Adsorption of Basic and Acid Dyes Using Alginate/Sericin Composite Beads

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Alginate/sericin composite beads were prepared from a blend between sericin and alginate by the gelation process. The composite beads were used as the adsorbent for the removal of basic and acid dyes from an aqueous solution. The morphological structure of the beads was characterized by scanning electron microscopy (SEM). Adsorption experiments were conducted using contact time, concentration of sericin, pH and adsorbent dosage. The equilibrium adsorption data was achieved within 360 min at 5 % w/v of sericin. The Langmuir isotherm model showed the maximum monolayer adsorption capacity for basic dye (pH 8) and acid dye (pH 2) was 5.98 and 7.60 mg/g, respectively. The adsorption kinetics were described by the pseudo second-order model. The desorption experiment was demonstrated and repeated three times, all of which exhibited that it was regenerative and reusable. This study demonstrates that the alginate/sericin composite beads are effective adsorbent for the removal of dyes from aqueous solution.

Keywords: Acid dye, Adsorption, Alginate, Basic dye, Composite, Sericin.

INTRODUCTION

Synthetic dyes such as cationic (all basic dyes) and anionic (direct, acid and reactive dyes) were found to be present in wastewater from industries such as textile, leather, paper, printing, plastic, cosmetics and other. This dye pollutant has become a major source of contamination which is toxic and has detrimental effect on human health [1,2]. Moreover, the dyes pollutants were determinant hindering photosynthesis effects on ecosystem [3]. As a result the removal of the dye from the wastewater is essential. The methods of dye removal from wastewater, include membrane filtration [4], coagulation-flocculation [5], advanced oxidation process [6], biodegradation [7], adsorption process [8] and ion exchange [9].

The adsorption process is considered to be very effective for the removal of dye from wastewaters. Activated carbons from natural wastes are a great adsorbent due to low-cost materials, waste materials and by-products [10,11]. However, there are many problems regarding the regeneration of the exhausted activated carbon [12]. Thus, an effort has been made to develop them by using adsorbent materials such as: polymers, clays [13], biosorbents [14] and composites [15].

Bombyx mori silk is composed of two major protein, fibroin and sericin constitute 70-75 %, 20-25 % of silk and 5 % other impurities [16]. Sericin can be extracted by various

methods such as the high pressure and high temperature method, boiling, alkaline degumming, acidic degumming, soap degumming and enzymes [17,18]. Sericin is a water soluble protein and has several important properties including its excellent moisture adsorption, UV resistance, anticoagulation, anti-carcinogenic qualities the inhibitory action of tyrosinase and low-cost biosorbent [19,20].

Alginate, or alginic acid, is unbranched polysaccharides extracted from brown seaweed, which is widely diffused in natural organisms. A linear anionic copolymer is comprised of α -L-guluronic acid and β -D-mannuronic acid. Alginate has been widely applied to the treatment of wastewater because of its biocompatibility, nontoxicity, high viscosity and low-cost relatively compared to other polymeric materials [2]. More importantly, alginate solution is used for cross-linking to make forming hydrogels (egg-box structure) of alginate beads by selective ionic interaction with the divalent metal such as Ca^{2+} or Na^+ [21,22]. Therefore, alginate and sericin were of interest to the researchers toward the properties and the composite material found to be adsorbent has been used for the treatment of a wide range of polluted wastewater [23]. In this study, alginate/sericin composite beads were prepared with a gelation process. The adsorption process of dye adsorption on composite beads was investigated. Additionally, the batch experiment was examined by assessment of the adsorption

isotherms, kinetic parameter and desorption and recycling studies.

EXPERIMENTAL

B. mori silkworm's cocoons were provided by the Queen Sirikit Department of Sericulture, the Ministry of Agriculture and Cooperatives. Sodium alginate was purchased from Sigma-Aldrich. Calcium chloride was purchased from Ajax Finechem Pty Ltd. Cationic dye (C.I. basic yellow 11) was obtained from DyStar Thai Ltd. and anionic dye (C.I. acid red 336) was secured from Clariant (Thailand) Ltd. The chemical structures of the basic and acid dyes are shown in Fig. 1.

