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Removal of acid dyes from aqueous solution using potato peel waste biomass: a kinetic and equilibrium study

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ABSTRACT

The goal of this study was to survey the feasibility of the biosorption of two acid dyes (Acid Blue 113 and Acid Black 1) from aqueous solution using biomass prepared from potato peel waste. Adsorption isotherms were constructed and the kinetics of dye adsorption were studied. Langmuir and Freundlich isotherms, pseudo-first-order, and pseudo-second-order kinetic models were studied. The maximum biosorption was observed at a pH of 2 and 3 for Acid Blue 113 and Acid Black 1, respectively. The biosorption of two dyes increased with increasing contact time and reached equilibrium after two hours, approximately. Acid dye removal efficiency decreased with an increase in the initial dye concentration. The sum of squares due to error and the coefficient of determination (R2) analysis showed that the pseudo-second-order kinetic and the Langmuir isotherm model are better fit for the adsorption of Acid Blue 113 and Acid Black 1 on used potato peel waste.

Keywords: Biomass; Acid dye; Potato; Isotherm; Kinetics; Biosorption

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1. Introduction

The increasing trend in industrialization and urbanization has led to the generation of higher amounts of aqueous effluents. Most of these effluents have high levels of toxic pollutants [1,2]. Every year, more than 7×10^5 metric tons of synthetic dyes are produced throughout the globe and used for printing and dyeing, 5–10% of which is discharged with wastewater [2]. Many industries are now using synthetic dyes for reasons such as being easy to use, having higher chemical stability compared with natural dyes, and the low price of their synthesis [1,3,4]. Therefore, considering the ever growing use of synthetic dyes in different industries, it is necessary to remove dyes from effluents. Since the toxic nature of

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dyes has negative impacts on aquatic life, the discharge of dye-containing wastewater into rivers and streams by industries such as textile, paper, carpet, leather, distillery, and printing can damage the esthetic nature of the environment. Many dyes used in such industries are resistant to aerobic digestion and are stable to light and oxidation.

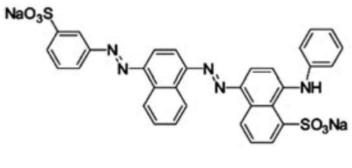
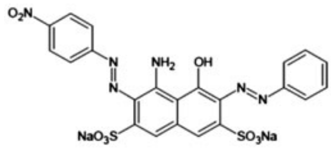
Acidic dyes are one of the best classes of soluble dyes used by textile industries [5]. It is necessary to treat dye-bearing effluents to remove the dye in an economical fashion to their normal concentration levels prior to discharge into the water bodies. The wastewater discharged from the textile industry is classified as the most polluting out of all the other industries with respect to discharge volume and effluent combustion. Therefore, a suitable and cost-effective dye removal technology should be used in this industry. Moreover, in the treatment of dyes, when the adsorbates have a low concentration, methods such as chemical precipitation or coagulation are less effective and more expensive [1,6–8].

As effective commercial adsorbents, activated carbons and ion exchange resins have been traditionally used to treat industrial wastewaters containing adsorptive pollutants; however, they have less practical use in different industries because of their high price and low efficiency [5,9,10]. Recently, environmental or bioresource technology has introduced a new method for removing toxic pollutants or recovering valuable resources from aqueous wastewaters with the use of biosorbents [2,3,9]. Compared with conventional techniques, the use of biosorbents has several advantages such as being more cost-effective and efficient, using less chemical or biological sludge, having regeneration ability, and the possibility of metal recovery following adsorption [2]. Biosorption is essential in developing countries because of its low cost and accessibility [2,11]. Availability and cheapness are two factors that need to be taken into account when selecting biomass for large-scale broad industrial uses [2,11–13]. Previous reports have mentioned that potato peel waste is able to remove reactive dyes and some heavy metal ions from aqueous solutions [2]. The use of cheap and environmentally friendly adsorbents has been studied as an ideal alternative to the existing expensive methods of removing dyes from solutions. Arami and colleagues used orange peels as a low-cost adsorbent for the removal of Direct Red 23 (DR23) and Direct Red 80 (DR80) from aqueous solutions [14]. Some published papers also reported some cheap adsorbents for dye removal such as used tea leaves [15]; methylene blue adsorption by used coffee grinds [16]; removal of crystal violet dye using agricultural waste cocoa (theobroma cacao) shell [17]; coffee wastes for removing synthetic dyeing effluents [18]; and removal of Direct Red 28, Direct Yellow 12, Direct Orange 26, and Direct Blue 1 with various bio/natural adsorbents (sugarcane bagasse pith (SB), saw dust (SD)—the plant origin products, and brick powder (BP)) [19].

