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Original article

Biosorption of acid blue 113 dyes using dried Lemna minor biomass

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ABSTRACT

Industrial wastewater is one of the major environmental pollutants. Discharge of the colorful industrial waste into the receptive waters leads to eutrophication and has mutagenic and carcinogenic properties. Therefore, the aim of this study was to investigate the removal of acid blue 113 (AB113) from aqueous solutions by dried Lemna minor. The experimental data were analyzed by the Langmuir, Freundlich, Temkin and Harkins- Jura isotherm models. Equilibrium data fitted well to the Langmuir model with a maximum adsorption capacity of 59.96 mg/g at temperature 333 K. The dimensionless separation factor RL revealed the favorable nature of the isotherm of the AB113-Lemna minor system. The pseudo-second-order kinetic model best described the adsorption process. The adsorption capacity increases with increasing temperature then the adsorption is an endothermic process. The results proved that the Lemna minor was an effective adsorbent for removal of AB113 from aqueous.

1. Introduction

Dyes are colour organic compounds which can colorize other substances (Balarak et al., 2015). Dye wastewater discharged from textile and dyestuff industries are important sources of contamination responsible for the continuous pollution of the environment (Madrakian et al., 2012). These substances are usually present in the effluent water of many industries, such as textiles, leather, paper, printing, and cosmetics (Zazouli et al., 2013). The need for the treatment of textile wastewater will be higher in near future because the presence of dyes in the receiving surface and underground waters is not safe, pleasant and welcomed (Fang et al., 2010). In addition, there are well documented facts that the ef fl uents of textile industry are containing suspected carcinogenic materials which are posing serious hazard to aquatic living organisms (Garg et al., 2003). The removal of colour from dye bearing effluents is one of the major problem due to the difficulty in treating such wastewaters by conventional treatment methods (Walker et al., 2003). Various physical, chemical, and biological methods, namely adsorption, biosorption, coagulation, precipitation, membrane filtration, solvent extraction, chemical oxidation have been used for the treatment of dye-containing wastewater (Chakravarty et al., 2008). However, of these, it has been found that adsorption technique is the most prominent for removal of dyes from wastewater (Dogan et al., 2008). The activated carbon is used as adsorbent for colour removal by adsorption (Zazouli et al., 2014). Due to high cost of activated carbon, lot of alternative adsorbents are developed and used for removal of colour from the aqueous solutions (Diyanati and Balarak, 2013). The number of non-conventional, low-cost agricultural materials are used as adsorbents for removal of pollutants from wastewater (Zazouli et al., 2013). Some of them are Azolla filiculdes, Canola and rice straw and etc (Balarak et al., 2015; Diyanati et al., 2014). Few data are available on the potential of dried Lemna minor for adsorbing dye (Diyanati et al., 2013). L. minor is a small plant that flourishes in quiescent, shallow water bodies, Including constructed wetlands, and grows in a variety of climate (Zazouli et al., 2013). The aim of the present work is to study the performance of dried L. minor, which is in great supply, inexpensive and easily found around wetland anzali, iran for removing Acid Blue 113 dyes from aqueous solution.

2. Materials and methods

The duckweed was collected from the Anzali wetland, Iran and was taxonomically classified as Lemna minor. It was then sun dried and using a disk mill to obtain material with an average particle size of 1 mm. The crushed particles were then treated with 0.1M HCl for 5 h followed by washing with distilled water and then kept for shaded dry. The resultant biomass was subsequently used in sorption experiments.

The dyes used in this study were AB113 dye obtained from Alvan Sabet Company in Hamadan, Iran and used without further purification. The chemical structures and general data of this dye are displayed in Fig. 1 and Table 1. The dye stock solutions were prepared by dissolving accurately weighted dyes in distilled water to the concentration of 1000 mg/L and the experimental solutions concentrations were obtained by dilution.

Table 1
Properties of AB113 dye (Zazouli et al., 2013).

C.I. name	Type	Molecular weight	λ_{max} (nm)	Molecular formula
Acid Blue 113	Anionic	681.66	565	$C_{26}H_{20}Br_2N_3O_6S$

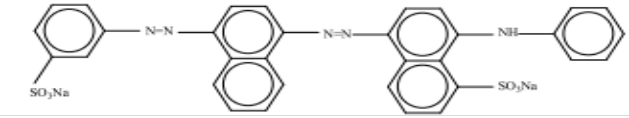

Fig. 1. Molecular structure of AB113 dye.

