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Research Article

Biosorption of Reactive blue 59 dyes using dried Azolla filiculoides biomass

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Abstract: The aim of this study is to evaluate adsorption kinetics, isotherms parameters of Reactive blue 59 (RB-59) onto Azolla filiculoides from aqueous solutions. The effects of pH, contact time, initial dye concentration and dose adsorbent were investigated in the experimentally. The percentage of color removal decreased with increase in initial dye concentration. Adsorption equilibrium of color removal was reached after 75 min of contact time. Equilibrium data were fitted to Langmuir, Freundlich, and Tempkin isotherms, and their constants were determined. Using the linear correlation coefficients showed that the Langmuir isotherm best fits the RB-59 adsorption data on biomass. The experimental data were fitted into the following kinetic models: pseudo-first-order, pseudo-second-order, and the intraparticle diffusion model. It was observed that the pseudo-second-order kinetic model described the adsorption process better than any other kinetic models. The results obtained indicate that Azolla filiculoides could be employed as a much more efficient adsorbent for dye removal from aqueous solution.
Keywords: Adsorption- Azolla Filiculoides - Reactive blue 59 - Isotherm- Kinetic

INTRODUCTION

Dyes are used extensively in the textile, leather, paper, plastic and other industries [1]. Reactive dye production is characterized by the great losses that are caused by the high solubility of the dyes, which also creates an economical and environmental problem [2]. Color removal from effluents polluted with dyes of textile industries has been considered a challenge due to the difficulty of treating such wastewaters by conventional methods [3]. Many physical and chemical treatment methods including coagulation, precipitation, filtration, electrodialysis, membrane separation and oxidation have been used for the treatment of dye containing effluents [4]. Due to low biodegradability of dyes, a conventional biological treatment process is not very effective in treating a dye wastewater [5]. It is usually treated by physical or chemical processes [6]. However these processes are costly and cannot effectively be used to treat the wide range of dye wastewaters [7]. Adsorption has been found to be superior to other techniques for treating wastewater: it is low-cost, highly efficient, simple, easy to perform and insensitive to toxic substances [8].

Adsorption on activated carbon has been found to be an effective process for dye removal, but it is too expensive [9, 10]. Consequently numerous low cost alternatives have been proposed including vermiculite; wood; peat; fly ash; sawdust; soil; china clay; waste coir pith; banana pith; and bagasse pith [11, 12]. New economical, easily available and highly effective adsorbents are still needed. Since the aquatic plant are capable to grow in contaminated water therefore they has been used in several study to remove the pollutants [13, 14]. Azolla filiculoides is one of this plants which it is commonly found in ditches, ponds and slow moving streams. Its high growth rate is considered as main feature of Azolla [15, 16]. Recently, the Azolla has been used in several studies to remove the pollutants [17, 18]. The present study was intended to remove Reactive blue 59 dyes from aqueous solutions using Azolla filiculoides as a new low cost adsorbent. The effect of various parameters like, adsorbent amount, dye concentration, contact time, pH, kinetics, equilibrium and thermodynamic studies was investigated.

MATERIAL AND METHODS:

Biosorbent and chemicals

A. filiculoides was collected from Anzali wetland, Iran. It was then sun dried and crushed to particle sizes in the range of 1–2 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 [19]. The morphological features and surface characteristics A. filiculoides before and after use were examined using an environmental scanning electron microscopy (ESEM) instrument (Philips XL30).

Reactive Blue 59 is an anionic dye with molecular weight 715 g/mol and Molecular Formula $C_{22}H_{20}Br\,N_6O_{12}S$ which was provided from Alvan Sabet Company of Iran and the other chemicals used in these experiments were the product of the Merck Company (Darmstadt, Germany). Double distilled water (DDW) was used throughout the study. Fig 1 shows the structure of the investigated dye. Stock solutions of dyes were prepared by dissolving the powder in double distilled water. Dye solutions of different initial concentrations were prepared by diluting the stock solution in appropriate proportions.

