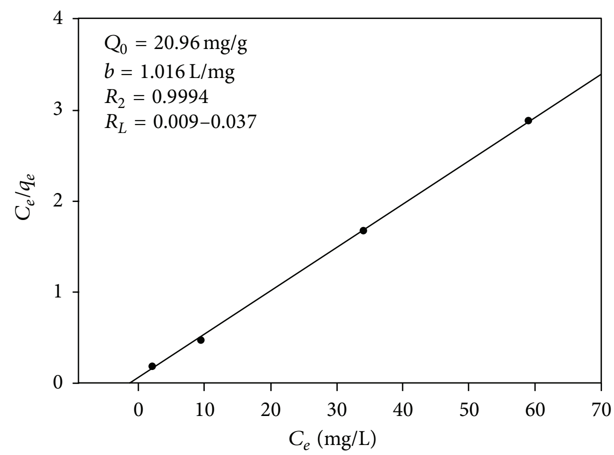

Figure 7: Langmuir isotherm for Maxilon Red GRL dye adsorption onto sugarcane stalks powder.

to be dominated by ion-exchange processes [35], the results may be explained in terms of the competition between dye and the cations in the added salt for the binding sites in the adsorbent. This increase is expected and previously reported [36].

3.6. Adsorption Isotherms. The adsorption data were analyzed using adsorption isotherm models, Langmuir and Freundlich. The Langmuir model is based on the assumption that maximum adsorption corresponds to a saturated monolayer of solute molecules on the adsorbent surface. The expression of the Langmuir model is given by the following equation [37]:

$$q_e = \frac{Q_0 b C_e}{1 + bC_e}, \tag{3}$$

where q_e (mg/g) and C_e (mg/L) are the amounts of adsorbed dye per unit mass of sorbent and dye concentration in solution at equilibrium, respectively. Q_0 is the maximum amount of the adsorbed dye per unit mass of sorbent to form a complete monolayer on the surface bound at high C_e (mg/g), and b (L/mg) is a constant related to the affinity of the binding sites on the adsorbent surface. The linear form of the Langmuir equation is written as follows:

$$\frac{C_e}{q_e} = \frac{1}{bQ_0} + \frac{C_e}{Q_0}. \tag{4}$$

A plot of (C_e/q_e) versus C_e should be a straight line with a slope of $1/Q_0$ and intercept $1/Q_0 b$. The essential characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor R_L that is given by the following equation [38]:

$$R_L = \frac{1}{1 + bC_e}, \tag{5}$$

where C_0 represents the initial concentration (mg/L) and b the Langmuir constant related to adsorption energy (L/mg). R_L value implies the shape of the isotherms to be either unfavorable (R_L), linear ($R_L = 1$), favorable ($0 < R_L < 1$), or irreversible ($R_L = 0$). As can be seen in Figure 7, for the sorption system, R_L values at different temperatures are between 0 and 1, showing favorable adsorption ($C_0 = 50$ mg/L). All R_L values at different temperatures and concentrations were between 0 and 1. The Freundlich model assumes heterogeneous adsorption due to the diversity of active sites on the surface. The Freundlich equation is expressed as [39]

$$q_e = K_F C_e^{1/n}, \tag{6}$$

where K_F (mg/g) is an indicator of the biosorption capacity and $1/n$ (L/mg) is the biosorption intensity. A value for $1/n$ below one indicates a normal Freundlich isotherm while $1/n$ above one is an indicative of cooperative adsorption. Equation (6) can be written in the logarithmic form as

$$\log q_e = \log K_F + \frac{1}{n} \log C_e. \tag{7}$$

A plot of $\log q_e$ versus $\log C_e$ is shown in Figure 8, where the values of K_F and $1/n$ are determined from the intercept and slope of the linear regressions. As seen, a very high regression correlation coefficient was shown by the Langmuir model ($R^2 = 0.9994$). This indicates that the Langmuir model was very suitable for describing the sorption of Maxilon Red GRL dye on sugarcane stalks powder compared to Freundlich model ($R^2 = 0.7649$).

The maximum capacity Q_0 determined from the Langmuir isotherm defines the total capacity of the sugarcane stalks for the dye as 20.96 mg/g sorbent. The fact that the Langmuir isotherm fits the experimental data compared to Freundlich isotherm may be due to the homogeneous distribution of active sites on the surface of sorbent.

