Adsorption of Benzo Fast Scarlet from Aqueous Solution by Parthenium L (Carrot Grass): Equilibrium and Kinetic Studies.

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Abstract

In present work, adsorption experiments were carried out for the removal of Benzo Fast Scarlet from aqueous solutions using Parthenium L. The results have shown that, the amount of dye adsorption increases with increasing the initial concentration of the dye and temperature. The adsorption kinetic data were analysed by using various kinetic models. The correlation coefficient and comparison between theoretical and experimental values of adsorption showed that the first order kinetic model was the most appropriate model, describing the adsorption kinetics. The statistical values explain the better fitting of first order model. The kinetic experimental results were fitted to adsorption diffusion models like film diffusion, intra-particle diffusion model one suggested by Weber-Morris and another by Dumwald-Wagner.Equilibrium data were fitted to the Langmuir, Freundlich and Temkin isotherm models. In all above three isotherm models the values of correlation coefficient of Freundlich isotherm are comparatively greater than Langmuir isotherm and Temkin isotherm. Freundlich isotherm model fits better for adsorption of Benzo Fast Scarlet onto Parthenium L other than two models.

Key words: Parthenium L, Benzo Fast Scarlet, adsorption, kinetic, thermodynamics.

Introduction

Synthetic dyes are one of the main pollutant groups of water. The presence of dyes contamination in water, even in very low quantities, is highly visible and undesirable; color interferes with penetration of sunlight into waters; retards photosynthesis; inhibits the growth of aquatic biota and interferes with gas solubility in water bodies[1-5]. The dyes cannot be

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decomposed easily[6-7]. Direct discharge of dyes containing effluents into environment may cause the formation of toxic carcinogenic breakdown products. The highest rates of toxicity were found amongst basic and diazo direct dyes[8-9]. Therefore, it is highly necessary to reduce dye concentration in the wastewater. The conventional methods for treating dye containing wastewaters are electrochemical treatment[10-11], coagulation and flocculation[12], chemical oxidation[13], liquid–liquid extraction[14] and adsorption[15-18]. Adsorption has been shown to be an effective way for removing organic matter from aqueous solutions in terms of initial cost, simplicity of design, ease of operation and insensitivity to toxic substances.

The main objective of present work was to evaluate the adsorption aptitude of Parthenium L for the removal of BSF as a model compound for basic dyes. BSF can be used for silk, wool, paper and pulp dyeing, also can be used in the manufacture colors which have pigment.BSF is irritant to eyes, skin, mucous membranes and respiratory system. It may be harmful by ingestion, skin absorption and inhalation. Adsorbents prepared from P. hysterophorus L are tested to remove heavy metals and dyes[19-21]. The effects of contact time, initial dye concentration and temperature on adsorption capacity were investigated. Moreover, kinetic and equilibrium models were used to fit experimental data and the adsorption thermodynamic parameters were determined.

Experimental

Preparation of adsorbent

Naturally dried plant leaves of Parthenium L (PL) are cut into tiny pieces and treated with 0.0025 M sulphuric acid. It stirred for half an hour vigorously using mechanical stirrer at room temperature. It then filtered and washed with distilled water repeatedly to remove free acid till the pH of residual solution is in the range of 4.5 to 5.0. It is then dried in air and finally dried in oven at 363-373 K for 8-10 hours and powdered homogeneously using electric grinder. The homogeneous powder was passed through mesh for desired particle size 800 -1,000 μm.

Preparation of sorbet

A dye Benzo Fast Scarlet (BSF) was chosen as sorbet its stock solution was prepared by dissolving accurately weighed BSF in double water to the concentration of 1×10^{-3} Molar. The

experimental solutions were obtained by diluting the dye stock solution in accurate proportions to required initial concentrations.
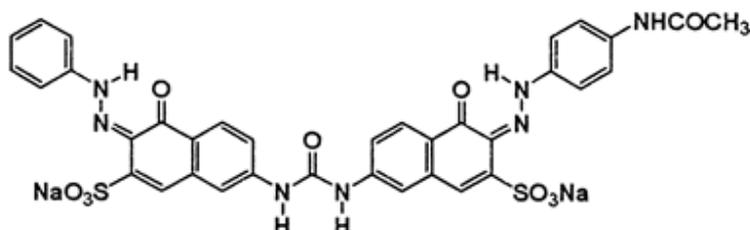

Fig. 1. Molecular structure of Benzo Fast Scarlet

Adsorption dynamics

i) Effect of initial concentration

The effect of initial concentration of BSF was studied using concentration range 5-25 mg.L^{-1}. A typical experimental procedure was conducted by measuring accurately 100 ml of the BSF solution of known concentration; 0.9 g of PL was added and agitated up to 80 min, based on the results obtained from the equilibrium time studies, carried out as a preliminary study. Samples were withdrawn, at fixed time intervals, centrifuged, and the supernatant was analyzed for residual BSF using a UV-visible spectrophotometer. The amount of BSF uptake by PL in each flask was calculated using the following equation:

$$q_e = \frac{(C_0 - C_e)}{W} \times V \qquad (2)$$

Where q_e is the equilibrium adsorption capacity of BSF adsorbed on unit mass of the PL (mg g^{-1}); C_0 and C_e are the initial and equilibrium BSF concentration (mg L^{-1}) respectively; V is the volume of the BSF solution (L); and W is the weight of the PL (g).