After China and India, Iran is the third largest producer of potatoes in Asia and the twelfth largest worldwide. Since 1961, the production rates have had a 15-fold increase with a record breaking harvest of 5.24 million tons in 2007 and potatoes are now one of Iran’s leading agricultural exports. Potatoes are grown mostly under irrigation around the southern shore of the Caspian Sea, in the Zagros Mountains, and in the southern lowlands, alternated with wheat, vegetables, sugar beets, and lying fallow in three- or four-year rotations [2].

We aimed to study the potential of potato peel waste in removing Acid Black 1 (AB1) and Acid Blue 113 (AB113) from aqueous solution as azo dyes (Table 1). In detail, we also aimed to determine the isotherm and adsorption kinetic, the effect of initial dye concentration, adsorbent dosage, contact time, and pH in the function of eliminating this pollutant.

Table 1
Characteristics of acid dyes (AB1 and AB113) used in this study

Color index name	C.I. Acid Blue 113 (AB113)	C.I. Acid Blue 1 (AB1)
Chemical structure		
Chemical class	Anionic, Azo	
Molecular formula	$C_{32}H_{21}N_5Na_2O_6S_2$	$C_{22}H_{14}N_6Na_2O_9S_2$
λ_{max} (nm)	566	**622**
M_w (g/mol)	681.65	616.5

2. Material and methods

2.1. Biosobent preparation

This experimental study was performed in two separate steps; adsorbent preparation and adsorption. The adsorbent used in this study was prepared according to procedures described in a previous study [2] in seven steps: (1) potato samples were washed completely; (2) potatoes were hulled with a huller to obtain peels without any potato tissue; (3) the peels were separated into fine parts (to accelerate drying) and dried at room temperature for 5–7 days; (4) biomass (1 mg) was emulsified into 5 mL of acidic solution using 0.01 M HCl at 25 ± 1°C for 30 min to remove impurities or soluble molecules that may interact with dye molecules; (5) samples were washed with distilled water until the pH rose above 6; (6) the processed biomass was dried in an oven at 90°C for 24 h; and (7) the final product was sieved with a 40–45 range mesh.

2.2. Experimental procedure

The adsorption test was performed in a batch system and effective factors on the process including the contact time (5, 10, 15, 30, 45, 60, and 120), pH of the dye solution (3, 5, 7, 9, and 11); adsorbent dosage (0.2, 0.4, 0.6, 0.8, and 1 g), and dye concentration (50–400 mg L^{-1}) were studied in separate stages. In this study, agitation rate (200 rpm), temperature (1 ± 25˚C), and adsorbate particle size were fixed in all stages. Solutions were prepared at a volume of 250 mL. Adsorbent particles were separated after the dye-removing process using centrifugation. The main part of the tests consisted of making other variables constant, changing each variable in each stage, and studying the effect of changes on the dye removal efficiency.

