



Biosorption of synthetic dyes from textile industrial effluent using waste papaya latex

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Abstract

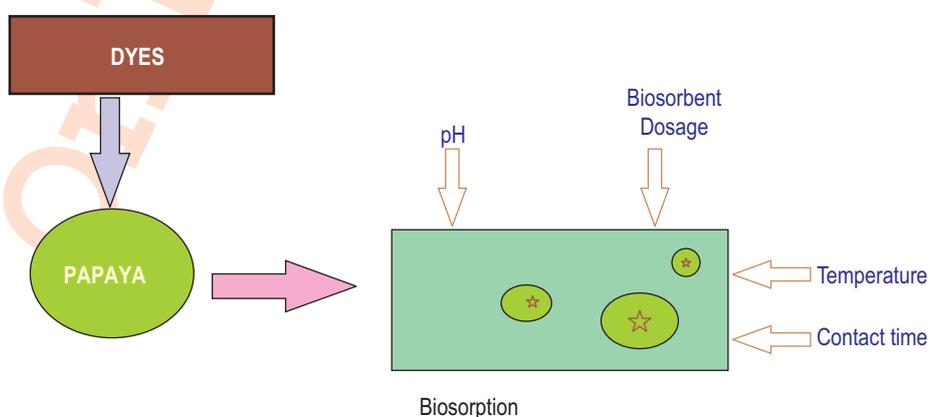
Aim: The effect of pH, adsorbent concentration, time, initial dye concentration and agitation speed on adsorption capacity of papaya latex as an adsorbent was investigated.

Methodology: The biosorption experiments were carried out in batch process by means of conical flask and agitated in a thermostated shaking incubator at rpm for a suitable contact time at different temperature range. The experiments were carried out at different pH (4-9), fixed biosorbent dosage, temperature, contact time, initial dye concentration and agitation speed. The samples were withdrawn at regular intervals and the concentration of dyes were analyzed using a UV-Visible spectrometer.

Results: The adsorption studies revealed that papaya latex removed about 83.18% of the reactive red dye and 87.97% of deep marine blue dye from aqueous phase under optimal conditions. The maximum adsorption of both dyes was found at pH 4. Adsorption isotherms such as Langmuir, Freundlich, Temkin and Dubin models were attempted to predict equilibrium data. The pseudo-first-order kinetics provide the best correlation of experimental data.

Interpretation: Waste papaya latex can be used as an eco-friendly and low-cost adsorbent in removal of reactive red dye and deep marine blue from textile effluent.

Key words: Deep marine blue dye, Kinetics, Isotherm, Papaya latex, Reactive red dye



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Introduction

Textile industry is one of the most important chemically intensive industries that has significance throughout the world. On earth, it is one of the leading water consumer industries and the highest polluter of clean water. About 2,000 different chemicals are used in processing of dye in textile industries, from dyes to transfer agents (Deniz and Kepekci, 2016). The pollution load on the liquid effluent increases with subsequent steps of chemical processing, where contribution of one single dyeing step amounts to about 8-20% of the total pollution load due to incomplete exhaustion of dye (Dallel et al., 2018). Water consumption in chemical processing operation varies from 50-300 l per kg of the processing goods, so that a mill processing 100,000 mt of cotton per day may require water varying from 2.5 to 4.0 million litre per day. About 40,000 to 80,000 tons of dyes are estimated to be discharged every year in textile processing units on global basis. Dye production in India is estimated to be around 64,000 MT, which is about 6.6% of the world production. There are around 700 varieties of dyes and dye intermediates produced in India (Hameed, 2009)

A huge amount of synthetic dyes are produced to meet the society needs, exports and as a basic material for various industries every day. About 50% of the dyes are lost to textile processing unit effluent during dyeing and finishing operations due to lack of proper methodology and inefficiency of dyeing process (Cengiz et al., 2012). Most dyes escape from convention treatment process as they are highly stable and it is difficult to remove them. Various methods like adsorption (Annadurai et al., 2002., Chiou and Li, 2002), oxidative and reductive treatments (Aplin and White, 2000; Tokuda et al., 1999; Yoshida et al., 2001; Alvarez and Pletcher, 1999), electrochemical treatment with flocculation (Marzinokowaki and Van Clewe, 1998) and membrane separation (Diaper et al., 1996) have been widely used for removal of dyes in textile and dyes effluents. Each technology has its own merits and demerits. The treated effluents contain high BOD and synthetic complex aromatic agents which adversely affect the aquatic environment (Bhunia, et al., 2001; Lin and Peng, 1994).

