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Removal of Congo Red from water by adsorption onto activated carbon prepared from coir pith, an agricultural solid waste

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Abstract

The adsorption of Congo Red by coir pith carbon was carried out by varying the parameters such as agitation time, dye concentration, adsorbent dose, pH and temperature. Equilibrium adsorption data followed both Langmuir and Freundlich isotherms. Adsorption followed second-order rate kinetics. The adsorption capacity was found to be 6.7 mg dye per g of the adsorbent. Acidic pH was favourable for the adsorption of Congo Red. Desorption studies suggest that chemisorption might be the major mode of adsorption. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Congo Red; Adsorption; Coir pith carbon; Isotherms; pH Effect; Desorption studies

1. Introduction

In developing countries like India, industries cannot afford to use conventional wastewater treatment chemicals like alum, ferric chloride, polymer flocculants and coal based activated carbon because they are not cost-effective. An inexpensive and more easily available adsorbent would make the removal of pollutants an economically viable alternative. Agricultural wastes like coir pith are discarded in the agricultural sector in India. Annual production is around 7.5 million tons [1]. Coir pith, which constitutes as much as 70% of the coconut husk, is a light fluffy material that is generated in the separation process of the fibre from the coconut husk. Coir pith is discarded as a waste and its accumulation around coir-processing centers is creating a menace.

Nowadays thousands of dyes are prepared for printing and dyeing industries from coal-tar based hydrocarbons such as benzene, naphthalene, anthracene, toluene, xylene, etc. 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 [2]. In particular, printing and dyeing unit wastewaters contain several types of coloring agents, which are difficult to be treated by biological methods [3].

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Dyes even in low concentrations affect the aquatic life and the food web. Some dyes are carcinogenic and mutagenic. Hence ways and means are required to remove the dyes from wastewater.

Activated carbon adsorption is the most popular physico-chemical treatment for the removal of dissolved organics from wastewaters. The various phenomena pertaining to decolorisation and adsorptive efficiencies of carbon have been correlated and shown to be directly proportional to the "activity" of the carbon. A number of non-conventional, low cost adsorbents have been tried for dye removal. These include wood [4], Fuller's earth and fired clay [5], fly ash [6], biogas waste slurry [7–11], waste Fe(III)/Cr(III) hydroxide [12], waste orange peel [13], banana pith [14,15], peat [16,17], chitin [18], chitosan[19], silica [20] and others [21–26]. Adsorption studies for dye removal have been carried out using activated carbon made from non-conventional sources as adsorbents [27–29]. However, as the adsorption capacities of the above adsorbents are not large, new absorbents are still under development. Activated carbons obtained from rubber seed coat, palm seed coat and myrobalan waste were investigated for the removal of a wide variety of impurities from water and wastewater. In general, these carbons will be as efficient in the adsorption of both organics and inorganics as the commercial activated carbons [30]. Commercial activated carbons are sophisticated in the sense that they are designed for a variety of applications. If low cost non-conventional sources are used to prepare activated carbons for a specific purpose, then they will be economical for wastewater treatment. The objective of this study was to investigate the feasibility of using carbonized coir pith for the removal of Congo Red, a toxic dye, from wastewater by adsorption method.

2. Experimental

2.1. Material

Coir pith was collected from nearby coir industries, dried in sunlight for 5 h and ground. The dried coir pith powder was sieved to 250–500 μm in size. It was subjected to carbonization at 700 °C for 1 h using a muffle furnace under closed condition. The carbonized material was taken out, sieved to 250–500 μm size again and used for adsorption studies.

2.2. Methods

2.2.1. Adsorption studies

Adsorption experiments were carried out by agitating 200 mg of adsorbent with 50 ml of dye solution of the desired concentration and pH at 200 rpm, 35 °C in a thermostated rotary shaker (ORBITEK, Chennai, India). Dye concentration was estimated spectrophotometrically by monitoring the absorbance at 495.7 nm using a UV–vis spectrophotometer (Hitachi, model U-3210, Tokyo). pH was measured using a pH meter (Elico, model LI-107, Hyderabad, India). The samples were withdrawn from the shaker at predetermined time intervals and the dye solution was separated from the adsorbent by centrifugation at 20,000 rpm for 20 min. The absorbance of supernatant solution was measured. Effect of pH was studied by adjusting the pH of dye solutions using dilute HCl and NaOH solutions. Effect of adsorbent dosage was studied with different adsorbent doses (100–900 mg) and 50 ml of 20, 40, 60, 80 mg/L dye solutions at equilibrium time. Langmuir and Freundlich isotherms were employed to study the adsorption capacity of the adsorbent.

2.2.2. Desorption studies

The adsorbent that was used for the adsorption of 20 and 40 mg/l of dye solution was separated from the dye solution by centrifugation. The dye-loaded adsorbent was filtered using Whatman filter paper and washed gently with water to remove any unadsorbed dye. Several such samples were prepared. Then the spent adsorbent was agitated with 50 ml of distilled water, adjusted to different pH values for 40 min. The desorbed dye was estimated as before.

