Uptake Hazardous Dye from Wastewater Using Water Hyacinth as Bio-Adsorbent

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ABSTRACT

The existing study demonstrates that water hyacinth (*eichhorniacrassipes*) is a potential adsorbent for the removal of Congo red dye from synthetic wastewater by batch process. The experiments were conducted to study the influence of various parameters such as initial dye concentration, pH, contact time and adsorbent dosage at different operating conditions. The effect of pH and dye concentration was found to be significant and the maximum removal was detected at pH 5 and concentration 100 ppm; considered to be optimum values. The removal of Congo red was consistent initially proportional to the adsorbent dosage. The adsorption process followed Langmuir adsorption isotherm model; point out that the process supported monolayer adsorption of Congo red on the adsorbent surface. Adsorption kinetics closely followed the pseudo-second-order model and mass transfer analysis indicated better transportation of adsorbate from solution phase to solid phase. These results point out suitability of the locally available low cost adsorbents in the niche area of wastewater treatment and can be implemented in commercial dye enriched industrial effluent.

Keywords: adsorption, water hyacinth, Congo red, kinetic study, isotherm, mass transfer

INTRODUCTION

Water resources, soil fertility, aquatic organisms and ecosystem integrity are adversely affected by toxic effluents which are discharged from various chemical industries. Textile industry is one of the chemical industries where coloring effluents are the major encountered problems. The textile wastewater is rated as the most polluting discharges among all industrial sectors (Awomeso et al., 2010, Vilaseca et al., 2010). The textile industry produces large amount of dye contained wastewater in dyeing and finishing processes. Dyes usually have complex aromatic molecular structure which makes them non-biodegradable (Gharbani et al., 2008, Taha, 2011). Due to non-biodegradability, the components of dye substances create severe problem not only to the ecosystem but also create toxicity to the biological life (Choi et al., 2004). Various physical and chemical treatment processes such as coagulation, flocculation, membrane filtration, etc. are commercially practiced and removal efficiency of these processes are satisfactory (Robinson et al., 2001), however, these processes are costly, large amount of sludge is produced during operation and ecologically unsafe. From these perspectives, adsorption is one of the most effective and alternative methods to remove dyes from wastewater (Deans et al., 1992, Nigam et al., 2000). Adsorption is the method for separation of mixtures on laboratory and industrial scale where it is a surface

phenomenon that can be defined as the increase in concentration of a particular component at the interface between two phases. In this study, we represent how dye can be removed from synthetic wastewater through adsorption by using water hyacinth as bio-adsorbent. Water hyacinth is one of the abundant weed in the world but it can be employed as a bio-adsorbent after some modification that is capable to remove various industrial dyes such as methylene blue, Congo red, crystal violet, malachite green from aqueous solution (Nath et al., 2013). In case of heavy metal removal from aqueous solution, water hyacinth showed great potentiality. Water hyacinth has the ability to uptake copper, cadmium, chromium, and zinc from textile wastewater with high efficiency (Mokhtar et al., 2011, Ajayi and Ogunbayo, 2012; Mahmood et al., 2005). Nowadays, a large number of low cost adsorbents have drawn attention to the researchers due to their capacity, availability and ease to operation.These are, for example, rice husk (Reddy et al., 2013), wood dust (Garg et al., 2004), tree bark powder (paul 2013), peat (Fernandes et al., 2006), lignin (Cotoruelo et al., 2010), wheat bran (Ata et al., 2012), brown sea weed (Vijayaraghavan et al., 2008), banana and orange peel (Annadurai et al., 2002), fly ash (Janos et al., 2003), pineapple stem waste (Hameed et al., 2009), tuberous pulp, sugarcane pulp, coconut pulp (Pramanik et al., 2011), etc. However, the search for simple, economic, eco-friendly and highly effective adsorbents are still challenging. From these perspectives, water hyacinth was selected as a bio-adsorbent to remove dye from synthetic wastewater.

