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Kinetic and Equilibrium Studies of Adsorption of Dye Congo Red from Aqueous Solutions on Bagasse Charcoal and Banana Peels

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Abstract — Batch adsorption experiments were carried out for the removal of dye congo red (CR) from aqueous solutions using bagasse charcoal (BC) and banana peels (BP). Experiments were carried out as a function of contact time, initial dye concentration, adsorbent dose and ionic strength. The results showed that the amount of dye adsorbed increased with increasing initial dye concentration and contact time. The kinetics of basic dye adsorption nicely followed the pseudo second-order rate expression and demonstrates that intraparticle diffusion plays a significant role in the adsorption process. The adsorption data fitted well with Langmuir, Freundlich, Temkin, Dubinin and Radushkevich and Generalized isotherms. The characteristic parameters for each model have been determined. The Langmuir isotherm gave the best correlation for the adsorption of dye congo red on both the adsorbents. The monolayer saturation capacity was found to be 45.3 and 44.4 mg/g for BC and BP respectively. The value of separation factor (R_L) insinuates that BC and BP can be used as effective adsorbents. The results of the present study corroborate that BC and BP are promising adsorbents for the removal of toxic dye CR.

Keywords : *Adsorption, congo red, adsorbents, kinetics, isotherms.*

INTRODUCTION

Dyes are one of the major constituents of the wastewater produced from dyestuff manufacture and some similar industries, which have posed a severe threat to the surrounding ecosystem because many of the dyes are extremely toxic. So the study of removal of dyes from wastewater is significant and required strongly [1]. The discharge of highly colored effluent is currently one of the world's major

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environmental problems as some of the dyes and their metabolites are either toxic or mutagenic and carcinogenic and pose a potential health hazard to all forms of life [2]. During the past 35 years, India has become a major producer of dyes and pigments to cater to the needs of not only the textile industries but also of other industries such as paper, rubber, plastics, paints, printing inks, art and craft, leather, food, drug and cosmetics [3]. Congo Red (CR) (1-napthalene sulfonic acid, 3, 3′-(4, 4′-biphenylene bis (azo)) bis (4-amino-) disodium salt) is a benzidine-based anionic disazo dye, this dye is known to metabolize to benzidine, a known human carcinogen [4].

There are a number of methods for dye removal which include chemical coagulation, flocculation, chemical oxidation, photochemical degradation, membrane filtration, aerobic and anaerobic biological degradation but all of these methods suffer from one or other limitations, and none of them were successful in completely removing the color from wastewater. Dyes can be effectively removed by adsorption process; in which dissolved dye compounds attach themselves to the surface of adsorbents [5]. Adsorption onto activated carbons has proven to be one of the most effective and reliable physicochemical treatment methodology. However, commercially available activated carbons are very expensive. Therefore, there is a need to produce low cost and effective carbons that can be applied to water pollution control [6]. A large variety of non-conventional adsorbent materials have been also proposed and studied for their ability to remove dyes [7]. Several natural adsorbents, such as saw dust [8], peat [9], bagasse pith [10], orange peel [11], nut shells [12], and deoiled mustard [13] etc have been tried for this purpose. The present investigation reports the adsorption of dye congo red onto bagasse charcoal and banana peels. The experimental data is analyzed by adsorption isotherm models namely, Langmuir, Freundlich, Dubinin and Radushkevich, Temkin equation and Generalized Isotherm. Kinetics experiments are also conducted to determine the rate at which congo red is adsorbed on bagasse charcoal and banana peels. To investigate the adsorption process of congo red (CR), various constants are determined using pseudo first-order, pseudo second order, intraparticle diffusion and Bangham's model and discussed.

