

## **Research Article**

# **Adsorption Kinetics and Thermodynamics Study for the Removal of Anionic Dye Eosin-B From Aqueous Solution by Anion Exchange Membrane: Adsorption Kinetics and Thermodynamics**

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**Abstract:** Batch adsorption of anionic dye Eosin-B (EB) from aqueous solution on anion exchange membrane (EBTAC) has been studied at ambient temperature. The influence of contact time, membrane dosage, ionic strength and temperature on the adsorption of dye from aqueous solution were investigated. Kinetic models such as pseudo-first-order, pseudo-second-order, Elovich model, liquid film diffusion model, modified Freundlich equation and Bangham equation were used to evaluate the experimental data. Endowments such as adsorption capacities, rate constant and related correlation coefficients for every model were measured and discussed. Thermodynamic characteristics illustrated that adsorption of EB on EBTAC followed pseudo-second-order kinetics. Moreover, change in Gibbs free energy ( $\Delta G^\circ$ ), EB on EBTAC revealed that the adsorption of EB dye on EBTAC is an exothermic process.

**Keywords:** Adsorption, Eosin-B, Anion exchange membrane, Kinetics, Thermodynamics, Exothermic process

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## **INTRODUCTION**

Modern industrial processes and households yield a lot of waste-water every year. Treatment and recycling of wastewater provides a solution for lack of water in the world. The wastewater contains several types of contaminants which are harmful to the environment. Dyes, one of the adverse pollutants, are usually detected in the effluent water from textile, leather, paper, printing, and cosmetic industries. Dye removal from water can be achieved via several techniques, such as physical, chemical and biochemical processes. An effective and attractive process for the treatment of these dyes bearing wastewater involves adsorption technique [1-3]. The adsorbents reported in the literature include natural or synthetic products such as zeolites, activated clay, activated carbons, activated slag, chitosan beads, cellulosic resins, polymeric resin, modified rice husk, modified cross-linked starch, palm kernel fiber, red mud, bottom ash, de-oiled soya, etc. [4-20]. Their price and efficiency change from one adsorbent to other.

In recent years, commercial anion exchange resins have been shown to possess excellent adsorption capacity and demonstrate efficient regeneration property for the removal and recovery of reactive dyes [4, 21]. As, packed-bed operations in anion exchange

resins would suffer from certain disadvantages (e.g. slow pore diffusion, low accessible flow rate, high pressure drop and flow channeling). Therefore, in order to eliminate the above limitations, anion exchange membranes, instead of resin particles, have been successfully used to remove the anionic reactive dyes, for example, Cibacron blue 3GA and Cibacron red 3BA against water [22]. The macroporous membrane system can not only remove the technical problems of packed-bed operation but also exhibit the property of scale-up by simple stacking several membranes together or by using a large membrane area. Thus, the ion exchange membrane becomes an excellent choice for industrial applications.

In this manuscript, commercial anion exchange membrane EBTAC was employed to remove anionic dye Eosin-B (EB) from aqueous solution. The effect of contact time, membrane dosage, temperature and ionic strength on the adsorption of EB from aqueous solution was studied in batch mode. Adsorption kinetics was revealed by employing pseudo-first-order model, pseudo-second-order model, Elovich model, liquid film diffusion model, the Bangham equation and the modified Freundlich equation. Thermodynamics has also been studied for adsorption of EB on to anion exchange membrane EBTAC.

**EXPERIMENTAL**

**Adsorbent**

The commercial anion exchange membrane EBTAC was provided by Chemjoy Membrane Co. Ltd, Hefei, Anhui, China. The ion exchange capacity (IEC) and water uptake ( $W_R$ ) of EBTAC membrane are 0.42 mmol/g and 164.31 % respectively. They were used as adsorbents for anionic dye Eosin-B (EB). Prior to experiment, it was conditioned with 1 M HCl and NaOH to withdraw impurities.

