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Efficient Removal of Cationic and Anionic Dyes from Aqueous Solutions using Regenerated Silk Fibroin Beads

WALAIKORN NITAYAPHAT[1,*] and THANUT JINTAKOSOL[2]

[1]Department of Home Economics, Faculty of Science, Srinakharinwirot University, Bangkok, Thailand
[2]Gems and Jewelry Program, College of Creative Industry, Srinakharinwirot University, Bangkok, Thailand

*Corresponding author: Fax: +66 2 2584000; Tel: +66 2 6495000; E-mail: walaikorn@g.swu.ac.th

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Regenerated silk fibroin beads (rSFB) were successfully prepared and applied to adsorbing methylene blue and Lanasyn Navy M-DNL. The effects of contact time, initial pH and initial concentration of dyes were analyzed. The experimental results showed that, the adsorption was pH dependent with a high removal efficiency of methylene blue in basic range and high removal of Lanasyn Navy M-DNL in acidic range. The adsorption process was analyzed by using Langmuir and Freundlich isotherm models and the Langmuir isotherm model showed the best fitting to the isotherm data. The maximum adsorption capacities for methylene blue and Lanasyn Navy M-DNL were 47.55 and 78.74 µmg/g, respectively. The kinetics study showed that the adsorption process followed the second order kinetic model. The SEM images demonstrated that after adsorption the dye was adsorbed onto the regenerated silk fibroin beads surface. The adsorbent was stable and active for up to five successive cycles. Regenerated silk fibroin beads showed to be an advantageous adsorbents in terms of availability, which is beneficial for the wastewater treatment.

Keywords: Adsorption, Silk fibroin, Methylene blue, Lanasyn Navy M-DNL, Biosorbent.

INTRODUCTION

Synthetic dyes are often discharged in the form of coloured wastewater in industries of cosmetics, food, leather, paper and textiles. These pollutants are environmentally hazardous to human beings and animals. Dyes and degradation products are toxic, carcinogenic, mutagenic and teratogenic [1,2]. Exposure to synthetic dyes can result in irritation of the respiratory tract, skin, eyes, sore throat, asthma and allergic contract dermatitis [3].

Many conventional techniques have been developed for the removal of dyes from wastewater *e.g.* coagulation, ion-exchange, precipitation, solid phase extraction, *etc*. Among these techniques, adsorption has attracted great interest because of its easy operation, high efficiency and low cost [4]. Many potential low cost adsorbents, such as activated carbon, mineral clays, biomaterials and polymers have been extensively used for dye removal [5-8].

Silk fibroin is a natural protein fiber that has long been used as a high quality textile fabric for its moderate moisture absorption and retention properties [9]. However, a large amount of leftovers during silk fiber production are wasted due to limited fiber reprocessing technology. The reuse of waste silk is becoming more and more important. With its hydrophilic nature and amphoteric properties, silk fibroin can interact with many dyes [10,11]. Thus, this makes it possible to remove both anionic and cationic dyes [12,13]. However, there are no studies exploring the potentiality of silk fibroin in the form of beads and using it for anionic and cationic dyes from aqueous solutions.

In the present work, regenerated silk fibroin beads (rSFB) were prepared and used to remove anionic and cationic dyes from aqueous solutions by a batch adsorption process. The influences of contact time, adsorbent concentration and initial pH on the adsorption efficiency were studied systematically. The efficiency of regeneration of the adsorbent was also evaluated. Additionally, the experimental data was examined by evaluating the adsorption isotherms and kinetics.

EXPERIMENTAL

Thai silk cocoons of Bombyx mori silkworms "Nang Lai" were supplied by the Queen Sirikit Department of Sericulture

Center, Thailand. Methylene blue is a cationic dye, with a molecular formula $C_{16}H_{18}N_3SCl$, and a maximum absorbance that equals 575 nm. Lanasyn Navy M-DNL is an anionic dye, with a molecular formula $C_{14}H_8N_2O_8S_2Na_2$ and a maximum absorbance that equals 576 nm. Analytical grade formic acid, sodium hydroxide, methanol and hydrochloric acid were purchased from Sigma-Aldrich, U.K.

