

Adsorption kinetics of reactive dye using agricultural waste: banana stem

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ABSTRACT

Removal of color from dye-bearing water has been a bothering task from the health point of view and in the aesthetic sense as well. Finding out the effectiveness of Remazole Red RGB dye removal is the aim of the present work using banana stem, an agricultural waste, as an activated carbon. The preparation of the banana stem activated carbon was done in the laboratory by carbonization followed by activation. To assess the impact of varied exploratory variables such as pH of adsorbate, contact time, initial dye concentration and dosage of adsorbent on the removal of Remazole Red RGB dye from aqueous solution adopting batch studies were performed. The Freundlich and Langmuir isotherm models were used to predict the adsorption capacity of adsorbent at equilibrium conditions, the kinetic parameters were also determined. The concentration of dye remaining in the system was measured using a UV visible spectrometer. The batch test resulted in the removal of the maximum of 87% of dye when the dye concentration was 2 mg/L, at an adsorbent dose of 0.5 gm/L and dye pH 4 in 35 minutes. It is established from the studies that the equilibrium data suited excellently both Freundlich and Langmuir isotherms. The maximum adsorption capacity of 14.28 mg/L was established from the Langmuir isotherm model. To state the sorption kinetics, pseudo-first-order and pseudo-second-order kinetic models were evaluated. The intraparticle diffusion model was checked for the adsorption data and it was observed that intraparticle diffusion is not the only rate-limiting step. The regression analysis results indicated that the linear regression model gave the best results. The above detection exhibits that banana stems can be used productively for the removal of color from dye-containing wastewater.

Key words: adsorption, banana stem, batch study, isotherm, kinetics, Remazol Red RGB

HIGHLIGHT

- Removal of color from dye bearing water has been a bothering task from the health point of view and in the aesthetic sense as well. Finding out the effectiveness of Remazole Red RGB dye removal is the aim of the present work using banana stem an agricultural waste as an activated carbon. The preparation of the banana stem activated carbon was done in the laboratory by carbonization followed by activation.

INTRODUCTION

The major causes for environmental pollution are effluents discharged from Industries. Effluents that come out from the dye bearing industries are highly colored. Dye wastewater contains various chemicals which may lead to harmful effects in surface and subsurface water bodies have been discussed by the high society section of public and government all around the world.

The main sources of dye are industries such as textile, food processing, tannery, printing and paint. Once the dye bearing wastewater comes in contact with water bodies, due to the presence of chemicals they are resistant to fading on exposure light and also very difficult to decolourize when released into the aquatic environment (Batzias & Sidiras 2007). So, successful secure treatment of dye-containing wastewater is all the time an inspiring assignment for the industries as well as for researchers.

Annual production of dye stuff is more than 7×10^5 tonnes with over 10,000 commercially dyes is available in the market and it is approximated 2% of dyes manufactured per year reach the aquatic system in different ways. More than 90% of dyes were consumed by textile industries for colouring their product (Bharathi & Ramesh 2013).

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Dyes are mainly classified into anionic, cationic and disperse dyes. Due to their high water-soluble nature, anionic dyes (reactive dyes) are commonly used in the textile industry (Li *et al.* 2009). After the dyeing of fibres, nearly 50% of reactive dyes may reach the effluent (Aksu & Alper Isoglu I 2006).

Due to the low biodegradability of color-causing pollutants (Bharath *et al.* 2020), physico-chemical methods are commonly adopted for treating color-bearing wastewater. Other conventional methods for treating wastewater containing dyes are chemical oxidation, radiation, ion exchange, reverse osmosis and adsorption. Among all treatment methods, adsorption looks to be a promising method of removing colors from aqueous effluent.

Most of the conventional adsorption systems use activated carbon, which is expensive, in addition to regeneration problems, trouble in segregation of used adsorbent after use and the complication in disposing of concentrated sludge. Several types of research have investigated the alternative adsorbent materials that though less efficient involve lower cost. Hence the researchers explore economically viable and easily available alternative adsorbents (Tamez Uddin *et al.* 2017). Usually, agricultural by-products are set on fire or blindly discarded because of low prices, which may cause environmental pollution as well as loss of resources (Gong *et al.* 2008).

In this research work, attempts have been made to make use of the agricultural by-product material as an alternative sorbent. Banana stem is considered as agricultural waste; after harvesting of bananas, it is to be dumped or burned. However, it can be translated into a potential adsorbent in that it is possible to bring down the volume of waste and produce a low-cost adsorbent.