In the present work, naturally available low–cost adsorbent, such as water hyacinth has been used as an adsorbent for Congo-red dye removal from aqueous solution. The consequence of the experimental parameters such as initial pH, concentration of dye, adsorbent dosage and contact time has been investigated. The outcome of the experiments was analyzed by Langmuir adsorption isotherm, mass transfer and kinetics viewpoints.

MATERIALS AND METHODS

Materials

Water hyacinth was collected from the local area of Jessore University of Science and Technology, Bangladesh. The chemical substances, e.g., Congo red (CR) dye, HCl and NaOH were purchased from Merck, Germany. Stock solution of Congo red was prepared by dissolving 696.665 g/mol of dye in doubled distilled water and different concentrations (25, 50,100, 200, 300, 400 and 500 ppm) of solution were made by dilution of the stock solution.

Preparation of Adsorbent

Water hyacinth was carefully washed by distilled water to remove the dirt and mud. It was then sun dried and acid washed to eliminate the soluble impurities. Then acid washed sample was dried again in an oven at 105°C for 2 hrs to leave out the moisture from it. The dried adsorbent was crushed in a micro-plant grinding machine and converted into fine powder which was stored in air tight plastic containers for further use in the whole experiment.

Adsorbent Characterization and Analysis

UV-1650 spectrophotometer (Shimadzu Co., Japan) was used to measure the concentration of solution where maximum absorbance wavelength was 450 nm. pH of the solutions were studied over the range from 4 to 8 through pH meter.

Adsorption Experiments

The batch experiments were run out to investigate the influence of various parameters including contact time (0 to 210 mins), pH (4 to 8), adsorbent dosages (0.5 to 5 g/L). In this study 250 ml conical flasks were used to keep 100 ml adsorbate solution of various concentrations (25, 50,100, 200, 300, 400 and 500 ppm) and known amount of adsorbent were added into each flask at different conditions which shaken continuously at a constant oscillation of 211 osc/min for 3.5 hrs. Then the solution was filtered and the remaining sample was analyzed to determine the adsorption percentage of Congo red which computed by the following equation (1):

$$\% \text{ Removal of CR} = \frac{C_0 - C_t}{C_0} \times 100\% \tag{1}$$

where, C_0 is the initial concentration (mg/L) and C_t is the concentration at time t. The amount of CR adsorbed per unit mass of the adsorbent, q_e (mg/g) was evaluated by the following mass balance equation (2):

$$q_e = \frac{(C_0 - C_e)V}{m} \tag{2}$$

where, C_0 and C_e are the concentrations (mg/L) of Congo-red at initial and at equilibrium, correspondingly. V is the volume (L) of the solution and m is the mass of the adsorbent (g).

Pseudosecondorder Model

Adsorption capacity of dye on the adsorbent particles was assumed proportional to the active sites of the surface. For pseudo–second order kinetic rate, the above equation is as in the following equation (3):

$$\frac{t}{q_1} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \tag{3}$$

If second–order kinetic is applied, the plot of t/q_1 against t of the above equation should demonstrate a linear relationship and q_e and k_2 can be determined from the slope and intercept.

Intra-particle Diffusion Study

Diffusion coefficient (K_{ip}) for the intra–particle transport of CR has been calculated at initial dye concentrations and pH by employing the rate equation (4) expressed by Vadivalen and Kumar, 2005:

$$q_t = \emptyset + K_{ip} \sqrt{t} \tag{4}$$

Langmuir Adsorption Isotherm

Langmuir represents the equilibrium distribution of dye molecules between the solid and liquid phases. Langmuir isotherm is valid for monolayer adsorption onto a surface containing a finite number of identical sites. Based upon these assumptions, Langmuir represented the following equation (5):

$$q_e = \frac{Q_o K_L C_e}{1 + K_L C_e} \tag{5}$$

Langmuir adsorption parameters were determined by transforming the Langmuir equation into linear form (6):

$$\frac{1}{q_e} = \frac{1}{Q_0} + \frac{1}{Q_0 K_L C_e} \tag{6}$$

where, C_e is the equilibrium concentration of adsorbate (mg/L), q_e is the amount of dye adsorbed per gram of the adsorbent at equilibrium (mg/g), Q_0 is the maximum monolayer coverage capacity (mg/g) and K_L is the Langmuir isotherm constant (L/mg).