3.7. Effect of Temperature and Thermodynamic Parameters. Adsorption experiments were carried out for 25 mL dye solution containing 50 mg/L at three different temperatures (21, 32, and 45°C) using 0.05 g of adsorbent in order to follow

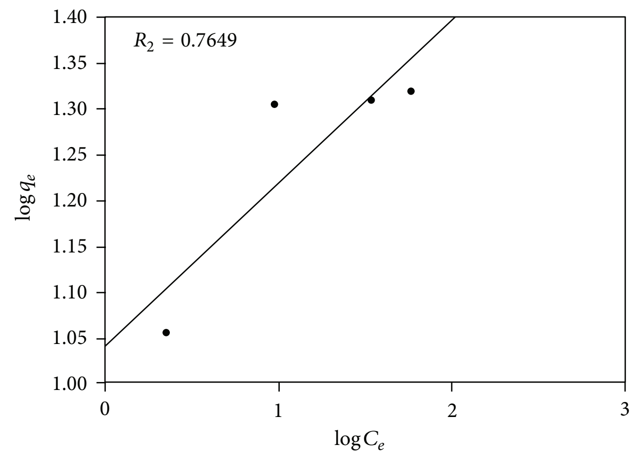

Figure 8: Freundlich isotherm for Maxilon Red GRL dye adsorption onto sugarcane powder.

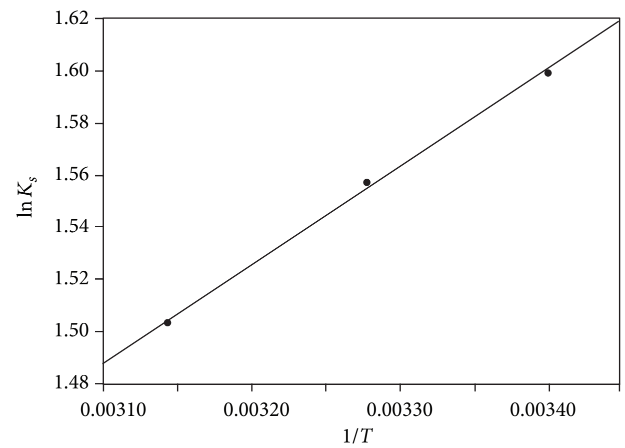

Figure 9: Plot of ln K_s versus $1/T$ for adsorption of Maxilon Red GRL on sugarcane stalks powder.

the effect of temperature on the adsorption capacity. It was observed that with an increase in temperature, adsorption capacity decreased. In order to explain and confirm the mechanism of Maxilon Red GRL adsorption onto sugarcane stalks, the thermodynamic parameters including Gibbs free energy change (ΔG°), enthalpy change (ΔH°), and entropy change (ΔS°) were calculated from the following equations [40]:

$$K_s = \frac{C_{\text{solid}}}{C_{\text{liquid}}},$$
$$\Delta G^\circ = -RT \ln K_s, \tag{8}$$
$$\ln K_s = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT},$$

where K_s is the equilibrium constant, C_{solid} is the solid phase concentration at equilibrium (mg/L), C_{liquid} is the liquid phase concentration at equilibrium (mg/L), T is the temperature in Kelvin, and R is the gas constant (8.314 J/mol K).

By plotting a graph of ln K_s versus $1/T$ (Figure 9), the values of ΔH° and ΔS° can be estimated from the slopes and intercepts, respectively (Table 1). The linear correlation coefficient value (R^2 = 0.9979) indicates good linearity. The negative value of ΔH° indicates that the sorption reaction is exothermic [41]. The positive value of ΔS° shows the increasing randomness at the solid/liquid interface during the sorption of Maxilon Red on sugarcane stalks.

The negative values of ΔG° (Table 1) indicate the feasibility of the process and the spontaneous nature of adsorption at studied temperatures. It has been resported that ΔG° values up to −20 kJ/mol are consistent with electrostatic interaction between sorption sites and the metal ion (physical adsorption), while ΔG° values more negative than −40 kJ/mol involve charge sharing or transfer from the adsorbent surface to the adsorbate to form a coordinate bond (chemical adsorption) [42]. The ΔG° values obtained in this study are

Table 1: Thermodynamic parameters for the adsorption of Maxilon Red GRL on sugarcane stalks powder.

ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)		
		294 K	305 K	318 K
−3.122	2.692	−3.908	−3.948	−3.973

<−10 kJ/mol, which indicate that physical adsorption was the predominant mechanism in the sorption process [43].

3.8. Kinetics of Adsorption. Many kinetic models have been proposed to elucidate the mechanism of solute adsorption. These models are useful for the design and optimization of effluent treatment process. In order to investigate the mechanism of Maxilon Red GRL dye adsorption by sugarcane stalks, two kinetic models were considered.

3.8.1. Pseudo First-Order Kinetic Model. The integrated linear form of pseudo first-order kinetic model proposed by Lagergren [44] is

$$\ln (q_e - q_t) = \ln q_e - k_1 t, \tag{9}$$

where q_e is the amount of dye adsorbed at equilibrium (mg/g), q_t is the amount of dye adsorbed at time t (mg/g), k_1 is the first order rate constant (min^{-1}) and t is the time (min). Hence, a linear trace is expected between the two parameters log($q_e - q_t$) and t (Figure 10), provided the adsorption follows first order kinetics. The values of k_1 and q_e can be determined from the slope and intercept. The pseudo first order rate constant ranged between 5.47×10^{-2} min^{-1} to 7.92×10^{-2} min^{-1}. The low R^2 values (Table 2) suggest that the adsorption data fitted poor to pseudo first-order kinetics. Hence, the adsorption of Maxilon Red GRL onto sugarcane stalks may not follow the pseudo first order rate expression.