Adsorption isotherm is very much useful in finding the adsorption capacity of adsorbent per unit weight and quantity of dye left over in the banana stem – Remazole Red RGB dye system at equilibrium condition. The adsorption isotherms are useful to evaluate the adsorption capacities of adsorbents (Parab *et al.* 2006). The equilibrium adsorption data is essential for the design and efficient operation of adsorption processes (Hameed *et al.* 2007).

To explore the adsorption mechanism, several kinetic models have been suggested (Alzaydien 2009). In this research work, some of the kinetic models were adopted to check out the foremost suited model for the banana stem – Remazole Red RGB adsorption system experimental data.

A wide variety of agricultural by-product based materials have been utilized for color removal from the aqueous solution, including rice husk, boiler bottom ash (Chaurasia & Kant 2008), maize shell carbon (Chakrapani *et al.* 2008), banana leaves (Sonawane & Shrivastava 2008), and *Musa paradisiaca* (Sonawane & Shrivastava 2009). In the present investigation, the application of banana (*Musaceae*) stems as a low-cost adsorbent has been reported to be viable for removing reactive dyes. The result of varying parameters such as concentration of dye, adsorbent dosage, time of contact and pH value has been studied.

After use of the banana stem, it is treated by using the solid state fermentation process to remove the dye bearing present in the adsorbent and after that disposed of safely. Solid state fermentation is a technique in which microorganisms are cultured on a solid matrix in near absence or absence of free water in the solid matrix (banana stem carbon) acts as a support for the growth of metabolism of the microorganisms; white -rot fungi are a commonly used microorganism for this purpose.

MATERIALS AND METHODOLOGY

Preparation of adsorbent: Banana stem activated carbon was manufactured by mixing 4 parts of the banana stem with 3 parts of concentrated sulfuric acid and keeping it in a hot air oven with the temperature being maintained in the range of 85–100 °C for 24 hours. After that, excess acid present in the carbonized material was removed by washing and dried at 101 °C. The dried material was subjected to thermal activation in a muffle furnace at 600 °C for 30 minutes.

Preparation of adsorbate: The stock solution was made ready by dissolving a suitable amount (1 mg/L) of Remazole Red RGB, a coloring reagent, in distilled water. The standard solution was prepared by diluting the stock solution as per requirement. The same was used throughout the experiment.

Specification of dye used:

Dye – Remazole Red RGB

Type – Reactive dye

Wavelength (λ) – 517 nm

The adsorption investigation was executed in a batch process by using an aqueous solution of Remazole Red RGB in the concentration range 1–6 mg/L, the other variable parameters were adsorbent dose (0.25–0.75 mg/L), contact time (0–60 minutes) and pH of the medium (2–10). The concentrations were determined with the help of a carefully prepared calibration curve with a standard Remazole Red RGB dye solution.

Studies on equilibrium

The quantity of dye adsorbed at equilibrium condition q_e (mg/g) was found by

$$q_e = (C_0 - C_e)(V/W)$$

The percentage of dye removal is found by using

$$= (C_0 - C_e) \times 100 / (C_0)$$

where C_0 = initial dye concentration in mg/L and C_e = dye concentration at equilibrium condition, respectively, 'V' is the bulk of the solution in the system (L) and 'W' is the mass of adsorbent added (g).

The kinetics examined are supervised using the same procedure which is used for equilibrium studies. By conducting the kinetic studies, it is possible to find the effect of time on the processes of adsorption and to evaluate the adsorption rate.

The sorption amount at time t , q_t (mg/g) was intended by

$$q_t = (C_0 - C_t) \cdot (V/W)$$

where, C_t = dye concentration at any time 't' in mg/L

RESULTS AND DISCUSSIONS

Initial dye concentration effect

The initial dye concentration effect on adsorption of Remazole Red RGB dyes onto banana stem carbon was evaluated to the extent of 1–6 mg/L of the initial dye concentration. The adsorbent dose and agitation speed were kept at 0.50 g/L and 100 rpm respectively for Remazole Red RGB dye at natural pH (existing pH), for a contact time of 60 minutes and at room temperature (27 ± 2 °C). The maximum dye removal of 85% at an initial dye concentration of 2 mg/L has been observed and it decreased to 80% for further increase in dye concentration. The dye removal decreased for further increase in initial dye concentration as observed in Figure 1.

The dye removal in percentage declines with the rising of the initial dye concentration. Initial dye concentration increases then available sites essential for adsorption may decrease.