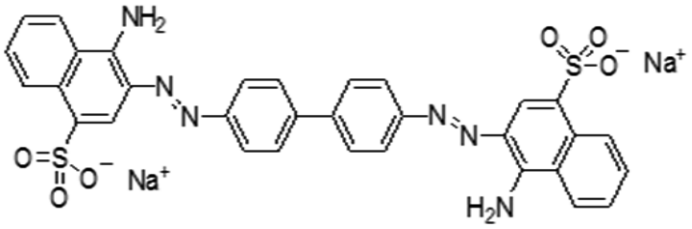
METHODS

MATERIALS

The dye Congo red supplied by S.D fine chemicals, Mumbai, India, was used as such without further purification. The physicochemical properties of the dye are presented in Table 1. For experimentation all the solutions were prepared after dilution of the

TABLE 1.

General characteristics of dye congo red.

Type	Congo red
Phase	solid
λ_{max}(nm)	497
Formula weight	696.65
Chemical structure	

stock solution, which was prepared in double distilled water. The adsorbents banana peels and bagasse are collected from local juice bar washed with distilled water. Banana peels was dried in sunlight, grinded then kept in hot air oven at 60°C for 24 hours. Bagasse Charcoal was prepared in a very economical way i.e by burning in the absence of free excess of air. Adsorbents were sieved through sieves having mesh number 240–200, to remove coarse particles, and the corresponding particle size of 70–75μm was obtained [14]. The adsorbents were dried at 100+5 °C for 24 h and kept in separate bags in vacuum desiccators.

Equilibrium studies :

All the batch adsorption studies were performed at room temperature (313 K). Different initial concentrations of the dye solution were taken in 250 ml conical flasks containing 0.1 g of adsorbent (bagasse charcoal or banana peels). The flasks were agitated at 120 rpm for a pre-determined time. Dye concentrations were measured using UV/VIS spectrophotometer (Shimadzu : UV 1800). The absorbance of colour was read at 497 nm and it was used to calculate the amount of dye adsorbed, q_t, mg/g. Control experiments were carried out for the adsorption of congo red in absence of adsorbents by the container walls. It was found that there was no degradation or adsorption of congo red by container walls. All the experiments were carried out in duplicate and the mean values are presented.

Calculation of dye concentration for isotherm studies :

The dye concentration on adsorbent at equilibrium, q_e, was calculated from

$$q_e = (C_o - C_e)V/m$$

where q_e = the dye concentration on the adsorbent at equilibrium (mg/g), C_0 = the initial dye concentration in the liquid phase (mg/L), C_e = the liquid-phase dye concentration at equilibrium (mg/L), V = the total volume of solution, and m = mass of adsorbent used (g).

RESULTS AND DISCUSSION

Effect of contact time :

Fig. 1 presents the effect of agitation time on the removal of congo red by bagasse charcoal and banana peels. The amount of dye adsorbed increased with increase in agitation time and reaches a constant value where no more dye is removed from the solution. At this point, the amount of dye being adsorbed onto the material is in a state of dynamic equilibrium with the amount of dye desorbed from the adsorbent. Curves were single, smooth and continuous leading to monolayer saturation of congo red dye on the adsorbent surface. Equilibrium times come out to be 100 and 70 minutes for BC and BP respectively.

