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Biosorption of Acid Red 88 dyes using dried *Lemna minor* biomass

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ABSTRACT

The present investigation deals with the utilization of Lemna minor algae as an adsorbents for the removal of Acid Red88 (AR88) dye from aqueous solution. A series of experiments were conducted in a batch system to evaluate the effect of system variables. The effect of pH, initial dye concentration, dose of adsorbents and contact time were considered.

Under optimum condition (pH 3, contact time 75 min, adsorbent dose 6 g/L and AR88 concentration 25mg/L), application by Lemna minor was able to remove 98% of AR88from aqueous solutions. The equilibrium data is best fitted on Langmuir isotherm and the adsorption kinetic model follows pseudo-second model. The obtained results showed that the dried Lemna minor can be used as a high efficiency and low-cost adsorbent to treat of textile effluents.

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I. Introduction

The textile industries are account as main industry to consume the great amount of water and synthetic dyes which it is concluded to generate high volume colorful wastewater by these industries (Ahmad et al., 2006; Won et al., 2006; Zazouli et al., 2014). It is expressed that 1000 ton/yr of dyes are discharged into the effluents by textile industries in the world (Hameed et al., 2007). The synthetic dyes are classified as anionic, cationic and nonionic which the acid is located in anionic dyes group (Won et al., 2006). The toxicity and harmful impacts of the dyes have been confirmed by many studies which were

performed in the world (Wang et al., 2013). The dyes can threaten the human health by their carcinogenic and mutagenic properties; also, the aquatic life can be influence by the blocking of sun light penetration and the reduction of oxygen in water bodies (Moussavi et al., 2009; Qin et al., 2009; Donia et al., 2009). Since the textile dyes have complex aromatic structure, therefore they are difficulty removable by conventional biological treatment which it is shown the necessity to use efficient and promising method to remove these dyes (Garg et al., 2003; Irem et al., 2013). The previous studies have been indicated that the adsorption onto activated carbon is a reliable, inexpensive and significantly effective technique to remove the dyes; however, the high cost of activated carbon limits the use of activated carbon (Dizge et al., 2008; Hoda et al., 2008; Lata et al., 2014; Batzias et al., 2007). Recently, various materials such as Azolla, fly ash, wheat straw, apple pomace, fungus, and orange peels, soy meal hull, eggshell membrane and etchas been applied to develop low-cost and effective adsorbent (Zazouli et al., 2014; Doulati et al., 2008; Arami et al., 2006). The *Lemna minor* is one of wide-spread aquatic plant which is belonged to duckweed species with special characteristics including rapid growth, high nutritional value, and high water purification capabilities have been used to remove the pollutant from water and wastewater. Several studies to remove the heavy metal, dyes and etc by the *Lemna minor* have been conducted by the scientists (Ge et al., 2012; Alvarado et al., 2008; Uysal et al., 2013). Therefore, the purpose of present study was to assess the *Lemna minor*'s ability to remove the Acid red 88 (AR88) dye. Also, the effect of several parameters including contact time, pH, adsorbent dose and initial dye concentration was investigated.

II. Materials and Methods

Adsorbent preparation: *Lemna minor* was collected from Sari city. It was sun dried then crushed and finally sieved to particle sizes in the range of 1–2 mm. The biomass was treated with 0.1 M HCl for 5 h followed by washing with distilled water and then dried in shade (Zazouli et al., 2014). The resultant biomass was subsequently used in sorption experiments.

The specific surface area of adsorbent was determined by the BET method using the Gemini2357 surface area analyzer of Micromeritics Instrument Corporation, USA. Scanning electron microscopy (SEM) of the modified *Lemna minor* were carried out using a Philips XL30 scanning electron microscope for morphological features and surface characteristics.

Materials: The Acid Red 88(AR88) dye was provided from Alvan Sabet Corporation, Hamadan, Iran. The stock solution (1000mg/L) was prepared by dissolving appropriate quantity of RR88 dye. Other experimental solutions were prepared by dilution of stock solution. General characteristics and chemical structures of AR88 are presented in Table 01 and Figure 01, respectively.