Currently, biosorption method is considered to be effective in removal of dyes from effluents as it is a simple, eco-friendly and easy handling. In this process, a soluble dye from effluent is transferred to the adsorbent solid surface (Zaharia and Susteu, 2012). However, choosing a suitable adsorbent is a major task. Adsorbent is produced from variety of materials such as cucumber peel, wheat shells (Bulut and Aydin, 2006; Ahalya et al., 2012), rice husk ash (Chowdhar et al., 2011) and agricultural wastes (Adegoke and Bello 2015; Nirmal Kumar and Cini Oommen, 2012).

The latex of *Carica papaya* is a rich source of cystein, endopeptidases, including papain, glycol endopeptidase,

chymopapain and caricain, which constitute more than 80% of the whole enzyme fraction. Papain (EC 3.4.22.2) is a minor constituent (5–8%) among the papaya endopeptidases. The enzyme is widely used as meat tenderizer, and have several other applications, e.g., defibrinating wounds, treatment of edemas, shrink proofing of wool, etc. (Thippeswamy et al., 2012)

The current work focus on removal of dye using waste papaya latex biomass and to study the effect of operating parameters such as pH, biosorbent dosage, initial dye concentration, contact time, agitation and temperature.

Materials and Methods

Commercial deep marine blue dye and reactive red dye were obtained from local industries. The chemicals such as sodium hydroxide, hydrochloric acid were obtained from Fissure chemicals. The materials such conical flasks, pipettes, test tubes, beakers etc, were obtained from Borosil Ltd. UV- Visible spectrometers as well as pH meters of the brands Shimadzu and Elico were used for the analysis part of the experiments.

Collection and preparation of sample: Reactive red and deep marine blue dyes were provided by Clariant Chemicals, Tiruppur, Tamil Nadu, India. The dye was stored at 4 °C for experimental studies. Papaya latex was collected from Senthil Papain and Food Products Pvt. Ltd.

Biosorption experimental procedure: Biosorption experiments were carried out by batch process and agitated in a thermostated shaking incubator for a suitable contact time of 45 min at different temperature (298-333 K). The experiments were carried out at initial pH of dye solution (1 to 8), fixed biosorbent dosage (0.05 to 3 g) using 50 ml of initial concentration of dye solution. At regular intervals, the samples were withdrawn and the residual concentration of Deep Marine Blue and Reactive Red were analyzed using a UV- Visible spectrometer.

The amount of dye biosorbed onto gram of absorbent was calculated by the equation given below

$$q_e = \frac{(C_0 - C_e)}{m} \times V \quad (1)$$

where, q_e is the amount of dye biosorbed at equilibrium (mg g^{-1}); V is the volume of dye solution (l) and m is the mass of biosorbent used (g).

Results and Discussion

The effect of pH was observed on adsorption of Red Dye and Deep Marine Blue Dye at 30°C and 25 mg l^{-1} dye concentration (Fig. 1). The maximum bioadsorption was found at pH 4 and removal of dye decreased with increase in pH (Belala et al., 2011). The results confirm that the dye possess negative

charge due to the presence of sulphonate groups. The percentage of dye was higher in acidic solution and lower in basic solution. The increase in pH increased the number of negative charged active sites on the adsorbent which decreases the possibility of sorption of dyes on negatively charged surface as dyes are anionic (McMullan, 2001). Interestingly, similar kind of results were not observed for metal ions (Adie Gilbert *et al.*, 2011).

The removal ability of waste Papaya latex was determined by biosorbent dosage. The effect of biosorbent was examined by varying the biosorbent dosage from 0.02 to 0.3 g at initial dye concentration of 25 mg l⁻¹ and initial pH 4.0 (Fig. 2). The results revealed that the increase in dosage decreased the biosorption capacity for both dyes due to increase in surface available for dye biosorption (Farkas *et al.*, 2013).