2.1. Batch equilibrium and kinetic studies

In adsorption equilibrium, experiments were conducted in a set of 200 mL Erlenmeyer flasks, where solutions of AB113 (100 mL) with different initial concentrations (25–200 mg/L) were added in these flasks. Equal masses of 0.20 g of L. minor of particle size 0.1 mm were added to AB113 solutions and each sample was kept in an isothermal shaker of 180 rpm at 30 ± 2 ◦C for 2 h to reach equilibrium of the solid–solution mixture. The pH of the

solutions was not adjusted. A similar procedure was followed for another set of Erlenmeyer flask containing the same AB113 concentration without L. minor to be used as a blank. The flasks were then removed from the shaker and the final concentration of AB113 in the solution was analyzed using a double beam UV–vis spectrophotometer (DR 5000) at 565 nm wavelength. The samples were filtered prior to analysis in order to minimize interference of the L. minor fines with the analysis. Each experiment was duplicated under identical conditions. The amount of adsorption at equilibrium, q_e (mg/g), was calculated by (Bulut et al., 2007):

$q_e = (C_O - C_e)\ V/m$

Where C_0 and C_e are the initial and equilibrium dye concentrations in solution, respectively (mg/L), V the volume of the solution (L) and m is the mass (g) of the adsorbent used.

3. Results and discussion

3.1. Effect of contact time

The effect of contact time on adsorption of dye can be carried out by preparing adsorbent–adsorbate solution with fixed adsorbent dose and initial dye concentration for different time intervals and shaken until equilibrium. Fig.1 reveals that the adsorption proceeded quickly and reached equilibrium within 60 min. This was probably resulted from the relatively low initial concentration of dye and the absence of micropores in the adsorbent (Diyanati and Balarak, 2013; Tan et al., 2010). Further increase in contact time does not increase the uptake due to deposition of dyes on the available adsorption site on adsorbent material.

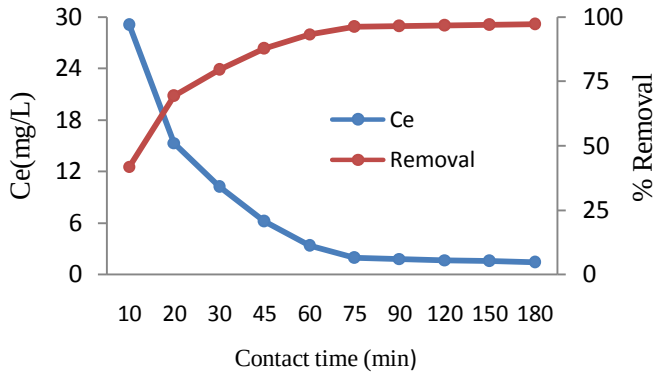

Fig. 1. Effect of contact time for AB 113 dye adsorption (Con = 25 mg/L, pH = 3, Biomass dose: 4 g/L).

3.2. Effect of temperature

A study of the temperature dependence of adsorption reactions gives valuable knowledge about the enthalpy and entropy changes during adsorption. Temperature is an indicator for the adsorption nature whether it is an exothermic or endothermic process. In order to investigate the effect of temperature on the adsorption, a series of adsorption experiments was carried out with different temperature at initial concentration of 25 to 200 mg/L without changing the volume of dye solution (100 ml) with constant speed of 180 rpm for 75 min. in this study adsorption capacity increases with increasing temperature then the adsorption is an endothermic process (Figure 2). This may be due to increasing the mobility of the dye molecules and an increase in the number of active sites for the adsorption with increasing temperature (Wang et al., 2005; Mall et al., 2006).