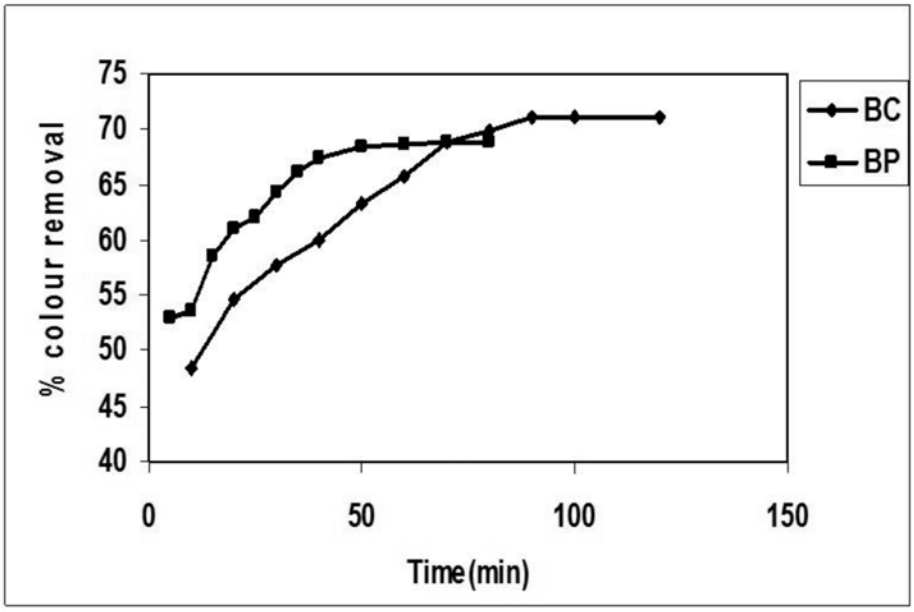


Fig. 1. Effect of contact time on the adsorption of dye congo red onto Bagasse charcoal (BC) and Banana peels (BP).

Effect of adsorbent dose :

Effect of adsorbent dose on removal of dye was studied by varying the dose of adsorbents (0.1 g to 0.9 g/100 ml) in the test solution while keeping the initial dye concentration as 60 mg L^{-1} at constant temperature (39 $\pm$ 1°C). Fig. 2 shows that adsorption percentage increases with increase in adsorbent dose. The maximum removal was found to be 93% for bagasse charcoal and 82% for banana peels. The increase in adsorption with adsorbent dosage can be attributed to the increased adsorbent surface and availability of more adsorption sites.

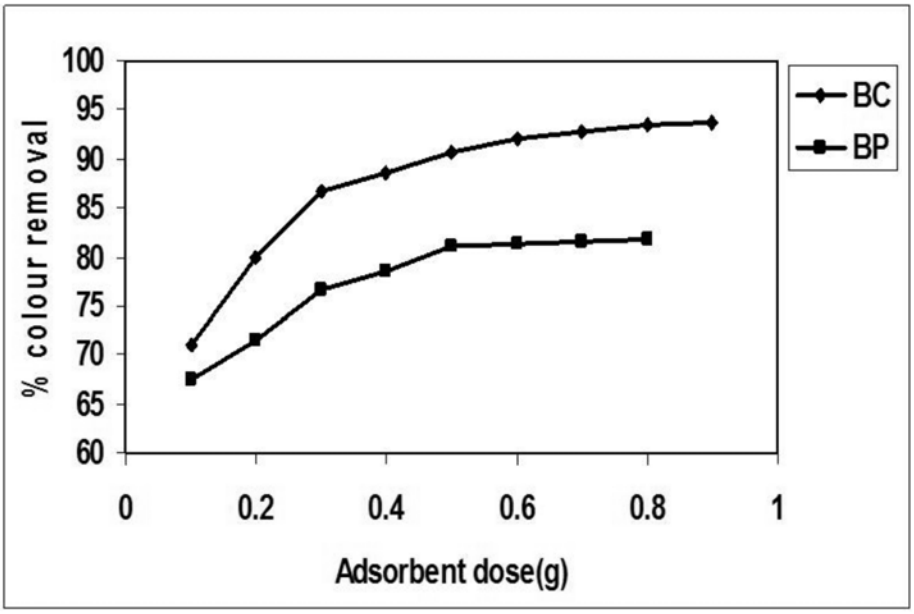


Fig. 2. Effect of adsorbent dose on the adsorption of dye congo red onto Bagasse charcoal (BC) and Banana peels (BP).

Effect of initial concentration :

The effect of initial concentration on the removal of CR by both the adsorbents is indicated in Fig. 3. Experiment was done at constant adsorbent dose 1g/L. It is evident from the figure that percentage CR removal decreases with increase in CR concentration. This is due to increase in CR concentration, surface area and active sites of the adsorbent were saturated and hence percentage adsorption decreases.

