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Original article

Adsorptive removal of acid blue 15 dye (AB15) from aqueous solutions by red mud: characteristics, isotherm and kinetic studies

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A B S T R A C T

The dyes are one of the main environmental pollutants in various industrial wastewaters. In the present study, the adsorption potential of Red mud in a batch system for the removal of Acid blue 15 dye from aqueous solutions was investigated. Batch kinetics and isotherm studies were carried out to evaluate the effect of contact time (10-150 min), initial pH of solution (3-11), initial phenol concentration (25-200 mg L-1) and adsorbent dose (0.5-8 g L-1) on sorption efficiency. Adsorption capacities (qe) increased with increasing of initial dye concentration and decreased with increasing adsorbent dose and pH. Maximum adsorption capacity of the red mud was 29.44 mg g-1 when 73.6% of the AB15 dye was removed. The adsorption equilibriums were analyzed by Langmuir, Freundlich, Temkin and BET isotherm models. It was found that he data fitted to Langmuir (R2=0.997) better than isotherm other models. Batch kinetic experiments showed that the adsorption followed pseudo-second-order kinetic model with correlation coefficients greater than 0.995. According to achieved results, it was defined that Red mud not only was an inexpensive absorbent, but also a quite effective factor in removal of Dyes from water and wastewater.

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1. Introduction

Dyes are colour organic compounds which can colorize other substances (Safa and Bhatti, 2011). These substances are usually present in the effluent water of many industries, such as textiles, leather, paper, printing, and cosmetics (Tan et al., 2010; Garg et al., 2003). The complex aromatic structures of dyes make them more stable and more difficult to remove from the effluents discharged into water bodies (Gong et al., 2005; Robinson et al., 2002). Approximately 10-15% of the overall production of dyes is released into the environment, mainly via wastewater (Lin et ., 2013; Crini and Badot, 2008). This is very dangerous because some of the dyes and their degradation products have a toxic, mutagenic or carcinogenic influence on living organisms (Oliveira et al., 2008; Namasivayam and Kavitha, 2002). Therefore, the dye containing wastewater should be treated before discharge (Padmesh et al., 2006). Due to low biodegradability of dyes, a conventional biological treatment process is not very effective in treating a dye wastewater (Zazouli et al., 2013; Ponnusami et al., 2007). Recently, physicochemical methods have been used for removal of dyes in wastewaters though, these methods are economically unviable, environmentally unfriendly and results in generation of residual sludge (Zazouli et al., 2013; Suna et al., 2010). Consequently, amongst numerous techniques, adsorption techniques seem to have the most potential for future use in industrial wastewater treatment because of their proven efficiency in the removal of organic and mineral pollutants and for economic considerations (Padmesh et al., 2005; Dogan et al., 2008). Activated carbon is the most widely used adsorbent for this purpose because it has a high capacity for adsorption of color but its use is limited because of high cost (Ferdag and Necip, 2009; Cestar et al., 2006). The removal of dyes from effluent using adsorption process provide an alternative treatment, especially if the adsorbent is inexpensive and readily available (Mall et al., 2006; Hilal et al., 2012). An adsorbent can be considered as cheap or low-cost if it is abundant in nature, requires little processing and is a byproduct of waste material from industrial waste (Diyanati et al., 2013; Zazouli et al., 2013). Many low-cost adsorbents have been investigated on dye removal, such as orange peel (Sivaraj et al., 2001), bottom ash (Wang and Zhu, 2005), Bentonite (Toor and Jin, 2011), dolomitic (Walker et al., 2003), chitosan (Dotto and Pinto, 2011), silica fume (Nadaroglu et al., 2013), clay (Ozdemir et al., 2044), zeolite (Tahir and Rauf, 2006), calcine alunite (Özacar and Şengil, 2003), Lemna minor (Balarak et al., 2015), Azolla filiculoides (Zazouli et al., 2013), canola (Balarak et al., 2014) and rice husk (Diyanati and Balarak, 2013). As an alternative low-cost absorbent material, solid wastes are generally used as adsorbent for the remediation of wastewater. One type of solid waste materials, Red mud, is largely produced from the alumina industry (Nadaroglu et al., 2014; Nadaroglu et al., 2010). The Red mud emerges as a by-product of the caustic leaching of bauxite to produce alumina (Nadaroglu and Kalkan, 2012). This material is principally composed of fine particles of silica, aluminum, iron, calcium and titanium oxides and hydroxides, which are responsible for its high surface reactivity (Gupta et al., 2004). In the present study describes the use of Nitric acid modified Red mud for removal of AB15 dye from aqueous solutions. The adsorption of AB15 dye has been investigated as a function of contact time, pH, dye concentration and adsorbent dose. Adsorption isotherm and kinetic studies have been performed to describe the adsorption process.