Mass Transfer Analysis

Mass transfer analysis for adsorption of Congo red on the water hyacinth was carried out using the Mckay et al. (1981) equation (7):

$$\ln\left(\frac{C_t}{C_o} - \frac{1}{1 + MK_{bq}}\right) = \ln\left(\frac{MK_{bq}}{1 + MK_{bq}}\right) - \left(\frac{1 + MK_{bq}}{MK_{bq}}\right)\beta S_s t \tag{7}$$

where, M is the mass of the adsorbent per unit volume (g/L), K_{bq} is the constant obtained by multiplying Q_o and K_L which are defined in 2.7 section, S_s is the external surface area of the adsorbent per unit volume (m^{-1}), β is the mass transfer coefficient (cm/min) and t is the contact time (min).

The plot of $\ln\{C_t/C_0 - 1/(1 + MK_{bq})\}$ versus t is carried out. The values of mass transfer coefficient, β were determined graphically from the slope $\{(1 + MK_{bq})/MK_{bq}\}\beta S_s$ of the individual plots.

RESULTS AND DISCUSSION

Removal Properties of CR

Effect of pH

The removal of Congo red dye from synthetic waste water with varying pH is illustrated in **Figure 1** where initial dye concentration 100 ppm and contact time 150 min.

From the figure, it can be seen that the maximum dye adsorption (87%) was obtained at pH 5 and gradual decrease in adsorption take place with increasing the value of pH. Only 58% dye adsorption take place at pH 8. This is due to the fact that, Congo red is anionic dye and at higher pH, OH$^-$ ions are plenty and it can compete with the anionic dye which accounts low adsorption. At lower pH, due to the electrostatic attraction between negative charged dye molecules and positive charged adsorbent surface higher adsorption can be observed (Aboul-Fetouh et al., 2010; Alam et al., 2014).For these reason, pH 5 was taken as optimum result for further whole experiment herein.
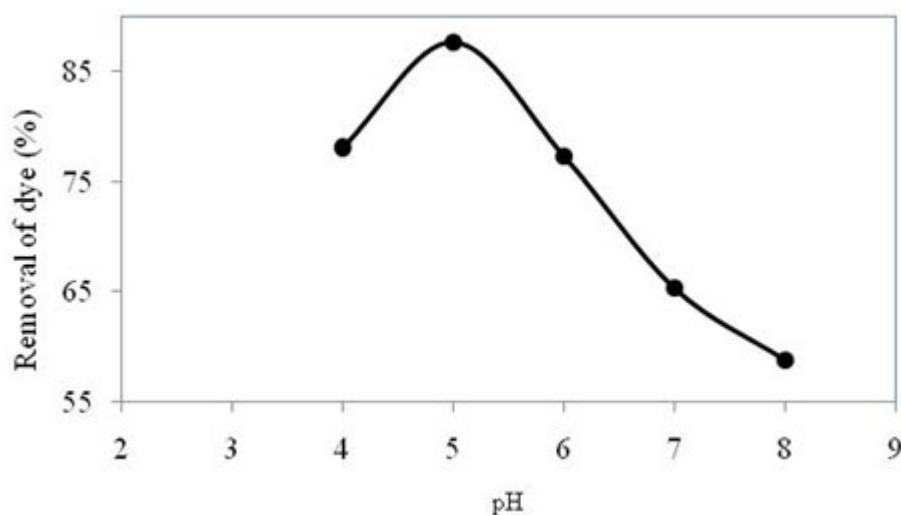
Figure 1. Effect of pH on adsorption of CR by the water hyacinth adsorbent at optimum concentration 100 ppm, adsorbent dosage 1.0 g/L and contact time 150 min
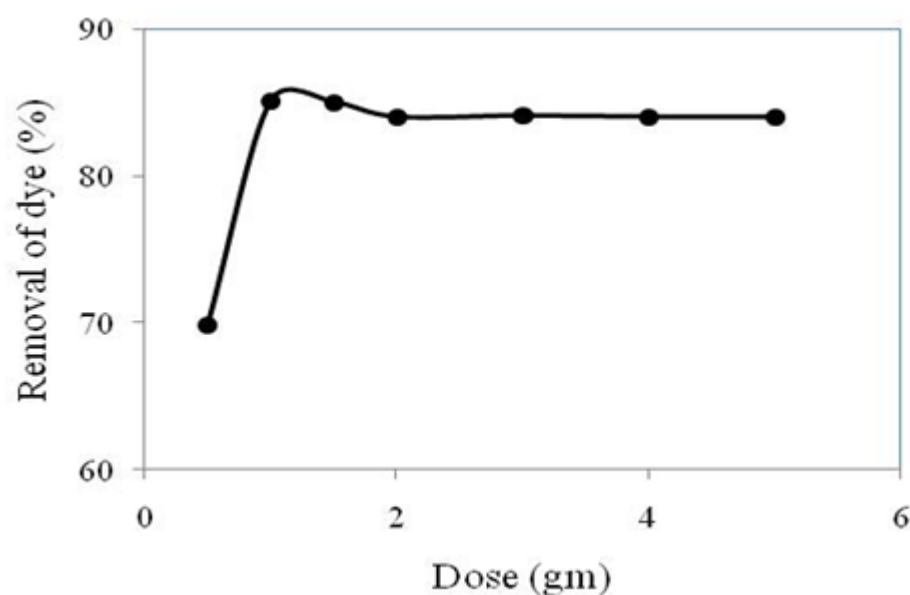