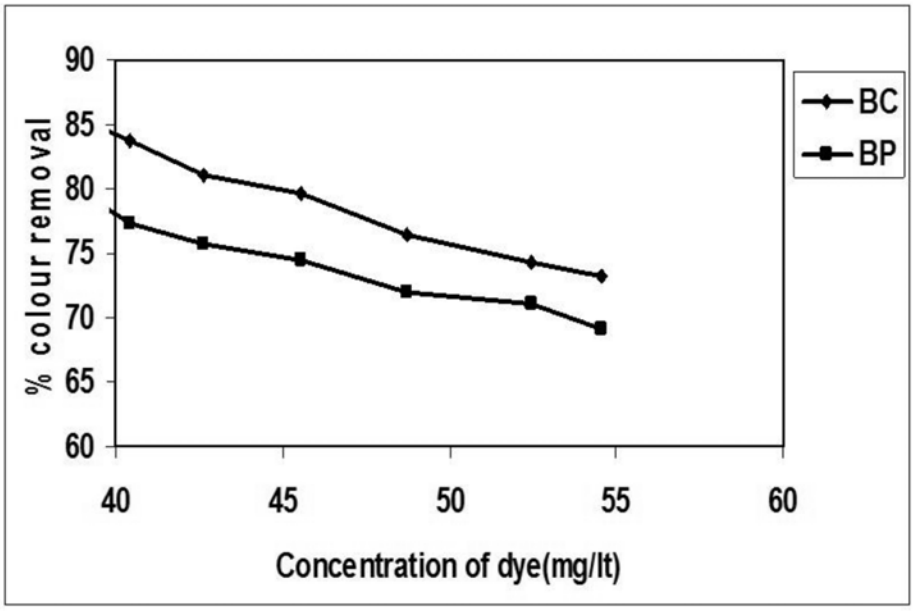


Fig. 3. Effect of initial concentration on the adsorption of dye congo red onto Bagasse charcoal (BC) and Banana peels (BP).

Effect of ionic strength :

Since large amounts of salts are generally utilized in the dyeing process, the effects of ionic strength on adsorption must be evaluated. The effect of ionic strength on the adsorption of the dye was investigated by adding 0.1 g of adsorbent (BC and BP) in 100 ml of the solution with varying concentration of NaCl and KCl (0.01–0.05 M). Fig. 4 shows that increase in the concentration of NaCl and KCl promotes the adsorption of dye. This can be attributed to the aggregation of dye molecules induced by the action of salt ions, i.e., salt ions force dye molecules to aggregate, increasing the extent of adsorption of dyes onto BC and BP [15].

Adsorption kinetic study :

Pseudo first order and Pseudo second order models :

The pseudo-first-order equation is given as [16] :