Table 01. Properties of AR88 (Padmesh et al., 2005)

C. I. name	Molecular weight	λmax (nm)	Molecular formula
AcidRed 88	400.39	503	$C_{20}H_{13}N_2NaO_4S$

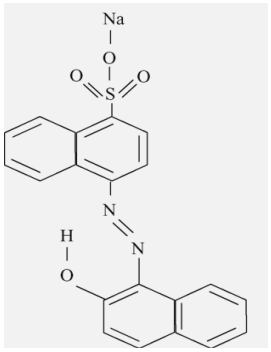

Figure 01. The chemical structures of AR88 (Padmesh et al., 2005)

Batch adsorption experiments: This study was an experimental-lab study which was conducted to evaluate the potential of *Lemna minor* in RR88 dye removal efficiency. The most effective variable including contact time (15-180min), pH (3-11), adsorbent dose (0.1-1 g/100mL) and initial dye concentration (25-500 mg/l) were selected and their effect on RR88 dye removal was investigated. The experiments were carried out in batch system by using 100mL in a 250 ml Erlenmeyer flask. The certain concentration of AR88 solution was poured into the flask. The specific amount of absorbent was added to the solution and then it was completely mixed with magnetic stirrer at 3600 rpm for 60 minutes. The prepared mixture was centrifuged at 3600 rpm for 10 minutes. Finally, the residual concentrations of samples were measured using spectrophotometer at maximum wavelength of 503nm (Padmesh al., 2005). The amount of adsorbed AR88 was calculated by following Equation 01 (Zazouli et al., 2013);

$$q_e = (C_0 - C_e)\, V/m \qquad (01)$$

Where q_e is the amount of adsorbed RR88 dye (mg/g), C_0 and C_e are the initial and equilibrium concentrations of dye (mg/L), respectively. V is the volume of the solution (L), and m is the mass of the adsorbent (g).

Adsorption isotherms: The equilibrium adsorption isotherm is importance in the design of adsorption systems. Although several isotherm equations are available, but four important isotherms including Langmuir, Freundlich, Tekmin and BET isotherms were selected. The Isotherm equations are presented the Table 02.

Table 02. The equations of isotherms

Model	Equation	References
Langmuir	$\frac{ce}{qe} = \frac{1}{q_m K_L} + \frac{ce}{q_m}$	Diyanati et al., 2012
Freundlich	$\mathrm{Log}\frac{x}{m} = \frac{1}{n}\log Ce + \log K_F$	Diyanati et al., 2012
Tekmin	$q_e = B_1 ln\,(k_t) + B_1 ln\,(c_e)$	Santhy et al., 2006
BET	$\frac{c_e}{(C_i - C_e)q} = \left(\frac{1}{q_m A}\right) + \left(\frac{A-1}{q_m A}\right)$	Santhy et al., 2006

Adsorption kinetics: The study of kinetic models was performed in contact time between 15-180 min with dye concentration of 25 and 50 mg/L and pH=3 and adsorbent dose of 6g/L. To evaluate the differences in the biosorption rates and uptakes, the kinetic data were described with Elovich, Intra particle diffusion, pseudo first, pseudo second order models. The linearized form of model is shown in Table 03.

Table 03. The equations of kinetics

Model	Equation	Reference
pseudo first order	$\log (q_e - q) = \log q_e - k_1 t / 2.3$	Zazouli et al., 2014
pseudo second order	$t/q = 1/k_2 q_e^2 + 1/q_e t$	Zazouli et al., 2014
Elovich	$q_e = (\frac{1}{\beta})\ln(\alpha\beta) + (\frac{1}{\beta}) \ln t$	Thinakaran et al., 2008
Intraparticle diffusion	$qt = k_{dif}\, t\, 0.5 + c$	Thinakaran et al., 2008

III. Results

Scanning electron microscopy (SEM) images were used to analyze the surface structure of *Lemna minor* (Figure 02). It was found that the adsorbent has heterogeneous surface structure with deep pores. The specific surface area of modified *Lemna minor* was determined in size of 30 m^2/gr.