2.2.3. Temperature studies

Adsorption of 20 mg/l of Congo Red dye by 100 mg of adsorbent was carried out at 35, 40, 50 and 60 °C in the thermostated rotary shaker.

3. Results and discussion

3.1. Effects of agitation time and concentration of dye on adsorption

Effects of agitation time and dye concentration on removal of dye by coir pith carbon are presented in Fig. 1. The amount of dye adsorbed (mg/g) increased with increase in agitation time and reached equilibrium after 10 min for the dye concentrations 20, 40, 60 and 80 mg/l used in this study. The adsorption density (mg/g) increased with increase in dye concentration. The per cent dye removal at equilibrium decreased from 66.5 to 30.5 as the dye concentration was increased from 20 to 80 mg/l. It is clear that the removal of dyes depends on the concentration of the dye. The removal curves are single, smooth and continuous leading to saturation.

3.2. Adsorption dynamics

3.2.1. Adsorbent rate constant

The rate constant of adsorption is determined from the first order rate expression given by Lagergren [31]:

$$\log(q_e\text{-}q) = \log q_e - k_1 t/2.303 \qquad (1)$$

where q_e and q are the amounts of dye adsorbed (mg/g) at equilibrium and at time t (min), respectively, and k_1 is the rate constant of adsorption (1/min). Values of k_1 were calculated from the plots of $\log(q_e - q)$ vs t (figures not shown) for different concentrations of the dye. The correlation coefficients were low. Also the experimental q_e values do not agree with the calculated ones, obtained from the linear plots (Table 1). This shows that the adsorption of dye onto coir pith carbon is not a first order reaction.

The second-order kinetic model [32,33] is expressed as

$$t/q = 1/k_2 q_e 2 + t/q_e \qquad (2)$$

where k_2 (min g/mg) is the rate constant of second order adsorption. If second-order kinetics is applicable, the plot of t/q vs t should show a linear relationship. There is no need to know any parameter beforehand and the equilibrium adsorption capacity, q_e can be calculated from Eq. (2). Also, it is more likely to predict the behaviour over the

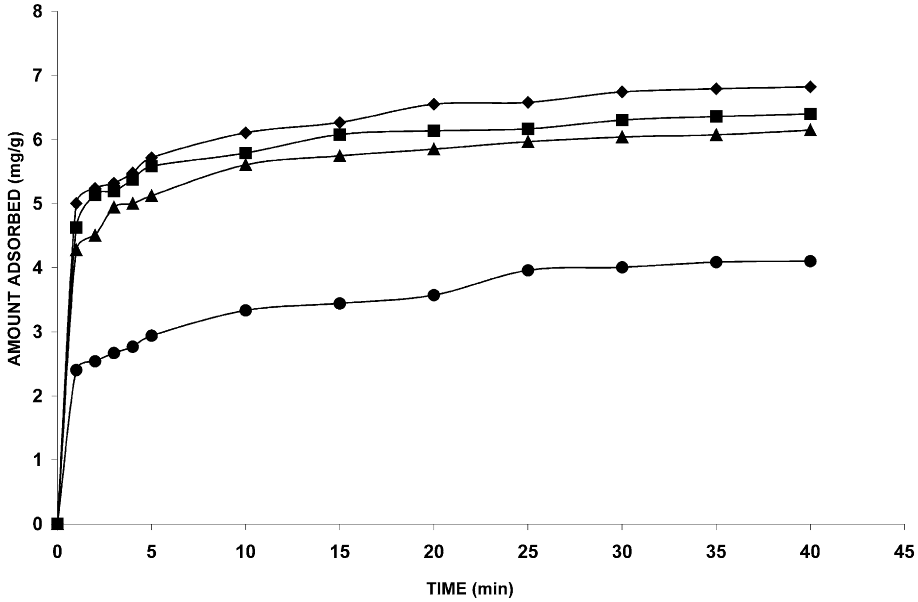

Fig. 1. Effect of agitation time and concentration of Congo Red on removal: adsorbent dose, 200 mg/50 ml; dye concentration (●) 20 mg/L, initial pH, 7.70, (▲) 40 mg/L, initial pH, 7.58 (■) 60 mg/L, initial pH 7.65, (O) 80 mg/L, initial pH, 8.19; temperature, 35 °C.