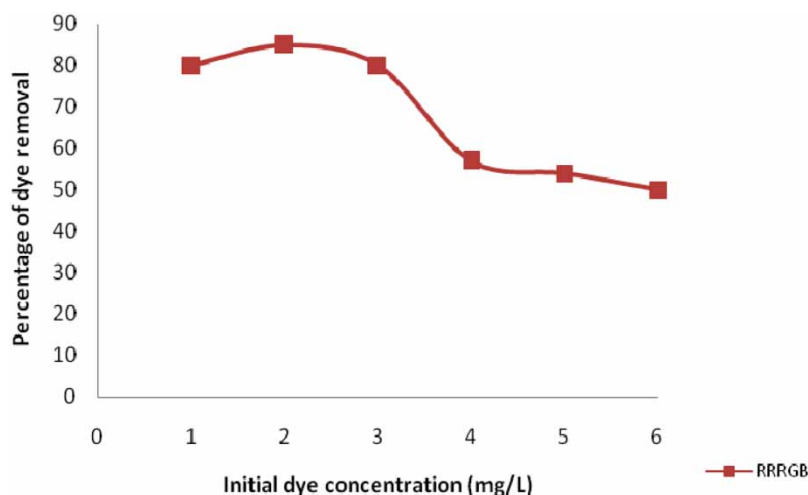


Figure 1 | Effect of dye concentration for the removal of dye.

Effect of dosage of adsorbent

The initial concentration of dye and stirring speed were kept at 2 mg/L and 100 rpm for a contact time of 60 minutes at natural pH (existing pH), and room temperature (27 ± 2 °C). The dosage of adsorbent was varied from 0.25 to 0.875 gm/L. It has been noticed that there is a considerable decrease in dye removal percentage as the dosage of adsorbent increases. The maximum dye removal efficiency of 85% was obtained at a dosage of 0.5 gm/L. It is as shown in Figure 2.

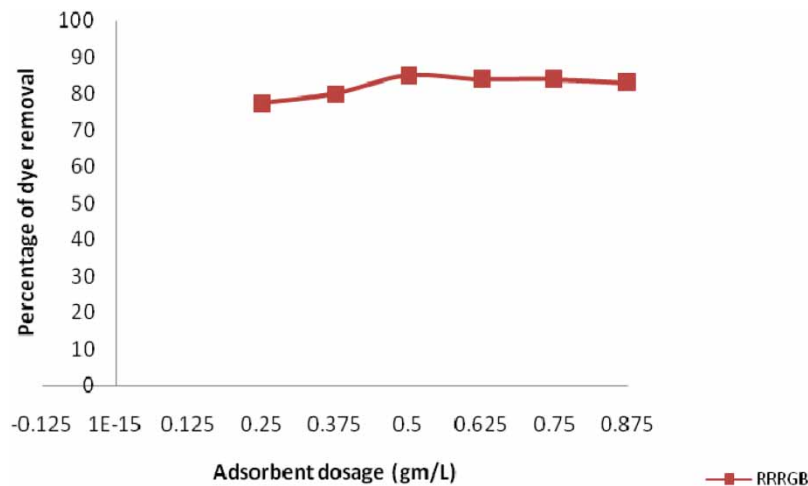


Figure 2 | Removal of dye as the adsorbent dosage varies.

The variability is noticed on account of the accumulation of adsorbent molecules in aqueous solution, because of which the functional surface area accessible for adsorption reduces. It has been noticed that there is an insignificant decrease in dye removal percentage as the dosage of adsorbent increases.

Generally, the percentage of dye removal increases with increasing adsorbent dosage. Initially, the rate of increase in the percent dye removal has been found to be rapid but to slow down as the dose increased. This phenomenon can be explained, based on the fact that at lower adsorbent dose the adsorbate (dye) is more easily accessible and because of this, the percentage of dye removal is high. With a rise in adsorbent dose, there is less commensurate increase in adsorption, resulting from many sites remaining unsaturated during the adsorption. But after a certain dosage the increase in removal efficiency is insignificant with respect to increase in dose. This is due to the fact that, at higher adsorbent concentration, there is a very fast superficial adsorption onto the adsorbent surface that produces a lower solute concentration in the solution than when the adsorbent dose is lower. Thus, with increasing adsorbent dose, the amount of percentage of dye removal is reduced.

Contact time effect

The outcome of the effect of contact time on adsorption of Remazole Red RGB dyes onto banana stem carbon was investigated in the range of 0–60 minutes. The adsorbent dose of 0.5 g/L for Remazole Red RGB dye was selected. The initial dye concentration and agitation speed were kept at 2 mg/L and 100 rpm respectively for the dyes at natural pH (existing pH) and room temperature (27 ± 2 °C). The color removal increases with increasing contact time. The color removal reached 87% within 35 minutes and became almost constant for further increase in contact time. The outcome of the results is present in Figure 3.