Preparation of sericin powder: The cocoons were cleaned to remove impurities and cut into small pieces (about 0.5 cm²). After cleaned, the cocoons were dried at 50 °C for 60 min. The degumming process was carried out at a high pressure and high temperature for a liquor ratio of 1:30 at 121 °C for 30 min. The filtrate solution used paper filtration to remove insoluble fibers and dialyzed against deionized water for 3 days. After dialysis, the sericin solution was dried and finally the sericin was crushed into sericin powder.

Preparation alginate/sericin composite beads: Alginate/sericin composite beads were prepared by adding sericin powder (1 to 5 g) in 100 mL of 2 % (w/v) alginate solution. The solution was magnetically stirred for 240 min. The alginate/sericin solution was dropped through syringe precipitation in an aqueous solution of CaCl₂ (1 % w/v). The composite beads were washed and preserved in deionized water and an aqueous solution for future use.

Characterization: The surface morphology of the alginate/sericin composite beads was characterized by scanning electron microscopy (SEM, JEOL JSM-6380 LV, USA).

Adsorption studies: The batch adsorption experiments were carried out with a fixed amount of adsorption (2 g) was dispersed in 100 mL of concentration and 50 mg/L of basic and acid dyes solution without adjusting the pH value. The dispersion was stirred at a shaking speed of 120 rpm at room temperature. The dye concentrations were measured using double beam UV-visible spectrophotometer (Shimadzu, model UV 2450, Japan) with the wavelength at 424 and 303.5 nm for basic and acid dyes, respectively. The effect of pH on the adsorption capacities was determined in the pH range from

1 to 10. The pH was adjusted with 0.1 M NaOH or 0.1 M HCl solution. A different dosage of the adsorbent in range of 2 to 14 g was used to examine the effect of an adsorbent dosage on the adsorption of basic and acid dyes.

Adsorption isotherm: The adsorption isotherm was obtained by a fixed 2 g of adsorbent and 100 mL of basic and acid dye solution with different concentrations, ranging from 10 to 50 mg/L. The maximum pH value of basic and acid dye solution for adsorption was equal to 8 and 2, respectively. The dispersion were stirred until they reached adsorption equilibrium at 360 min.

Adsorption kinetics: The batch adsorption kinetic experiment was carried out by different concentration of dyes solution from 10 to 50 mg/L and by stirring in the dye solution batch shaker at room temperature for different time between 60 to 360 min. At the end of each adsorption period the dye solution concentrations were analyzed similarly.

Desorption and recycling studies: For the desorption experiment, 2 g of adsorbent was used in 100 mL of dye aqueous solution, with 50 mg/L at pH equal to 8 for basic dyes and 2 for acid dyes. After the adsorption, the mixture was washed thoroughly in deionized water. The desorption experiment used 100 mL of deionized water and the pH was adjusted by 0.1 M NaOH or 0.1 M HCl solution at room temperature. The desorption experiments were determined in a pH range from 1 to 10 with constant stirring (120 rpm) at room temperature for 360 min. The removal of dyes in deionized water was measured at 424 and 303.5 nm for basic and acid dyes, respectively. The desorption can be calculated from the amount of dyes adsorbed according to an equation:

$$\text{Desorption ratio} = \frac{\text{Amount of dyes desorbed}}{\text{Amount of dyes adsorbed}} \times 100 \quad (1)$$

RESULTS AND DISCUSSION

Characterization of alginate/sericin composite beads:

Fig. 2 showed the SEM images of dried alginate/5 % sericin composite beads. The surface of alginate/5 % sericin composite beads before dye adsorption exhibited a slightly rough and homogeneous blend composite as shown in Fig. 2a. After acid dye adsorption, the composite bead showed a smooth surface (Fig. 2b). Fig. 2c shows that the surface roughness of the