**Adsorbate**

The commercial anionic dye Eosin-B (EB), obtained from Fluka chemicals was used as adsorbate. A stock solution of 1000 mg/L was prepared by dissolving 1.0 g of accurately weighed for each dye into 1 litre of deionized water and required concentrations were obtained by further dilution of stock solution. All the chemicals used in the experiments were of analytical reagent grade. The structures of EB are shown in Fig.1.

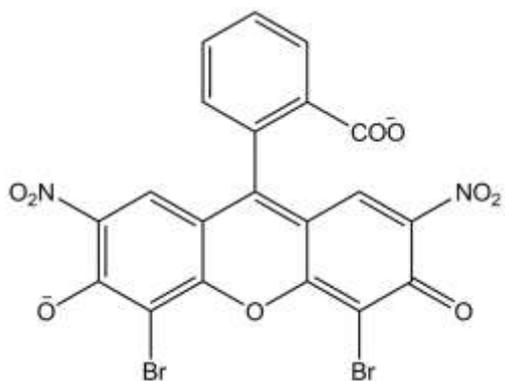


Fig. 1 Chemical structure of Eosin-B (EB) dye

**Adsorption**

Batch adsorption of Eosin-B (EB) dye was carried out by immersing anion exchange membrane (EBTAC) into measured volume of aqueous dye solution at room temperature. The bottles were shaken at a constant

speed of 120 rpm. EB concentration was determined by UV/Vis spectrophotometer (UV-2550, SHIMADZU) and related calibration curves were obtained. The wavelength used for EB was 514 nm. The EB adsorption on EBTAC at time  $t$ , was calculated by equation 1.

$$q_t = \frac{C_o - C_t}{W} \times V \tag{1}$$

Where  $C_o$  and  $C_t$  are the concentration of EB at initial state and at time  $t$  respectively. Similarly  $V$  and  $W$  are volume of EB aqueous solution and weight of adsorbent respectively.

**RESULT AND DISCUSSION**

**Effect of Operational Parameters**

Herein, the influence of operational parameters such as contact time, membrane dosage, ionic strength and temperature on the removal of EB dye from aqueous solution has been discussed. Their details are given below.

**Effect of contact time**

The effect of contact time on the percentage removal of EB from aqueous solution by anion exchange membrane EBTAC was investigated keeping membrane dosage (0.1 g), concentration of dye (25 mg/L and 50 mg/L), volume of solution (40 ml) and stirring speed (120 rpm) at room temperature and results are shown in Fig. 2. It is shown in Fig. 2 that the uptake of EB dye was very rapid initially and then continued to increase with contact time at relatively slow rate until it reached a state of equilibrium after 24 hrs. It is attributed to the fact that a lot of vacant active sites are present for adsorption of dye on the membrane surface in the initial stage of reaction. But with the passage of time, the available active sites are hard to be occupied due to repulsive forces between solute molecules on the solid and bulk phase.

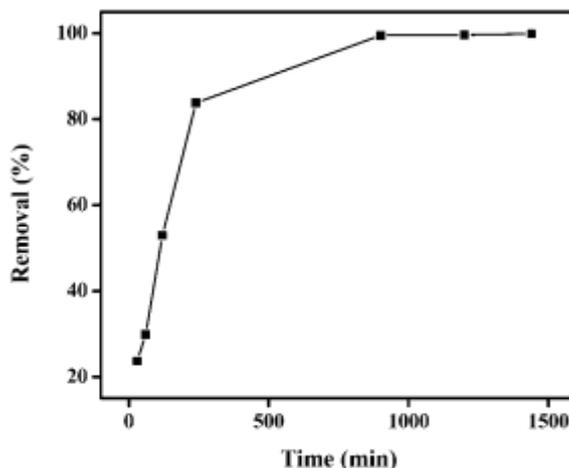
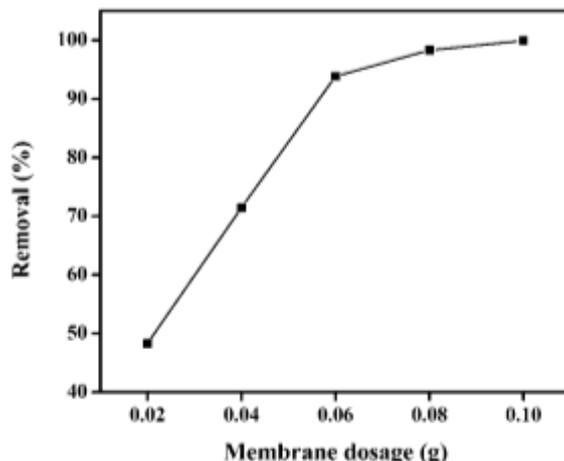