Figure 2. Effect of adsorbent dosage on % of dye removal by the adsorbent at optimum concentration 100 ppm, pH 5 and contact time 150 min

Effect of Adsorbent Dosage

The result of Congo red dye removal where initial concentration 100 ppm, contact time 150 min and optimum pH 5 with varying amount of adsorbent dosages is depicted in **Figure 2**.

From the Figure, it can be seen that adsorption of dye increased from 69% to 85% with increasing adsorbent dose from 0.5 to 1.0 g/L. After that when we increased the adsorbent dose, the dye removal% decreased due to the fact that there might be formation of particle aggregation resulting in a decrease in the total surface area which responsible for decreasing adsorbed amount per unit mass (Khan et al., 2015). Even if the uptake of dye increased by increasing the adsorbent dose beyond a dose of 1.0 g/L, the rise of dye removal% is insignificant and capacity of adsorbent is low. Therefore, further increase of dose result the much production of sludge and wastage of material. Similar result was found from lead ion removal by bamboo based activated carbon which is reported in Khan et al., 2015. Thus 1.0 g/L of adsorbent dose was taken as an optimum dose for further experiments.

Effect of Dye Concentration

This study was performed by changing the initial dye concentration in the range of 25 to 500 ppm (**Figure 3**) with optimum conditions received from previous experiments (adsorbent dose 1.0 g/L, contact time 150 min and pH 5).