The equilibrium technique was used to determine the pH_{ZPC} in the batch system [1,2,7,8]. This technique is based on estimating the concentrations of H^+ and OH^- ions, which determine the electric potential. Biosorbent (0.1 g) was stirred with 20 mL of KNO_3 solution in 11 closed containers in an incubator for 24 h. The initial pH was set at 2–12 using HNO_3 and KOH (0.1 M). The final pH of each container was recorded with a pH meter (Hach Co. USA), and the values were plotted against initial pH values. The Langmuir and Freundlich models were used to construct adsorption isotherms. Kinetic studies of uptake dye using pseudo-first-order and pseudo-second-order kinetic models were performed. Table 2 depicts the formulas used in the presented models [9,20]. The most suitable model was selected considering the coefficient of determination (R^2) and the sum of squares due to error (SSE) analyses (Eq. 1).

$$\mathrm{SSE} = \sum \left[\frac{(q_{t,expt} - q_{t,theo})^2}{q_{t,expt^2}} \right] \qquad (1)$$

The percentage of dye removed by biomass was calculated using the following equation:

$$\text{Removed dye } (\%) = [(C_0 - C)/C] \times 100 \qquad (2)$$

where C_0 is the initial dye concentration before adding the sorbent and C is dye concentration after adsorption with potato peel waste biomass.

3. Results and discussion

3.1. Biomass characteristics

The processed biomass was characterized using scanning electron microscopy (SEM) and energy dispersion spectroscopy (EDS) at the Materials Engineering Laboratory of Majlesi University, Iran (kV 20.0, takeoff angle 25.0˚, and elapsed live time 10.0). A SEM micrograph and an EDS spectrum of the biomass are shown in Figs. 1(a) and 2(b). As shown in Table 3, the main elements of the biomass structure are carbon, oxygen, and potassium.

Potato peels have phenolic compounds, glycoalkaloids, and cell wall polysaccharides [21]. There are many chemical groups on the sorbent which can react chemically to form a chemical bond; acid groups on the sorbent could provide ion exchange sites for metal ions and ionic dyes, amine groups can provide a single pair of electrons for chelating with metal ions, etc. Relatively inert sorbent surfaces may only provide

Table 2
Formulas used in models for kinetic study and isotherm construction [2,8]

Kinetic equations		Isotherm equations	
Pseudo-first-order	$\ln\left(1 - \frac{q_t}{q_e}\right) = -k_1 t$	Langmuir	$\frac{c_e}{q_e} = \frac{1}{q_m K} + \frac{1}{q_m} c_e$
Pseudo-second-order	$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$	Freundlich	$\log q_e = \log k + \frac{1}{n} \log c_e$

