Optimisation and kinetics of biosorption of Coomassie Brilliant Blue G250 dye from synthetic effluent using *Pennisetum purpureum* biosorbent

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Abstract: Water-soluble reactive acid dyes are most problematic and susceptible to oxidative catabolism. Hence, their removal is of great importance and necessitates cost effective, environmentally sound techniques for the treatment of waste water containing dyes and metal. In the present study, the effect of the biosorbent dosage, pH, agitation rate, temperature and time was studied and their optimum values were found as 1 g/l, 2, 160 rpm, 30°C and 140 minutes, respectively. It was found that the *Pennisetum purpureum* biomass acts as a potential biosorbent to remove an industrial dye Comassie Brilliant Blue G250. The kinetics of the biosorption of Comassie Brilliant Blue G250 dye by *Pennisetum purpureum* adsorbent was studied at optimal conditions and Pseudo-second order model was found to be best described in this adsorption kinetics. The mechanism and the potential rate controlling step on biosorption have been investigated and were found to follow external diffusion model.

Keywords: biosorption; external diffusion; isotherms; kinetics; optimisation; wastewater.

Reference to this paper should be made as follows: Baskar, G., Sriharini, C., Sripriya, R. and Pavithra, S.K. (2016) 'Optimisation and kinetics of biosorption of Coomassie Brilliant Blue G250 dye from synthetic effluent using *Pennisetum purpureum* biosorbent', *Int. J. Environment and Sustainable Development*, Vol. 15, No. 3, pp.241–253.

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1 Introduction

In an effluent treatment, among the many types of organics present, the most difficult one to remove is the colour of the dyes (Reid, 1996). Dyes affect the aesthetic value of water, transparency in water, gas solubility, and penetration of light and hence reduce photosynthesis and also cause toxicity from heavy metals associated with pigments. Dyes cause carcinogenesis, mutagenesis, chromosomal fractures and respiratory toxicity. Dyes are used in textile, leather tanning, paper production, food technology, agriculture research and in cosmetic preparations. Many dyes are visible in water at concentrations as low as 1 mg/l. Textile processing wastewaters, typically with dye content in the range from 10 to 200 mg/l, are therefore highly coloured and discharged in the open waters presents an aesthetic problem. As dyes are designed to be chemically and photolytically stable, they are highly persistent in natural environments. The release of dyes may therefore present an eco-toxic hazard and introduce the potential danger of bioaccumulation that may eventually affect man by transport through the food chain (Forgacs, Cserhati and Oros, 2004; Aksu and Tezer, 2005).

Water-soluble, brightly coloured acidic and reactive dyes are the most problematic as they tend to pass through conventional decolourisation systems without being affected. Nearly 50% of the reactive dyes are lost in effluence, after dyeing which is very high when compared to the 2% loss of basic dyes in effluence and their removal is also significant (Moran, Hall and Howell, 1997; Robinson et al., 2001). Acid dyes are anionic compounds that are mainly used for dyeing nitrogen-containing fabrics like wool, polyamide, silk and modified acryl which are bound to the cationic NH_4^+ ions of those fibres. The adjective acid refers to the pH in acid dye baths rather than to the presence of acid groups (sulphonate, carboxyl) in the molecular structure of these dyes. Most acid dyes are azo (yellow to red, or a broader range of colours in case of metal complex

azo-dyes); anthraquinone or triarylmethane (blue and green) compounds (Churchley, 1994).

Reactive dyes are dyes with reactive groups that form covalent bonds with OH-, NH-, or SH-groups in fibres (cotton, wool, silk, nylon). The reactive group is often a heterocyclic aromatic ring substituted with chloride or fluoride, e.g. dichlorotriazine. During the process of dying with the reactive dye hydrolysis (inactivation) of the reactive groups is an undesired side reaction that lowers the degree of fixation. In spite of the addition of high quantities of salt and ureum (up to respectively 60 and 200 g/l) to raise the degree of fixation, it is estimated that 10 to 50% will not react with the fabric and remain hydrolysed in the water phase. The problem of coloured effluents is therefore mainly identified with the use of reactive dyes. Mostly 80% of the reactive dyes are azo or metal complex azo compounds but also anthraquinone and phthalocyanine reactive dyes are applied, especially for green and blue (Mishra and Tripathy, 1993; Forgacs, Cserhati and Oros, 2004; Renganathan et al., 2008).