In the beginning percentage of dye, removal is higher because of the availability of more surface area for adsorption. Once the contact time increases, the amount of dye removal is dominated by the amount at which the dye molecules move from the outermost to the inner sites of the banana stem adsorbent.

Outcome of pH-values

The outcome of pH 2–10 on adsorption of Remazole Red RGB dye from solution onto banana stem carbon was investigated for contact time in the range of 0–60 minutes while the dosage of adsorbent, concentration of dye and stirring speed were kept at 0.50 g/L, 2 mg/L and 100 rpm for Remazole Red RGB dye, at room temperature

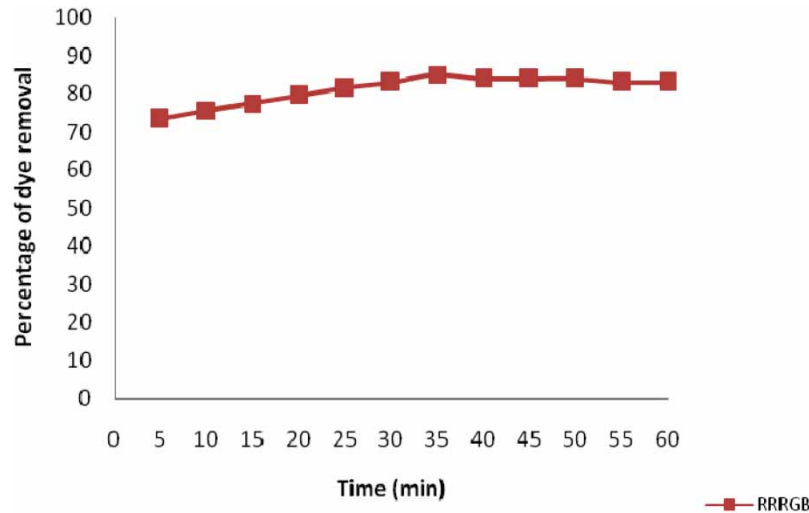


Figure 3 | Removal of dye as the contact time increases.

(27 ± 2 °C). The removal percentage of Remazole Red RGB dye by banana stem carbon increased from 45% to 87% for a decrease in pH of the solution from 10 to 2 and the optimal pH value for the maximum dye removal was found to be 4 for a contact time of 35 minutes. The results are shown in Figure 4.

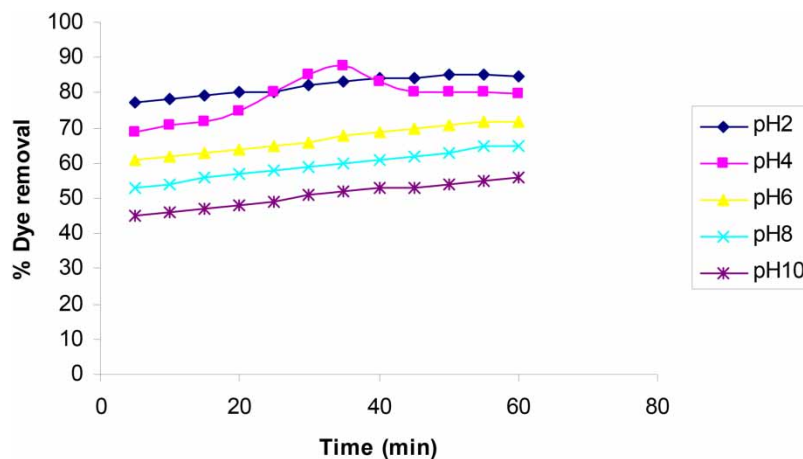


Figure 4 | Percentage of dye removal by varying pH.

When the pH of the aqueous media decreased then the amount of available negative charged surface sites declined and at the same time the amount of positively charged sites shot up. The amount of adsorption increases because of the electrostatic force of attraction between the positively charged surface sites on the adsorbent and the dye anions.

pH has an influence on the charge of the dye molecule since the pH of the dye solution plays an important role in the whole adsorption process and particularly on the adsorption capacity, influencing not only the surface charge of the adsorbent, the degree of ionization of the material present in the solution and the dissociation of functional groups on the active sites of the adsorbent, but also the solution dye chemistry. As a charged species, the degree of its adsorption onto the adsorbent surface is primarily influenced by the surface charge on the adsorbent, which in turn is influenced by the solution pH. It is a commonly known fact that anions are favorably adsorbed by an adsorbent at lower pH values due to the presence of H^+ ions. At high pH values, cations are adsorbed due to the negatively charged surface sites of adsorbents.