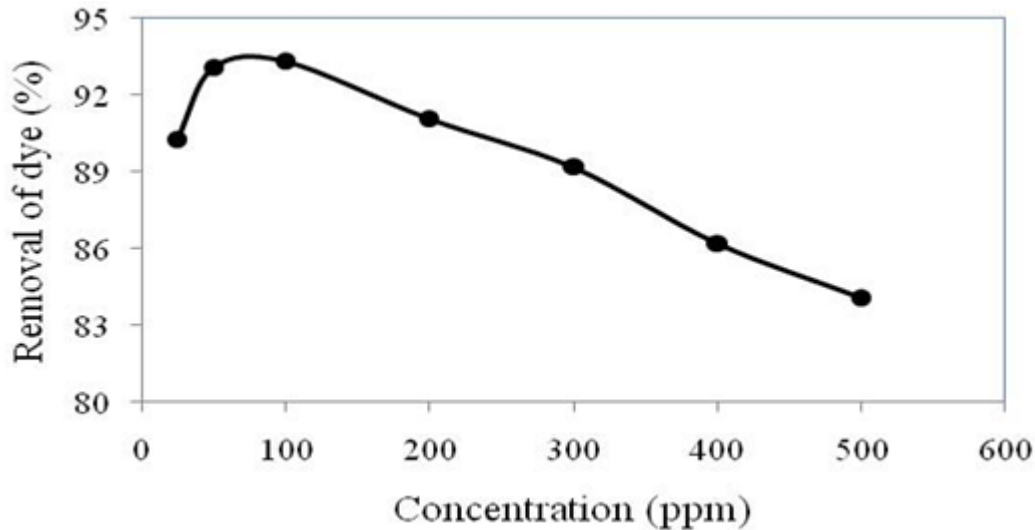
Figure 3. Effect of dye concentration on the adsorbent at optimum adsorbent dosage of 1.0 gm/L, pH 5 and contact time 150 min
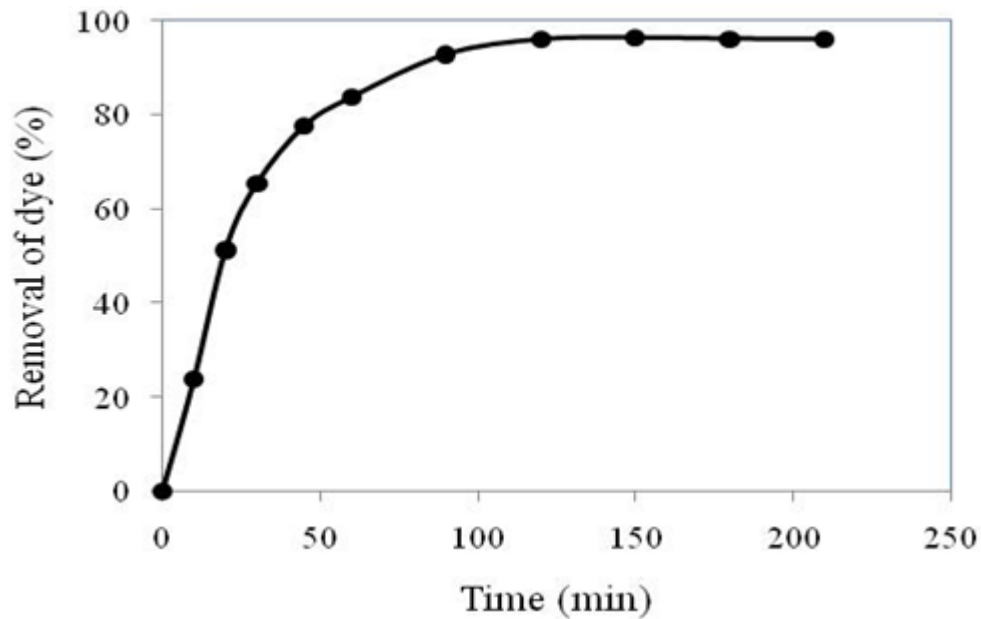

Figure 4. Effect of contact time of the CR dye on the adsorbent at optimum concentration 100 ppm, adsorbent dosage of 1.0 gm/L and pH 5

The initial dye concentration provided significant driving force to overcome all mass transfer resistances of the dye between the aqueous and solid phases (Ho et al., 2005). In this study, when the initial concentration of dye increased from 25 to 100 ppm, the percentage of dye removal using water hyacinth increased from 90 to 93% and after that the removal percentages fall with increasing dye concentration. The reasons behind that, the increase of adsorption capacity might be due to the increment of driving force that is concentration gradient which causes an increased number of dye particles coming in contact with the adsorbent. On the other hand, number of available adsorption sites in adsorbent is the same for all concentrations. Thus concentration increases with the more number of dye particles but adsorbent dose remain same, therefore, dye competes with the same adsorption sites. As a result, some dye particles without being adsorbed retain in the system and decrease the removal percentage upon increases the Congo red dye concentration.