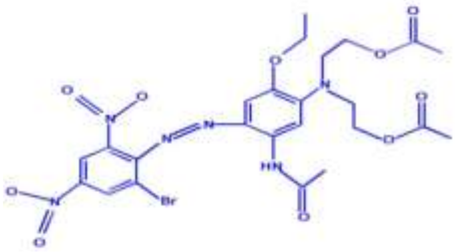

Fig-1: Molecular structure of RB-59 dye(20)

Batch adsorption studies

Various experimental conditions which may influence the biosorption of RB-59 on dried A. filiculoides, including initial solution pH, biosorbent dosage, initial RB-59 concentration and Contact time were tested using batch experiments. Initial RB-59 solutions with different concentrations were prepared by diluting a RB-59 stock standard solution of 1000 mg/L with distilled water. The solution pH was adjusted using either diluted 0. 1M HCL or 0.1M NaOH solution. In adsorption equilibrium, experiments were conducted in a set of 250 mL Erlenmeyer flasks, where solutions of dye (100 mL) with different initial concentrations (25–200 mg/L) were added in these flasks. Equal masses of 5 g of biomass were added to dye solutions and each sample was kept in an isothermal shaker of 180 rpm at 25±1 ◦C for 10 to 180 min to reach equilibrium of the solid-solution mixture. At the end of the equilibration time, the solution was centrifuged at 3600 rpm for 10 min. The supernatant was then analysed for RB-59 using a UV-Spectrophotometer (DR-4000) at λ_{max} 585 nm [20]. The content of RB-59 biosorbed on dried A. filiculoides was estimated by the difference between the initial and final RB-59 concentrations remained in the supernatant. All batch experiments were carried out in triplicate.

The amount of adsorption at equilibrium, q_e (mg/g), was calculated by [21].

$$q_e = \frac{(C0 - Ce)V}{W} \quad (1)$$

Where C_0 and C_e (mg/L) are the liquid-phase concentrations of dye at initial and equilibrium, respectively. V (L) is the volume of the solution and W (g) is the mass of dry sorbent used.

The dye removal percentage can be calculated as follows [22]:

$$R= \frac{C0 - Ce}{C0} \times 100$$

Batch kinetic studies

The procedures of kinetic experiments were basically identical to those of equilibrium tests. The aqueous samples were taken at preset time intervals, and the concentrations of dye were similarly measured. All the kinetic experiments were carried out at 30 ◦C. The amount of sorption at time t, qt (mg/g), was calculated by [23].

$$q_t = \frac{(C0 - Ct)V}{W} \quad (3)$$

Where C_t (mg/L) is the liquid-phase concentrations of dye at any time.

Isotherm models

Adsorption isotherm is basically important to describe how solutes interact with adsorbents, and is critical in optimizing the use of adsorbents. The Langmuir [24], the Freundlich [25] and the Temkin [26] were employed in the present study. The linearized forms of the three isotherms are

$$\frac{C_e}{q_e} = \frac{1}{q_m K} + \frac{C_e}{q_m} \quad (4)$$

$$\text{Log } q_e = \frac{1}{n} \log Ce + \log K_F \quad (5)$$

$$q_e = B\ln(A) + B\ln(C_e) \quad (6)$$

The Langmuir constants q_m (mg/g) and K (L/mg) are Langmuir constants related to adsorption capacity and energy of adsorption, respectively. The constants q_m and K can be calculated from the plot between $1/q_e$ versus $1/C_e$ (Eq. (4). C_e (mg/L) and q_e (mg/g) are the equilibrium concentration, and the amount of dye adsorbed at equilibrium, respectively. Similarly the Freundlich isotherm constants K_F and 1/n can be calculated from the plot of ln (q_e) versus ln (C_e) (Eq. 5). K_F and n are the Freundlich constants, which are indicators of adsorption capacity and adsorption intensity, respectively. The Temkin isotherm has generally been applied in the form given by Eq. (6). Therefore, by plotting q_e versus ln C_e (Eq. 6), enables the determination of the constants A and B. B is the Temkin constant related to heat of sorption (J/mol), A is the Temkin isotherm constant (L/g), R the gas constant (8.314 J/mol K), B is Temkin isotherm constant and T the absolute temperature (K).