In this research work, experiments were conducted to find the percentage of dye removal efficiency of Remazole Red RGB dye at same working condition for both agricultural by product-based activated carbon

(banana stem carbon) and commercial activated carbon and for banana stem carbon a maximum removal of 87% of dye was observed when the dye concentration was 2 mg/L, at an adsorbent dose of 0.5 g/L at dye pH 4 in 35 minutes. For commercial activated carbon maximum removal of 96% of dye was achieved when the dye concentration was 4 mg/L, at an adsorbent dose of 0.625 g/L at dye pH 4 in 35 minutes. Hence the banana stem carbon finding is in line with commercial activated carbon results.

Some similar findings have been observed by researchers as follows

- (1) Studies were carried out on adsorption of Methylene blue from aqueous solution onto low-cost natural Jordanian Tripoli by [Alzaydien 2009](#) and it was observed that 97% of dye was removed when the dye concentration was 100 mg/L, at an adsorbent dose of 5 g/L at dye pH 8. (9)
- (2) Studies were carried out on removal of Methylene Blue by maize shell carbon by [Chakrapani et al. 2008](#) and it was observed that 98.60% of dye was removed when the dye concentration was 10 mg/L, at an adsorbent dose of 0.8 g/L at dye pH 6.5. (11)
- (3) Studies were carried out on adsorption kinetics for the removal of Malachite green from aqueous solution by using banana leaves and it was observed that maximum removal of 92.75% of dye when the dye concentration was 100 mg/L, at an adsorbent dose of 2 g/L at dye pH 8 within 50 minutes. (12)
- (4) Studies were carried out on preparation and utilization of rice straw bearing carboxyl groups for removal of basic dyes (Basic green 4) from aqueous solution by [Gong et al. 2008](#) and it was observed that 97.33% of dye was removed when the dye concentration was 250 mg/L, at an adsorbent dose of 2 g/L at dye pH 6. (6)

Therefore, from the above it is noticed that the banana stem carbon findings are in line with research done by the above scholars and also performance is high compared to that at lower initial dye concentration.

Adsorption isotherms

The dye removal efficiency of the adsorbent is evaluated by using Freundlich and Langmuir isotherms.

The Freundlich isotherm is expressed as follows:

$$\log (q_e) = \log (k_f) + (1/n)\log (C_e) \quad (1)$$

where q_e = amount of anion dye absorbed per unit weight of the banana stem adsorbent (mg/gm), C_e = concentration of the dye at equilibrium in the banana stem – Remazole Red RGB adsorption system (mg/L), k and n are constants subject to the adsorbent nature and amount of dye absorbed (Table 1). The graph (Figure 5) between $\log q_e$ and $\log C_e$ exhibit the usefulness of Freundlich isotherms. The value of n in the middle of 0–10 shows that adsorption is effective.

Table 1 | Freundlich and Langmuir isotherm parameters for Remazole Red RGB (RRRGB) dyes on banana stem carbon

Freundlich isotherm				Langmuir isotherm			
Dye	k_f (mg/g)(L/g) ^{1/n}	n	R^2	Q_0 (mg/g)	b (L/mg)	R^2	R_L
RRRGB	2.28	1.404	0.906	14.28	1.257	0.816	0.1355

The Langmuir isotherms are expressed as follows

$$(C_e/q_e) = (1/(Q_0b) + (C_e/Q_0) \quad (2)$$

where C_e = initial dye concentration in mg/L, q_e is the quantity of dye absorbed at equilibrium condition for banana stem adsorbent in mg/gm and Q_0 and b are the Langmuir constants connected to the capacity of adsorption and adsorption energy respectively. The graph (Figure 6) between C_e/q_e and C_e exhibit the usefulness of Langmuir isotherms. R_L is a dimensionless factor and R_L is equal to $1/(1 + bC_0)$, where C_0 = initial dye concentration. From Table 1, it is observed that the value of R_L lies between 0 and 1 indicating that the adsorption of anion dyes on the banana stem carbon is favourable.

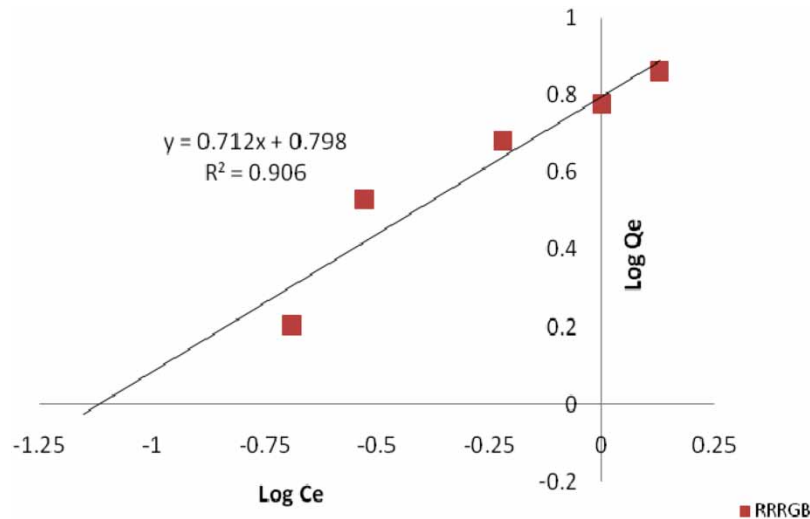


Figure 5 | Freundlich isotherms for banana stem carbon.