Effect of Contact Time

Effect of contact time on the removal of dye by the water hyacinth as adsorbent is illustrated in **Figure 4**. It showed that the removal percentage increased (0 to 96%) with the increase in contact time (0 to 150 min). Highest removal (96%) was obtained at and above 150 min. It means that the removal rate becomes slower with time and finally a saturation stage is obtained. Similar result was discussed in Kumar and Bilal, 2018.
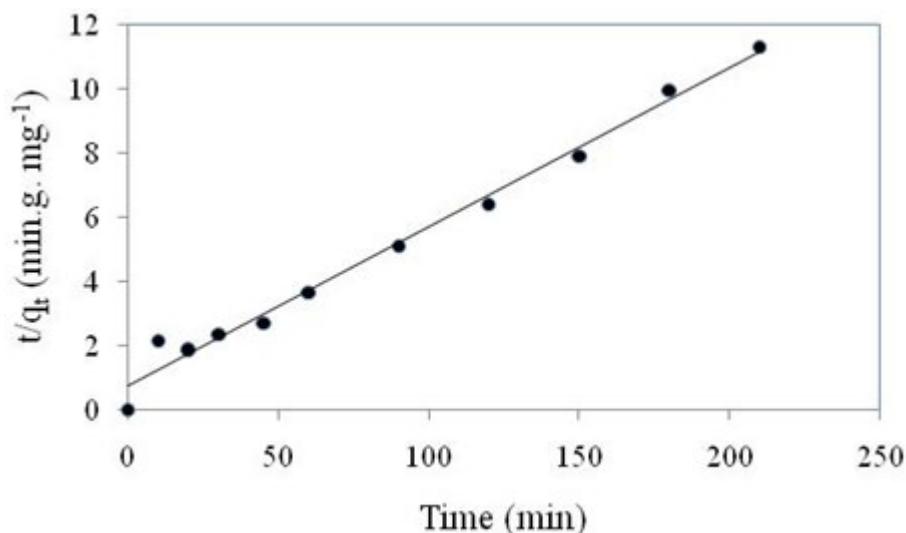
Figure 5. Pseudo second order kinetic model for the removal of CR using water hyacinth at optimum concentration 100 ppm, adsorbent dosage 1.0 g/L and pH 5

Table 1. Kinetics results for adsorption of CR onto water hyacinth

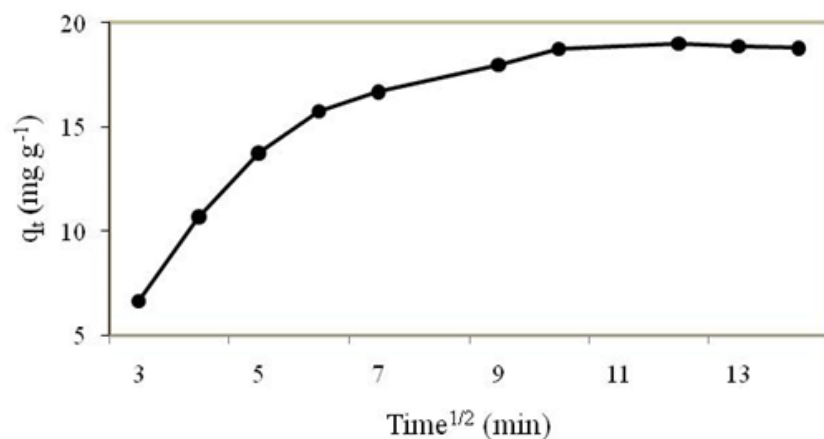| Pseudo second order				Intra–particle diffusion	
$K_2 (g/mg)$	$q_e (mg/g)$		R^2	$K_{ip} (g/mg/min^{1/2})$	$\emptyset (mg/g)$
1.77×10^{-3}	Exp.	Calc.	0.9865	0.9512	7.6938
	8.77	20.12			