The effect of initial dye concentration was investigated by varying the initial dye concentration in the range of 10-50 mg l⁻¹ at biosorbent dosage of 0.1 g for DMB, 0.05 g for RR and 4.0 initial pH (Fig. 3). Previous studies have reported that biosorption capacity increases with initial dye concentration due to larger

driving force offered by initial concentration of dye to overcome the mass transfer resistance (Georgieva *et al.*, 2015).

To investigate the effect of contact time on adsorption process, the contact time varied from 25 to 120 min at minimum dosage of 1g 100 ml⁻¹, pH 4, temperature of 30°C and initial concentration of 25 mg l⁻¹ (Fig. 4). It was observed that dye removal efficiency increased with contact time up to 60 min for reactive red dye (Belala *et al.*, 2011) and remains constant on further increase of contact time due to the availability of vacant sites during the initial stage and non-availability of vacant sites after the attainment of equilibrium (Kamranifar *et al.*, 2018). In case of deep marine blue removal efficiency increased up to 45 min and remained constant on further increase in time.

In order to determine the effect of temperature on biosorption capacity, the experiments were carried out at temperatures ranging from 303 to 323 K with the initial dye concentration of 25 mg l⁻¹ (Fig. 5). It was observed that biosorption increased with increase in temperature as the rise of temperature enhances the mass transfer diffusion rate and raises the number of active sites (Iqbal and Saeed, 2007).

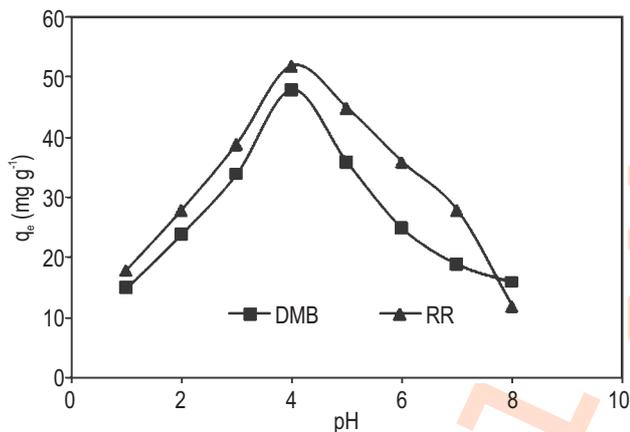


Fig. 1 : Effect of pH.

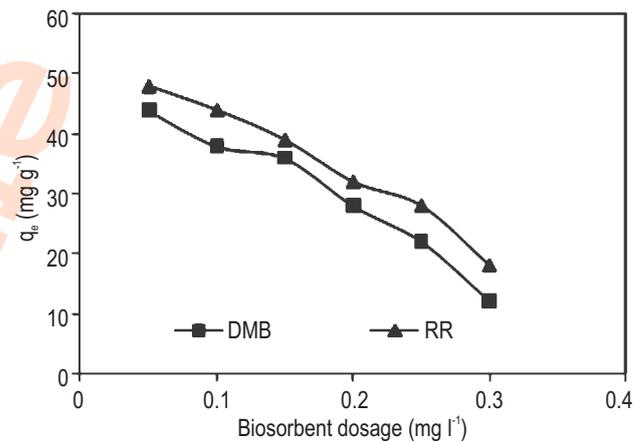


Fig. 2 : Effect of biosorbent dosage.

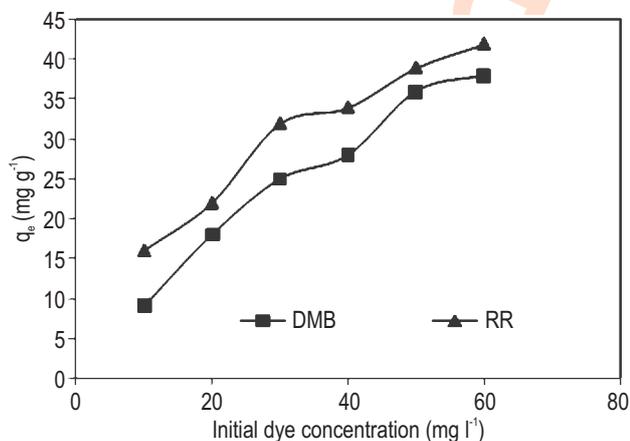


Fig. 3 : Effect of initial dye concentration.