Table 1
Comparison of the first- and second-order adsorption rate constants and calculated and experimental q_e values for different initial dye concentrations and temperatures

Parameter	First-order kinetic model				Second-order kinetic model		
	q_e (exp) (mg/g)	k_1 (1/min)	q_e (cal) (mg/g)	R^2	k_2 (g/ mg/ min)	q_e (cal) (mg/g)	R^2
Initial dye concentration (mg/l)							
20	3.333	0.21	1.188	0.966	0.83	3.096	0.996
40	5.606	0.26	1.708	0.950	0.59	5.408	0.998
60	5.789	0.39	1.680	0.942	0.61	5.821	0.999
80	6.107	0.24	1.462	0.934	0.71	5.886	0.998
Temperature (°C)							
35	4.527	0.36	1.343	0.956	0.70	4.546	0.999
40	4.746	0.17	1.140	0.990	1.05	4.413	0.999
50	4.866	0.56	0.581	0.986	2.30	4.904	1.000
60	5.137	0.25	0.780	0.978	1.50	5.023	0.999

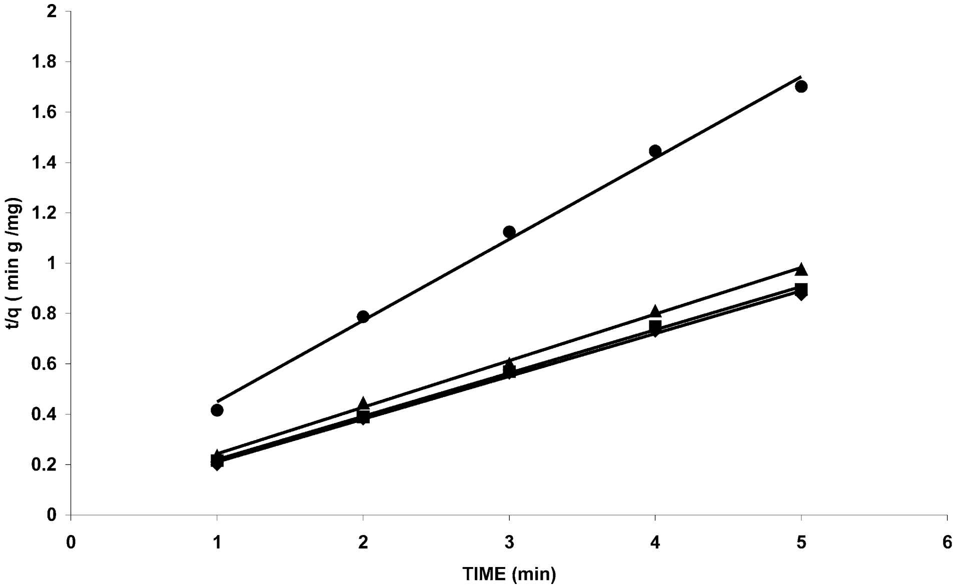

Fig. 2. Plots of the pseudo second-order model at different initial dye concentrations: adsorbent dose, 200 mg/50 ml; dye concentration (●) 20 mg/l, initial pH, 7.70, (▲) 40 mg/l, initial pH, 7.58 (■) 60 mg/l, initial pH, 7.65, (O) 80 mg/l, initial pH, 8.19; temperature, 35 °C.

whole range of adsorption [32,33]. The second-order rate constant, k_2 and q_e were calculated from the intercept and slope of the plots of t/q vs t. The linear plots of t/q vs t show a good agreement of experimental data with the second-order kinetic model for different initial dye concentrations (Fig. 2). The correlation coefficients for the second-order kinetic model are greater than 0.995 (Table 1). Also, the calculated q_e values agree very well with the experimental data. These indicate that the adsorption system belongs to the second-order kinetic model. Similar phenomena are observed in the biosorption of dye Remazol Black B on biomass [25,26].

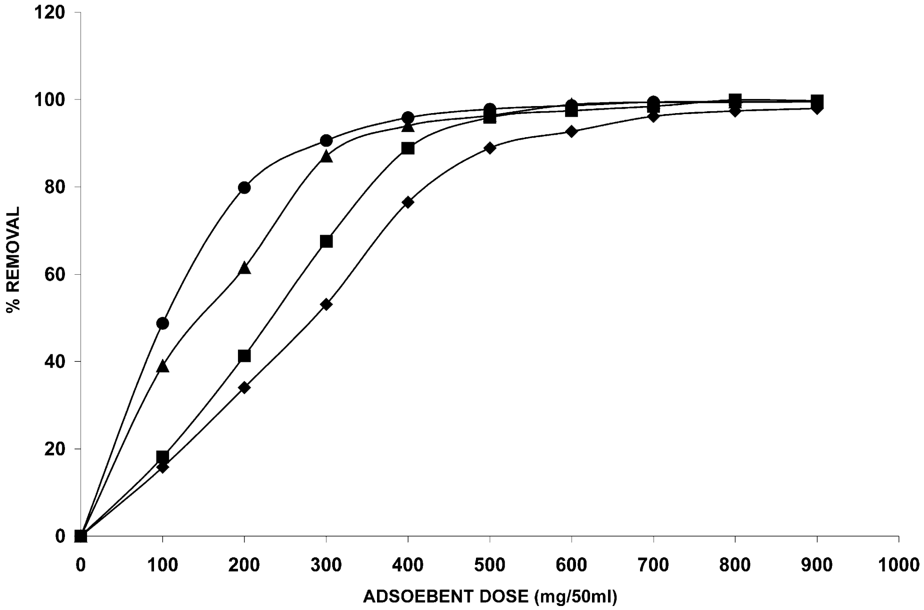

Fig. 3. Effect of adsorbent dosage on removal of Congo Red by coir pith carbon: agitation time, 40 min; phenol concentration (●) 20 mg/l, initial pH, 7.70, (▲) 40 mg/l, initial pH, 7.58 (■) 60 mg/l, initial pH, 7.65, (O) 80 mg/l, initial pH, 8.19; temperature, 35 °C.