Fig. 2 : Effect of contact time on the removal of EB from aqueous solution by anion exchange membrane EPTAC

**Effect of membrane dosage**

The influence of membrane dosage on the percentage removal of EB from aqueous solution was studied keeping the other conditions constant and results are represented in Fig. 3. It has been observed that the percentage removal of EB increases with increasing the membrane dosage. The percentage removal of EB is found to be increased from 48.31 % to 99.91 % with increasing membrane dosage from 0.02 g to 0.1 g. This increase in adsorption of EB dye was attributed to the increase in the number of available sorption sites on the

surface of the anion exchange membranes. It can be seen from the Fig. 3 that the removal of EB was rapid in the initial stage, and then it becomes almost constant with further increase in the membrane dosage. It can be seen that the removal of EB was maximum at membrane dosage of 0.1 g. Therefore, 0.1 g was selected as the optimum quantity and used in further experiments to get better results. The observed two stage-dependent adsorption behaviour has also been reported in the literature [23].

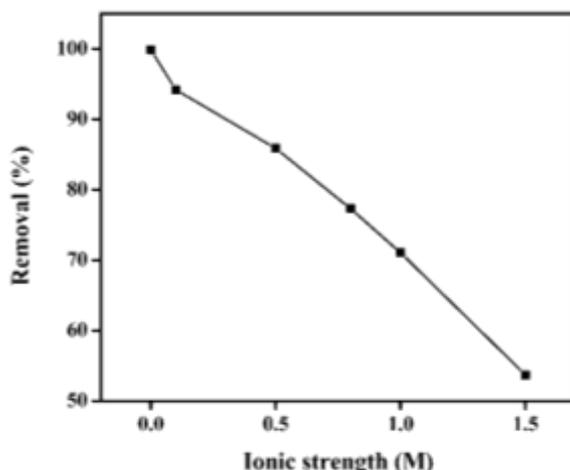


**Fig. 3 Effect of membrane dosage on the removal of EB from aqueous solution by anion exchange membrane EPTAC**

**Effect of ionic strength**

The ionic strength of the solution is an important parameter that governs both the electrostatic and non-electrostatic interactions between the dye and the membrane surface. The effect of ionic strength on the removal of EB from solution was investigated by adding different quantity of sodium chloride to the dye

solution and results are given in Fig. 4. It can be seen from Fig. 4 that the removal of EB dye decreases with increasing salt concentration. The removal of dye is found to be decreased from 94.21% to 53.70 % with increasing salt concentration from 0 M to 1.5 M. This could be due to the competition between EB anions and Cl<sup>-</sup> for the active sorption sites [24].



**Fig. 4 Effect of ionic strength on the removal of EB from aqueous solution by anion exchange membrane EPTAC**

**Effect of temperature**

The effect of temperature on removal of EB from aqueous solution was studied by keeping the contact

time, membranes dosage, stirring speed, solution volume and concentration (25 mg/L to 50 mg/L) constant and results are shown in Fig. 5. It has been

observed that the removal of EB decreases with increasing temperature. The removal of EB is found to be decreased from 99.91 % to 90.51 with increasing the

temperature from 303K to 323 K. These results show that adsorption of EB on anion exchange membrane EBTAC is an exothermic process.