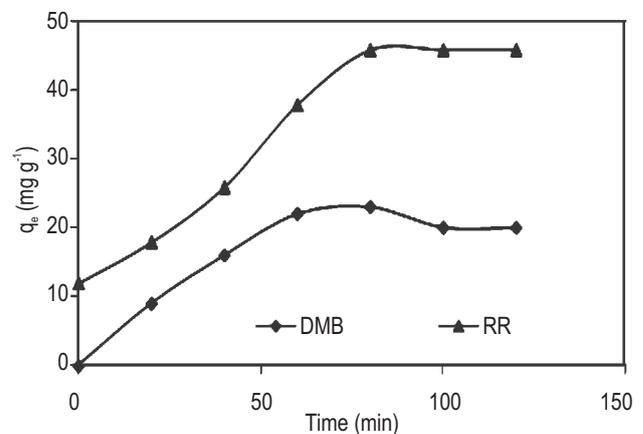


Fig. 4 : Effect of contact time.

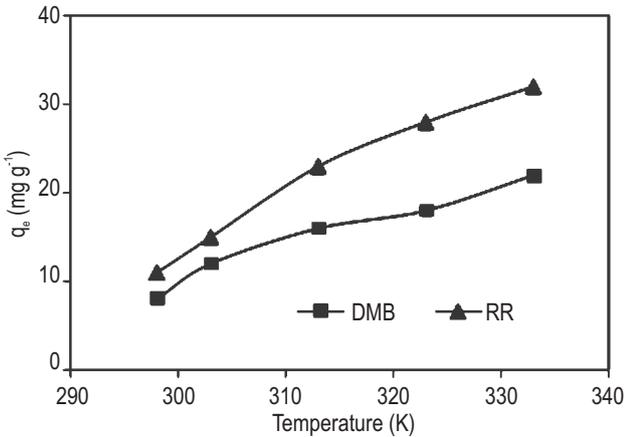


Fig. 5 : Effect of temperature.

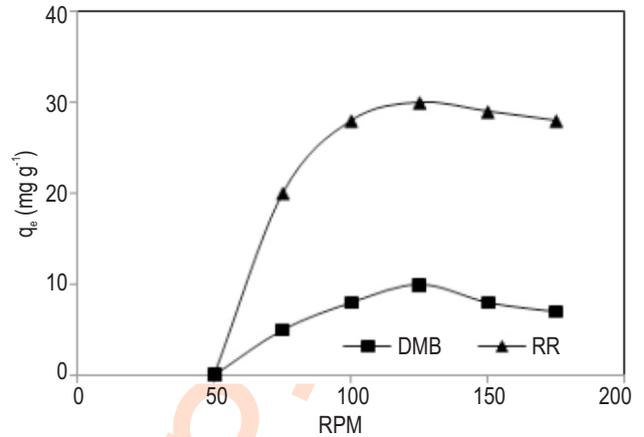


Fig. 6 : Effect of agitation speed (rpm).

To investigate the effect of agitation on biosorption capacity, the agitation time varied from 50 to 175 rpm. The biosorption capacity increased with agitation up to 125 rpm for both cases of dye and decreased on further increase on agitation (Fig. 6). The results revealed that the agitation has a significant effect on removal of dye due to the effect of higher diffusion rate of the adsorbate into the intraparticle pores of the adsorbent (Khan et al., 2018).