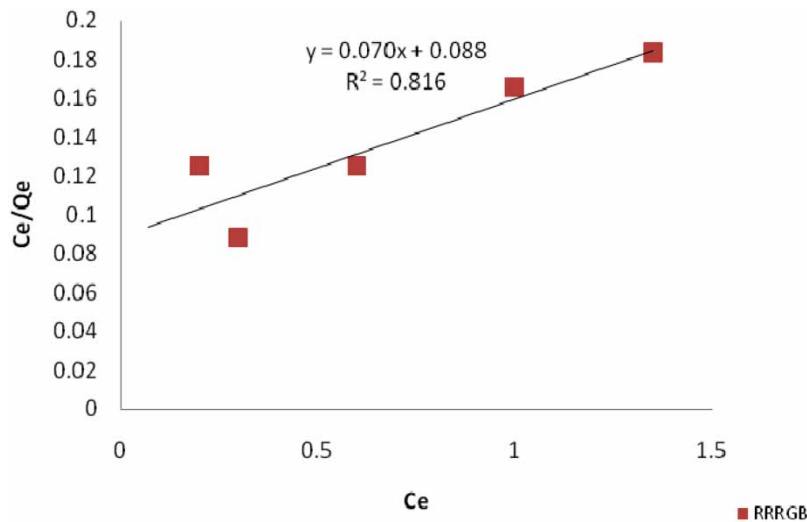


Figure 6 | Langmuir isotherms for banana stem carbon.

From the Freundlich isotherms study, it is observed that the values of k_F , n and R^2 were 2.28, 1.404 and 0.906, respectively, and it is noted that the value of n is more than one, which signifies that adsorption is favourable.

From Table 1, values of Q_o , b , R_L and R^2 for adsorption of Remazole Red RGB are found to be 14.28 (mg/g), 1.257 (L/mg), 0.1355 and 0.816 respectively and it is noted that the value of R_L is 0.1355, signifying a favourable adsorption process.

The above results established that both Freundlich and Langmuir isotherms fitted very well with the banana stem – Remazole red RGB equilibrium data. The adsorption capacity of banana stem carbon for Remazole Red RGB dye is 14.28 mg/g.

Kinetic studies

Kinetic models are very much useful to evaluate the adsorption process rate and rate-controlling step. In this research study, equilibrium data acquired from the batch adsorption system were examined by employing the following models.

The first order kinetic equation of Lagergren is expressed as:

$$\log (q_e - q_t) = \log (q_e) - (k_t/2.303)t \quad (3)$$

where q_e = quantity of dye removed in mg/g at equilibrium condition and q_t = quantity of dye removed in mg/g at time t in a minute, respectively, and k_1 = rate of adsorption constant in min^{-1} . The value of k_1 is found from the graph of $\ln(q_e - q_t)$ vs. t as presented in Figure 7.

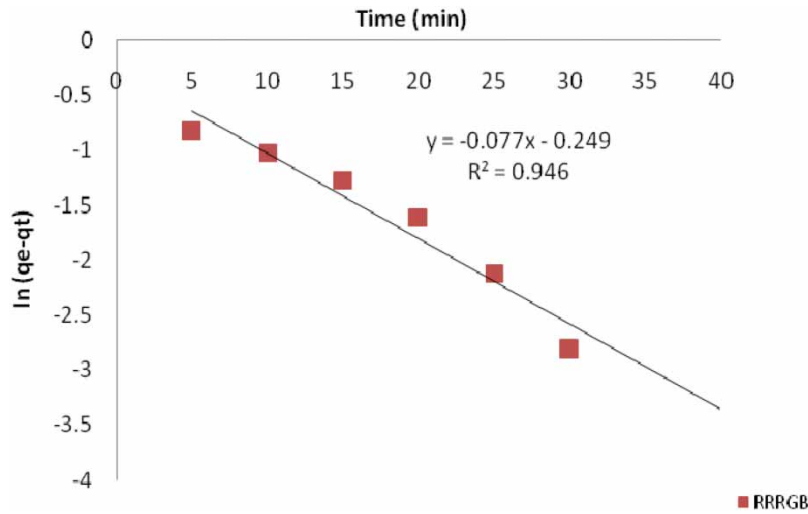


Figure 7 | Graph of the adsorption of RRRGB on banana stem carbon for the pseudo-first-order kinetic model.