3.3. *Effect of adsorbent dosage*

Fig. 3 shows the removal of Congo Red by coir pith carbon at different adsorbent doses (100–900 mg/50 ml) for the dye concentrations 20, 40, 60 and 80 mg/l. Increase in adsorbent dosage increased the per cent removal of dye, which is due to the increase in absorbent surface area of the adsorbent.

3.4. *Adsorption isotherms*

Langmuir isotherm is represented by the following equation [34]:

$$C_e/q_e = 1/Q_0b + C_e/Q_0 \tag{3}$$

where C_e is the concentration of dye solution (mg/l) at equilibrium. The constant Q_0 signifies the adsorption capacity (mg/g) and b is related to the energy of adsorption (l/mg). The linear plot of C_e/q_e vs C_e shows that adsorption follows a Langmuir isotherm (Fig. 4). Values of Q_0 and b were calculated from the slope and intercept of the linear plots and are presented in Table 2. The applicability of the Langmuir isotherm suggests the monolayer coverage of the dye on the surface of the coir pith carbon.

Table 2
Langmuir constants

Dye concentration (mg/l)	Q_0 (mg/g)	b (l/mg)	R_L
20	6.72	0.18	0.21
40			0.12
60			0.08
80			0.06

The essential characteristics of the Langmuir isotherm can be expressed by a dimensionless constant called equilibrium parameter, R_L [35] defined by

$$R_L = 1/(1 + bC_0) \tag{4}$$

where b is the Langmuir constant and C_0 is the initial dye concentration (mg/l), R_L values indicate the type of isotherm. An R_L value between 0 and 1 indicates favorable adsorption [36]. The R_L

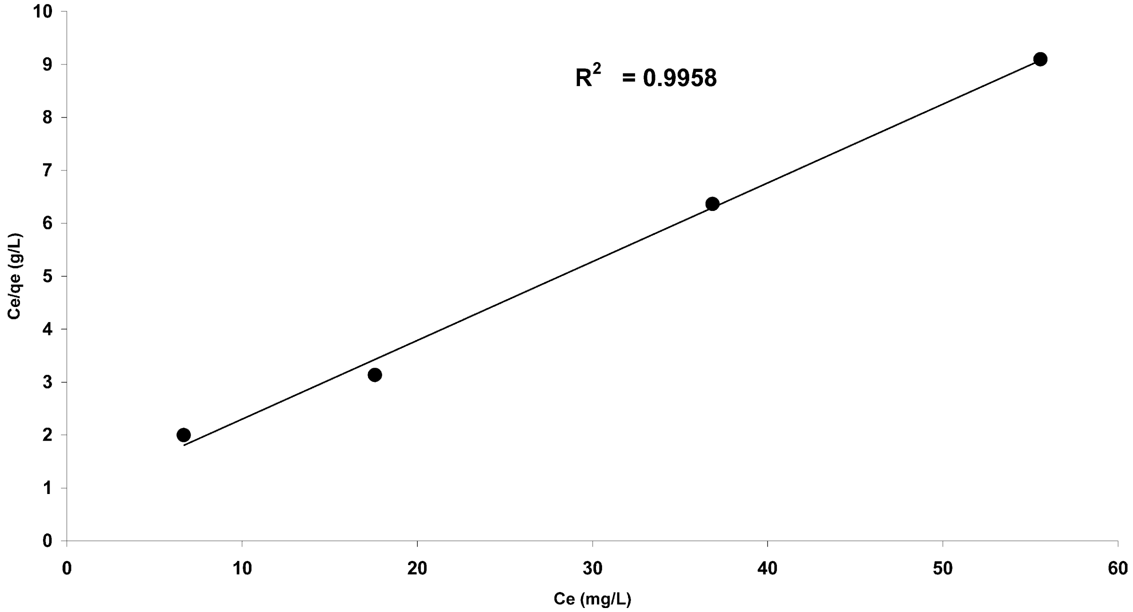

Fig. 4. Langmuir plot for the adsorption of Congo Red by coir pith carbon.

values were found to be between 0 and 1 for dye concentrations of 20, 40, 60 and 80 mg/L (Table 2).