Different isotherms such as Langmuir isotherm (Fig. 7a), Freundlich isotherm (Fig. 7b), Temkin isotherm (Fig. 7c) and Dubin- Radushkevich (D-R) isotherm (Fig. 7d) are used to represent the kinetic behavior of dye adsorption. The isotherms Langmuir, Freundlich were identified as best adsorption isotherms for dye removal for both reactive red and deep marine blue dyes. Adsorption process is described by isotherms which connect the amount of adsorbate on the adsorbent (Singh et al., 2018). Distribution of metal ions between the adsorbate and adsorbent and the equilibrium data were analyzed by several isotherms such as Langmuir, Freundlich and Tempkin models. Langmuir adsorption model is derived for single adsorbate case and adsorbate sites with homogeneous surface (Dada et al., 2012). Langmuir adsorption model is given by the equation below:

$$\frac{1}{q_e} = \frac{1}{q_0} + \frac{1}{q_0 K_L C_e} \quad (2)$$

Where, q_e is the amount of metal adsorbed per gram of the adsorbate at equilibrium (mg g^{-1}), q_0 is the maximum monolayer coverage capacity (mg g^{-1}), K_L is the Langmuir isotherm constants (L mg^{-1}), and C_e is the equilibrium concentration of the adsorbate (mg l^{-1}). The values of q_0 and K_L were computed from the slopes and intercepts of Langmuir plot of $1/q_e$ vs. $1/C_e$. The value of R_L decides the type of isotherm and it was determined to be less than 1, which indicates that Langmuir isotherm is suited for the adsorption (Morosanu et al., 2018).

$$R_L = \frac{1}{1 + (1 + K_L C_e)} \quad (3)$$

Freundlich isotherm is significant multisite adsorption isotherm for uneven surfaces and is given by:

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad (4)$$

where, K_f is the Freundlich isotherm (mg g^{-1}), n is the adsorption intensity, C_e is the equilibrium concentration of adsorbate (mg l^{-1}), and q_e is the amount of metal adsorbed per gram of adsorbate at equilibrium (mg g^{-1}) (Nayak and Pal, 2017). The constant K_f is an indicator of adsorption capacity, while $1/n$ is a function of strength of adsorption in adsorption process which was found by linear plot. From the data obtained, the value of $1/n$ was 0.706 and the value of n was found to be greater than unity ($n = 1.416$), confirming higher affinity between the sorbent and sorbate (Khalaf, 2008).

Temkin isotherm is derived for heterogeneous surface energy system:

$$q_e = \frac{RT}{b} \ln A_T C_e \quad (5)$$

Where, A_T = Temkin isotherm equilibrium binding constant (L g^{-1}); T = Temkin isotherm constant; R = universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$); T = Temperature at 298K; b = Constant related to heat of sorption (J mol^{-1}) (Singh et al., 2017). The heat of sorption (b) and A_T values were calculated to be 7.038 J mol^{-1} and 0.1686 l mg^{-1} from the slope and the intercept of linearized Temkin plot which indicates that it was a physical adsorption (Ncibi et al., 2009).

Dubin–Radushkevich isotherm is given by:

$$q_e = q_s \exp(-k_{ad} \varepsilon^2) \quad (6)$$

Where, q_e , q_s , K_{ad} , are constants; q_e is the amount of adsorbate in the adsorbent at equilibrium (mg g^{-1}); q_s is the theoretical isotherm saturation capacity (mg g^{-1}); K_{ad} is the Dubinin–Radushkevich isotherm constant ($\text{mol}^2 \text{ kJ}^{-2}$) and - Dubinin–Radushkevich isotherm constant (Shakoor and Nasar, 2017).

The adsorption phenomenon is determined by the kinetics of adsorption process. Kinetic mechanism of the adsorption is evaluated by means of pseudo- first order models. The validity of order of reaction was determined by regression coefficients, the predicted Q_e values and Chi- square (Rangabhashiyam et al., 2018). The correlation coefficients express the predicted values for the model. A model with higher correlation coefficient suits the adsorption kinetics successfully. Biosorption is found to follow pseudo first order kinetics (Unuabonah et al., 2009).

The rate expression for pseudo first order kinetics is given by

$$\log C_e = \frac{k_1}{2.303} t + \log C_0 \quad (7)$$

Where, C_0 and C_e are the concentration (mg l^{-1}) of chromium ions at initial and at t time, respectively, and k_1 is the first order rate constant (min^{-1}) and t is the time of adsorption process. The graph of $\log C_e$ vs. t gives the value of rate constant and regression coefficients. The slope of the plot predicts the rate constant and the intercept of the plot gives the regression coefficient (Wanyonyi et al., 2014).