The q_e of experimental values did not concur with the calculated ones, as found from Figure 7 (as shown in Table 2), which suggested that adsorption kinetics of dye on banana stem carbon does not replicate the first-order model.

Table 2 | Pseudo-first order and pseudo-second order models kinetic constants for the adsorption of Remazole Red RGB dyes onto banana stem carbon

Dyes	q_e exp (mg/g)	k_1 (1/min)	q_e cal (mg/g)	R^2	k_2 (g mg ⁻¹ min ⁻¹)	q_e cal (mg/g)	R^2
RRRGB	3.38	0.0777	1.28	0.946	0.827	3.41	0.998

The second order kinetic equation is expressed as:

$$(t/q_t) = (1/(k_2q_e^2)) + (1/q_e)t \tag{4}$$

where k_2 (min gm g^{-1}) is the second-order adsorption rate constant.

The values of k_2 and q_e can be secured from the graph of (t/q_t) vs. t as shown in Figure 8. The correlation coefficient is close to 1.0 (Table 2). The q_e calculated values suit well with the experimental q_e .

Since the banana stem – Remazole Red RGB system adsorption data fit well with the above models it shows evidence that the adsorption processes are chemisorption in nature.

Model for intra particle diffusion

The intra-particle diffusion model is indicated as below:

$$q_t = k_p t^{0.5} + C \tag{5}$$

where q_t = quantity of Remazole Red RGB dye adsorbed in mg/g at time t , k_p = intra-particle diffusion rate constant in $\text{mg}/(\text{g min})$ and C = the intercept in mg/g , by drawing the graph of q_t versus $t^{0.5}$ to find the values of k_p in terms of slope and C in terms of intercept as shown in Figure 9.

Table 3, the values of k_p , C and the regression coefficients R^2 are tabulated.

From the result, it is observed that the value of $C \neq 0$, intra-particle diffusion is not the only controlling step and this adsorption process is complex. The value of intercept C shows the result of boundary layer thickness (Nandi et al. 2009).

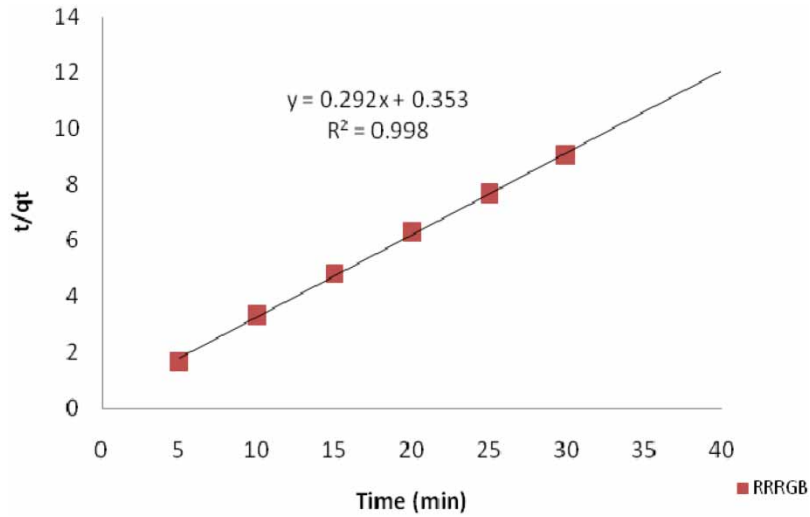


Figure 8 | Graph of the adsorption of RRRGB on banana stem carbon for the pseudo-second order kinetic model.