Fig. 1. Chemical structure of (a) basic and (b) acid dyes

Fig. 2. SEM image of alginate/5 % sericin composite beads a) before adsorption, b) after basic dye adsorption (pH 8) and c) after acid dye adsorption (pH 2)

Fig. 3. Effect of contact time and sericin concentration in bead on dye removal of (a) basic and (b) acid dye (adsorbent dosage: 2 g; dye concentration: 50 mg/L; Volume: 100 mL; pH 7)

composite bead after acid dye adsorption. The surface of the composite beads after acid dye adsorption had a rougher surface than with basic dye. It could be explained that with acidic condition dye adsorption, the degradation arose from the alginate mostly due to acid catalyzed hydrolysis [24].

Effect of contact time and concentration on sericin:

The inclusion of contact time and different sericin powder concentration on the removal of basic and acid dyes by alginate/sericin composite beads is shown in Fig. 3. It can be seen that the basic dye removal was rapidly adsorbed in the first 60 min and then the dye adsorption gradually increased from 60 to 360 min. After 360 min, the adsorption rate decreased (Fig. 3a). Meanwhile, the acid dye was rapidly adsorbed in the first 360 min and then the dye removal decreased between 360 to 480 min (Fig. 3b). Therefore, 360 min was chosen to be the equilibrium state contact time for basic and acid dyes adsorption with alginate/sericin composite beads. The dye removal of alginate/sericin composite beads improved with the increasing of sericin powder in the composite beads. However, sericin powder of more than 5 % in the alginate/sericin composite beads, hindered the formation of the alginate/sericin composite beads. Therefore, alginate/5 % sericin composite beads were chosen as adsorbent of basic and acid dyes throughout this study.

Effect of pH: The pH of the aqueous solution was a significant factor controlling the adsorption process by affecting both the adsorbent surface charge (protonated or deprotonated)

and the degree of ionization of dye molecules [25,26]. The adsorption of basic and acid dyes by alginate/5 % sericin composite beads was studied at different pH solutions from 1 to 10, as shown in Fig. 4.

Fig. 4. Effect of pH of solution on basic and acid dyes adsorption onto alginate/5 % sericin composite beads (adsorbent dosage: 2 g; dye concentration: 50 mg/L; Volume: 100 mL; 360 min)

The basic dye was cationic dye with NR_3^+ groups in its chemical structure. The results showed that the alkaline conditions

supported the adsorption of basic dye on alginate/5 % sericin composite beads. Electrostatic interaction occurred between cationic groups of basic dye and negatively charged the adsorbent surface. The maximum basic dye removal ratio reaches over 45.11 % at pH equal to 8. The dye removal was increased sharply as the pH solution increases from 5 to 6. As a result, the low pH solution seemed to be connected with a high concentration of H^+ ions (proton) in the dye solution, the dye removal of basic dye decreased. The electrostatic repulsions between cationic groups and positively charged adsorbent surface in acidic condition. When, the pH solution increased, the removal of basic dye increased due to larger electrostatic interaction between cationic groups and a negatively charged adsorbent surface.

As shown in Fig. 4, the acid dyes have anionic properties meaning they have a negative charge of SO_3^- groups (sulfonate groups) in the chemical structure. This result showed that the acidic condition supports the adsorption of acid dye. The maximum removal of acid dye appeared to be at a pH value equal to 2, at which about 45.01 % of the acid dye could be removed. The dye removal of the adsorbent rapidly increased with the increasing of the pH solution from 5 to 6. A pH solution below 5 can be attributed to a high concentration of H^+ ions in the dye solution due to increases electrostatic interactions between SO_3^- groups of acid dyes and positively charged adsorbent surface. With a pH solution above 6, the removal of acid dye decreased due to the solution showing the amount of OH^- groups (hydroxyl) increased. Thus, the electrostatic repulsions between SO_3^- groups and negatively charged adsorbent surface in alkaline conditions. On the other hand, with a pH solution below 2, the dye removal decreased. The reasons for this may be because the solution was too acidic and the adsorbent began to degrade, thus leading to weakened electrostatic interactions between the anionic groups and the positively charged adsorbent surface.