The Freundlich isotherm was also applied for the adsorption of the dye [12]:

$$\log_{10}(x/m) = \log_{10}k_f + (1/n)\log_{10}C_e \quad (5)$$

where x is the amount of dye adsorbed (mg), m is the weight of the adsorbent used (g), C_e is equilibrium concentration of dye in solution (mg/l), k_f and n are constants incorporating all factors affecting the adsorption process (adsorption capacity and intensity of adsorption). Linear plots of $\log_{10}(x/m)$ vs $\log_{10} C_e$ for the dye concentrations, 20 and 40 mg/l show that the adsorption also follows the Freundlich isotherm (Fig. 5). For the dye concentrations 60 and 80 mg/l, the correlation constants are very low (0.1687 and 0.2394, respectively) and hence the adsorption does not follow the Freundlich isotherm at these high concentrations. In general, as the k_f value increases the adsorption capacity of the adsorbent for a given dye increases. Values of k_f and n were calculated from the intercept and slope of the plots and are presented in Table 3.

3.5. pH effect

Effect of pH on the removal of Congo Red is shown in Fig. 6. For 20 mg/l dye concentration the per cent removal decreased from 70 to 57, when the pH was increased from 2 to 4 and then the per cent removal remained almost the same up to pH 10. For 40 mg/l dye concentration the per cent removal decreased from 50 to 42, when the pH was increased from 2 to 4 and then the per cent removal remained almost the same up to pH 10. Two possible mechanisms of adsorption of Congo Red on coir pith carbon may be considered: (a) electrostatic interaction between the protonated groups of carbon and acidic dye and (b) the chemical reaction between the adsorbate and the adsorbent. At pH 2.0 a significantly high eletrostatic attraction exists between the positively-charged surface of the adsorbent and anionic dye. As the pH of the system increases, the number of negatively charged sites increases and the number of positively charged sites decreases. A negatively charged surface site on the adsorbent does not favour the adsorption of dye anions due to the electrostatic repulsion. Also, lower adsorption of Congo Red at alkaline pH is due to the presence of

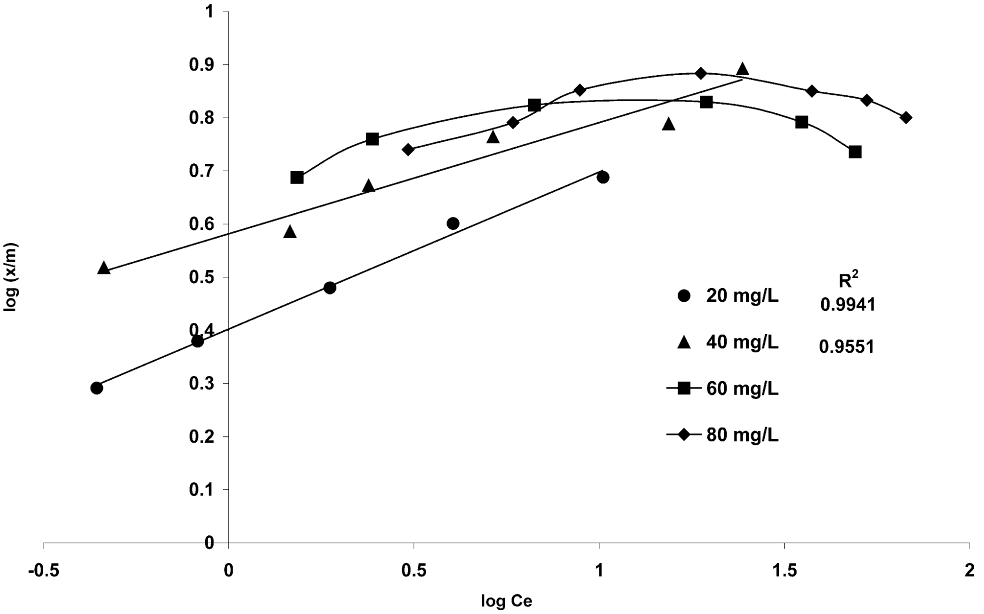

Fig. 5. Freundlich plots for adsorption of Congo Red by coir pith carbon.

excess OH^- ions competing with the dye anions for the adsorption sites. At alkaline pH significant adsorption of the anionic dye on the adsorbent still occurred. This suggests that the second mechanism, i.e. chemisorption, might be operative. A similar trend was observed for the adsorption of Congo Red [7], Acid Brilliant Blue [10], and Acid Violet [11] on biogas residual slurry and adsorption of Congo Red on waste orange peel [13] and banana pith [14] and adsorption of Acid Violet on coir pith carbon [37].