2. Materials and methods

2.1. The characteristics of dye

The used dye was the analytical grade which were purchased from alvan sabet CO. it's molecular weight is 775.96 mg/mol. it's chemical fomula is C42H46N3NaO6S2 (Zazouli et al., 2014). The chemical structure of AB15 is shown in Figure1. The stock solution (1000mg/L) was prepared and desired concentration of dye solution was prepared by dilution of stock solution.

2.2. Adsorbent characterization

The adsorbent is a waste By-product of alumina production process. It contains the oxygen, iron and calcium and etc with various contents. Elemental analysis was performed by electron microscope (SEM) using Oxford instrument (Stereo Scan S360).

2.3. Preparation procedure of adsorbent

The first, the red mud was provided from Tabriz-Iran Aluminum Co. Red mud was washed thoroughly with distilled water, dried at 110 o C for 24 hours. The each 10 gr of Red mud was activated by using of 20 ml Nitric acid for 24 h. Then, it was rinsed 3 times by the distilled water. It was dried in 103oC for 6 h. After that the red mud was grinded and sieving by using a 100 mesh sieve (Zazouli et al., 2013).

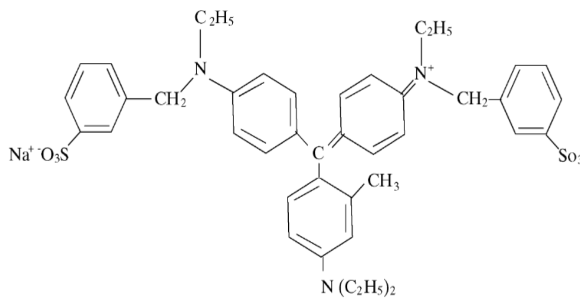

Fig. 1. The chemical structures of Acid blue 15 (Zazouli et al., 2014).

2.4. Batch adsorption experiments

The batch adsorption system was employed in present study and the most effective factors for adsorption process including contact time (10-150 min), pH (3-11), adsorbent dose (0.5-8 g L-1) and initial concentration of dye (25- 200 mg/L) were assessed.

The optimum pH was determined by varying the pH in range of 3-11 and keeping constant of other variables (contact time, adsorbent dose and initial dye concentration). The experiments were preformed in 200 milliliter beaker with a constant concentration of dye. Then this mixture was shaked with a shaker device of enforce model with 180 rpm and the room temperature of 20 – 25°C. HCl and NaOH were used to adjust the pH solution. In the next step, the optimum adsorbent dose was estimated by keeping constant of contact time and initial day concentration and also the obtained optimum pH. After determination of the optimum pH and adsorbent dose, the various concentrations of the dye in the specified times of contact was investigated. The final dye concentration in solution was measured by the UV-Visible spectrophotometer at a wavelength of 565 nm and with regarding to standard curve (Padmesh et al., 2006). The equilibrium experiments of adsorption process was occurred after determination of equilibrium time in order to evaluate the effect of adsorbent mass on dye removal to obtain the adsorption isotherms.

3. Results and discussion

3.1. Red mud characterization

SEM has been a primary tool for characterizing the surface morphology and fundamental physical properties of the adsorbent surface. It is useful for determining the particle shape, porosity and appropriate size distribution of the adsorbent (Zhang et al., 2010). SEM of Red mud after and before dye adsorbed are shown in Fig. 2.