$$\frac{dq_t}{dt} = k_f (q_e - q_t) \qquad (1)$$

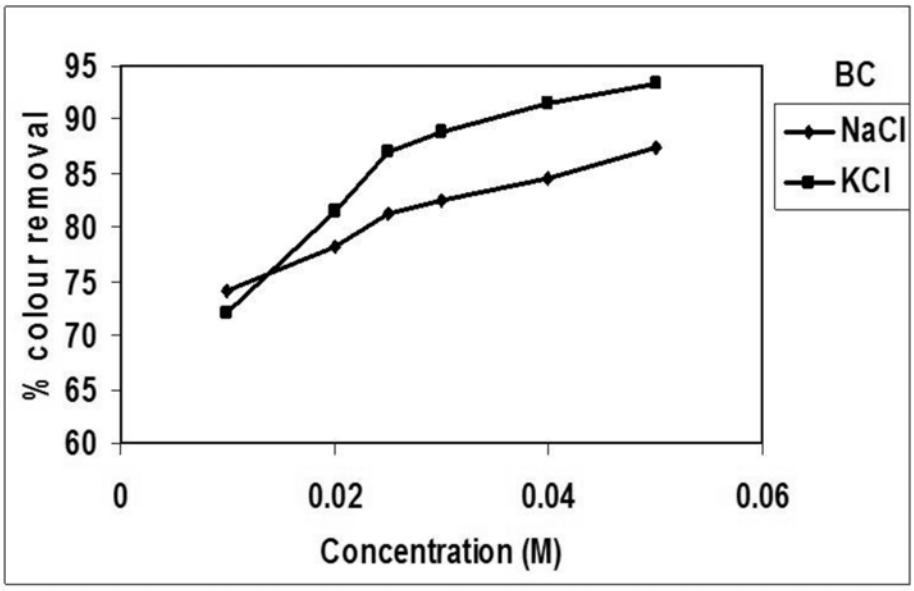


Fig. 4. (a) Effect of NaCl and KCl on adsorption of dye congo red onto Bagasse charcoal (BC).

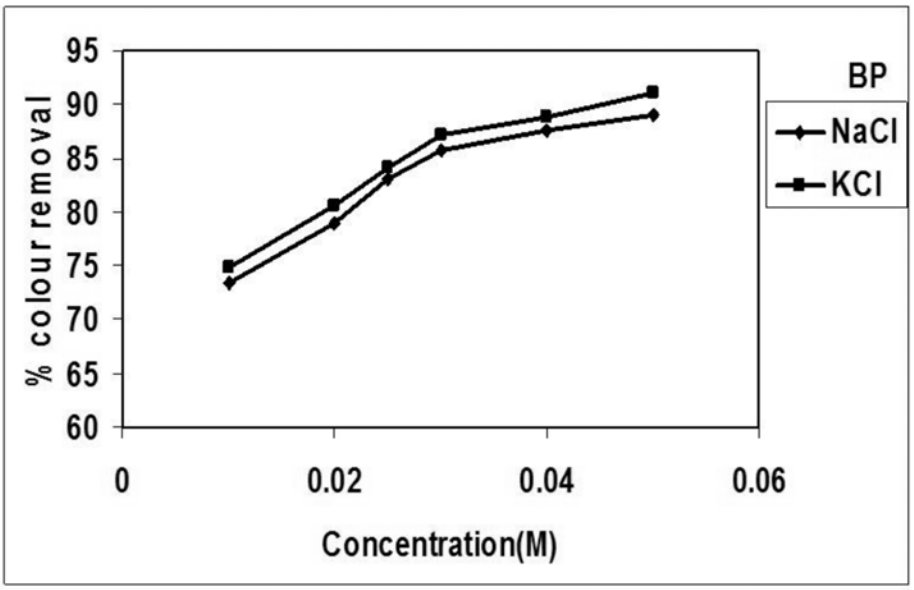


Fig. 4. (b) Effect of NaCl and KCl on adsorption of dye congo red onto Banana peels (BP).

Where q_t (mg/g) is the amount of dye adsorbed at time t. q_e (mg/g) is the adsorption capacity at equilibrium, k_f (min^{-1}) is the pseudo first order rate constant, and t is the contact time (min). The integration of eq (1) with initial condition ($q_t = 0$ at $t = 0$) leads to following equation :

$$\log (q_e - q_t) = \log q_e - \frac{k_f}{2.303} t \quad (2)$$

The values of k_f, calculated from the linear plots of log (q_e-q_t) vs t, for the adsorption of dye CR on both the adsorbents are given in Table 2. These curves are linear, however linearity of these curve does not necessarily assure first order mechanism [17] due mainly to the inherent disadvantage of correctly estimating equilibrium adsorption capacity (q_e). The q_e values obtained from Lagergren plots is differed from the experimental q_e values, therefore first order kinetic is less likely to explain the rate processes.