Figure 6. Intra-particle diffusion model for the removal of CR onto water hyacinth

Adsorption kinetics

Pseudo second order kinetic model was studied at optimized operating conditions such as concentration 100 ppm, pH 5 and the results were described in **Figure 5**. The figure (t/q_t versus t) showed linear plot from which the value of q_e and k_2 were calculated (**Table 1**). The result revealed that R^2 value is close to 1 and linearity of this curve indicates that the kinetic data fitted well with the pseudo second order model. Similar results were also reported in Cheng et al., 2015 to uptake CR dye via activated carbon.

For the intra-particle diffusion analysis, if the plot of q_t versus $t^{1/2}$ gives a straight line that pass through the origin which indicates the intra-particle diffusion model contributes in the rate determining step (Ghorai and Pant 2005) but in **Figure 6**, this linear relationship does not pass through the origin that imply the intra-particle diffusion model was not rate controlling step.

Adsorption Isotherm Study

In the present study, the plot of $1/q_e$ against $1/C_e$ gives straight line shown in **Figure 7** with a slope of $1/Q_0 K_L$ and intercept of $1/Q_0$. From the plot we can notice that the value of regression correlation co-efficient (R^2) is 0.9899 which is very close to 1 and indicates that the obtained data are well fitted in Langmuir isotherm model and also suggests that monolayer sorption exists under the experimental conditions.
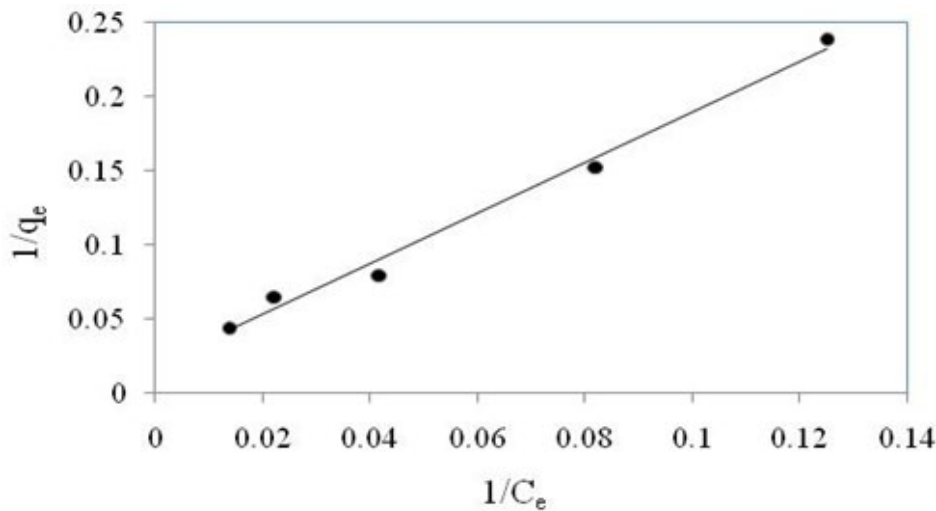
Figure 7. Langmuir plot for the adsorption of congo red dye by the adsorbent at optimum concentration 100 ppm, adsorbent dosage 1.0 g/L, pH 5 and contact time 150 min