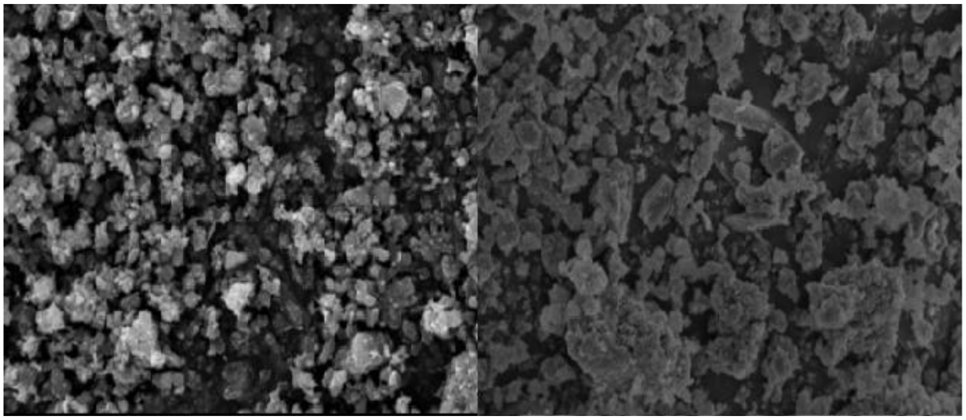

Fig. 2. SEM image of Red mud before and after dye adsorbed.

3.2. Effect of contact time

The contact time between adsorbate and adsorbent is one of the most important design parameters that affect the performance of adsorption processes. Figure 3 shows the effect of contact time on the adsorption capacity and percent removal efficiency of AB15 dye onto the Red mud at a constant initial dye concentration (25 mg/L) and 5 g/L adsorbent dosage. The uptake of AB15 on Red mud was rapidly in the first 30 min (%89.7) and then the adsorption rate decreased gradually from 30 to 45 min and finally reached equilibrium in about 60 min (%98.8).Then gradual decrease in capacity after 30 minutes can be the due to the reduction of the adsorption sites on the absorbent surface and these sites' being satiated (Tor and Cengeloglu, 2006). On the other hand a large number of surface sites are available for adsorption at the initial steps and after a lapse of time the remaining surface sites are difficult to be occupied because of repulsion between the solute molecules of the solid and bulk phases (Namasivayam and Yamuna, 2001).

3.3. Effect of pH

One of the most important factors affecting the capacity of adsorbents in wastewater treatment is pH. The pH of the solution affects the surface charge of the adsorbents as well as the degree of ionization of different pollutants. The pH value of the solution was an important controlling parameter in the adsorption process, as can be seen from Fig. 4. It shows that the adsorption capacity of AB15 onto Red mud increases significantly with decreasing pH. The maximum removals of AB15 for contact time 60 min were carried out at pH 3. The hydrogen ion and hydroxyl ions are adsorbed quite strongly and therefore the adsorption of other ions is affected by the pH of the solution (Cardoso et al., 2011). Change of pH affects the adsorptive process through dissociation of functional groups on the adsorbent surface active sites (Kumar et al., 2010). At strongly acidic pH, a significantly high electrostatic attraction exists between the positively charged surface of the adsorbent and acid (anionic) dyes. As the pH of the system increases, the number of negatively charged sites increases and the number of positively charged sites decreases (Balarak et al., 2015). A negatively charged surface site on the adsorbent does not favor the adsorption of dye anions, due to the electrostatic repulsion (Zazouli et al., 2014). Also, lower adsorption of acid dyes at alkaline pH is due to the presence of excess hydroxyl ions competing with the dye anions for the adsorption sites (Padmesh et al., 2005).

3.4. Effect of initial dye concentration

To determine the effect of initial dye concentration on the adsorption process the initial concentration of AB15 was varied from 25 to 200 mg/L at the fixed PH=3, adsorbent dose 5 g/L and contact time 60 min. As presented in figure 5, dye removal efficiency decreased with increasing of dye concentration, so maximum efficiency was achieved at initial dye concentration 25 mg/L (%98.9). This may be attributed to surplus dye molecules being present in the solution, with respect to the available active adsorption sites owing to fixed surface area of red mud (Gulnaz et al., 2004). At the low concentration, there will be unoccupied active sites on the adsorbent surface and when the dye concentration increases, the active sites required for adsorption of the dye molecules will lack (Kanawade and Gaikwad, 2011). On the other hand, the increase in dye concentration will cause an increase in the loading capacity of the adsorbent and this may be due to the high driving force for mass transfer at a high initial dye concentration (Zazouli et al., 2014).