Similarly, alginate had an adsorption effect on basic and acid dyes because the pH decreases with H^+ ions and they will protonate the carboxylate group within the composite beads. Accordingly electrostatic interaction between H^+ and the carboxylate group of alginate has the ability to remove dyes.

Effect of adsorption dosage: Fig. 5 shows the effects of an adsorbent dosage on the basic and acid dyes removal efficiency. The dosage of adsorbent was 2 to 14 g at 360 min and the dye removal increased from 42.27 to 60.07 % and 43.74 to 74.22 % for basic and acid dyes, respectively. The adsorption amount of dye increased with an increasing amount of adsorbent until it reached the equilibrium state. The rapid increase of adsorption with a higher adsorbent dosage was attributed to the availability of a larger surface area and more adsorption sites.

Adsorption isotherms: The adsorption isotherm models studies were completed by alginate/5 % sericin composite beads in basic and acid dyes solution with concentration of dyes at 50 mg/L. In this study, the two adsorption isotherm models, Langmuir and Freundlich isotherms were used for clarifying the adsorption mechanism.

The Langmuir model assumes that the adsorption process occurs at a homogeneous surface. Meanwhile, the Langmuir isotherm describes quantitatively the formation of a monolayer of adsorbate on the surface of the material.

The Langmuir isotherm equation [27]:

$$\frac{C_e}{q_e} = \frac{1}{bX_m} + \frac{C_e}{X_m} \quad (2)$$

where q_e is the amount of dye adsorbed per unit of adsorbent (mg/g), C_e is the equilibrium concentration of adsorbate (mg/L). The constant X_m is the monolayer adsorption capacity (mg/g) and b is the Langmuir constant (L/mg). The Langmuir isotherm of alginate/5 % sericin composite beads and linear plot of C_e/q_e versus C_e as described in Fig. 6a. The correlation coefficient (R^2) was found to be 0.98 and 0.94 for basic and acid dyes, respectively. The results showed the adsorption capacity (X_m) listed in Table-1 gives the adsorption coefficients. The adsorption isotherms of basic and acid dyes could be described very well by the Langmuir isotherm equation. The adsorption capacity was found to be 7.60 and 5.98 mg/g for acid and basic dyes, respectively.

The Freundlich isotherm assumes that the multilayer of the adsorption process occurs on a heterogeneous surface.

The Freundlich isotherm equation [28]:

Fig. 5. Effect of adsorption dosage on dye removal of (a) basic dye (pH 8) and (b) acid dye (pH 2) (alginate/5 % sericin composite beads; dye concentration: 50 mg/L; Volume: 100 mL; 360 min)

Fig. 6. Isotherm plot of the adsorption of alginate/5 % sericin composite beads (a) Langmuir adsorption model and (b) Freundlich adsorption model

TABLE-1
ADSORPTION KINETIC PARAMETERS FOR BASIC AND ACID DYES ADSORPTION ON ALGINATE/5 % SERICIN COMPOSITE BEAD (ADSORBENT DOSAGE: 2 g; VOLUME: 100 mL; CONCENTRATION 50 mg/L)

Dyes type	$q_{e(\text{exp})}$	Pseudo-first order			Pseudo-second order		
		$q_{e(\text{cal})}$ (mg/g)	K_1 (min^{-1})	R^2	$q_{e(\text{cal})}$ (mg/g)	K_2 (g/mg min)	R^2
Basic dye	3.167	0.6767	-0.0072	0.8673	3.971	0.0001	0.9317
Acid dye	5.731	0.7159	-0.0041	0.8907	6.506	0.0002	0.9673