Various conventional techniques can be employed to remove colour from dye containing wastewaters. Removal of dyes using physical or chemical treatment processes membrane filtration, coagulation/flocculation, precipitation, flotation, adsorption, ion exchange, ion pair extraction, ultrasonic mineralisation, electrolysis, advanced oxidation (chlorination, bleaching, ozonation, Fenton oxidation and photocatalytic oxidation) and chemical reduction but in general, each technique has its limitations. The above methods are involved with high initial investment and operation costs. The adsorption employing activated carbon and various other adsorbents has been widely applied, but the cost of carbon confines its application (Clarke and Anliker, 1980; Banat et al., 1996; Chen et al., 2003).

Biosorption is the process of the uptake of a solute (dye) using biomaterials (plant parts, microbial cells), whether dead or alive. So, cheap and low cost adsorbents such as bagasse, peat, soil, tree and eucalyptus barks, rice husk, fly ash, waste residues, wheat straw, corn cobs barley husks, agricultural residues, apple pomace and quaternised would have been used for the removal of dyes using adsorption techniques. Biosorption's main advantage is its cost-effective performance on removal of the colour. Biosorbents must be hard enough to withstand the application pressures, porous and/or transparent to metal ion-sorbate species, and have high and fast sorption uptake (Nigam et al., 2000; Fu and Viraraghavan, 2001; Robinson, Chandran, and Nigam, 2002; Baskar et al., 2014). In the present study the effect of the biosorbent dosage, pH agitation rate, temperature and time on biosorption of Comassie Brilliant Blue G250 was studied using *Pennisetum purpureum* biomass and the kinetics and mechanism of biosorption were also analysed.

2 Materials and methods

2.1 Preparation of biosorbent

Pennisetum purpureum also called as Napier Grass or Elephant Grass is native to the tropical regions of Africa. This is a perennial plant and grows from 2 to 4.5 m tall with 30 to 120 cm long leaves. This is found in abundance in India and grows on a wide range of soils and needs good moisture for growth. Hence it serves as a cheap source of biosorbent. The leaves of *P. purpureum* were collected from the water lake in Velachery

area, Chennai and were dried under sunlight. The dried leaves were then milled to a fine powder and sieved using mesh number 72 and 80, which is used for the current adsorption studies.

2.2 Dye used

Comassie Brilliant Blue G250 which is used in this study is an acid dye which has a molecular weight of 833.048 (g/mol) with the molecular formula as $C_{47}H_{50}N_3O_7S_2^+$. This dye has the maximum absorption level at 595 nm.

2.3 Batch adsorption studies

Each batch adsorption experiment was carried out in 250 ml Erlenmeyer flask containing 50 mg/ml of Comassie Brilliant Blue G250 dye solution for different sorbent concentration in a rotary shaker incubator. The effect of the factors such as rpm, temperature, pH and time were also studied by fixing the sorbent concentration to 1 g/l. Initial readings were taken immediately after the pH adjustment and the final readings were taken after subjecting the sample in the cooling microfuge at 10000 rpm for 15 minutes for the settling of the fine particles.

2.4 Estimation of the dye uptake

The Absorbance of the dye was observed in UV-VIS Double Beam Spectrometer Systronics-2201 at 595 nm. The dye uptake capacity of the biosorbent was calculated using Eq. 1.

$$q = ((C_o - C_{eq}) \times V) / W \tag{1}$$

where q (mg/g) is the amount of dye adsorbed by biosorbent, C_o and C_{eq} (mg/l) are the initial and equilibrium liquid phase concentrations of the dye respectively, V (l) is the initial volume of dye solution and W (g) is the weight of biosorbent (Zhou and Banks, 1993).