The integrated form of pseudo-first-order-kinetics is expressed by:

$$\log(q_o - q_e) = \log(q_o) - \frac{k_1}{2.303} t \quad (8)$$

Where, q_o and q_e are the amount (mg g^{-1}) of ions at initial and at time t , respectively, and k_1 is the first order rate constant (min^{-1}) and t is the time of adsorption process. The linear plot of $\log(q_o - q_e)$ vs. t predicts the applicability of process (Saeed et al., 2009).

FTIR analysis of the adsorbent papaya latex showed two peaks at wave numbers OH stretching and -C=C stretching, respectively (Fig. 8). The spectrum showed peaks at 3354.715cm^{-1} which confirms the presence of free hydroxyl groups of carboxylic acids, alcohols and phenol on the surface of the adsorbent and the peak at 1633.03 cm^{-1} represents the bending vibrations of O-C-H, C-C-H and C-O-H groups (Pavan et al., 2014).

The study revealed that papaya latex could be effectively used to remove Reactive red dye and deep marine blue dye from

Langmuir isotherm model

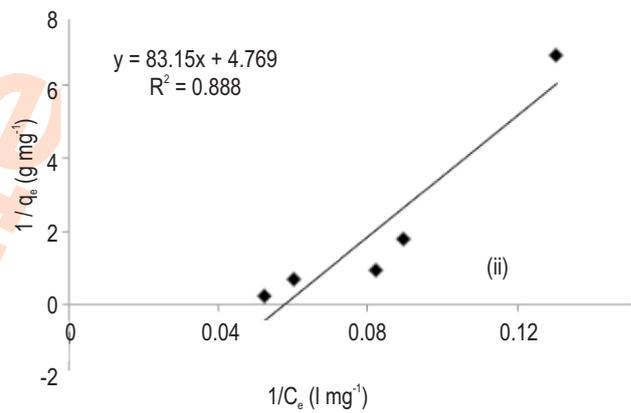
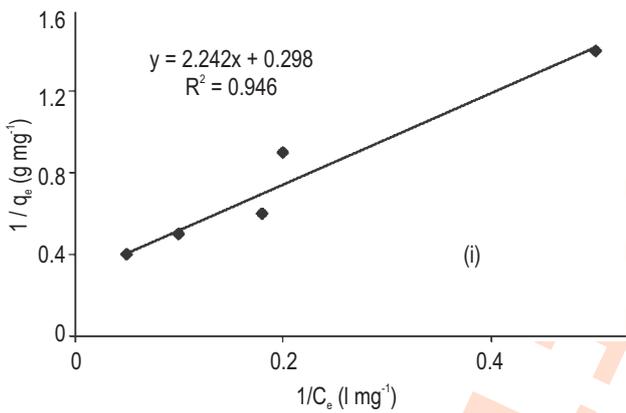


Fig. 7(a) : Langmuir plots for dyes removal (I) and RR (ii) DMB.

Freundlich isotherm model

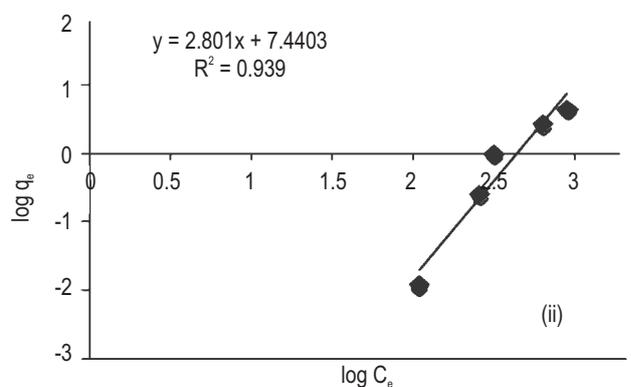
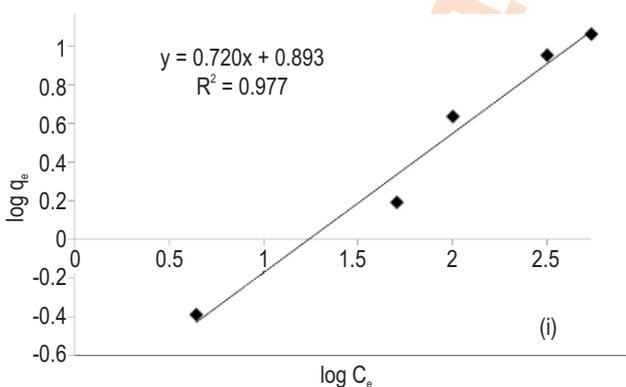