3.8.2. Pseudo Second-Order Kinetic Model. The adsorption may also be described by a pseudo second-order kinetic

Table 2: Comparison of the first-order and second-order adsorption rate constants and calculated and experimental q_e values for different initial Maxilon Red GRL concentrations on sugarcane stalks powder.

C_0 (mg/L)	Pseudo first-order kinetics			Pseudo second-order kinetics		
	$q_{e,\text{cal}}$ (mg/g)	k_1 (min^{-1})	R^2	$q_{e,\text{cal}}$ (mg/g)	k_2 (g/mg min)	R^2
25	8.91	7.24×10^{-2}	0.9544	23.92	1.27×10^{-2}	0.9999
50	14.55	5.47×10^{-2}	0.9104	42.01	0.72×10^{-2}	0.9994
75	30.72	7.92×10^{-2}	0.9708	44.64	0.40×10^{-2}	0.9982
100	17.30	6.27×10^{-2}	0.9389	34.48	0.31×10^{-2}	0.9717

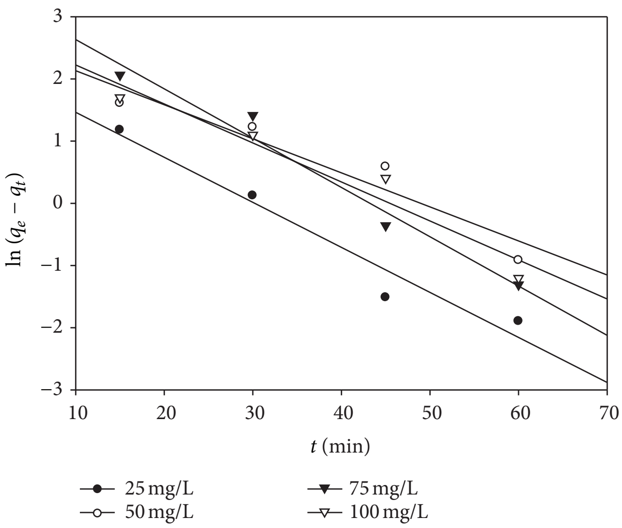

Figure 10: The pseudo first-order kinetics for the adsorption of Maxilon Red GRL on sugarcane stalks powder.

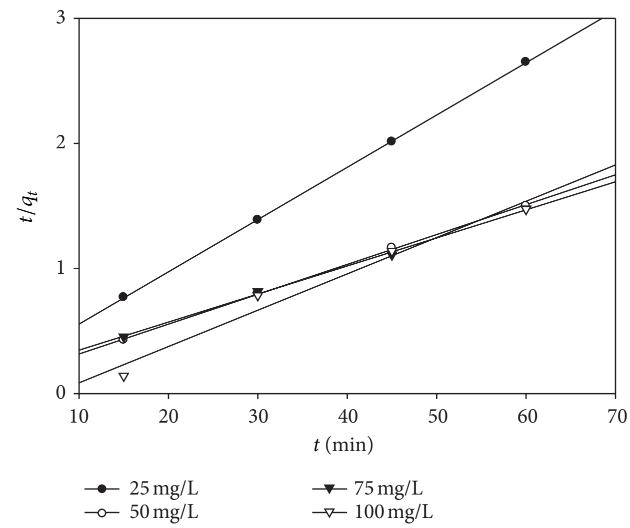

Figure 11: The pseudo second-order kinetics for the adsorption of Maxilon Red GRL on sugarcane stalks powder.

model [45]. The linearized form of the pseudo second-order model is

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t, \tag{10}$$

where k_2 is the second order rate constant (g/mg min). A plot of t/q_t and t should be linear if the adsorption follows second order. q_e and k_2 can be calculated from the slope and intercept of the plot (Figure 11). The correlation coefficients (R^2) for the second-order kinetic model are between 0.9717 and 0.9999 (Table 2); therefore, the adsorption system is not a first-order reaction but fits the pseudo second-order kinetic model. The decrease of R^2 by increasing dye concentration indicates a possible change in the kinetic mechanism of adsorption with increasing concentration [46].

4. Conclusion

The results of different experiments showed that natural sugarcane stalks powder has an ability to adsorb Maxilon Red GRL dye from aqueous solution. The sorption process was found to be pH dependent, and the adsorption process has nearly reached equilibrium in 60 min. The experimental data are fitted well to Langmuir isotherm model, and the maximum adsorptive quantity of Maxilon Red GRL was 20.96 mg/g according to Langmuir model. These values obtained from thermodynamic parameters demonstrated that adsorption process is physical in nature and the adsorption process is exothermic and spontaneous. The kinetic evaluation of the adsorption showed that the adsorption is governed by pseudo second-order kinetics.

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