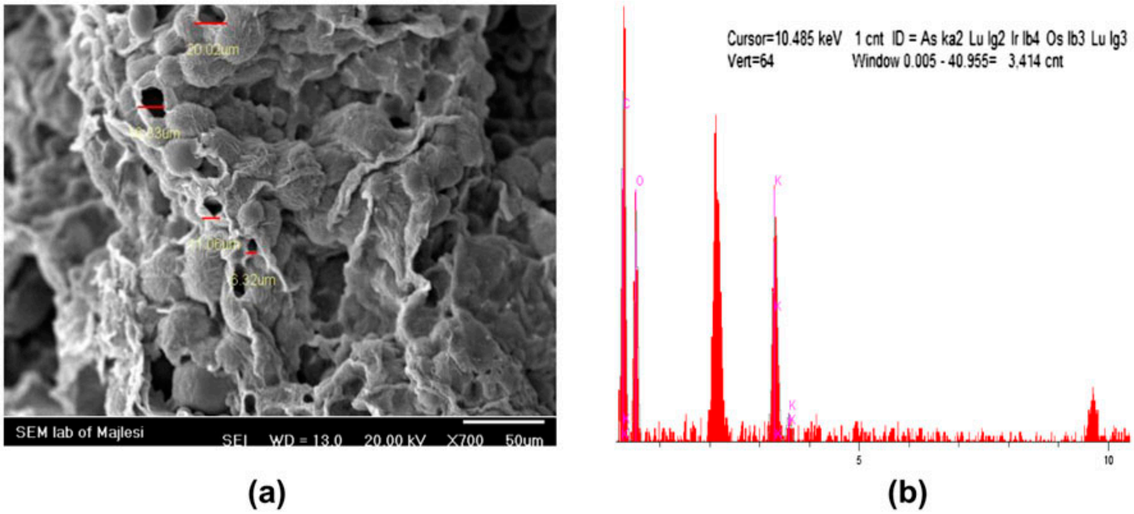


Fig. 1. (a) EDS surface chemical properties of the biomass and (b) SEM micrograph of potato peel waste biomass.

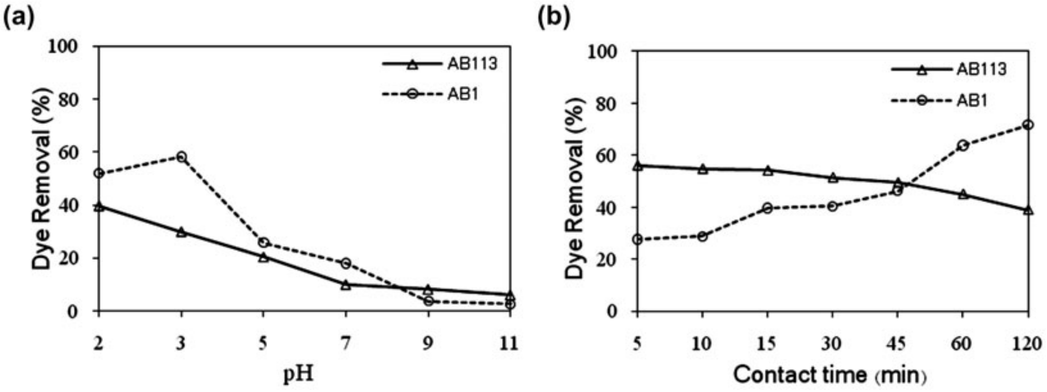


Fig. 2. Effects of pH and contact time on removal of acid dyes, initial dye concentration was 300 mg L^{-1}: (a) Effect of pH; (b) Effect of contact time.

Table 3
EDS analysis result of the used biomass (kV 20.0, takeoff angle 25.0°, and elapsed live time 10.0)

	Units	Conc.	Error sig-2	Intensity (c/s)	Line	Elt.
Total	wt.%	36.402	4.312	46.51	Ka	C
	wt.%	54.657	3.357	28.20	Ka	O
	wt.%	8.941	3.948	38.99	Ka	K
	wt.%	100.000				

physical sites for diffusion-controlled bond formation. Although FTIR analysis can determine the presence of functional groups, we could not use this technique for this study. Oxygen was the most concentrated element in the biomass structure and pollutants have been strongly linked with functional groups containing oxygen and nitrogen in biosorbent structures [2–4], [22].

3.2. Effect of pH and contact time on dye removal

The pH of the dye solution is one of the effective factors on dye removal through adsorption. pH can affect the dye and surface charge on the used biomass. As shown in Fig. 2(a), the biosorption of both AB1 and AB113 on the processed biomass decreased with an increasing pH. The highest uptake was obtained at pH of 3 and 2 for AB113 and AB1, respectively, while Prasad et al. reported a maximum adsorption efficiency of nickel onto processed potato peel waste biomass at a pH of 7. Hoseinzadeh et al. reported biosorption of RB5 on the processed biomass decreased with increasing pH, with the highest uptake at a pH of 3. This increase may be related to the dye structure and the pHzpc of the biomass [2,4]. The pHzpc of the biomass was 6.59 in our study. The effect of pH on the biosorption process is possibly related with interactions between the dye anions in the solution and adsorbent surface charge contaminants [2,23]. When the pH of the solution is higher than the pHzpc, the biosorbent surface has more negative charge and therefore dye anion adsorption is decreased. Acid dyes are known to ionize to some extent in aqueous solutions to form colored anions, which could then exchange with the hydrogen ions of the biomass. During the adsorption experiments, the solution pH slightly increased, possibly because of the transfer of OH^- from the adsorbent's surface into the solution. In other words, the results of this stage showed the strong reaction power between the dye and the elements present in the biosorbent structure. Also, the H^+ and OH^- ions can influence adsorption capacity. The adsorption amount at higher pH than pHzpc reached its minimum amount because the surface of the adsorbent has negative charge, which causes mutual repulsion between dye ions and the adsorbent surface.