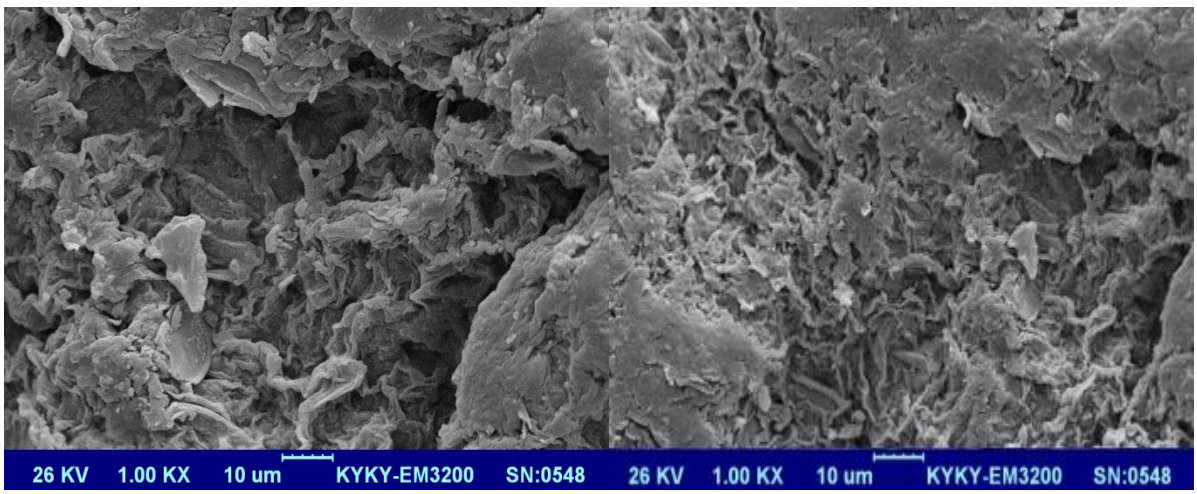

Figure 02. Scanning electron microscopy image of modified *Lemna minor* before and after used as adsorbent

Effect of contact time and initial AR88 concentration

Contact time is an important factors influencing adsorption of AR88 on dried *Lemna minor*. It is well known that the adsorption capacity and removal efficiency of dye by biosorbents increases with increasing contact time.As shown in Figure 03, biosorption of AR88 on dried *Lemna minor* increased rapidly within the first 60 min and then slowed from 60 min to 75min, reaching equilibrium after 75 min.The influence of initial AR88 concentrations, ranging from 25 to 500 mg/L, on AR88 biosorption on *Lemna minor* was investigated. As shown in Figure 04, biosorption increased significantly from 4.14 to 59.33 mg/g with increasing initial concentrations of AR88 from 25 to 500 mg/L, while the AR88 removal efficiency decreased from 99.5% to 71%.

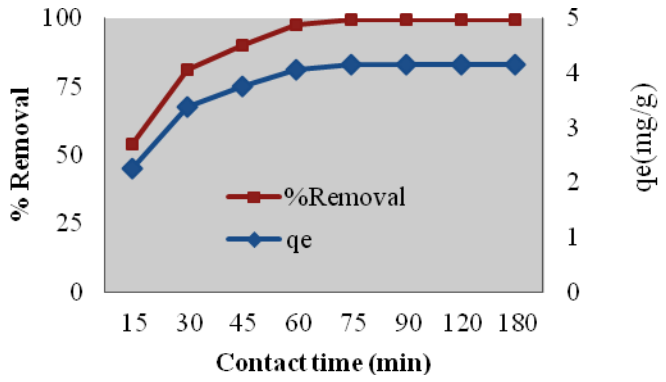

Figure 03. Effect of contact time on removal efficiency of AR88 (pH = 3, dosage 6g/L, AR88 concentration: 25 mg/L)

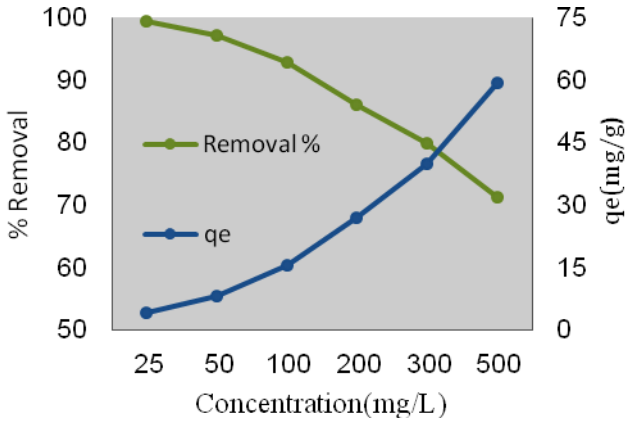

Figure 4. Effect of AR88 concentration on removal efficiency of AR88 (time = 75 min, dosage: 6 g/L, pH = 3)

Effect of adsorbent dosage and pH

The effect of pH on dye uptake in the batch process was studied by varying the pH from 3 to 11(Figure 05). The biosorption of AR88 on dried *Lemna Minor* decreased significantly with increasing solution pH from 3 to 11.The effect of biosorbent dosage on biosorption of AR88 on to dried *Lemna minor* was

studied to determine an optimum biosorbent dosage. The tested biosorbent dosages varied from 1 to 10 g/L using an initial AR88 concentration of 25 mg/L and contact time of 75 min. As shown in Fig. 6, the biosorption capacity of AR88 on the biomass decreased from 10.25 to 2.48 mg/g, while the AR88 removal percent increased from 41% to 99.5% when biosorbent dosage increased from 1 to 6 g/L. Biomass dosage increased to 6 g/L and remained approximately constant with further increases in biosorbent dosage. On the basis of both biosorption capacity and the removal percentage, an optimum biosorbent dosage of 6 g/L was selected for all further experiments.