Adsorption Isotherms and Kinetic Models

Kinetic Models

The kinetics of Benzo Fast Scarlet adsorption on the PL was analysed using following different kinetic models.

First-order kinetic model

The integrated form of pseudo-first order model[22] is

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$$log(q_e - q_t) = log q_e - \frac{K_1}{2.303} t \qquad (3)$$

Where q_e and q_t (mg.g^{-1}) are the amount of adsorption at equilibrium and time t (min), respectively. K_1 (min^{-1}) is the rate constant of the pseudo-first-order kinetic model.

Second-order kinetic model

The integrated and simplified form of second order kinetic equation[23] is

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \qquad (4)$$

Where K_2 is the rate constant of pseudo second-order adsorption (mg·g^{-1} min^{-1}).

The Elovich model

The integrated and simplified Elovich equation[24] is

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

Where α is the initial adsorption rate (mg·g^{-1}·min^{-1}) and β is the desorption constant (g·mg^{-1})

Adsorption Diffusion models

The adsorption diffusion models used were

Film diffusion mass transfer rate equation

The liquid diffusion model presented by Boyd[25-26] may be applied as follows

$$ln[1 - F] = -K_{fd} t \qquad (6)$$

Where F is the fractional attainment of equilibrium $F = q_t/q_e$ and K_{fd} is the adsorption rate constant.

Weber-Morris model

Most widely applied intra particle diffusion equation for sorption system is given by Weber and Morris[27] as

$$q_t = K_{int} t^{1/2} \qquad (7)$$

Where K_{int} is the intra-particle diffusion rate constant.

Dumwald-Wagner model

Dumwald-Wagner proposed another intra-particle diffusion model[28] as

$$log(1 - F^2) = -\frac{K}{2.303}t \qquad (8)$$

where K (min^{-1}) is the rate constant of adsorption.

Adsorption Isotherms -

Langmuir Isotherm model

The simplified form the Langmuir isotherm model[29] can be represented as

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m K_a} \qquad (9)$$

Where C_e is concentration of dye solution at equilibrium (mg.L^{-1}), q_e is amount of dye adsorbed at equilibrium (mg.g^{-1}), q_m is Langmuir constant related to adsorption capacity (mg.g^{-1}) and K_a is Langmuir adsorption intensity, (L.mg^{-1}).

The effect of isotherm shape can be used to predictwhether an adsorption system is favourable" or "unfavourable"both in fixed-bed systems[30] as well as inbatch processes[31]. According to Hall et al.[32], theessential features of the Langmuir isotherm can be expressedin terms of a dimensionless constant separationfactor or equilibrium parameter R$_L$, which is defined bythe following relationship:

The essential characteristics of the Langmuir isotherm can be expressed in terms of dimensionless constant separation factor of equilibrium parameter, which is defined by:

$$R_L = \frac{1}{1 + K_a C_0} \qquad (10)$$

The parameter R$_L$ indicates the nature of the adsorption isotherm.

R$_L$ > 1   Unfavorable adsorption;

0 < R$_L$ < 1   Favorable adsorption

R$_L$ = 0   Irreversible adsorption

R$_L$ = 1   Linear adsorption

Freundlich Isotherm model

In linear form the Freundlich adsorption isotherm[33] is represented by the equation

$$\log q_e = (1/n)\, log C_e + \log K_f \qquad (11)$$

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Where q_e is dye concentration in solid at equilibrium (mg.g^{-1}), K_f is measure of the adsorption capacity and n is adsorption intensity

Temkin model

The nonlinear form of Temkin isotherm[34] is

$$q_e = \frac{RT}{b} \ln(AC_e) \qquad (12)$$

Where, A is the equilibrium binding constant (Lg^{-1}), b is Temkin constant, R is the universal gas constant and T is the absolute temperature.