3.5. Effect of adsorbent dose

The effect of adsorbent dose on removal of AB15 was studied by varying the dose of adsorbent from 0.5 to 8 g/L. From fig 6, it is evident that adsorbent dose significantly influences the amount of adsorbed. In fact the percentage of AB15 removal steeply increases with the adsorbent loading up to 5 g/L, but dye adsorbed decreased as the dose of adsorbent increases from 0.5 to 8 g/L. This result can be explained by the fact that the biosorption sites remain unsaturated during the biosorption reaction whereas the number of available sites for biosorption increases by increasing the biosorbent dose (Ratnamala et al., 2012). Similarly, Zazouli et al (2013) investigated the effect of biosorbent dose on the removal of reactive red 198 by Red mud from aqueous solution. The results showed that the removal efficiency increased from 38% to 96%, while the qe decreased from 5.2 to 1.6 mg/g with an increase in the Biosorbent dose from 2 to 10 g/L.

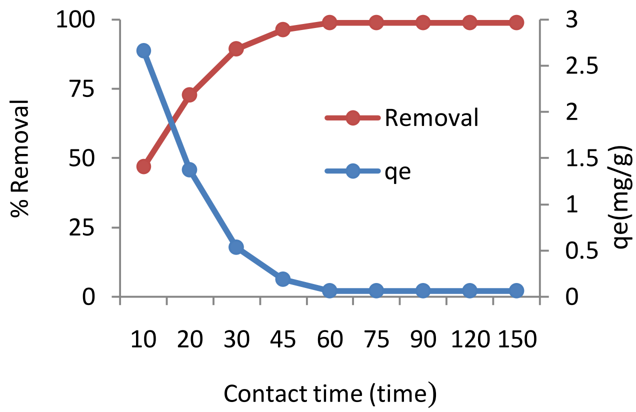

Fig. 3. Effect of adsorbent dosage on removal of AB15 by Red mud.

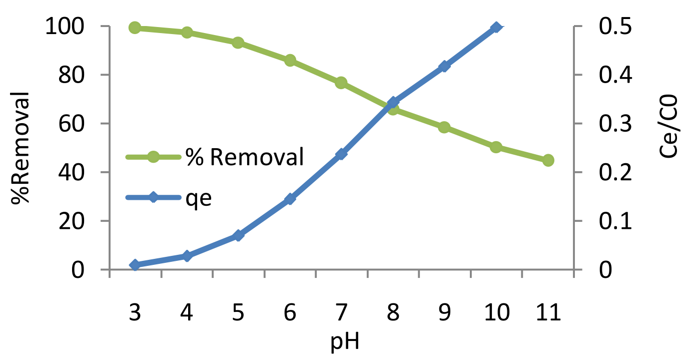

Fig. 4. Effect of pH on AB15 removal efficiency (C_0 = 25 mg/L, adsorbent dose of 5 g/L, contact time = 60 min).

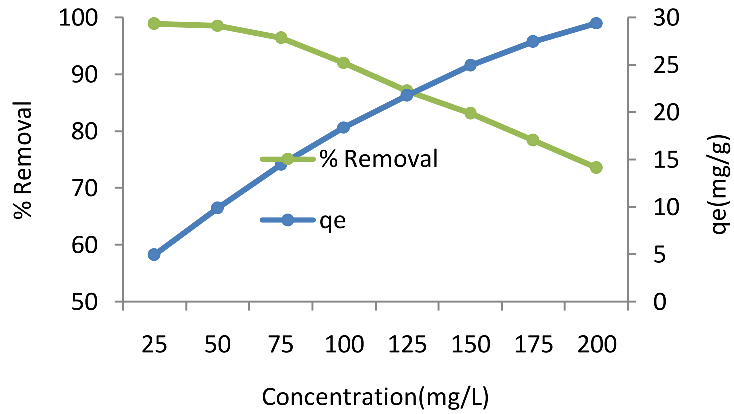

Fig. 5. Effect of initial AB15 concentration on removal efficiency.