2.5 Kinetics modelling of biosorption by P. purpureum biosorbent

It is important to understand the rate at which the dye is adsorbed from the aqueous solutions/waste waters in order to design appropriate adsorption treatment plants. The kinetics of adsorption describes the equilibrium solute uptake rate, which in turn governs the residence time of the adsorption reaction. The biosorption kinetics has been investigated at optimal conditions using the Pseudo-first order and Pseudo-second order models (Hall et al., 2009). Pseudo-first order kinetic model was described by Lagergren as given in Eq. 2 (Lagergren and Svenska, 1898).

$$q_t = \ln q_e \left(1 - e^{-k_1 t}\right) \tag{2}$$

$$\ln\left(q_e - q_t\right) = \ln q_e - k'_t\, t \tag{3}$$

The slope and intercept of ln $(q_e - q_t)$ versus time yield the values of k_1 values and predicted q_e. Pseudo-second order assumes that the adsorption capacity is proportional to the number of active sites occupied on the sorbent, and the kinetic model was described as given in Eq. 4 (Ho and McKay, 1999).

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \tag{4}$$

$$\frac{t}{q_t} = \frac{1}{h} + t\left(\frac{1}{q_e}\right) \tag{5}$$

Where h is equal to k_2/q_e^2. The values $(1/h)$ and q_e can be determined from the intercept and slope, respectively, from a linear plot of t/q_t versus time.

2.6 Mechanisms of biosorption by P. purpureum biosorbent

The biosorption mechanism and potential rate controlling steps have been investigated at optimal conditions using external diffusion and Weber-Morris intra-particle diffusion models (Hall et al., 1966). The external diffusion model is given as given in Eq. 6.

$$\ln \frac{c_t}{c_0} = -k_3 t \tag{6}$$

where k_3 is the external diffusion rate of the constant and c_0 is the constant. The plot of $\ln(c_t)$ versus time gives a linear relationship for external diffusion model. Weber and Morris stated that if intra-particle diffusion is the rate-controlling factor, the uptake of the adsorbate varies with the square-root of time; hence the rates of adsorption are usually measured by determining adsorption capacity as a function of the square root of the time. Weber-Morris intra-particle diffusion model is given by Eq. 7.

$$q_t = k_4 \sqrt{t} + C \tag{7}$$

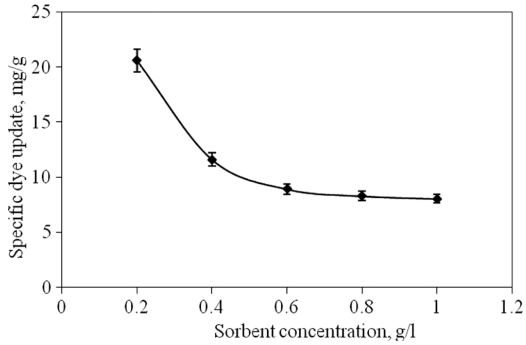
where k_4 is the intra-particle diffusion rate constant (mg g^{-1} $min^{-0.5}$) and C is the constant (Hall et al., 2009; Daifullah, Yakout, and Elreefy, 2007; Alkan et al., 2008).

3 Results and discussion

3.1 Effect of sorbent dosage on biosorption of Comassie Brilliant Blue G250

To determine the effect of sorbent concentration on its dye uptake capacity, the dried powder of *P. purpureum* at different concentrations such as 1, 2, 3, 4, 5 g/l was added to five different 250 ml Erlenmeyer flasks. The volume was made up to 50 ml with distilled water dye at a concentration of 50 mg/l. The flasks were kept on a rotary shaker at an agitation rate of 140 rpm for 100 minutes, room temperature was maintained. It was observed that the amount of dye uptake was decreased with increase in sorbent dosage from 0.2 to 1 g/l (Figure 1). The percentage of dye removal was increased with increase in sorbent dosage from 0.2 to 1 g/l and it was found maximum at 1 g/l.

Figure 1 Effect of sorbent dosage on biosorption of Coomasie Brilliant Blue G250 *P. purpureum* biosorbent

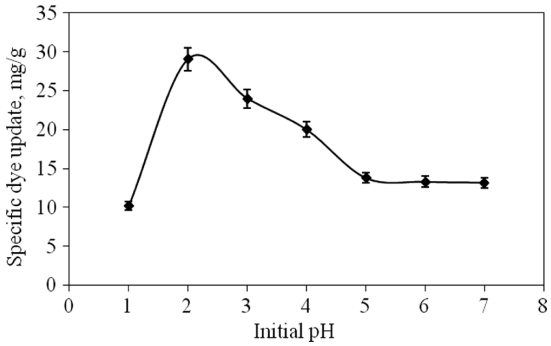


Note: (dye concentration of 50 mg/l; temperature 30°C; agitation rate 140 rpm).