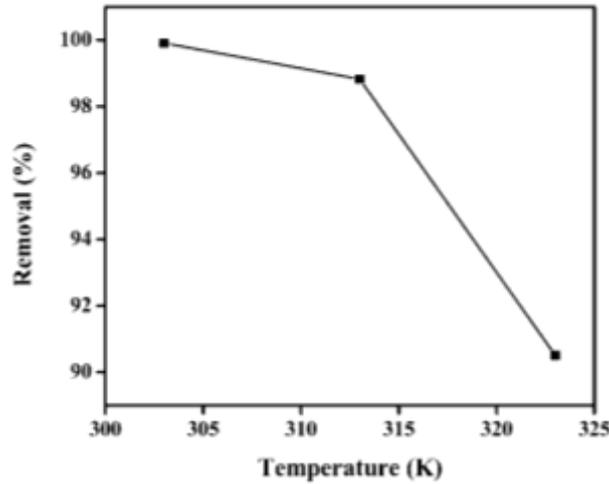


Fig. 5: Effect of temperature on the removal of EB from aqueous solution by anion exchange membrane EPTAC

**Adsorption Kinetics**

Several adsorption models have been utilized to study the controlling mechanism of adsorption process such as chemical reaction and diffusion control.

**Pseudo-first-order model**

The linearized form of the Lagergren Pseudo-first-order rate equation is given by [25]

$$\log(q_e - q_t) = \log q_e - \frac{K_1 t}{2.303} \quad (2)$$

Where  $q_e$  and  $q_t$  are the amount of adsorbate adsorbed at equilibrium and time  $t$  respectively, and  $k_1$  (/min) is the

rate constant of pseudo-first-order adsorption model. The plot of  $\log(q_e - q_t)$  vs time for pseudo-first-order model is given in Fig. 6. The value  $K_1$  is calculated from slope of eq. 2 and given in Table 1. These plots are linear, however the linearity of these curves does not necessarily assure the mechanism due to the inherent disadvantage of correctly estimating the equilibrium adsorption capacity [26]. Furthermore, there is a large difference between experimental adsorption capacity value ( $q_{e,exp}$ ) and calculated adsorption capacity value ( $q_{e,cal}$ ), therefore, pseudo-first-order model does not explain the rate process.

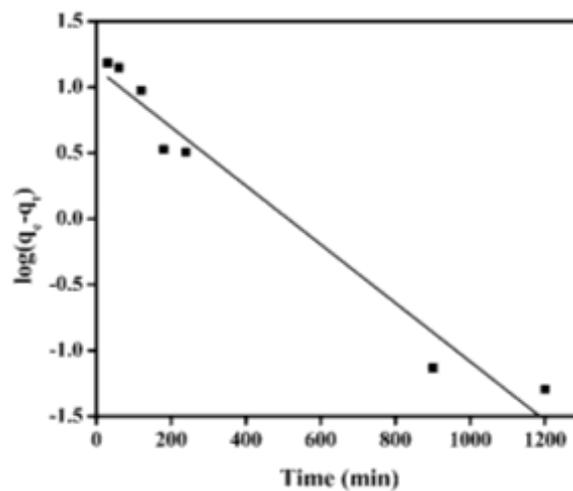


Fig.6: Pseudo-first-order kinetics for adsorption of EB on anion exchange membrane EPTAC

**Pseudo-second-order model**

The linearized form of pseudo-second kinetic model is expressed as [27]

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (3)$$

where  $k_2$  (g/mg.min) is the rate constant of pseudo-second-order model. The graphical representation of pseudo-second-order model is depicted in Fig. 7. The values of adsorption capacity ( $q_e$ ) can be determined from slope of Fig. 7 and are given in Table 1. It has

been observed that value of experimental ( $q_{e, \text{exp}}$ ) adsorption capacity is very close to the calculated adsorption capacity ( $q_{e, \text{cal}}$ ). Moreover, the value of

correlation coefficient ( $R^2 > 0.995$ ) is close to unity which indicates that experimental data fits well to the pseudo-second-order model.