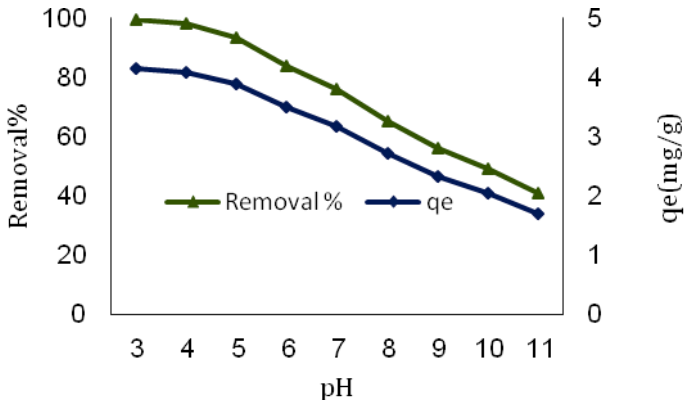

Figure 07. Effect of pH on removal efficiency of AR88 (contact time = 75 min, dosage: 25g/L, AR88 concentration: 25 mg/L)

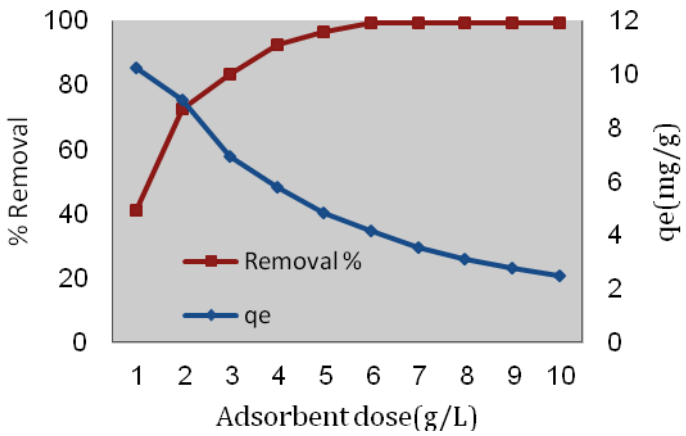

Figure 06. Effect of adsorbent dose on removal efficiency of AR88 (contact time = 75 min, PH = 3, AR88 concentration: 25 mg/L)

Adsorption kinetics and isotherms:

The results obtained by the adsorption of dye were analyzed by the well- known models of Langmuir, Freundlich and Tekmin and BET. The results showed that AR88 dye on dried *Lemna minor* fitted according to Langmuir Model isotherm model (R^2=0.998). Furthermore it agreed with BET isotherm (R^2=0.971) that better than Freundlich (R^2=0.913) and Temkin model (R^2=0.942). The isothermal models and adsorption kinetics in Table 04 and 05. The R^2 of kinetic models suggested that the pseudo

second-order model mechanism is predominant which means the uptake process follows the pseudo-second-order expression with correlation coefficients was always greater of 0.99.

Table 04. The adsorption isotherms constants for the removal Acid red 88 dye

Langmuir model			**Freundlich model**			**Temkin model**			**BET model**		
q_m	K_L	**R^2**	n	K_F	**R^2**	B	k_t	**R^2**	**A**	**q_m**	**R^2**
7.8	0.94	0.998	1.18	2.49	0.913	14.4	1.21	0.942	24.4	1.197	0.971

Table 05.The adsorption kinetic model constants for the removal Acid red 88 dye

Pseudo second-order model				Pseudo first-order model			Elovich			Intraparticle diffusion		
C_o	k_2	R^2	q	K_1	R^2	q	α	β	R^2	K_{dif}	C	R^2
25	0.14	0.998	5.17	0.084	0.917	4.45	3.21	0.46	0.945	1.12	6.71	0.895
50	0.191	0.999	9.72	0.122	0.932	7.76	5.79	0.27	0.917	1.89	4.45	0.909

IV. Discussion

The specific surface area of adsorbent was one of the most important parameters on adsorption ability. The specific surface area is related to the number of active adsorption sites of dried *Lemna minor* (Padmesh et al., 2005). The adsorption increased with the specific surface area and pore volume of the sorbent. The surface area of dried *Lemna minor* was 30 m^2/gr. which it indicated that the modified Lemna Minor area have relatively good ability to remove the pollutants.