3.2 Effect of pH on biosorption of Comassie Brilliant Blue G250

In order to study the dye uptake capacity of the sorbent under the influence of the parameter pH, the adsorption experiments were carried out for different pH ranging from 1 to 7. The optimised sorbent concentration of 1 g/l was taken from the above study. A total of seven Erlenmeyer flasks of 250 ml were prepared, each containing a powdered mass of *Pennisetum purpureum* at a concentration of 1 g/l with the dye at a concentration of 50 mg/l making up the total volume to 50 ml with distilled water. The flasks were kept on a rotary shaker at an agitation rate of 140 rpm for 100 minutes and the room temperature was maintained. It was observed that the amount of dye adsorbed was maximum at pH 2 (Figure 2) and then it prominently decreased for other values of pH. This may be due to the electrostatic attractions between negatively charged dye anions to the positively charged cell surface. At lower pH values the biomass will have a net positive charge. As the pH of the system increases, the number of negatively charged sites increase and the number of positively charged sites decrease in the biomass. A negatively charged surface site on the sorbent does not favour the adsorption mechanisms with respect to pH (Zhou and Banks, 1993, Renganathan et al., 2008).

Figure 2 Effect of pH on biosorption of Coomasie Brilliant Blue G250 *P. purpureum* biosorbent

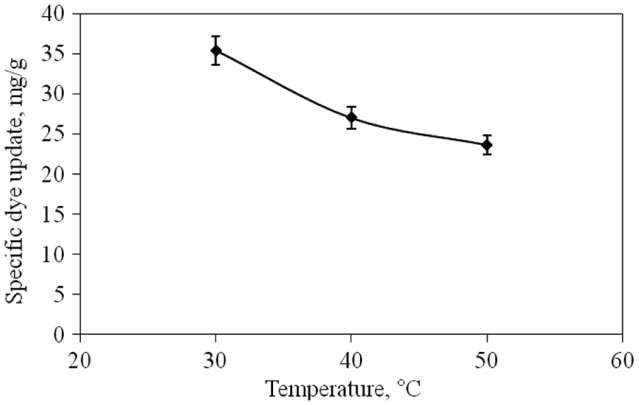


Note: (sorbent dosage 1 g/l; dye concentration of 50 mg/l; temperature 30°C; agitation rate 140 rpm).

3.3 Effect of temperature on biosorption of Comassie Brilliant Blue G250

For the optimised sorbent concentration, pH and rpm values; the effect of temperature (30, 40 and 50°C) was analysed for the dye uptake capacity of the sorbent. A total of three Erlenmeyer flasks of 250 ml were prepared, each containing a powdered mass of *P. purpureum* at a concentration of 1 g/l with dye at a concentration of 50 mg/l making up the total volume to 50 ml with distilled water. The optimised value of pH 2 was maintained in all the three flasks and they were placed on a rotary shaker at an agitation rate of 140 rpm for 100 minutes with each flask maintained at different temperature such as 30, 40 and 50°C. It was found that the dye uptake capacity of the sorbent decreased as the temperature increases and the optimum temperature for maximum dye uptake was found to be 30°C (Figure 3).

Figure 3 Effect of temperature on biosorption of Coomasie Brilliant Blue G250 *P. purpureum* biosorbent

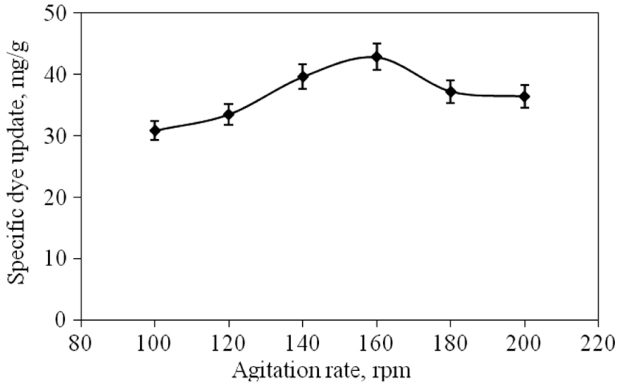


Note: (sorbent dosage 1 g/l; pH 2; dye concentration of 50 mg/l; agitation rate 140 rpm).