Table 2. Maximum adsorption capacities of different types of bio-adsorbents

| Adsorbents	Adsorption capacity (mg/g)	Type of Dyes	References
Water hyacinth	53.76	Congo red	Present study
Water hyacinth	17.58–46.35	Methylene blue	Murali and Uma, 2016
Water hyacinth root	46.15	Congo red	Kumar and Bilal, 2018
Water hyacinth root	8.04	Methylene blue	Soni et al., 2012
Rice Husk	4.29	Congo red	Taha et al., 2014
Sugarcane bagasse	39.8	Congo red	Zhang et al., 2011
Cashew nut shell	5.18	Congo red	Senthil et al., 2010
Kaolin	5.44	Congo red	Vimonses et al., 2009
Bagasse fly ash	11.89	Congo red	Mall et al., 2005
Neem leaf powder	41.20	Congo red	Bhattacharrya and Sharma, 2004
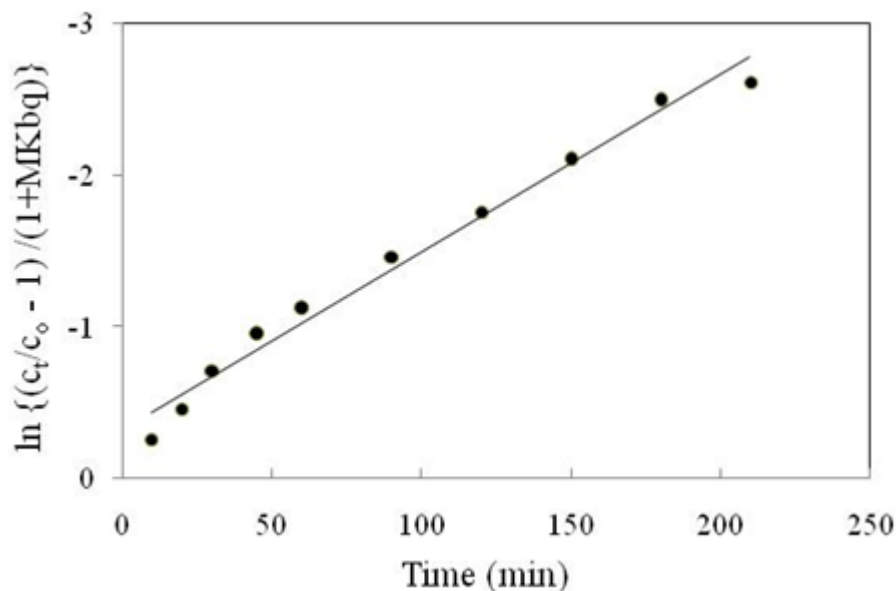
Figure 8. Mass transfer plot for the adsorption of Congo red by the adsorbent at optimum concentration 100 ppm, adsorbent dosage 1.0 g/L and pH 5

The adsorption capacity (Q_0) of the adsorbent was found to be 53.76 mg/g. The adsorption capacity of the adsorbent under study is comparable to other bio-adsorbents reported in **Table 2**.

Mass Transfer Analysis

The plot of $\ln\{C_t/C_0 - 1/(1 + MK_{bq})\}$ versus t resulted a straight line shown in **Figure 8** representing the applicability of the model.

Table 3. Mass transfer analysis for adsorption of congo red on water hyacinth

Adsorbents	Mass transfer constants, β_s (cm/min)	Correlation coefficient, R^2
Water hyacinth	62.39×10^{-12}	0.9825

The value of mass transfer coefficient (β) were determined graphically from the slope $\{(1 + MK_{bq}/MK_{bq})\beta S_s\}$ of the individual plots and presented in **Table 3**.

The values obtained from the experimental analysis shown that the velocity of the adsorbate Congo red for transporting from bulk i.e. solution phase to solid phase was quite good (Bhattacharya et al., 2008).

CONCLUSION

In this study, Congo red dye was removed from aqueous solution quite successfully by water hyacinth as a natural adsorbent. The effect of various parameters like pH, adsorbent dosage, initial dye concentration and contact time were evaluated on the uptake capacity of the adsorbent. The optimum dye removal by using water hyacinth as bio-adsorbent was achieved at pH 5, concentration 100 ppm and for contact time 150 min. Monolayer adsorption capacity of the bio-sorbent was obtained to be 53.76 mg/g, confirmed by Langmuir fitting. The results of the kinetic study suggests that adsorption of Congo red on the selected adsorbent followed pseudo second order kinetic model and the intra-particle diffusion model showed larger surface adsorption. The mass transfer analysis indicated advanced movement of adsorbate toward water hyacinth adsorbent.

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