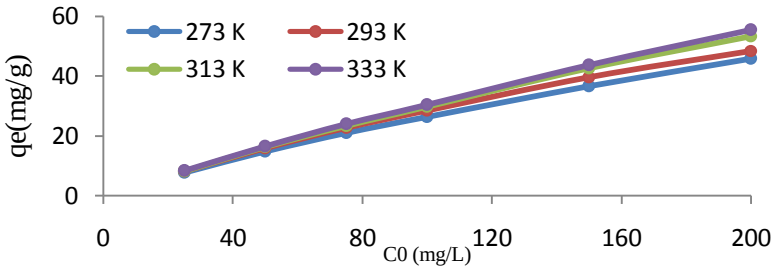

Fig. 2. Effect of temperature for AB 113 dye adsorption.

3.3. Adsorption isotherms

The analysis of the isotherm data is important to develop an equation, which accurately represents the results and could be used for designing process. The equilibrium of a solute separated between liquid and solid phase is described by various models of adsorption isotherms such as Langmuir, Freundlich, Temkin and Harkins-Jura isotherm models. The applicability of the isotherm equations was compared by judging the correlation coefficient, R^2.

3.4. Langmuir isotherm

Langmuir adsorption isotherm model is usually adopted for homogenous adsorption and it is used successfully in monomolecular adsorption processes. Linear form of Langmuir model is expressed by (Ponnusami et al., 2007):

$$\frac{Ce}{qe}=\frac{1}{q_m K_L}+\frac{Ce}{q_m}$$

Where C_e (mg/L) is the concentration of AB 113 dye at equilibrium, q_e (mg/g) is the amount of AB 113 dye adsorbed by the L. minor at equilibrium, qm (mg/g) is the maximum adsorption capacity corresponding to monolayer coverage, and K_L (L/mg) is the Langmuir constant. In order to determine if the dsorption process is favorable or unfavorable, a dimensionless constant separation factor or equilibrium parameter R_L, is defined according to the following equation (Balarak et al., 2015):

$$R_L=\frac{1}{1+K_L C_O}$$

R_L indicates the type of isotherm to be irreversible (R_L= 0), favorable (0 < R_L< 1), linear (R_L = 1) (or) unfavorable (R_L > 1). In the present investigation, the R_L values were less than one which shows the adsorption process was favorable.

3.5. Freundlich isotherm

The Freundlich isotherm model was chosen to estimate the adsorption intensity of the adsorbate on the adsorbent. Linear form of Freundlich model is expressed by (Dogan et al., 2008):

$$\text{Log } q_e=\frac{1}{n}\log Ce + \log K_F$$

K_F and 1/n are Freundlich constants related to adsorption capacity and adsorption intensity, respectively. 1/n values indicate the type of isotherm to be irreversible (1/n = 0), favorable (0< 1/n<1), unfavorable (1/n>1). The value of 1/n was less than one indicating the favorable adsorption.

3.6. Temkin isotherm

Temkin isotherm suggests that the heat of adsorption of all the molecules in the layer would decrease linearly with coverage due to adsorbate/adsorbent interactions. The linear form of Temkin isotherm is expressed as (Low et al., 2000):

$$q_e = \beta \ln \alpha + \beta \ln C$$

where β = (RT)/b, T is the absolute temperature in K, R the universal gas constant, 8.314 J/molK, α the Temkin isotherm constant (L/g) and b is related to the heat of adsorption(J/mol).

3.7. Harkins- Jura isotherm

Harkins- Jura isotherm assumes the presence of multilayer adsorption with the existence of heterogeneous pore distribution. The linear form of Harkins-Jura isotherm is expressed as (Gok et al., 2010):

$$\frac{1}{q_e^2}=\frac{B}{A}-\frac{1}{A}\log C_e$$

where C_e is the equilibrium concentration of the dye in solution (mg/L), q_e is the amount of dye adsorbed onto the adsorbent (mg/g), A and B are the isotherm constants.

The results of the isotherm parameters/constants are displayed in Table 2. Based on the correlation coefficient for all the isotherm models studied, Langmuir model gave the highest R^2 value showing that the adsorption of AB 113 onto L. minor was best described by this model followed by Temkin, Freundlich and Harkins-Jura isotherm models.

Table 2
Results of isotherm parameters for the adsorption of AB 113 onto L. minor.