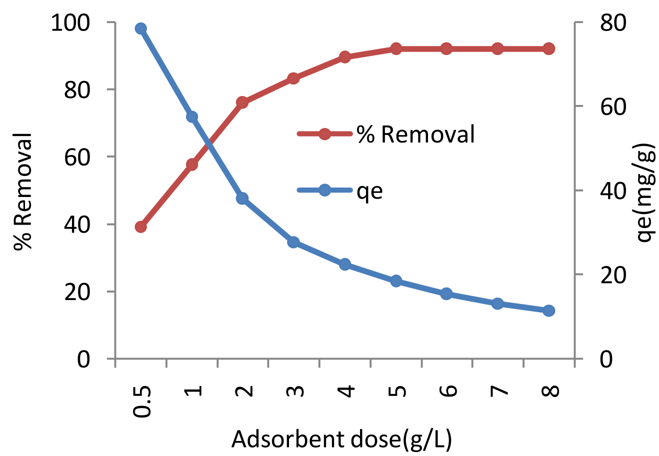

Fig. 6. Effect of biomass dose on AB15 biosorption (C_0 = 100 mg/L, pH=3 and Contact time=60 min).

3.6. Adsorption kinetic study

Kinetic models have been used to investigate the mechanism of sorption and potential rate controlling steps, which is helpful for selecting optimum operating conditions for the full-scale batch process. Pseudo-first-order, pseudo-second-order and intraparticle diffusion kinetic models were used. The pseudo-first-order rate expression based on solid capacity is generally expressed follows (Chakravarty et al., 2008): $\log (q_e - q_t) = \log q_e - \frac{K_1}{2.303} t$ Where q_e and q_t are the amounts of dye (mg g^{-1}) adsorbed at equilibrium and time t, respectively, and k_1 is the rate constant of adsorption (min^{-1}) biosorption. The pseudo-second-order kinetic model is expressed as (Ofomaja and Ho, 2007): $\frac{t}{q_t} = \frac{1}{k_2 qe^2} + \frac{1}{qe\ t}$ Where q_e is the biosorbed dye amount at equilibrium (mg g^{-1}) for the pseudo-second-order biosorption, q_t is the amount of dye biosorbed at time t (mg g^{-1}) and k_2 is the pseudo-second-order kinetic rate constant (g mg^{-1} min^{-1}). The intraparticle diffusion equation can be written as follows (Diyanati et al., 2014): $q_t = k_{dif}\, t^{0.5} + c$ where C is the intercept, and kdif is the intraparticle diffusion rate constant (mg g^{-1} min^{-1}). The results of the kinetic parameters for biosorption are given in Table 1. The correlation coefficients of all examined data were found very high (R^2> 0.99) for pseudo-second-order kinetic. This shows that the model can be applied for the entire adsorption process and confirms that the sorption of AB15 dye onto Red mud follows the pseudo-second-order kinetic model.This kinetic study confirmed that biosorption of AB15 onto Red mud 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.

Table 1
Kinetic parameters for AB15 dye adsorption onto Red mud.

Con (mg/L)	q_e exp (mg/g)	Pseudo-first order			Pseudo-second order			Intraparticle diffusion		
		K_1	q_e	R^2	K_2	q_e	R^2	k_{dif}	C	R^2
25	4.94	0.067	2.21	0.912	0.0094	4.57	0.998	2.60	8.15	0.901
50	9.86	0.094	6.45	0.894	0.0073	9.65	0.999	2.41	6.24	0.894
100	18.42	0.121	13.24	0.872	0.0049	18.06	0.999	1.77	4.37	0.876
200	29.44	0.156	22.92	0.881	0.0016	29.94	0.998	1.25	2.65	0.912

3.7. Adsorption equilibrium study

An adsorption isotherm describes the relationship between the amount of adsorbate taken up by the adsorbent and the adsorbate concentration remaining in the solution after the system has attained equilibrium. The biosorption of sorbate (AB15) onto the adsorbent (Red mud) was modeled using the Langmuir, Freundlich, Tekmin, D-Radushevich equations. The Langmuir equation is given as follows (Gok et al., 2010):

$$\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{q_{max}\ K_L} \times \frac{1}{C_e}$$