3.6. Desorption studies

To make the adsorption process more economical, it is necessary to re-generate the spent carbon and dye. As the desorbing pH was increased, the per cent desorption increased from 0.5 at pH 2 to 9.0 at pH 10 for the dye concentration of 20 mg/l. The per cent desorption increased from 1.2 at pH 2 to 11 at pH 10 for the dye concentration of 40 mg/l (Fig. 7). Very low desorption of dye suggests that chemisorption might be the major mode of dye removal by the adsorbent. This is also substantiated by the significant adsorption of the dye in the alkaline pH though electrostatic attraction does not favour the adsorption.

Table 3
Freundlich constants

Dye concentration (mg/l)	k_f	n
20	2.53	3.39
40	3.82	4.78

3.7. Effect of temperature

Effect of agitation time and temperature on Congo Red is shown in Fig. 8. Increase of temperature slightly increased the per cent removal. The change in standard free energy, enthalpy and entropy of adsorption were calculated using the following equations:

$$\Delta G^0 = -RT\ln K_c$$

where R is gas constant and K_c is the equilibrium constant and T is the temperature in ^{0}K.

According to the van't Hoff equation,

$$\log_{10} K_c = \Delta S^0/2.303R - \Delta H^0/2.303RT \quad (7)$$

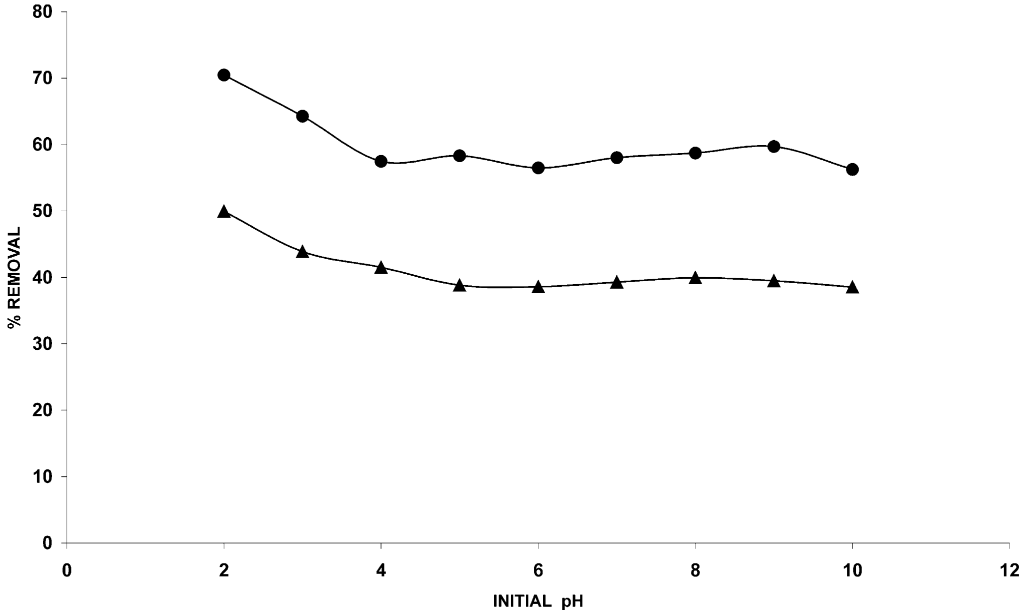

Fig. 6. Effect of pH on removal of Congo Red by coir pith carbon. Adsorbent dose, 200 mg/50 ml; agitation time, 40 min; dye concentration (●) 20 mg/l, (▲) 40 mg/l; temperature, 35 °C.