To study contact time effect on dye removal, we initially determined equilibrium which was 120 min (solution pH = 3 and 2, biomass dosage = 0.6 g in 250 cc, and initial dye concentration = 200 mg L^{-1}). After reaching equilibrium, the experiments were performed by keeping three variables constant and changing one variable.

As shown in Fig. 2, when the contact time increased from 5 to 120 min (equilibrium time), the removal dye percentage increased from 27.56 to 71.58% for AB1; while by increasing contact time from 5 to 120 min, the removal percentage of AB113 decreased. On the other hand, we found that the highest absorption percentage for AB113 was achieved in the first five minutes and after that, the biosorption rate decreased. Generally, in biosorption processes, if adsorption sites are easily accessible for adsorbate molecules, the adsorption rate will not increase with increasing contact [1,2,8]. In the previous study [2], the biosorption of RB5 dye also increased with increasing contact time, reaching equilibrium after two hours. The difference results we reached for AB1 and AB113 may be because of their different structures.

3.3. Effects of bioadsorbent dosage and initial dye concentration on dye removal

Acid dye removal percentage decreased by increasing the dyes' concentration (Fig. 3). The highest dye removal percentage for both dyes occurred at a dye concentration of 50 mg L^{-1}. The decrease in dye removal efficiency with an increase in dye concentration (Fig. 4) may be related to the initial amount of dye molecules being lower than the available surface sites, making the adsorption ratio independent of the initial dye concentration. At higher concentrations, there would be fewer available sites than dye molecules in the solution, and the AB1 and AB113 removal percentage would depend on the initial dye concentration [1,3,12,23]. As shown in Fig. 3(a), the rate of dye adsorption increased with increasing biomass dosage. The increasing rate of dye adsorption was possibly due to increase in the available surface for adsorption [2–4]. Several researchers have reported similar results [2–4,11,23]. A previous study showed that the adsorption rate of Reactive Black 5 (RB5) dye increased with increasing biomass dosage.

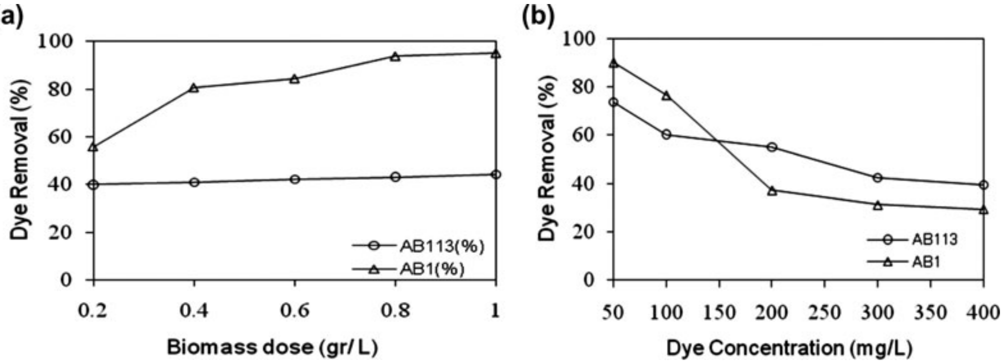


Fig. 3. Effect of biomass dosage (a) and initial dye concentration and (b) on acid dyes removal percentage.