Where q_e (mg/g) and C_e (mg/L) are the amount of adsorbed dye per unit mass of sorbent and unadsorbed dye concentration in solution at equilibrium, respectively. Qmax is the maximum amount of the adsorbed dye per unit mass of sorbent to form a complete monolayer on the surface bound at high C_e (mg/g), and K_L (L/mg) is a constant related to the affinity of the binding sites. The essential features of Langmuir can be expressed in terms of dimensionless constant separation factor R_L which is calculated using the following equation equation (Khaled et al., 2009):

$$R_L = \frac{1}{1+K_L C_0}$$

Values of R_L indicate the shapes of isotherms to be either unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$). The Freundlich isotherm model equation is expressed as (Low et al., 2000):

$$\text{Ln } q_e = \ln K_F + \frac{1}{n} \ln Ce$$

Where qe is the equilibrium dye concentration on the adsorbent (mg g^{-1}); Ce, the equilibrium dye concentration in solution (mg L^{-1}); and K_F is the Freundlich constant. The Tempkin isotherm model equation is expressed as (Arami et al., 2005):

$$q_e = B\ln A + B \ln Ce$$

Where B = RT/b, T is the absolute temperature in K, R the universal gas constant (8.314 JK^{-1} mol^{-1}), A the equilibrium binding constant and the constant B is related to the heat of adsorption. Doubinin-Radushevich model is a more generalized model as compared to the Langmuir isotherm. The linear form of (D–R) isotherm model can be seen below (Dincer et al., 2007):

$$\text{Ln } q_e = \ln q_m - K\varepsilon^2$$

Where K is a constant corresponding to the biosorption energy, q_m the theoretical saturation capacity and ε is the Polanyi potential which is calculated from equation below (Mane and Mall, 2007):

$$\varepsilon = RT \ln (1 + 1/C_e)$$

Where R (kJ mol^{-1} K^{-1}) is the gas constant and T (K) is the absolute temperature. E was calculated from the K value by the following relation (Diyanati et al., 2014):

$$E = 1/ (2K)^{1/2}$$

The results of the isotherm constants are displayed in Table 2. As shown in Table 2 that the correlation coefficients for the Langmuir isotherm model were close to 1.0 for all adsorbent dose. The correlation coefficients for Tempkin and D-Radushevich isotherms were low and it can be said that the experimental data was not fitted better to the Temkin and D-Radushevich isotherms model. 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, D-Radushevich equations are based on the hypothesis of multi-layer biosorption. As shown in table 2, of the Langmuir isotherm was greater than that of the other isotherms for the adsorption of dye investigated. This suggested that the biosorption of AB15 onto Red mud may be due to biosorption of monolayer to the functional groups as binding sites on the surface of the biomass. The similar results were reported for dye biosorption by Red mud (Zazouli et al., 2013).

Table 2
Isotherm constants for AB15 dye adsorption onto red mud.

Isotherm models	2.5 g/L	5 g/L	Isotherm models	2.5 g/L	5 g/L
Langmuir			**D-Radushevich**		
q_m(mg/g)	3.11	4.84	q_m(mg/g)	6.19	9.64
K_L (L/mg)	0.41	0.58	$K.10^{-4}$(mol^2 KJ^{-2})	1.94	3.06
R_L	0.041	0.059	E (kJ mol^{-1})	0.627	0.945
R^2	0.995	0.997	R^2	0.831	0.867
Freundlich			**Tempkin**		
K_F (mg/g)	1.74	1.02	A (L/g)	0.68	1.04
n	2.54	3.11	B	14.71	26.19
R^2	0.921	0.944	R^2	0.741	0.789

4. Conclusion

In this study, modified Red mud using as adsorbent for adsorption of AB15 dye from textile wastewater was investigated. The adsorption was highly dependent on various operating parameters, like: pH, biosorbent dose, time and initial dye concentration. The maximum percentage adsorption of 98.9% was obtained at pH 3, initial concentration 25 mg/L and with red mud dosage of 5 g/l and contact time 60 min. For equilibrium studies, four isotherm models were used in this study and it is found that Langmuir fitted experimental data very well. The kinetic data indicated that the adsorption process was controlled by pseudo-second-order equation. Based on the results, Red mud can be used as a relatively efficient and low cost absorbent for the removal of AB15 dye from textile wastewater.

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