The pseudo-second-order model is represented as [18] :

$$\frac{dq_t}{dt} = k_s (q_e - q_t)^2 \quad (3)$$

Where k_s is the pseudo second order rate constant (g mg^{-1} min^{-1}). Integrating equation (3) and noting that $q_t = 0$ at $t = 0$, the following equation is obtained.

$$\frac{t}{q_t} = \frac{1}{k_s q_e^2} + \frac{1}{q_e} t \quad (4)$$

The equilibrium adsorption capacity, q_e is obtained from the slope and k_s is obtained from the intercept of the plot t/q_t vs t. The values are given in Table 2. The q_e experimental and the q_e calculated values from the pseudo second-order kinetic model are very close to each other. The calculated correlation coefficients are also close to unity ($R^2 = 0.99$) for pseudo second order model. Results imply that adsorption of congo red on studied adsorbents nicely followed the pseudo second order kinetic model.

Intra-particle diffusion study :

An empirically found functional relationship common to most adsorption process is that the uptake varies almost proportionally with $t_{½}$, the Weber-Morris plot, rather than with the contact time, t [19].

$$q_t = k_{id} t_{½} + C \quad (5)$$

TABLE 2.

Kinetic parameters for the adsorption of dye congo red onto Bagasse charcoal (BC) and Banana peels (BP).

Equations	Parameters	Adsorbents	
		Bagasse charcoal	Banana peels
Pseudo first order	q_e exp (mg/g)	41.11	39.60
	q_e cal (mg/g)	77.99	33.17
	k_f (min^{-1})	0.069	0.099
	R^2	0.91	0.96
Pseudo-second-order	q_e exp (mg/g)	41.11	39.60
	q_e cal (mg/g)	44.96	41.87
	k_s (g/(mg min))	2.15×10^{-3}	6.39×10^{-3}
	R^2	0.99	0.99
Intra-particle diffusion	k_{id}(mg/g $min^{1/2}$)	2.35	2.33
	C(mg/g)	19.32	23.61
	R^2	0.99	0.98
Bangham	k_o(g)	27.84	45.11
	α	0.31	0.21
	R^2	0.99	0.97
Elovich	a	1.07×10^{5}	2.34×10^{9}
	b	0.15	0.20
	R^2	0.99	0.97

where k_{id} is the intra-particle diffusion rate constant. The values of k_{id} and C obtained from the slope and intercept of linear plots (q_t vs $t_{1/2}$) are listed in Table 2. Values of the intercept (C) gives an idea about the thickness of boundary layer i.e. larger the intercept the greater is the boundary layer effect [20]. This is attributed to the instantaneous utilization of the most readily available adsorbing sites on the adsorbent surface.

Bangham's equation :

Kinetic data were further used to know about the slow step occurring in the present adsorption system using Bangham's equation [21].

$$\text{Log log}(C_o/C_o - q_t m) = \log(k_o m/2.303V) + \alpha \log t \quad (6)$$

Where C_o is the initial concentration of dye in solution (mg/L). V is the volume of the solution (ml), m is the weight of adsorbent per liter of solution (g/L). q_t (mg/g) is amount of dye adsorbed at time t. $\alpha(<1)$ and k_o are constants, values given in Table 2. The linear double logarithmic plot (Log log $(C_o/C_o - q_t m)$ vs log t) reveals that the diffusion of adsorbate into pores of adsorbents is not the only rate controlling step [22].

Elovich model :

The most interesting model to describe the activated chemisorptions is Elovich equation [23]

$$q_t = \frac{1}{b} \ln(ab) + \frac{1}{b} \ln t \quad (7)$$

where q_t (mg/g) is the amount of dye adsorbed at time t (min) and a and b are constants. The constant 'a' is considered as the initial sorption rate (mg/(g min)) and b is related to the extent of surface coverage and activation energy for chemisorption (g/mg). The values of these constants obtained from the slope and intercept of the linear plot of q_t vs ln t, are given in Table 2.

Adsorption Isotherms :

To describe the equilibrium nature of adsorption various isotherm equations viz Langmuir, Freundlich, Dubinin and Radushkevich, Temkin equations and Genralized Isotherm have been used.