TABLE-2
PARAMETERS OF THE LANGMUIR AND FREUNDLICH ISOTHERM MODELS FROM BASIC AND ACID DYES FOR ALGINATE/5 % SERICIN COMPOSITE BEADS

Dye type	Langmuir isotherm model			Freundlich isotherm model		
	X_m (mg/g)	b (L/mg)	R^2	K (mg/g)	$1/n$ (g/L)	R^2
Basic dye	5.98	1.14	0.9809	0.29	0.68	0.9798
Acid dye	7.60	5.31	0.9482	0.96	0.57	0.9119

$$\log q_e = \log K + \left(\frac{1}{n}\right) \log C_e \quad (3)$$

where K is the Freundlich constant related to the adsorption capacity of the adsorption and n measures the surface heterogeneity. The Freundlich isotherm of alginate/5 % sericin composite beads the plot of $\log q_e$ versus $\log C_e$ for the dyes gave straight lines with a slope of $1/n$ and intercepted by $\log K$ (Fig. 6) Values of Freundlich constants are given in Table-2 listed by the Freundlich isotherm equation. The $1/n$ values are equal to 0.68 and 0.57 for basic and acid dyes, respectively. In this study, the slope of $1/n$ for basic and acid dyes onto alginate/5 % sericin composite bead was less than 1 and showed favourable sorption and confirmed the heterogeneity of the adsorption. The Freundlich isotherm parameter $1/n$ is a measure of the deviation from linearity of the adsorption.

The correlation coefficient (R^2) of the Langmuir isotherm model was better than that of the Freundlich isotherm model. This indicates that the adsorption of basic and acid dyes onto alginate/5 % sericin composite beads can occur on a homogeneous surface by monolayer adsorption and can be explained in terms of chemisorption as the formation of ionic bonds between adsorbent and dye solution.

Table-3 showed that the maximum adsorption of basic and acid dyes by alginate/sericin composite beads have been

TABLE-3
COMPARISON AMONG ADSORPTION OF DIFFERENT BIOSORBENTS FOR BASIC AND ACID DYES

Dyes	Biosorbents	Adsorption capacity (mg/g)	Ref.
Basic dyes	Coffee residues	4.68	[29]
	Pinewood	5.56	[30]
	Alginate/sericin composite bead	5.98	This study
	Banana pith	8.5	[31]
	Banana peel	20.8	[32]
	Eggshell	94.9	[33]
Acid dyes	Coir pith	1.6	[34]
	Banana pith	4.42	[35]
	Alginate/sericin composite bead	7.60	This study
	Peanut hull	13.99	[36]
	Orange peel	19.88	[37]
	Seed husk	41.66	[38]

compared with various natural adsorbents. In Table-3, it can be summarized that the adsorption of basic and acid dyes onto alginate/sericin composite beads is effective comparatively to other adsorbents. This result could be described as the adsorption mechanism is believed to have material properties because of the electrostatic interaction between the adsorbent and dye solutions. Moreover, both alginate and sericin powder are

abundant relatively low cost biosorbents with biocompatibility, excellent moisture adsorption and relatively non-toxic properties. Therefore, alginate and sericin powder provide efficient material for remediating dyes from contaminated wastewater.