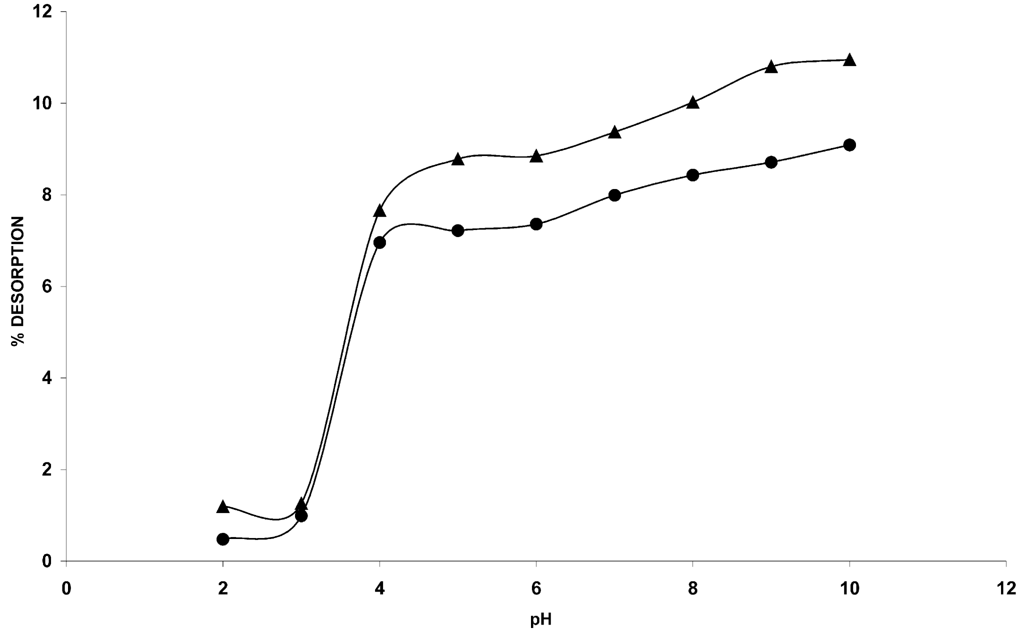

Fig. 7. Effect of pH on desorption of dye from dye-loaded adsorbent. Adsorbent dose, 200 mg/50 ml; agitation time 40 min; dye concentration (●) 20 mg/l, (▲) 40 mg/l; temperature, 35 °C.

where ΔS^0 and ΔH^0 are change in entropy and enthalpy of adsorption, respectively. Plots of log K_c vs $1/T$ is linear (Fig. 9). Values of ΔH^0 and ΔS^0 were evaluated from the slope and intercept of van't Hoff plots (Table 4). The positive values of ΔH^0 confirm the endothermic nature of adsorption. The endothermic adsorption has also been reported for the adsorption of Direct Red 12B on biogas waste slurry [9]. The negative value of ΔG^0 at 60 °C indicates spontaneous nature of

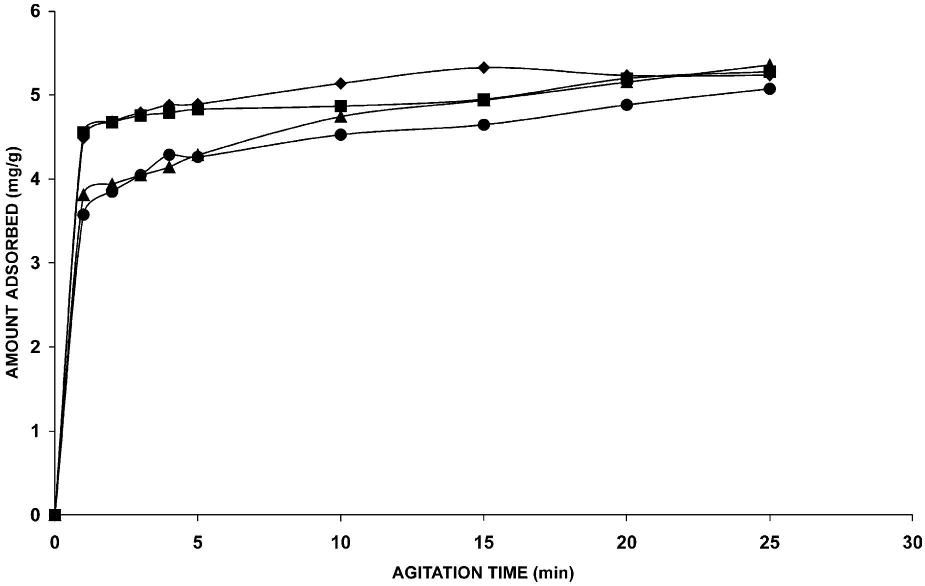

Fig. 8. Effect of temperature on removal of Congo Red by coir pith carbon: adsorbent dose, 100 mg/50 ml; dye concentration 20 mg/l; initial pH, 6.3; temperature (●) 35 °C, (▲) 40 °C, (■) 50 °C, (◆) 60 °C.

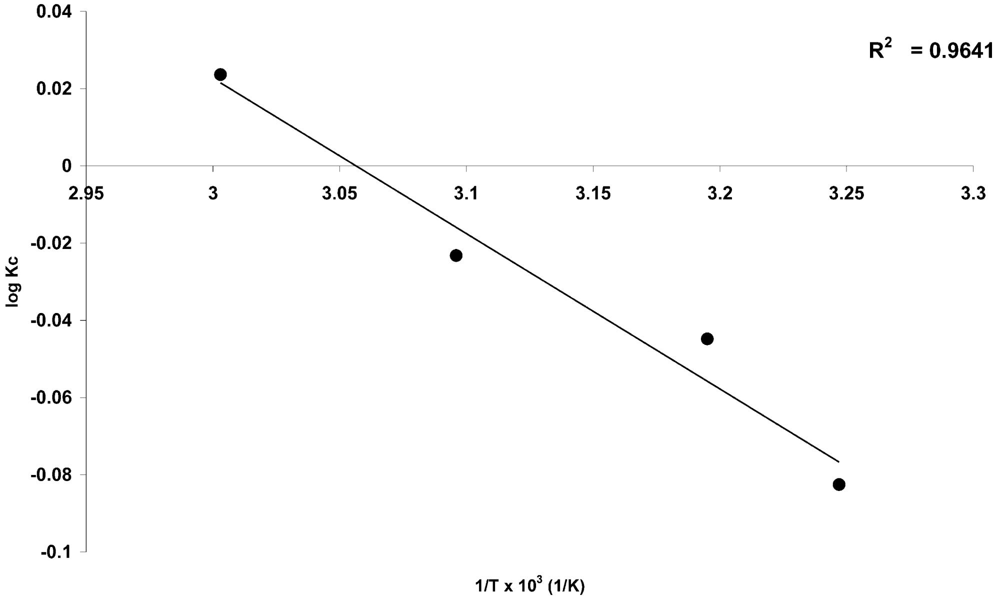

Fig. 9. van't Hoff plot for Congo Red adsorption.