Freundlich isotherm :

This isotherm is an empirical equation employed to describe heterogeneous system. Freundlich isotherm is also applied to plot the equilibrium data of the adsorption. The linear form of Freundlich equation can be expressed as [24].

$$\log x/m = \log K_F + 1/n \log C_e \quad (8)$$

Where x is the amount of dye adsorbed (mg), m is the weight of the adsorbent used (g), C_e is the equilibrium concentration of the dye in solution (mg/L). K_F and n are Freundlich constant. n is heterogeneity factor and K_F indicates the adsorption capacity. The value of $n>1$, reflecting the favorable adsorption. Freundlich plots are linear and linearity suggests that fit is well for the adsorption system under the studied concentration range. The values of n and K_F are calculated from the slopes and intercepts of the linear plots (log (x/m) vs log q_e) and are given in Table 3. Adsorption capacity of BC is slightly more than BP.

TABLE 3.

Isotherm parameters for the adsorption of dye congo red onto Bagasse charcoal (BC) and Banana peels (BP).

Equations	Parameters	Adsorbents	
		Bagasse charcoal	Banana peels
Freundlich	K_F (mg/g)(L/mg)$^{1/n}$	23.91	19.71
	n	5.37	4.46
	R^2	0.98	0.96
Langmuir	K_L (L/mg)	0.45	0.30
	C_m (mg/g)	45.31	44.38
	R^2	0.99	0.98
Dubinin Radushkevih	q_s (mg/g)	39.46	36.73
	E(KJ/mol)	0.72	0.61
	R^2	0.90	0.86
Temkin	K_T (L/mg)	23.77	8.42
	B_1	6.72	7.47
	R^2	0.97	0.95
Generalized	N	0.96	0.94
	K_G (mg/L)	2.04	2.87
	R^2	0.96	0.93

Langmuir Isotherm :

The Langmuir isotherm model is valid for monolayer adsorption onto a surface containing a finite number of identical sites. The linear form of Langmuir Isotherm is represented by the following equation

$$C_e/q_e = C_e/C_m + 1/K_L C_m \quad (9)$$

Where C_e is the concentration of dye solution (mg/L) at equilibrium and q_e(mg/g) is the adsorption capacity at equilibrium. The constant C_m signifies the adsorption capacity (mg/g) when monolayer is complete and K_L is related to the affinity of the binding sites. The values of K_L and C_m (monolayer concentration) were calculated from the intercept and slope of the plots (Ce/q_e vs C_e) are given in Table 3. In the present study monolayer concentration was found to be higher for basic dye congo

red as compare to acidic dyes studied elsewhere [10]. In the present study, maximum adsorption capacity was found to be 45.3 and 44.4 mg/g for BC and BP respectively, however activated carbon shows the adsorption capacity of 300 mg/g [25] and activated carbon prepared from coir pith have capacity of 6.7 mg/g [26] for the removal of dye congo red. So results suggest that BC and BP can be used as an effective adsorbent for the dye removal from industrial effluents.

The essential feature of the Langmuir isotherm to identify the feasibility and favorability of the adsorption process can be expressed by a dimensionless constant called separation factor (R_L) was adopted. The separation factor (R_L) was calculated in each case using the following equation :

$$R_L = 1/(1 + K_L C_o) \quad (10)$$

Where C_o is the initial dye concentration (mg/L).The value of R_L lies between 0 and 1 for favorable adsorption, while $R_L > 1$ represent unfavorable adsorption, and $R_L = 1$ represent linear adsorption while the adsorption process is irreversible if $R_L = 0$ [27]. The values of 'R_L' were found to be less than unity for the studied adsorbents shown in Table 4, indicates highly favorable adsorption for the dye congo red on studied biosorbents.

Temkin Isotherm :

The Temkin isotherm equation suggests a linear decrease of sorption energy as the degree of completion of the sorptional centers of an adsorbent is increased. This model

TABLE 4.

Values of separation factor (R_L) at various initial concentrations of dye congo red.