Kinetics models

The most common models used to fit the kinetic sorption experiments are pseudo-first-order model [27] and pseudo-second-order model [28] were used:

$$\log(q_e - q_t) = \log q_e - k_1 t / 2.303 \quad (7)$$

$$t / q_t = 1/k_2 q_e^2 + 1/q_e\, t \quad (8)$$

Where q_e (mg/g) and q_t (mg/g) are the amount of adsorbate adsorbed at equilibrium and at time t, respectively. k_1 (min^{-1}) and k_2 (gmg^{-1} min) are the pseudo-first-order and pseudo-second order adsorption rate constants, respectively.

Intraparticle diffusion model

In order to investigate the mechanism of the RB-59 adsorption onto A. filiculoides, intraparticle diffusion-based mechanism was studied. The most commonly used technique for identifying the mechanism involved in the adsorption process is by fitting an intraparticle diffusion plot. It is an empirically found functional relationship, common to the most adsorption processes, where uptake varies almost proportionally with $t^{1/2}$ rather than with the contact time t. According to the theory proposed by Weber and Morris [29]:

$$q_t = k\, t^{1/2} + C \quad (9)$$

Where kid (mg g^{-1} $min^{1/2}$), the rate parameter of stage i, is obtained from the slope of the straight line of q_t versus $t^{1/2}$. C is the intercept.

RESULTS AND DISCUSSION:

The Dried A. filiculoides was also examined before and after use using environmental scanning electron microscopy. Fig. 2(a) clearly shows the pore textural structure of dried A. filiculoides before use. However, as shown in Fig. 2(b), clear pore textural structure is not observed on the surface of dried A. filiculoides after use which could be due to either agglomeration on the surface or the incursion of RB-59 into the pores of dried A. filiculoides.

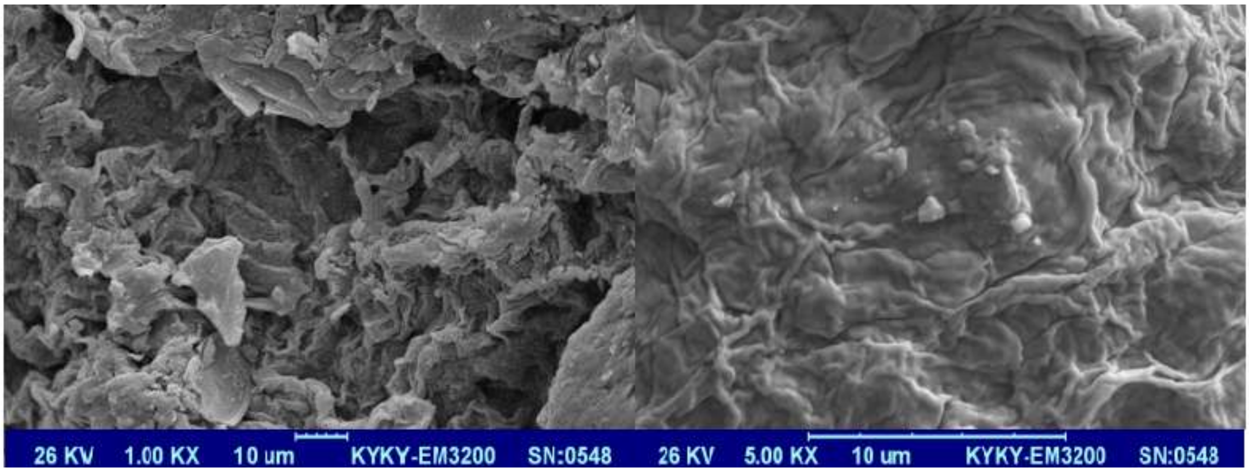

Fig-2: SEM image of Azolla Filiculoides before and after used

Effect of contact time:

The known of the equilibrium contact time are important for designing adsorption process [8]. Adsorption studies were carried out for 180 min in order to determine the effect of contact time on the percentage removal of RB-59 dye as shown in Fig. 3. Dye adsorption onto Azolla Filiculoides increased from 37.1% to 98.4% when the contact time was increased from 10 to 75 min, which was chosen as the experimental contact time for this study. The percentage removal of dye was rapid in the beginning, but it gradually decreased with time until it reached equilibrium. The rate of removal is higher in the beginning due to larger surface area available of adsorbent [30]. After adsorption, the rate of dye uptake is controlled by the rate of dye transported from the exterior to the interior sites of the adsorbent particles [31].