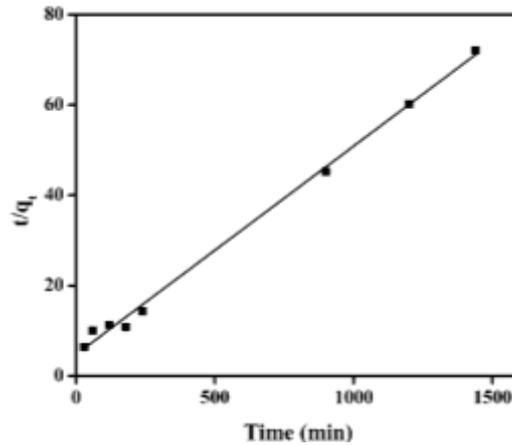


Fig.7: Pseudo-second-order kinetics for adsorption of EB on anion exchange membrane

**Elovich model**

The most interesting model to describe the activated chemisorption is the Elovich equation[28].

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t \quad (5)$$

Where  $\alpha$  and  $\beta$  are constants. The parameter  $\alpha$  is considered as initial sorption rate (mg/g.min) and  $\beta$  is

related to the extent of surface coverage and activation energy for the chemisorption. The plot of  $q_t$  vs  $\ln t$  for Elovich model is given in Fig.8. The values of  $\alpha$  and  $\beta$  are determined from intercept and slope of Fig. 8 and are given in Table 1. The values of correlation coefficient ( $R^2$ ) was 0.873 lower than that of pseudo-second-order model.

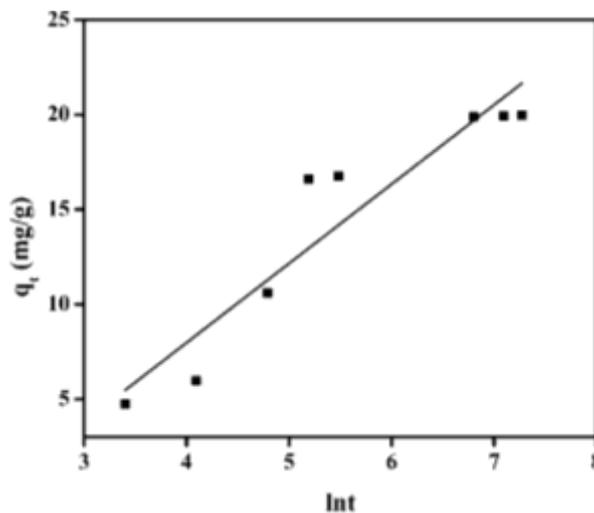


Fig.8 Elovich model for adsorption of EB on anion exchange membrane EPTAC

Table 1: Pseudo-first-order, pseudo-second-order and Elovich model rate constants ( $q_e$ :mg/g;  $k_1$ : (/min);  $k_2$ :g/mg.min;  $\alpha$ : mg/g.min;  $\beta$ : g/mg)

Pseudo-first-order				Pseudo-second-order			Elovich model		
$q_{e(\text{exp})}$	$q_{e(\text{cal})}$	$k_1 \times 10^{-3}$	$R^2$	$q_e$	$k_2 \times 10^{-4}$	$R_2$	$\alpha$	$\beta$	$R^2$
19.98	13.80	2.22	0.959	21.73	4.46	0.995	0.51	0.24	0.873