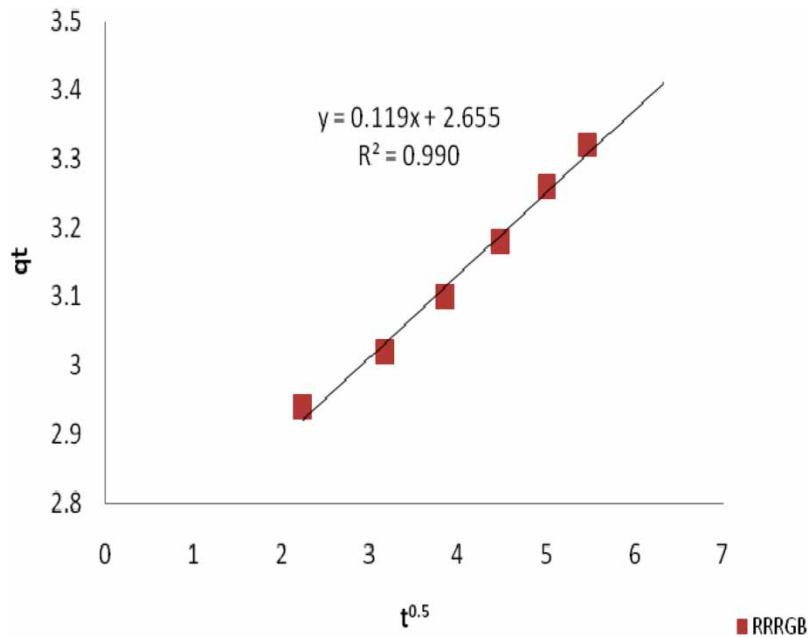


Figure 9 | Intraparticle diffusion kinetic model for the adsorption of RRRGB on banana stem carbon.

Table 3 | Parameters of intraparticle diffusion for the adsorption of Remazole Red RGB dyes onto banana stem carbon

DYE	K_p ($\text{mg g}^{-1} \text{min}^{-0.5}$)	C	R^2
RRRGB	0.119	2.655	0.990

Regression analysis

The estimated linear model interrelating percentage of color removal with the controlling parameters is expressed in the following Equations (6) and (7) for Remazole Red RGB dye.

$$Y = A_0 + A_1X_1 + A_2X_2 \tag{6}$$

$$Y = 84.35 + 0.199X_1 - 4.0583X_2 \tag{7}$$

X1 = contact time in minutes, X2 = pH of dye solution and Y = percentage of color removal.

From the model, the most significant controlling parameter of the system influencing dye removal percentage is the pH value of the dye solution and the least important parameter is the contact time for Remazole Red RGB dyes for banana stem carbon.

From correlation matrix correlation of the percentage of color removal to pH is -4.0583 and for contact time is 0.199 for Remazole Red RGB dye adsorption onto banana stem carbon.

From Figure 10, the R^2 is evaluated as 0.957 for Remazole Red RGB dye adsorption onto banana stem carbon. The results indicate that the linear regression model gives the best results (R^2 nearly equal to 1).

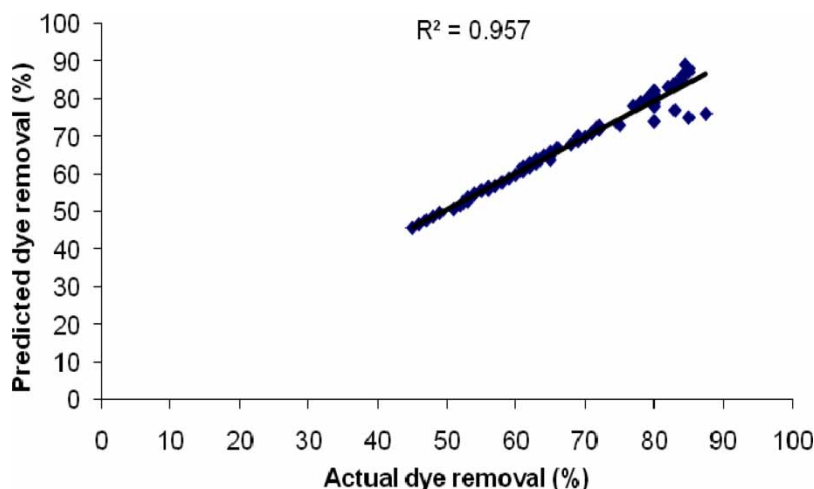


Figure 10 | Linear regression plot showing observed and predicted percentage color removal for RRRGB on banana stem carbon.

CONCLUSION

The banana stem nearly new cost-effective adsorbent for removal of dye from effluents was found to be efficient and capable of removing 87% of dye at a dye concentration of 2 mg/l at a dosage 0.5 gm/L, large solute removal takes place immediately after mixing and agitation and equilibrium conditions are nearly obtained after around 35 minutes. The adsorption capacity of banana stem carbon for Remazole Red RGB dye is 14.28 mg/g. Kinetics which follow pseudo-second-order models designate the chemisorption nature of the adsorption processes. In this adsorption system, the intra-particle diffusion is not the only rate-limiting step. The results indicate that the linear regression model gives the best results (R^2 nearly equal to 1). The results obtained from this study reveal that the low-cost adsorbent prepared from the banana stem removes the dye selected for this investigation efficiently and effectively. The material used for adsorption, which is an agricultural waste, is cheap and locally available. Therefore, banana stem carbon can be used for removing color in textile wastewater for a better environment.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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