Effect of pH:

pH plays a major role in the adsorptive removal of dyes from aqueous solution and is an important factor controlling the surface charge of the adsorbent and the degree of ionization of the materials in the solution [3]. The results showed that the maximum biosorption of RB-59 was observed at low pH 3 (Fig.4). At lower pH, the biosorbent surface turned out to be positively charged and electrostatic attraction develops between positively charged biomass and negatively charged anionic dyes [32]. However, at basic pH, adsorption decreases due to the presence of hydroxyl ions which show competition with dye anions for binding sites[33]. Ardejani et al. [34] examined the effect of initial pH on adsorption of Direct Red 80 from aqueous solution onto almond shells. As pH increased from 3 to 12, the adsorption capacity decreased from 20.5 to 18.8 mg/g. The maximum uptake of dye was observed at pH 3.0. Hence, pH 3.0 is selected in the subsequent experiments.

Effect of dye initial concentration:

The concentration of the dye in solution can strongly affect the sorption kinetics. Fig. 5 showed the effect of initial dye concentration (C_0) (10–160 mg/L) on the adsorption capacity and removal efficiency by the A. filiculoides. The increase in initial dye concentration from 25 to 200 mg/L causes an increase in the amount of dye adsorbed (q_e) from 6.15 to 32.95 mg/g, but %removal decreased from 98.4 to 65.9. The decrease in percentage removal can be explained by the fact that all the adsorbents had a limited number of active sites, which would have become saturated above a certain concentration [35]. The increase in adsorption capacity with increase in RB-59 concentration may be due to the higher adsorption rate and utilization of all active sites available for the adsorption at higher concentration [36].

Effects of Adsorbent Dosage:

The effect of different adsorbent dosage 1-8 g per 1000 ml solution under identical operating conditions for the removal of the dye is shown in Fig. 6. It was observed that at higher adsorbent dose, removal increases sharply within 75 minutes of adsorption. 90.4% removal is observed within initial 75 min of contact with 4 g of biomass. This is due to the fact that, increase in adsorbent dosage increase area available for adsorption [37]. At lower adsorbent doses, increase in removal is proportional to the increase in adsorbent dose. At a higher dose increase in removal less than proportional dose due to continuous depletion of reactive dye molecules in aqueous phase [38].

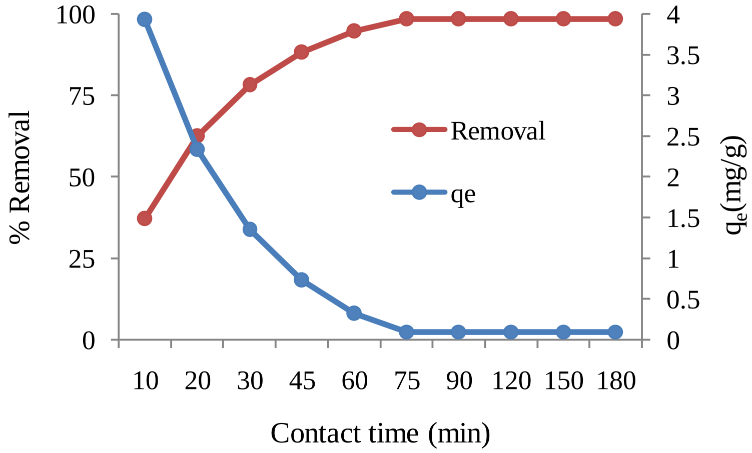

Fig-3: Effect of contact time for RB-59 adsorption (Con = 25 mg/L, pH = 3, Biomass dose: 4 g/L)

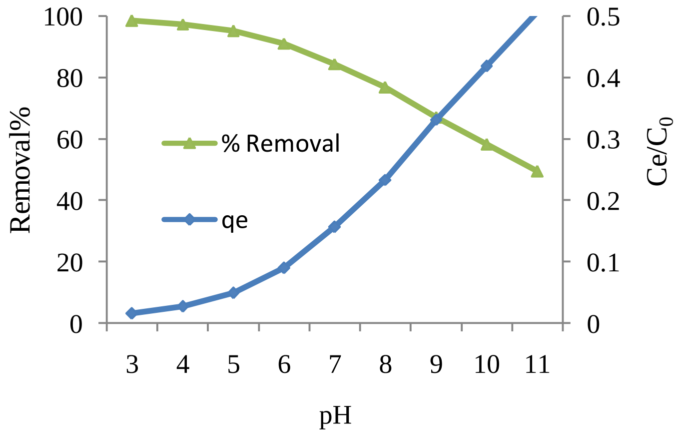

Fig-4: Effect of pH on adsorption of RB-59 Dye (Con = 25 mg/L, Contact time = 75 min, Biomass dose: 4 g/L)