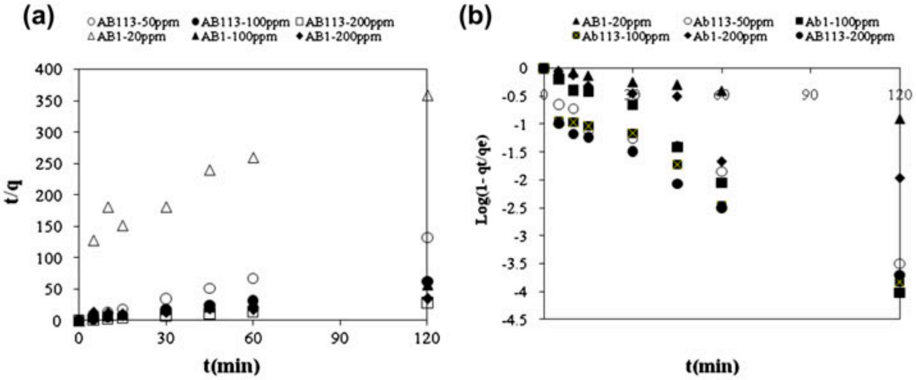


Fig. 4. Experimental uptake curves based on (b) the pseudo-first-order reaction model (a) the pseudo-second-order reaction model with three initial dye concentrations.

Table 4
Isotherm study results: Freundlich and Langmuir parameter values obtained for AB1 and AB113 uptake by potato peel waste

Isotherm model	Dye	K	n	q_m	R^{2*}	SSE**	SE***
Freundlich	AB113	19.44	8.77	–	0.963	5.398	0.00356
	AB1	2.88	6.27	–	0.9758	2.8662	0.0057
Langmuir	AB113	0.35	–	11.71	0.9998	0.4003	0.02375
	AB1	1.07	–	1.79	0.9987	0.516	0.0711

*Coefficient of determination. **Sum of the errors squared. ***Standard error.

3.4. Adsorption isotherm studies

It is important to understand the mechanism of how pollutants are removed by biosorbents in order to develop biosorption processes for the concentration, removal, and recovery of pollutants from aqueous solutions [1,3]. Adsorption isotherms play a fundamental role in the optimization of adsorbent consumption and explain how pollutants react with adsorbent material [23]. Choosing of the best fit isotherm was done by linear regression analyses and a comparison of their correlation coefficients (R^2). Equilibrium studies which express the adsorbent capacity for the dye are explained by adsorption isotherms. The data obtained from experiments were fitted to linear models of Langmuir and Freundlich isotherms (Table 2): in the Freundlich isotherm equation, q_e is equilibrium dye concentration on the adsorbent ($\mathrm{mg\,g^{-1}}$), C_e is the equilibrium concentration of dye ($\mathrm{mg\,L^{-1}}$), k and n are constants that describe the capacity and the intensity for the adsorption, respectively. For the Langmuir model, $q_{\max}$ is the maximum adsorption capacity ($\mathrm{mg\,g^{-1}}$) and b is a constant in the model ($\mathrm{mg\,L^{-1}}$), respectively. The parameters calculated based on the isotherm equations are given in Table 4. Prasad et al. reported maximum adsorption capacities of 13.09 and 7.874 $\mathrm{mg\,g^{-1}}$ for the adsorption of pollutants onto processed biomass from potato peel waste [1], Hoeinzadeh et al. obtained the maximum adsorption capacity for the RB5 dye 3.612 $\mathrm{mg\,g^{-1}}$, and the Langmuir constant (k_{ads}) for RB5 dye was 4.55 $\mathrm{mg\,L^{-1}}$, which is higher than q_m for AB1 (4.55 vs. 1.79 $\mathrm{mg\,L^{-1}}$) and lower than AB113 (4.55 vs. 11.71 $\mathrm{mg\,L^{-1}}$) in our study.