3.4. Effect of agitation rate on biosorption of Comassie Brilliant Blue G250

For different ranges of agitation rate (100, 120, 140, 160, 180 and 200 rpm) the adsorption experiments were performed by employing optimised values of pH and sorbent concentration from the above study in order to study the dye uptake capacity of the sorbent under the influence of the parameter agitation rate. *P. purpureum* concentration was 1 g/l; 50 ml of dye with a concentration of 50 mg/l was used. The optimised value of pH 2 was maintained in all the six flasks. Each of the flasks was placed on a rotary shaker at different agitation rates such as 100, 120, 140, 160, 180 and 200 rpm for 100 minutes. The maximum adsorption of dye uptake was observed at 160 rpm (Figure 4).

Figure 4 Effect of agitation rate on biosorption of Coomasie Brilliant Blue G250 *P. purpureum* biosorbent

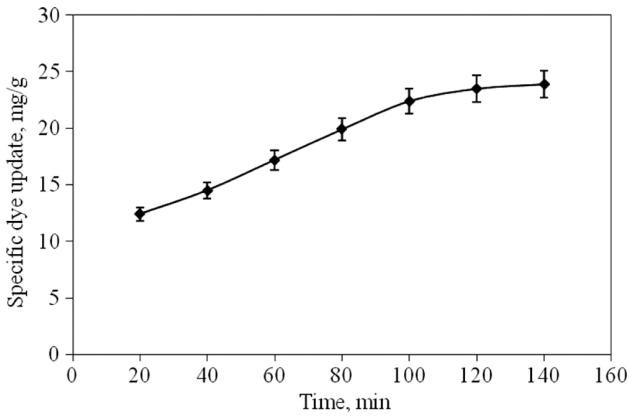


Note: (sorbent dosage 1 g/l; pH 2; dye concentration of 50 mg/l; temperature 30°C).

3.5 *Effect of time on biosorption of Comassie Brilliant Blue G250*

The adsorption experiment was carried out to study the effect of time on the dye uptake capacity of the sorbent by employing the optimised values of the parameters such as sorbent concentration, agitation rate, pH and temperature from the above studies. A total of eight Erlenmeyer flasks of 250 ml were prepared, each containing a powdered mass of *P. purpureum* at a concentration of 1 g/l with 50 ml of dye solution at a concentration of 50 mg/l. The optimised value of pH 2 was maintained in all the flasks and they were placed on a rotary shaker at an agitation rate of 160 rpm with each flask kept for different time such as 20, 40, 60, 80, 100, 120, 140 and 160 minutes. From the analysis of the dye under UV-VIS Double Beam Spectrometer at 595 nm, it was found that the maximum dye uptake by the sorbent was in 140 minutes (Figure 5).

Figure 5 Effect of time on biosorption of Coomasie Brilliant Blue G250 *P. purpureum* biosorbent



Note: (sorbent dosage 1 g/l; pH 2; dye concentration of 50 mg/l; agitation rate 160 rpm; temperature 30°C).

3.6 Kinetics modelling of biosorption of Comassie Brilliant Blue G250 by P. purpureum biosorbent

The kinetics values obtained for Pseudo-first order and Pseudo-second order models for the biosorption of Comassie Brilliant Blue G250 by *P. purpureum* biosorbent were summarised in Table 1. The plot of ln ($q_e - q_t$) versus t (Figure 6) represents the Pseudo-first order model with an R^2 values of 0.96, k_1 value of 0.0298 the predicted equilibrium dye uptake q_e of 56.08 mg/g. The plot of (t/q_t) versus t (Figure 7) represents the Pseudo-second order model with an R^2 values of 0.998, k_2 value of 0.00049 the predicted equilibrium dye uptake q_e of 60.97 mg/g. Based on R^2 values, it was confirmed that the Pseudo-second order model fits the experimental data very well in comparison with Pseudo-first order model. Hence the biosorption of Comassie Brilliant Blue G250 by *P. purpureum* biosorbent is the best way described by the Pseudo-second order kinetic model and the adsorption capacity *P. purpureum* biosorbent is found to be proportional to the number of active sites on the sorbent. Low k_2 value (0.00049) in Pseudo-second order model indicates that the *P. purpureum* biosorbent towards Comassie Brilliant Blue G250. The experimental and predicted dye uptake by Pseudo-first order and Pseudo-second order models using calculated kinetics values are shown in Figure 8. It was observed that the Pseudo-second order models shows good agreement between the experimental $q_{e,exp}$ and the predicted $q_{e,\,pred}$ equilibrium dye uptake for the bio-sorption of Coomasie Brilliant Blue G250 by *P. purpureum* biosorbent.