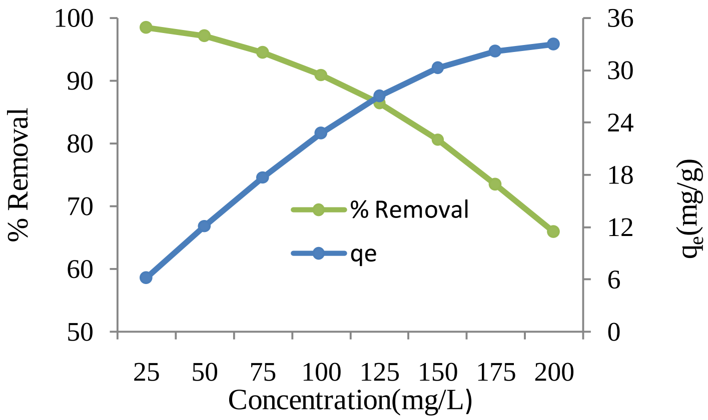

Fig-5: Effect of solute concentration (Contact time = 75 min, Biomass dose:4 gr/L, pH = 3)

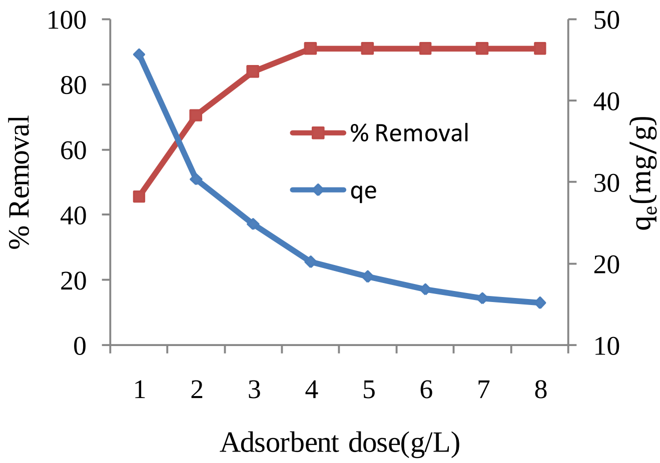

Fig-6: Effect of adsorbent dose (Contact time = 75 min, pH = 3, Con: 100 mg/L)

Isotherm analysis:

Equilibrium data, commonly known as adsorption isotherms, are basic requirements for the design of adsorption systems. In this work, the equilibrium data for RB-59 on A. filiculoides were modeled with the Langmuir, Freundlich and Temkin models.

The Langmuir constants q_m and K were determined from the slope and intercept of the plot and are presented in Table 1. The value of the coefficient of correlation (R^2 = 0.996 and 0.999) obtained from Langmuir expression indicates that Langmuir expression provided a better fit to the experimental data of RB-59 on A. filiculoides.

The essential characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor R_L that is given by the following equation [36]:

$$R_L = \frac{1}{K_L C_0} \quad (10)$$

Where C_0 (mg/L) is the highest initial concentration of adsorbate, and K_L (L/mg) is Langmuir constant. The value of R_L indicates the shape of the isotherm to be either unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$), or irreversible ($R_L = 0$). The R_L value for the adsorption of RB-59 on A. filiculoides was 0.224 and 0.416 to dose adsorbent of 2.5 and 5 g/L , indicating that the adsorption was a favorable process.

The calculated Freundlich isotherm constants and the corresponding coefficient of correlation are shown in Table 1. The coefficient of correlation was high (R^2 = 0.912 and 0.947) showing a good linearity. The result shows that the value of n is greater than unity indicating that the dye is favorably adsorbed on A. filiculoides. This is in great agreement with the findings regarding to R_L value. The magnitude of Freundlich constant indicates easy uptake of RB-59 from aqueous solution.