		Temperature (K)			
Models	**Parameters**	**273**	**293**	**313**	**333**
Langmuir	q_m	53.11	56.39	58.08	59.96
	K_L	8.25	11.45	13.42	15.18
	R_L	0.271	0.347	0. 259	0.628
	R^2	0.994	0.995	0.997	0.998
Freundlich	K_F	2.94	3.81	4.65	5.74
	1/n	0.37	0.21	0.63	0.46
	R^2	0.912	0.904	0.885	0.868
Temkin	α	0.092	0.121	0.164	0.179
	β	9.81	11.29	13.72	19.65
	R^2	0.845	0.869	0.894	0.816
Harkins- Jura	A	181.6	169.8	154.9	134.8
	B	1.17	1.69	2.32	2.78
	R^2	0.901	0.868	0.879	0.854

3.8. Adsorption kinetics

Kinetic studies are necessary to optimize different operation condition for the adsorption of dyes. The kinetics of AB 113 onto L. minor was analyzed using pseudo-first order kinetic, pseudo- second order kinetic, Elovich and intraparticle diffusion models.

3.9. Pseudo first order kinetic model

The model was suggested for the adsorption of solid/liquid systems. The integrated linear form of the model is as follows (Ofomaja and Ho, 2007):

$$\text{Log } (q_e - q_t) = \log q_e - \frac{K_1}{2.303}t$$

Where q_e is the amount of adsorbate adsorbed per unit mass of adsorbent at equilibrium(mg/g), q_t is the amount of adsorbate adsorbed at contact time t (mg/g), k_1 is the pseudo-first order rate constant (min^{-1}). The values of the correlation coefficient obtained at all the studied concentrations are low, in the range 0.845–0.881. This suggests $_{that}$ the pseudo-first-order kinetic model is not suitable to describe the adsorption process. The pseudo-second-order kinetic model can be expressed in linear form as follows (Balarak et al, 2014; Nadaroglu et al., 2013):

$$\frac{t}{q_t} = \frac{1}{k_2 . q^2} + \frac{t}{q_e}$$

where k_2 (g/mg min) is the rate constant of the pseudo second order. The parameters are listed in Table 3. The values of the correlation coefficient are higher than 0.994 suggest that adsorption of AB 113 dye onto L. minor predominantly follows the pseudo-second-order kinetic model. The intraparticle diffusion equation is written as follows (Balarak et al., 2015; Zazouli et al., 2014):

$$q_t = K\, t^{0.5} + C$$

Where C is the intercept which describes the foundry layer thickness and K (mg/g $min^{1/2}$) is the rate constant of intraparticle diffusion. According to results, values of coefficients of determination from Table 3 are also low. From these results one can conclude that the biosorption process of AB 113 dye onto the L. minor biosorbent is

not only depended on intraparticle diffusion but other mechanisms might be involved. Therefore, the data is not fitted well to the intraparticle diffusion model.

Table 3
Kinetic parameters for AB 113 dye adsorption onto L. minor.

Pseudo second-order model				Pseudo first-order model			Intraparticle diffusion		
C_o(mg/L)	k_2	R^2	q_e (mg/g)	K_1	R^2	q_e (mg/g)	K	C	R^2
25	0.029	0.998	4.25	0.124	0.851	2.89	2.48	1.76	0.794
50	0.054	0.996	10.73	0.235	0.872	7.93	3.97	2.98	0.806
100	0.072	0.994	25.19	0.411	0.845	16.45	5.15	3.72	0.832
200	0.093	0.997	44.66	0.659	0.881	38.14	7.86	2.19	0.859

4. Conclusion

The Lemna minor can be effectively used as adsorbent for the removal of Acid blue 113 from aqueous solutions. The selection of activation depends on contact time and temperature. The adsorption capacity increases with increasing contact time and temperature, then the adsorption is an endothermic process The equilibrium data have been analyzed using Langmuir, Freundlich, Temkin and Harkins-Jura isotherms. The Langmuir isotherm was demonstrated to provide the best correlation for the adsorption of AB113 onto L. minor. The kinetics of adsorption was studied by using pseudo-first order, pseudo- second order and intraparticle diffusion models. The results demonstrate that adsorption mechanisms in the system follow pseudo-second order kinetics. Analysis of the results with the intraparticle diffusion model showed that intraparticle diffusion is not the rate limiting step.

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