Fig. 7(b) : Freundlich plots for dyes removal (I) RR and (ii) DMB.

Temkin isotherm model

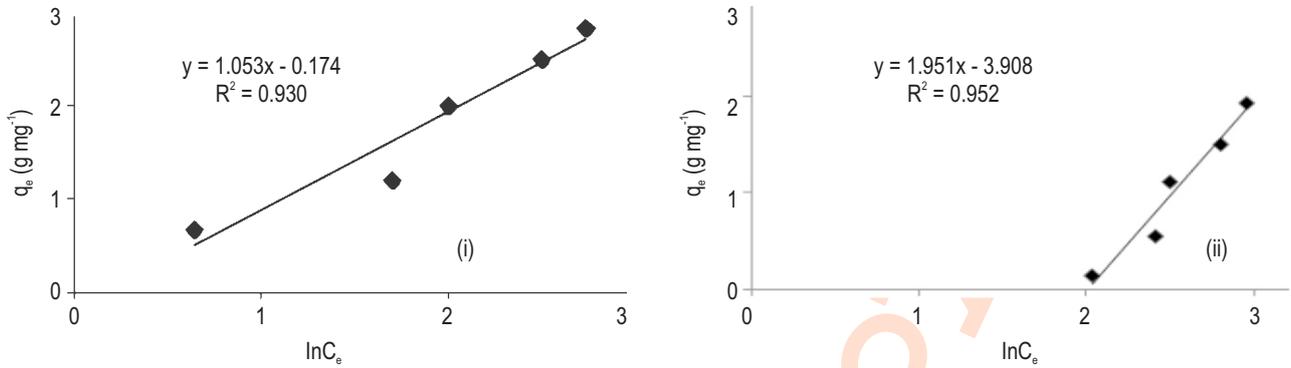


Fig. 7(c) : Temkin plots for dye removal (i) RR and (ii) DMB.

Dubin – Radushkevich (D-R) isotherm model

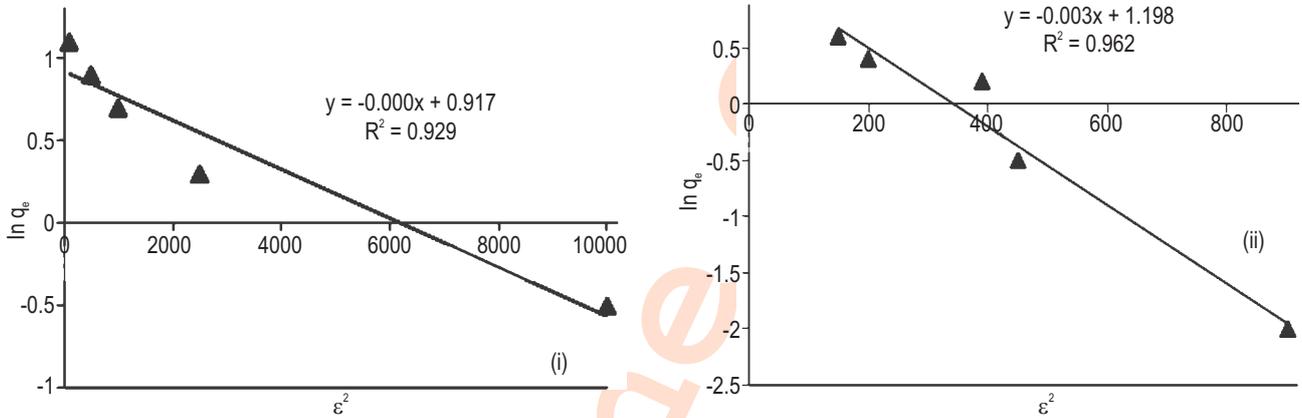


Fig. 7(d) : Dubin –Radushkevich (D-R) plots for dye removal (i) RR and (ii) DMB.