Adsorption kinetics: Kinetic models are used to predict the rate of adsorption. The dyes solution was concentrated at 50 mg/L at room temperature at a constant contact of 360 min. There were two kinetic models: pseudo-first-order and pseudo-second-order, while are represented by the following equation:

Pseudo-first order model [39]:

$$\log(q_e - q_t) = \log q_e - \frac{K_1}{2.303} t \quad (4)$$

Pseudo-second order model [40]:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

where q_e and q_t (mg/g) are the amount of dye adsorption equilibrium times (mg/g) and the amount of dye adsorbed at any time (min), K_1 (min^{-1}) and K_2 (g/mg min) are the constant rate of the pseudo-first order and pseudo-second order adsorption models. The values of K_1 and q_e were calculated from the slope and intercepts of the plot of the $\log(q_e - q_t)$ versus t (Fig. 7a), while the values of K_2 and q_e were evaluated from the intercept and slope of a plot t/q_t versus t . (Fig. 7b). The kinetic parameters were summarized in Table-1 and show a good agreement between the experiment q_e value [$q_{e(\text{exp})}$] and the calculated q_e value [$q_{e(\text{cal})}$] and linear correlation coefficients (R^2). It was observed that the correlation coefficients for the pseudo-first order kinetic model less was than that of pseudo-second order kinetic model for acid and basic dyes. Meanwhile, the $q_{e(\text{cal})}$ value for the pseudo-second order kinetic model was consistent with $q_{e(\text{exp})}$ value. Thus, the adsorption of alginate/5 % sericin composite beads for basic and acid dyes in this study could be described better by the pseudo-second-order kinetic model. This result indicated that the sorption process was complex and involved more than one mechanism.

Desorption and recycling studies: The desorption studies are the most important factor causing the economic success of the adsorption process. Fig. 8 shows the maximum desorp-

tion efficiency of basic and acid dyes within 360 min. The adsorption basic dye on adsorbent surface can represent desorption by acidic condition (pH 4). It can be seen that the maximum desorption of saturated adsorbent was 42.16 % for alginate/5 % sericin composite beads due to a low pH solution, concentrations of H^+ ions in deionized water were protonated. Thus, the desorption efficiency of basic dye increased with the decrease of the electrostatic interactions between cationic groups of basic dye and the adsorbent surface. The adsorption acid dye on adsorbents can cause desorption by alkaline condition (pH 8). The maximum desorption of acid dye was observed at 53.65 %. A high pH solution showed the amount of OH^- groups increased. Therefore, the desorption efficiency of acid dye increasing can be attributed to the electrostatic interactions between cationic groups of basic dye and OH^- groups. The higher desorption efficiency of the dyes can be explained by the fact that the amphoteric property of sericin in alginate/sericin composite beads can undergo desorption by acidic and alkaline conditions.

Fig. 8. Desorption study of basic and acid dyes at different pH (volume: 100 mL; contact time: 360 min)

Table-4 shows the recycle process of alginate/5 % sericin composite bead adsorbents was attributed to successively

Fig. 7. Fitting with a different kinetic model (a) Pseudo-first order model and (b) Pseudo-second order model for alginate/5 % sericin composite beads of adsorption basic and acid dyes

TABLE-4
REGENERATION CYCLE FOR BASIC AND ACID
DYES ADSORPTION ON ALGINATE/5 % SERICIN
COMPOSITE BEADS (ADSORBENT DOSAGE: 2 g;
VOLUME: 100 mL; 360 min)

Regeneration cycle	Dye removal (%)	
	Basic dye	Acid dye
1	42.16	70.39
2	2.07	62.84
3	1.04	6.34

completing the adsorption-desorption cycles for three times at pH 4 for basic dye and pH 8 for acid dye. Alginate/5 % sericin composite beads made possible the regeneration and good reusability.

Conclusion

The present study described the preparation of alginate/sericin composite beads and was used for basic and acid dye removal from an aqueous solution using the batch technique. The adsorption isotherm investigated could be expressed by the Langmuir model and Freundlich model. The kinetic study shows that the process can be described by a pseudo-second order model. The dye removal of basic and acid dyes can reach 5.98 and 7.60 mg/g, respectively. The pH value of dye removal changed significantly at 8 and 2 for basic and acid dyes, respectively. Finally, the desorption and recycling studies to alginate/5 % sericin composite beads indicated reusability after dye adsorption and desorption were repeated three times.

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