Based on the coefficient of determination (R^2) and the sum of SSE for the experiment data, the adsorption of both AB1 and AB113 dyes proceed in accordance with the Langmuir isotherm model. Langmuir isotherm model indicates a homogeneous process and each adsorbate molecule has equal adsorption activation energy. When the adsorption process follows the Type L isotherm of the Langmuir model, it shows that the ion substrate adsorption (cation or anion) is confronted with weak resistance of the solution molecules [2,11]. This also indicates that the processed biomass is homogeneous [1].

3.5. Adsorption kinetics

The reaction rate is described by adsorption kinetics. Adsorption kinetics are important in characterizing the efficiency of a sorbent for use in the

Table 5
Kinetic study results; values of pseudo-first-order and pseudo-second-order kinetic models

Dye	Pseudo-first-order kinetic parameters					Pseudo-second-order kinetic parameters			
	C (mg L^{-1})	Q_e (mg g^{-1})	k_1 (1/min)	R^2	SSE	Q_e (mg g^{-1})	k_2 (g/mg min)	R^2	SSE
AB113	50	0.911	0.059	0.9751	1.096	0.919	0.715	0.9996	1.105
AB1	20	0.381	0.017	0.992	3.84	0.425	0.053	0.794	4.11
AB113	100	1.911	0.0656	0.9473	1.037	1.92	0.477	0.9997	1.041
AB1	100	2.12	0.077	0.990	1.76	2.25	0.064	0.991	1.88
AB113	200	4.275	0.0619	0.9214	1.019	4.293	0.364	0.9999	1.021
AB1	200	3.51	0.0409	0.876	2.83	4.293	0.364	0.9999	3.31

biosorption process. In the used kinetic models (presented in Table 1), k_1 and k_2 are the rate constants for the pseudo-first-order and the pseudo-second-order models. q_e and q_t are the amounts of adsorbed material based on mg g^{-1} at equilibrium time and specified time, respectively, and k_m is the speed based on min L^{-1}. The results of the kinetic study and the parameters associated with each model are shown in Table 4 and Fig. 4.

They found the optimum pH value (3) and then analyzed the kinetic experiments at this value. The models were compared on the basis of SSE and R2 analyses. The analyses' results are presented in Table 5, which demonstrate that for AB113 dye, the pseudo-second-order kinetic model fits the experimental data significantly better than the pseudo-first-order kinetic model. Moreover, for AB1 dye, the pseudo-first-order kinetic model fits better. Hoseinzadeh et al. found RB5 biosorption onto peel waste to be consistent with pseudo-second-order kinetic model, too. The conclusion fits the experimental system in which dyes with different structures and characterizations are being adsorbed by same or different kinetic model(s) onto potato peel waste biomass.

As a very general guideline, if equilibrium is achieved within three hours, the process is usually kinetic controlled and when achieved above 24 h, it is diffusion controlled. Either or both kinetic and sorption processes may be rate controlling in the 3–24 h period. Hoseinzadeh and Rahmani [1] found that diffusion within solid particles is related to the ratio of molecular diameters of the diffusing particles to the diameters of pores. In this study, the average pore diameter of the biomass was 50 µm. As the (molecule diameter/pore diameter) ratio becomes larger, multidirectional interactions between pore walls and sorbate molecules become significant and the sorption of the dye slows down. They are therefore responsible for the slow approach to equilibrium.

4. Conclusions

Biosorption of AB1 and AB113 were highest under acidic conditions (pH of 2 and 3, respectively); the biosorption of the two dyes increases with increasing biomass dosage and contact time, but biosorption of the AB113 decrease with increasing contact time. The analyses demonstrate that the pseudo-second-order and pseudo-first-order kinetic models are better fit for the adsorption of Acid Blue 113 and Acid Black 1, respectively, on the potato peel waste biomass. Evaluation of the adsorption results obtained on the basis of isotherm models showed that biosorption of acid dyes by potato peel waste was best described by the Langmuir model.

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