S.No.	Adsorbents			
	Bagasse charcoal (K_L = 0.45 L/mg)		Banana peels (K_L = 0.30 L/mg)	
	C_o(mg/L)	R_L	C_o(mg/L)	R_L
1.	54.56	0.039	54.56	0.057
2.	52.47	0.041	52.47	0.059
3.	48.75	0.043	48.75	0.063
4.	45.53	0.046	45.53	0.068
5.	42.61	0.049	42.61	0.072
6.	40.45	0.052	40.45	0.076
7.	37.90	0.055	37.90	0.080

takes into account the presence of indirect adsorbate/adsorbent interactions and suggests that because of these interactions the heat of adsorption of all molecules in the layer would decrease linearly with coverage [28]. The Temkin isotherm has generally been applied in the following form :

$$q_e = B_1 \ln K_T + B_1 \ln C_e \quad (11)$$

The constant K_T and B_1 are calculated from a linear plot of q_e verses ln C_e and are given in Table 3. K_T is the equilibrium binding constant (L/mg) corresponding to maximum binding energy and constant B_1 is related to heat of adsorption.

Dubinin and Radushkevich (D-R) Isotherm :

D-R isotherm is generally used to describe the sorption isotherms of single solute system. The D-R isotherm, apart from being analogue of Langmuir isotherm, is more general than Langmuir isotherm as it rejects the homogenous surface or constant adsorption potential [29]. It is expressed as :

$$\ln q_e = n\, q_s - B\epsilon^2 \quad (12)$$

Where q_s is D-R constant and ϵ can be correlated as

$$\epsilon = RT \ln (1 + 1/C_e) \quad (13)$$

Where q_s is the maximum amount of adsorbate that can be adsorbed on adsorbent, B is the constant related to energy, and C_e is the equilibrium concentration (mg/L). R is Universal gas constant, 8.314 J mol^{-1} K^{-1}, T is the temperature (K). The mean free energy E of the adsorption per molecule of adsorbate can be calculated using the following equation :

$$E = 1/ (2B)^{1/2} \quad (14)$$

The D-R isotherm is plotted against experimental values and the calculated D-R constant are given in Table 3. Mean free energy is more for BC.

Generalized Isotherm :

The Generalized Isotherm has been used in the following form [30].

$$\ln[(q_{max}/q_{e)} - 1] = \ln K_G - N \ln C_e \quad (15)$$

Where K_G is the saturation constant (mg/L), N is the cooperative binding constant, q_{max} is the maximum adsorption capacity of the adsorbent (mg/g). q_e (mg/g) and C_e (mg/L) are the equilibrium dye concentrations in the solid and liquid phase, respectively. The constants N and K_G are calculated from the slope and intercept of the linear plot of ($\ln[q_{max}/q_e - 1]$ verses ln C_e)and the values are given in Table 3.

CONCLUSION

Equilibrium and kinetics studies were made for the adsorption of dye congo red onto bagasse and banana peels. The percentage removal of the dye increased with increasing the ionic strength of the solution, this was attributed to dye aggregation in solution. The adsorption of the dye CR onto BC and BP follows pseudo second order model. The fit of the Langmuir model in the present system shows the formation of a monolayer covering of the adsorbate at the surface of the adsorbents. The maximum adsorption capacities were found to be 45.3 and 44.4 mg/g for bagasse and banana peels respectively and is greater than acidic dyes for bagasse. It can be recapitulated that adsorbent BC is selective towards basic dyes. High value of monolayer concentration (C_m, Langmuir constant), adsorption capacity (K_F, Freundlich constant) and equilibrium binding constant (K_T) alluded that BC is better adsorbent than BP. It is also strengthened by high percentage removal of congo red (72%) by BC. The values of dimensionless equilibrium parameter separation factor (R_L) demonstrate the favorability of the adsorption of basic dye congo red on both the studied adsorbents. Hence BC and BP can be an attractive option for the dye removal from local dye industry.

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