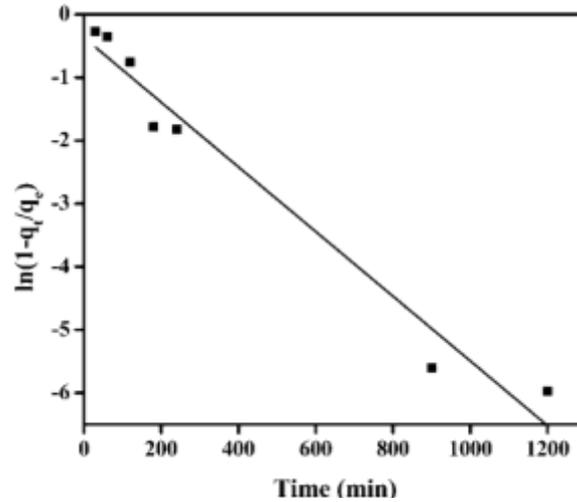
**Liquid film diffusion model**

The liquid film model is expressed as [29].

$$\ln(1 - F) = -K_{fd}t \tag{4}$$

Where  $K_{fd}$  is liquid film diffusion rate constant, and  $F=q_t/q$ . The plot of  $\ln(1-F)$  vs time for liquid film model is given in Fig.9. The values of  $K_{fd}$  are calculated from

slope of Fig. 9 and are given in Table 1. The value of correlation coefficient ( $R^2$ ) is found to be 0.959 which was lower than pseudo-second-order model. It indicates that liquid film diffusion model is not suitable to explain the experimental data.



**Fig. 9 Liquid Film diffusion model for adsorption of EB on anion exchange membrane EPTAC**

**Modified Freundlich Equation**

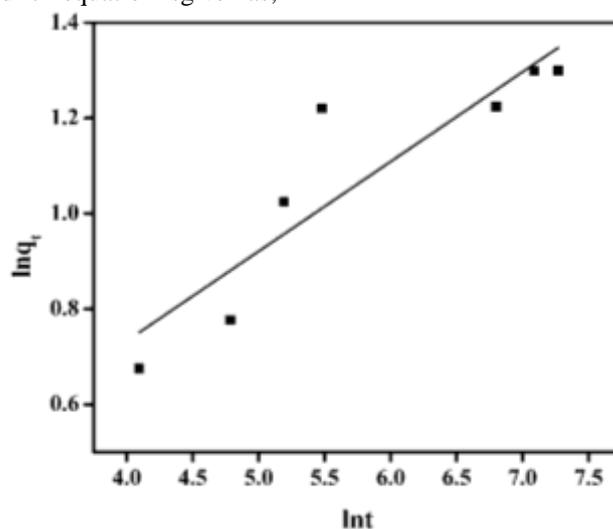
The modified Freundlich equation was originally developed by Kuo and Lotse [30]

$$q_t = kC_o t^{1/m} \tag{7}$$

where  $q_t$  amount of adsorbed dye (mg/g) at time  $t$ ,  $k$  apparent adsorption rate constant (L/g.min),  $C_o$  the initial dye concentration (mg/L),  $t$  the contact time (min) and  $m$  is the Kuo-Lotse constant. The values of  $k$  and  $m$  were used to evaluate the effect of dye surface loading and ionic strength on the adsorption process. Linear form of modified Freundlich equation is given as;

$$\ln q_t = \ln(kC_o) + \frac{1}{m} \ln t \tag{8}$$

The graphical representation of modified Freundlich model is given in Fig. 10. The parameters  $m$  and  $k$  were determined from slope and intercept of Fig. 10 and are given in Table 2. The value of  $m$  and  $k$  increases with increasing the initial concentration of dye. The correlation coefficient values was found to be 0.991 which is lower than pseudo-second-order model. Therefore, it is not suitable to explain experimental data.