The results showed that the dye removal density (mg/g) increased with increase in time. In the start, the biosorption rate was rapid due to adsorption of dye molecules on the upper surface of the biosorbent. Then it became slow due to slow passing of dye molecules into the inner structure of the biosorbent. Another reason was that a large number of exchanging sites in the start helped the biosorption process and then saturation occurred (Padmesh et al., 2005). Zazouli et al. (Zazouli et al., 2013) observed the effect of contact time on the Acid Blue 113 dye biosorption onto Canola. The biosorption capacity (mg/g) increased speedily in the beginning and equilibrium was attained after 75 min.

In the biosorption mechanism, in the start, the dye molecules were adsorbed externally and the biosorption rate increased rapidly. When the external surface became saturated, the dye molecules adsorbed into the porous structure of the biomass (Fang et al., 2005). The initial concentration of the dyes provides an important driving force to overcome the mass transfer resistance of all molecules between the aqueous and solid phases. Ponnusami et al. (Ponnusami et al., 2007) observed that the amount of dye sorbed per unit mass of biosorbent increased with an increase in initial dye concentration from 50 to 250 mg/L. It is estimated that the binding sites of biosorbent stay unsaturated during the biosorption mechanism.

Lower biosorption capacity of dyes at a higher dose of biosorbent is probably due to the decrease of the surface area of the biosorbent by the overlapping or aggregation during the sorption. However, the

higher the dose of the biosorbent in the solution, the greater the availability of active sites for dyes, leading to the higher dyes removal (Cardoso et al., 2013).

Dye biosorption is a pH dependent process. The pH of the solution influences the properties of biomass materials, affects the adsorption mechanisms and dissociation of the dye molecules. At lower pH, the biosorbent surface turned out to be positively charged and electrostatic attraction developed between positively charged biomass and negatively charged anionic dyes. However, at basic pH, adsorption decreased due to presence of hydroxyl ions which showed competition with dye anions for binding sites (Suna et al., 2010). Diyanati et al. (Diyanati et al., 2013) examined the effect of initial pH on adsorption of Acid orange 7 from aqueous solution onto Rice Stem. As pH increased from 3 to 11, the adsorption capacity decreased from 2.81 to 1.35 mg/g at dye concentration 10 mg/l. Maximum uptake of dye was observed at pH 2.0.

The biosorption of sorbate (AR88) onto the adsorbent (*Lemna minor*) was modeled using the Langmuir, Freundlich, Tekmin, BET equations. The Langmuir isotherm assumes monolayer coverage of a sorbate on to the solid surface of adsorbent, uniform energy of sorption, and no transmigration of sorbate in the plane of the surface. While freundlich, Tekmin, BET equations are based on the hypothesis of multi-layer biosorption (Toor et al., 2011). The correlation coefficient (R^2) for Freundlichisotherm was 0.913, which was slightly poorer than the R^2 value obtained from the Temkin and BET and Langmuir equations indicating that the Langmuir model better fitted the equilibrium obtained in this study. This suggested that the biosorption of AR88 onto dried *Lemna minor* may be due to biosorption of monolayer to the functional groups as binding sites on the surface of the biomass. The results obtained were similar and supported by other researchers (Diyanati al., 2013; Zazouli et al., 2013).

The correlation coefficient (R^2) in pseudo second-order model was better than the first order model, Elovich, Intraparticle diffusion, correlation coefficients. This kinetic study confirmed that biosorption of AR88 on dried *Lemna* minor was a multi step process, involving in biosorption on the external surface and diffusion into the interior with external surface chemical sorption being the rate-controlling step (Wang et al., 2013; Senthil Kumar et al., 2013).Conclusion: Based on the results, the dried Lemna Minor can be used as an effective and low cost adsorbent to treat effluent containing dye. The removal efficiency depends upon parameters such as initial dye concentration, adsorbent dose, pH, and contact time. The data were best fitted on Langmuir isotherm pseudo second- order kinetic.

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VII. References

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