The linear form of Temkin model can be presented as follows:

$$q_e = B\ln(A) + B \ln C_e \qquad (13)$$

Where B is RT/b, B is constant related to heat of adsorption (J mol^{-1})

Test for validity of kinetic Models: Various test used for the validity of models are

1. To evaluate the fitness of kinetic experimental data, the residual root mean square residual error (RMSE) was used to measure the kinetic and isotherm constants. The RMSE can be defined as

$$RMSE = \sqrt{\frac{1}{N-2} \sum_{i=1}^{N} \left(q_{e(exp)} - q_{e(cal)} \right)^2} \qquad (14)$$

The validity of models was also determined by calculating

2. The average relative error (ARE) using

$$ARE = \frac{100}{N} \sum_{i=1}^{N} \left[\frac{\left| \left(q_{e(exp)} - q_{e(cal)} \right) \right|}{q_{e(exp)}} \right] \qquad (15)$$

3. The sum of error squares (SSE%)

$$\% SSE = \frac{\sqrt{\Sigma \left(q_{e(exp)} - q_{e(the)} \right)^2}}{N} \qquad (16)$$

Where the subscripts '*the*' and '*exp*' refer to the theoretical (calculated) and experimental data and N is the number of data points.

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Result and Discussion

Effect of initial dye concentration and contact time

The initial concentrations of BSF solution was varied from 5 to 25 mg L^{-1} and batch adsorption experiments were carried out with 0.9 g of the adsorbent at 303 K. The effect of initial dye concentration and contact time on the adsorption rate of BSF is shown in Fig.2. It is observed that percentage adsorption decreases from 88 to 67 % with increase in initial dye concentration (Fig. 3). It means that the adsorption is dependent on initial concentration of dye. At lower concentration, the ratio of the initial number of dye molecules to the available surface area is low subsequently the fractional adsorption becomes independent of initial concentration. However, at high concentration the available sites of adsorption becomes fewer and hence the percentage removal of dye is dependent upon initial concentration [35-36]. Equilibrium have established at 80 minutes for all concentrations. Figure 2 and 3 reveals that the curves are smooth, and continuous, leading to saturation.

Adsorption dynamics

To investigate the kinetics of adsorption of BSF onto PL, pseudo-first order, pseudo-second order and Elovich models were used to fit the experimental data by using linear regression analysis. The straight lines of plot of $\log(q_e - q_t)$ versus t for pseudo-first order reaction (Fig. 4), t/q_t versus t for pseudo-second order reaction (Fig. 5), and q_t versus $\ln(t)$ for the Elovich equation (Fig. 6) for adsorption of BSF on PL has also been tested to obtain the rate parameters. The kinetic parameters K$_1$, K$_2$, $q_e(exp)$ and $q_e(the)$ were calculated from these plots and are summarized in Table 1. In above three models the most of the correlation coefficient exceeds 0.9 suggesting that all models are closely fitted the experimental results. While comparing the experimental and theoretical values of equilibrium adsorption capacity for all kinetic models it is observed that the values of $q_e(exp)$ are closer to the $q_e(the)$ for first order kinetic model (Table 1). This shows that the first order kinetic model fits better as compared to other kinetic models. The values of % SSE, RMSE and ARE (Table 3) are lower for first order in comparison to second order and Elovich model that explains the better fitting of first order model.

Adsorption mechanism

In order to get insight into mechanism the kinetic experimental results were fitted to adsorption diffusion models like film diffusion, intra-particle diffusion model one suggested by Weber-Morris and another by Dumwald-Wagner. In film diffusion model a plot of $-\ln(1-F)$ versus t (Fig. 7) gives straight line passing through origin with slope equal to liquid film diffusion constant and the results are given in Table 2. Since the plot passes through origin indicates fitting of model. Weber Morris models plot (Fig. 8) shows that the sorption process tends to be followed by two phases. These two phases suggest that the sorption process proceeds by surface sorption and intra-particle diffusion[37-38]. The initial curve portion of the plot indicates a boundary effect while the second linear portion is due to intra-particle or pore diffusion. The slop of second linear portion of the plot has been defined as the intra-particle diffusion parameter K_{id2} (mg g^{-1}.$min^{1/2}$). The calculated intra particle diffusion constants K_{id1} and K_{id2} values are summarized in Table 2. In intra-particle diffusion model of Dumwald-Wagner a plot of $-log(1-F^2)$ versus t gives a straight line (Fig. 9). The rate constant of adsorption is calculated from slope and given in Table 2.

Adsorption Isotherm

The analysis of isotherm data by fitting them to different isotherm model is an important step to find the suitable model that can be used for design purpose. The experimental analysed according to linear form of Langmuir isotherm, Freundlich isotherm and Temkin isotherm[30, 35].