**Fig. 10 Modified Freundlich equation plot between logt vs logqt for adsorption of EB on anion exchange membrane EPTAC**

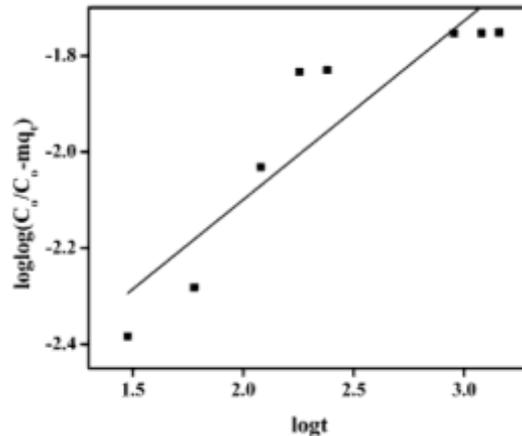
**Bangham Equation**

Bangham equation [31] is given as

$$\log \log \left( \frac{C_o}{C_o - q_t m} \right) = \log \left( \frac{k_o m}{2.303V} \right) + \alpha \log t \quad (6)$$

Where  $C_o$  is the initial concentration of dye solution (mg/L),  $V$  is volume of solution (mL),  $q_t$  is the amount of dye adsorbed (mg/g) at time  $t$ ,  $m$  is the weight of adsorbent used (g/L).  $\alpha$  (<1) and  $k_o$  (mL/(g/L) are

constants. The plot of  $\log \log(C_o / C_o - q_t m)$  Vs  $\log t$  is given in Fig. 11. The values of  $\alpha$  and  $m$  were calculated from slope and intercept of Fig. 11 and are given in Table 2. The double logarithmic plot did not give linear curves for EB removal by anion exchange membrane (EBTAC) indicating that the diffusion of adsorbate into pores of the sorbent is not the only rate controlling step [32, 33]. It may be concluded that both film and pore diffusion were important to different extent in the removal of EB from aqueous solution.



**Fig. 11** Bangham equation plot between  $\log t$  vs  $\log \log(C_o / C_o - q_t m)$  for adsorption of EB on anion exchange membrane EPTAC

**Table 2:** Liquid film diffusion model, modified Freunlich equation and Bangham equation rate constant ( $k_{fd}$ : (/min);  $k$  : L/g.min;  $k_o$ : mL/g/L)

Liquid film diffusion model			Modified Freunlich equation			Bangham equation		
$k_{fd} \cdot 10^{-3} C_{fd}$	$R^2$		$m$	$k$	$R^2$	$k_o$	$\alpha$	$R_2$
5.1	-0.39	0.959	0.76	0.005	0.991	1.33	0.37	0.805

**Adsorption Thermodynamics**

Thermodynamic parameters show the feasibility and spontaneity of adsorption process. The parameters namely change in Gibb’s free energy ( $\Delta G^\circ$ ), enthalpy ( $\Delta H^\circ$ ) and entropy ( $\Delta S^\circ$ ) were determined from given equations

$$\ln K_c = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \quad (9)$$

$$K_c = \frac{C_a}{C_e} \quad (10)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (11)$$

where  $K_c$ ,  $C_a$ ,  $C_e$ ,  $R$ ,  $T$  are the the equilibrium constants, amount of dye (mol/L) adsorbed on the adsorbent per litre (L) of the solution at equilibrium, equilibrium concentration (mol/L) of dye in solution, general gas constant (8.31 J/mol.K) and absolute temperature (T) respectively. Similarly  $\Delta G^\circ$ ,  $\Delta H^\circ$  and  $\Delta S^\circ$  are the change in Gibb’s free energy (KJ/mole), enthalpy