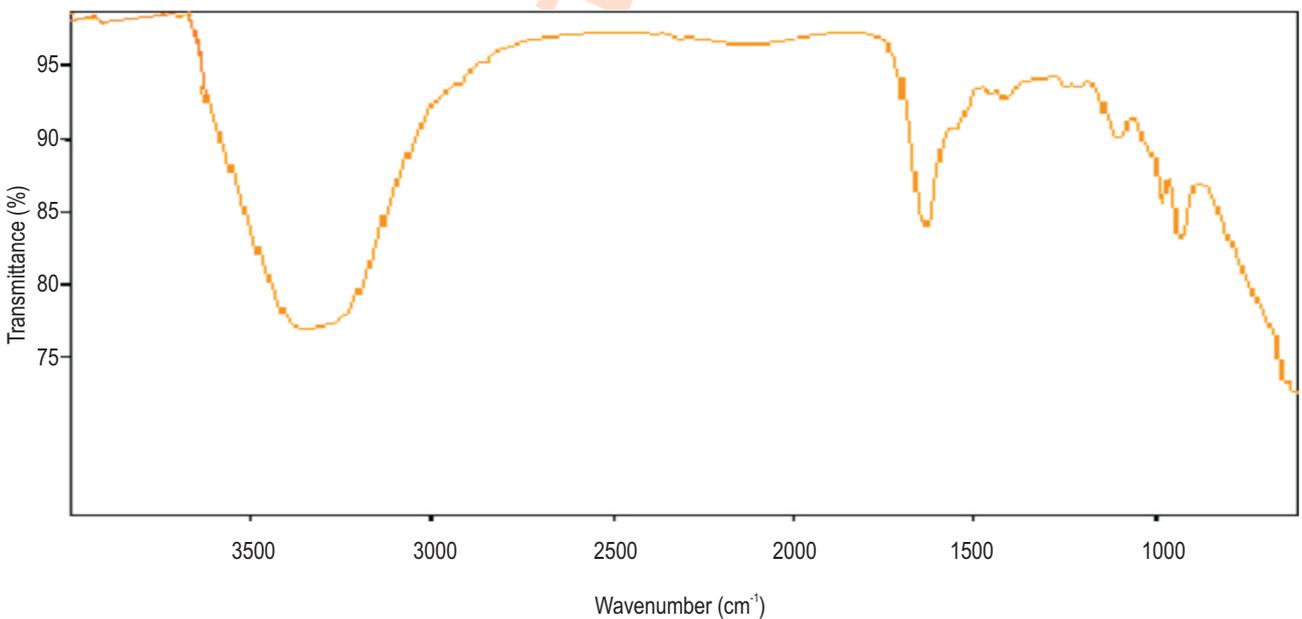


Fig. 8 : FTIR analysis for adsorbent (papaya latex).

an aqueous solution. At pH 4, adsorbent concentration of 1.6 g 100 ml⁻¹ and incubation of 45 min maximum removal was observed for deep marine blue dye. At pH 4, adsorbent concentration of 1.2 g 100 ml⁻¹ and incubation of 30 min maximum removal was observed for reactive red dye. It was found that the adsorption process by papaya latex as an adsorbent could be well described by both Freundlich and Langmuir isotherm. Furthermore, a pseudo-first-order kinetics showed to be well suited with the rate of adsorption. Although, the generation of effluents containing direct dyes by textile industry is almost unavoidable, the results of the experimental studies can help to design an appropriate remediation plan to minimize the unfavorable impacts caused by industrial effluents. Further studies on natural absorbents like latex of papaya their characteristics and efficiency on large scale would reveal whole new prospect for remediation of industrial pollutants.

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