Langmuir Isotherm

The linear plots of C_e/q_e versus C_e suggest the applicability of Langmuir isotherm (Fig. 10). The values of q_m and K_a were determined from slope and intercept and are presented in Table 4. From the results it is clear that the value of adsorption efficiency and adsorption energy of PL increases on increasing temperature. To confirm the favorability of adsorption process, the separation factor (dimensionless factor) (R$_L$) was calculated and presented in Table 5. The values of separation factor (R$_L$) were found to be between 0 and 1 and thus again confirmed that the Langmuir isotherm model was favorable for adsorption of BSF onto PL.

Freundlich Isotherm

Freundlich adsorption plot for the adsorption for BSF onto PL is shown in Fig. 11 and the values of adsorption capacity (K_f) and intensity of adsorption (n) are given in Table 4. The values of $1/n$ were between 0 and 1 indicating that the adsorption was favorable at studied condition.

Temkin Isotherm

In the Temkin plot q_e against $\ln C_e$ (Fig. 12) also shows linearity. The values of A and B are calculated from intercept and slope and are presented in Table 4. In all above three isotherm models the values of correlation coefficient of Freundlich isotherm are comparatively greater than Langmuir isotherm and Temkin isotherm. Freundlich isotherm model fits better for adsorption of BSF onto PL other than two models.

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Fig. 2. Effect of Initial Concentration

Fig. 3. Percentage removal

Fig. 4. Pseudo First order

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Fig. 5. Pseudo second order

Fig. 6. Elovich Model

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Table 1 Rate constants for First-order, Second-order and Elovich model

Dye Conc. (mg L^{-1})	First order				Second order				Elovich model		
	K$_1$ (min^{-1})	$q_{e(exp)}$ (mg g^{-1})	$q_{e(cal)}$ (mg g^{-1})	R^2	K$_2$ (min^{-1})	$q_{e(exp)}$ (mg g^{-1})	$q_{e(cal)}$ (mg g^{-1})	R^2	β (mg g^{-1})	α (mg g^{-1} min^{-1})	R^2
5	0.04395	0.775	0.838	0.998	0.0281	0.775	1.096	0.997	4.152	0.0883	0.9984
10	0.0466	1.438	1.489	0.998	0.0191	1.440	1.9432	0.996	3.7021	0.0992	0.994
15	0.0496	2.006	2.056	0.998	0.0164	2.01	2.6257	0.996	2.7110	0.1353	0.993
20	0.0530	2.591	2.659	0.998	0.0552	2.97	3.168	0.996	2.1271	0.1724	0.990
25	0.0564	3.110	3.193	0.999	0.0136	3.11	3.917	0.996	1.8080	0.2080	0.987

Table 2 Rate constants for diffusion model

Dye Conc. (mg L^{-1})	Dumwald model		Film diffusion model		Intra Particle diffusion model		
	K$_1$ (min^{-1})	R^2	K$_{fd}$ (min^{-1})	R^2	K$_{id1}$ (min^{-1})	K$_{id2}$ (min^{-1})	R^2
5	0.0398	0.988	0.0466	0.998	0.0790	0.0283	0.955
10	0.0425	0.990	0.0489	0.998	0.1543	0.0498	0.949
15	0.0454	0.989	0.0521	0.998	0.2230	0.0650	0.942
20	0.0478	0.988	0.0542	0.997	0.2778	0.0741	0.932
25	0.0505	0.992	0.0567	0.998	0.3242	0.0741	0.919

Table 3 Statistical Parameters of Kinetic Models

Kinetic models	%SSE	RMSE	ARE
Pseudo first order	0.0286	0.0068	3.8707
Pseudo second order	0.2394	0.4777	4.704
Elovich model	0.0237	0.0047	2.6860

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Table 4: Langmuir, Freundlich and Temkin isotherm parameters for adsorption of BSF on PL

Temp (K)	Langmuir isotherm			Freundlich isotherm			Temkin isotherm		
	Ka (L mg-1)	q_m (mg g-1)	R^2	N	Kr (mg g-1)	R^2	A (L mg-1)	B (J mole-1)	R^2
303	0.276	2.645	0.984	1.1584	0.1858	0.999	3.524	0.539	0.994
308	0.296	2.710	0.986	1.1748	0.1918	0.999	3.792	0.553	0.995
313	0.344	2.733	0.980	1.1829	0.1971	0.997	4.482	0.553	0.996
318	0.376	2.759	0.991	1.9440	0.1980	0.999	5.114	0.553	0.990

Table 5

Dye conc. (mg L^{-1})	Temperature (K)			
	303	308	313	318
5	0.9398	0.9361	0.9331	0.9342
10	0.8864	0.8798	0.8746	0.8765
15	0.8387	0.8300	0.8230	0.8255
20	0.7959	0.7854	0.7771	0.7801
25	0.7573	0.7454	0.7361	0.7395