(KJ/mol) and entropy (J/mol.K) respectively. The plots of  $\ln K_c$  versus  $1/T$  for adsorption of EB on anion exchange membrane is shown in Fig. 12. The adsorption enthalpy ( $\Delta H^\circ$ ) and entropy ( $\Delta S^\circ$ ) were determined from slope and intercept of Fig. 12 and are given in Table 3. The values of Gibb’s free energy ( $\Delta G^\circ$ ) were positive for adsorption of EB on anion exchange membrane at all temperatures studied. Its values are found to be increased with increasing the temperature as represented in Table 2. It might be owing to interaction between adsorbent and adsorbate, with unbalanced competition imputed to heterogeneity of membrane surface and system got energy from external source at higher temperatures. The negative value of enthalpy ( $\Delta H^\circ$ ) indicates that the adsorption of EB on anion exchange membrane is an exothermic process. Similarly, the negative values of entropy ( $\Delta S^\circ$ ) represent decrease in randomness at the dye-membrane interface during the adsorption of EB on anion exchange membrane.

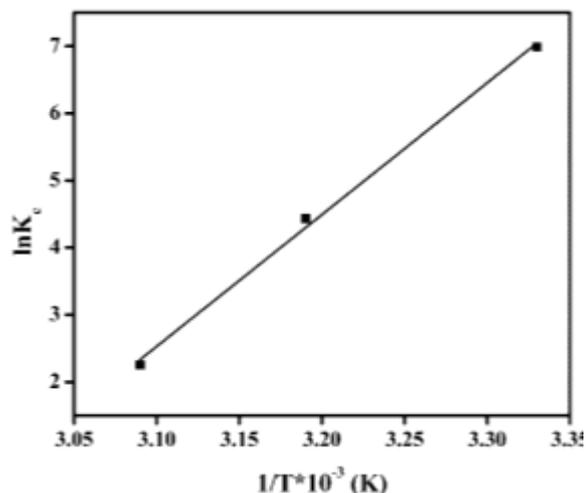


Fig. 12: Plot of 1/T vs lnKc for adsorption of EB on anion exchange membrane EPTAC

Table 3 :Thermodynamic parameters for adsorption of EB on anion exchange membrane EPTAC

$\Delta H$ (KJ/mol)	$\Delta S$ (J/mol)	$\Delta G$ (KJ/mol)		
		303 K	313 K	323 K
-163.04	-484.60	14.66	15.15	15.64

**CONCLUSION**

In this manuscript, the adsorption of anionic dye EB from aqueous solution on anion exchange membrane was investigated using different initial dye concentrations. The influence of contact time, membrane dosage, ionic strength and temperature on the removal of EB from aqueous solution was studied. These results revealed that removal of EB increases with increasing contact time, membrane dosage but decreases with ionic strength and temperature. Adsorption Kinetics showed that experimental data fitted well with the pseudo-second-order model. Moreover, thermodynamic study indicated that the adsorption of EB on anion exchange membrane was exothermic process. The negative value of  $\Delta S^\circ$  suggested that the randomness decreases between dye-membrane interface during adsorption. Thus, anion exchange membrane (EBTAC) is an excellent adsorbent for EB dye removal from waste water.

**Nomenclature**

- Code: Full name
- EB: Eosin-B
- IEC : Ion exchange capacity (mmol/g)
- $W_R$  : Water uptake (%)
- $C_o$ : Initial Concentration of dye (mg/L)
- $C_t$  : Concentration of dye at time t (mg/L)
- W : Weight of adsorbent (g)
- V : Volume of adsorbate (dm<sup>3</sup>)
- $k_1$  : Rate constant of pseudo-first-order model (/min)
- $k_2$  : Rate constant of pseudo-second-order model (g/mg.min)
- $\alpha$  : Initial sorption rate (mg/g.min)
- $\beta$  : Extent of surface coverage and activation energy for the Chemisorption (g/mg).
- $K_{fd}$  : Liquid film diffusion rate constant

- K : Apparent adsorption rate constant (L/g.min)
- m : KuoLotse constant.
- $\Delta G^\circ$  : Change in Gibb's energy
- $\Delta S^\circ$  : Change in entropy
- $\Delta H^\circ$  : Change in enthalpy

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