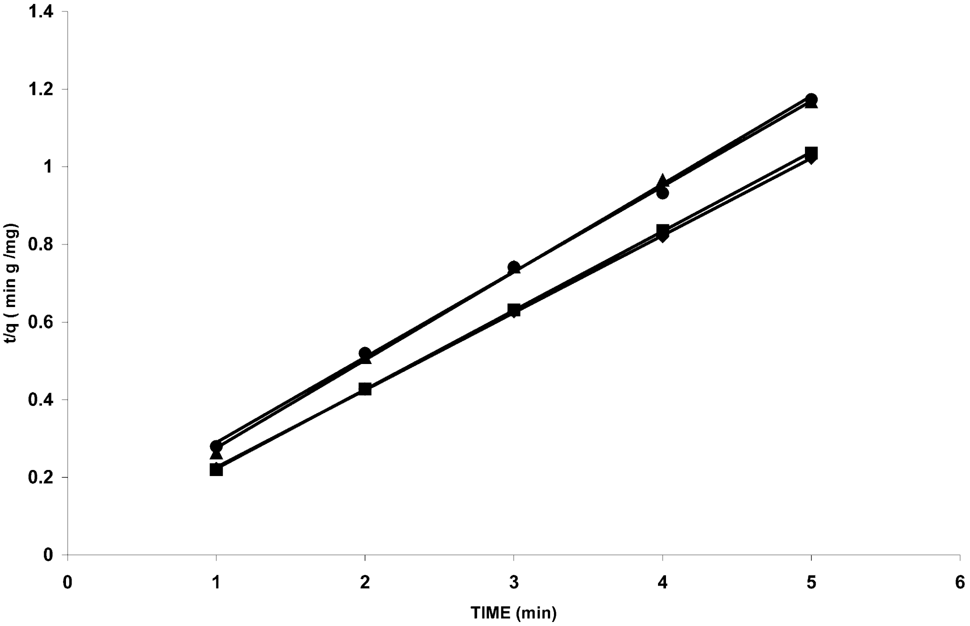

Fig. 10. Plots of the pseudo second-order model at different temperatures. Adsorbent dose, 50 mg/50 ml; dye concentration, 20 mg/l; initial pH, 7.70; temperature (●) 35 °C, (▲) 40 °C, (■) 50 °C, (O) 60 °C.

Table 4
Thermodynamic parameters

Temperature (°C)	K_c	ΔG^0 (kJ/mol)	ΔH^0 (kJ/mol)	ΔS^0 (J/K/mol)
35	0.827	0.487	7.705	23.549
40	0.902	0.269		
50	0.948	0.144		
60	1.056	−0.151		

adsorption for Congo Red at 60 °C. Positive ΔG^0 values were observed at 35, 40 and 50 °C, indicating that spontaneity is not favoured at low temperatures. A similar trend has been observed for the adsorption Fe(II) by waste Fe(III)/Cr(III) hydroxide at 24 °C [38]. The positive values of ΔS^0 suggest the increased randomness at the solid/solution interface during the adsorption of dye on coir pith carbon.

The first order rate constants of adsorption (k_1) were evaluated from the plots of log (q_e–q) vs t for different temperatures using the Lagergren first order rate expression. The correlation coefficients obtained were low. Also the calculated q_e values obtained from the first-order kinetic model do not agree with the experimental q_e values (Table 1). This shows that the adsorption of dye on coir pith carbon at different temperatures does not follow the first-order kinetic model. Linear plots of t/q vs t corresponding to the second-order kinetic model were obtained (Fig. 10). The correlation coefficients are greater than 0.998. Also, the calculated values of q_e agree very well with the experimental data. This indicates that the adsorption follows second-order kinetic model at different temperatures used in this study.

4. Conclusion

The present study shows that the coir pith carbon is an effective adsorbent for the removal of Congo Red from aqueous solution. Adsorption followed both the Langmuir and Freundlich isotherms. Kinetic data follows second-order kinetic model. The adsorption capacity was found to be 6.72 mg/g. Complete removal of the dye can be achieved using an appropriate dosage of the adsorbent and pH for wastewaters. The results would be useful for the fabrication and designing of wastewater treatment plants for the removal of dye. Since the raw material coir pith is freely available in large quantities in coir industries the treatment method seems to be economical.

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