Table 1 Pseudo-first order and Pseudo-second order kinetics values of biosorption of dye by *P. purpureum* biosorbent

Parameters	*Pseudo-first order*	*Pseudo-second order*
$q_{e,exp.}$	49.16	49.16
R^2	0.960	0.998
K	0.0298	0.00049
$q_{e,pred.}$	56.07	60.97

Figure 6 Pseudo-first order kinetic for biosorption of Coomasie Brilliant Blue G250 by *P. purpureum* biosorbent

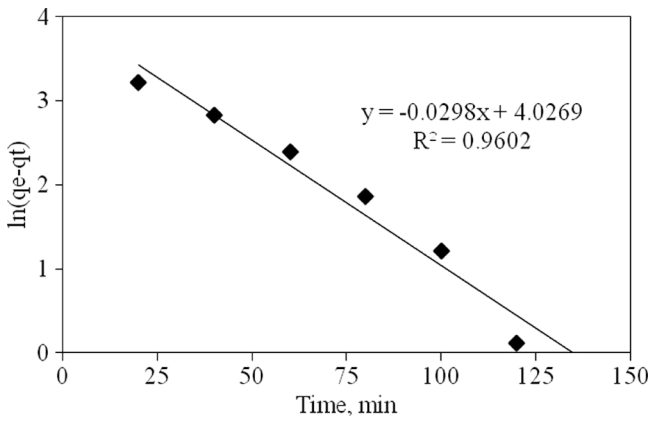


Figure 7 Pseudo-second order kinetic for biosorption of Coomasie Brilliant Blue G250 by *P. purpureum* biosorbent

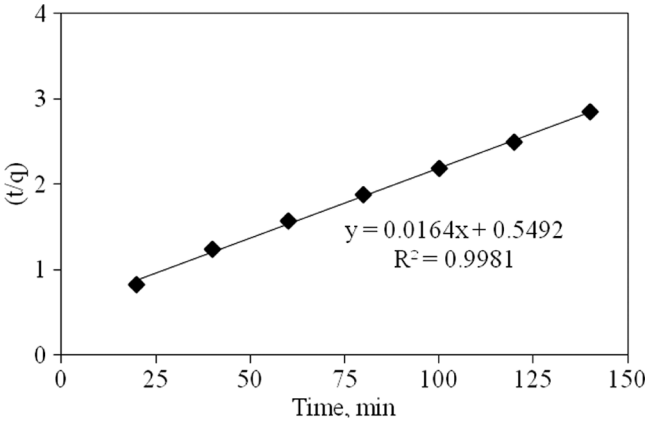
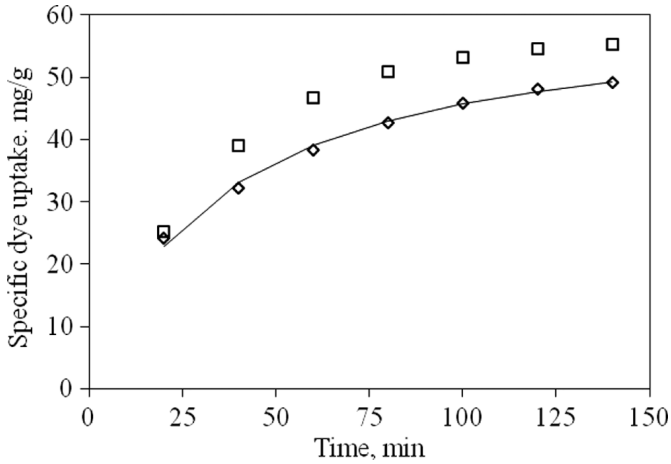