The adsorption data for RB-59 on A. filiculoides were analyzed by a regression analysis to fit the Temkin isotherm model. The parameters of Temkin model as well as the correlation coefficient are listed in

Table 1. The coefficient of correlation was high (R^2 = 0.876 and 0.893) showing a good linearity. From Table 1, it can be conclude that the Langmuir isotherm model was more suitable for the experimental data than other isotherms because of the high value of correlation coefficient. This indicates that the adsorption of RB-59 on A. filiculoides takes place as monolayer adsorption on a surface that is homogenous in adsorption affinity. A similar result was reported for the adsorption of Reactive Red 198 and Acid Blue 15 on Azolla filiculoides [14, 21] .

Adsorption kinetics:

The kinetics of RB-59 adsorption on A. filiculoides was studied with respect to different initial concentrations. For evaluating the adsorption kinetics of RB-59, the pseudo-first-order and pseudo-second-order kinetic models were used to fit the experimental data.The pseudo-first-order model data do not fall on straight lines for most initial concentrations indicating that this model is less appropriate. The pseudo-first-order rate constant (k_1) and $q_{e,cal}$ determined from the model are presented in Table 2 along with the corresponding correlation coefficients. It was found that the correlation coefficients for the pseudo-first order model are low and a wide range of variations are recorded for the q_e, obtained from the experimental and calculated. This indicates that the adsorption of RB-59 on A. filiculoides does not follow pseudo first- order kinetics. Therefore, the experimental kinetic data were further analyzed using the pseudo-second-order model. The $q_{e,exp}$ and the $q_{e,cal}$ values along with correlation coefficients for the pseudo-second-order models are shown in Table 2. The values of correlation coefficient were very high (R^2 >0.998) and the theoretical $q_{e,cal}$ values were closer to the experimental $q_{e,exp}$ values at different initial RB-59 concentrations (Table 2). From Table 3, it can be concluded that the pseudo-second-order kinetic model provided a good correlation for the adsorption of RB-59 on A. filiculoides at different initial RB-59 concentrations compared to the pseudo-first-order model. A number of authors have reported pseudo-second-order kinetics for adsorption of dyes on Lemna minor [36], A. filiculoides [21] and Canola biomass [39].

The intraparticle diffusion models constant are presented in Table 2 along with the corresponding correlation coefficients. According to results, values of coefficients are also low. From these results one can conclude that the biosorption process of RB-59 onto the A. filiculoides 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-1:The adsorption isotherms constants for the removal RB-59

	Langmuir model				Freundlich model			Temkin model		
Dose (g/L)	q_m	R_L	K_L	R^2	n	K_F	R^2	B	A	R^2
2.5	21.18	0.224	0.0026	0.996	3.17	1.761	0.912	28.17	2.41	0.876
5	18.45	0.416	0.0041	0.999	2.46	2.893	0.947	19.76	4.34	0.893

Table-2: kinetic parameters for RB-59 adsorption onto A. filiculoides

Pseudo second-order model					Pseudo first-order model			Intraparticle diffusion		
C_o	$q_{e,cal}$	k_2	R^2	$q_{e,exp}$	K_1	R^2	$q_{e,exp}$	K	C	R^2
25	6.15	0.022	0.998	6.72	0.091	0.841	3.11	2.07	1.14	0.812
50	12.13	0.048	0.999	11.49	0.352	0.883	8.94	2.07	1.83	0.796
100	22.72	0.069	0.998	22.06	0.654	0.892	15.61	4.18	3.09	0.874
200	32.95	0.095	0.998	32.24	0.812	0.901	24.17	9.71	4.49	0.832

CONCLUSIONS

This study investigated the adsorption of RB-59 dye from aqueous solutions onto dried A. filiculoides. An increase in the pH of solutions leads to an decrease in the sorption capacities of dye on the sorbent under study. The sorbed amounts of dye increase with increase in contact time, reaching a maximum value after 75 min. The Langmuir adsorption isotherm was found to have the best fit to the experimental data with maximum adsorption capacity of 21.18 mg/g. The pseudo-second-order kinetic model provided the best correlation of the experimental data. This study demonstrated that the A. filiculoides biomass could be used as an effective biosorbent for the treatment of wastewater containing dyes. A. filiculoides is natural abundant environmental biomass and it may be alternative to more costly materials.

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