Figure 8 Experimental and model predicted dye uptake (-) Experimental; (□) Pseudo-first order model; (◊) Pseudo-second order model



3.7 Mechanism of biosorption of Coomasie Brilliant Blue G250 by P. purpureum biosorbent

The mechanism of biosorption of Coomasie Brilliant Blue G250 by *P. purpureum* biosorbent was studied using external diffusion model and Weber-Morris intra-particle diffusion model. The calculated parameters of the biosorption mechanism are given in Table. 2. The plot of ln (Ct) versus t (Figure 9) represents the external diffusion model with an R^2 value of 0.983 and the external diffusion rate constant (k_3) of 0.0253. The plot of (q_t) versus $t^{½}$ (Figure 10) represents the Weber-Morris intra-particle diffusion model with an R^2 values of 0.978 and the intra-particle diffusion rate is constant (k_4) of 3.459. The higher R^2 and lower k_3 values proved that the experimental data fits very well for external diffusion model when compared to intra-particle diffusion model. Therefore the

biosorption of Coomasie Brilliant Blue G250 dye by *P. purpureum* biosorbent was found to be limited by external diffusion mass transfer.

Table 2 Pseudo-first order and Pseudo-second order kinetics values of biosorption of dye by *P. purpureum* biosorbent

Parameters	*External diffusion*	*Intra-particle diffusion*
R^2	0.983	0.978
k	0.0253	3.459
Constant (C)	48.584	10.264

Figure 9 External diffusion model for biosorption of dye by *P. purpureum* biosorbent

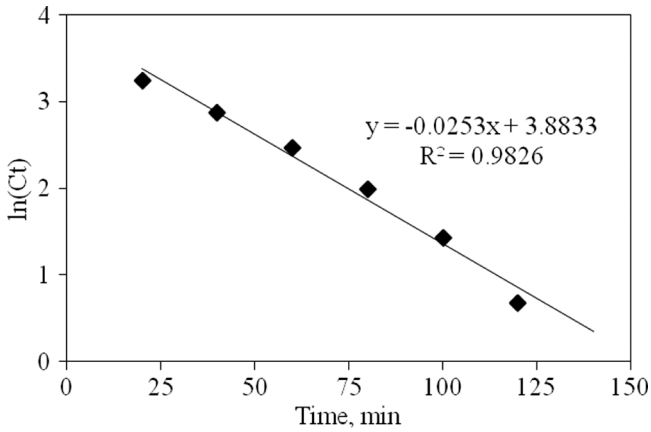
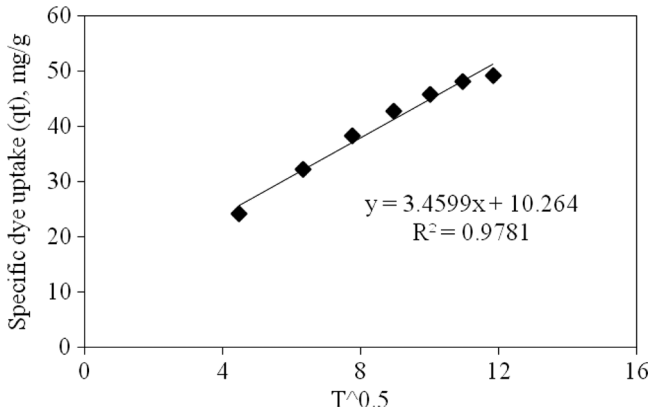


Figure 10 Weber-Morris intra-particle diffusion model for biosorption of dye by *P. purpureum* biosorbent



4 Conclusion

The biosorption capacity of *P. purpureum* biomass was studied for Coomasie Brilliant Blue G250 and found be a potential biosorbent. The kinetics of the biosorption of Coomasie Brilliant Blue G250 dye by *P. purpureum* adsorbent was agreed well with Pseudo-second order model and the rate of adsorption was found to be proportional to the active site in the adsorbent. The external diffusion mechanism was found as the potential rate controlling step for the biosorption of Coomasie Brilliant Blue G250 dye by *P. purpureum* biosorbent. Thus the *P. purpureum* biomass was found to be a low cost biosorbent for the removal of dyes from textile industrial effluents.

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