Preparation of regenerated silk fibroin beads: The silk cocoon were cleaned to remove impurities and then dried at 50 ºC for 60 min. The degumming process was carried out in an autoclave at 121 ºC and pressure 5 psi for 30 min. The material-to-liquid ratio used was 1:30. The material was filtered and the obtained degummed silk was rinsed with deionized water to remove sericin. The degummed silk was then directly dissolved in formic acid at room temperature to form a 25 wt.% silk fibroin solution. This solution was dropped through a syringe into a methanol bath and gave rise to the regenerated silk fibroin beads (rSFB). The beads were washed with deionized water and preserved in an aqueous environment for future use.

Characterization of regenerated silk fibroin beads (rSFB): The surface morphology and microstructure of rSFB were analyzed using a scanning electron microscope (SU3500, Hitachi, Japan). Before observing the SEM, the samples were fixed on aluminum stubs and coated with gold.

Adsorption experiments: The adsorption of dyes onto rSFB were carried out in a thermostatic bath, by adding an amount of rSFB into a flask that contained 50 mL of 50 mg/L methylene blue and Lanasyn Navy M-DNL by varying the contact time from 0 to 9 h, pH of solution from 4 to 10 and adsorbent concentration of 1 to 5 mg/L at 30 ºC and 120 rpm stirring condition. The pH was adjusted to a given value by adding of HCl (0.1 M) or NaCl (0.1 M) solutions. After well-established intervals of time, the adsorbate was separated from the adsorbent. The concentration of residue methylene blue and Lanasyn Navy M-DNL was determined by measuring the absorbance at maximum wavelengths by UV-vis spectrophotometer (UV2450, Shimadzu, Japan). The dye removal efficiency and equilibrium adsorption were calculated by the following equations:

$$\text{Dye removal efficiency (\%)} = \frac{C_o - C}{C_o} \times 100$$

$$\text{Adsorption capacity } (q_e\text{, mg/g}) = \frac{C_o - C_e}{W} \times V$$

where, C_o (mg/L) is the initial dye concentration, C (mg/L) is the concentration of dye at time t (min), C_e is the equilibrium dye concentration (mg/L), V is the volume of dye solutions (L), q_e is the adsorption amount per unit gram of adsorbent at equilibrium (mg/g), and W corresponds to dry weight adsorbents (g).

Isotherm studies: The adsorption isotherms experiments were carried out by using 1 mg/L of rSFB and 50 mL of dye solutions with the dye initial concentrations in 10-50 mg/L range. These solutions were adjusted at an optimum pH for adsorption, *i.e.* pH 10 (for methylene blue) and pH 4 (for Lanasyn Navy M-DNL). The dispersions were stirred at 30 ºC and 120 rpm until adsorption equilibrium was reached (7 h).

Adsorption kinetics: Adsorption of dye solutions with concentrations in the range of 10-50 mg/L on the rSFB was conducted at time intervals of 1-7 h. The same procedures as explained in the isotherm part were applied.

Desorption and regeneration experiments: The efficiency of regeneration was evaluated using five consecutive cycles. Each cycle (adsorption-desorption) consisted of an adsorption equilibrium test followed by a desorption test. Procedures similar to the adsorption tests as described above were adopted. After completion of the adsorption equilibrium test, the adsorbate was separated from the adsorbent. The concentration of residual dye in the solution was then determined. The dye-saturated-rSFB was transferred immediately to a deionized water, and proceeded with the desorption test. The mixture was stirred in a pH range from 4 to 10 with a speed of 100 rpm at 30 ºC for 7 h. The rSFB was separated again from the mixture upon completion of desorption and the concentration of desorbed dyes in the solution was determined.

RESULTS AND DISCUSSION

SEM analysis: SEM was used to characterize the surface morphology and microstructure of rSFB. Fig. 1 shows the SEM images of rSFB surface. The surface of rSFB before adsorption had a slight roughness, while that of rSFB after dye adsorption was smooth. The SEM images exhibited that there was a considerable change in the surface after adsorption of dye molecules, which were supposed to adhere to the rSFB surface.

The micrographs of surface of rSFB and rSFB/dyes are shown in Fig. 1. The micrograph of neat rSFB Fig. 1a shows a smooth surface. In rSFB and rSFB/dyes a very uniform distri-

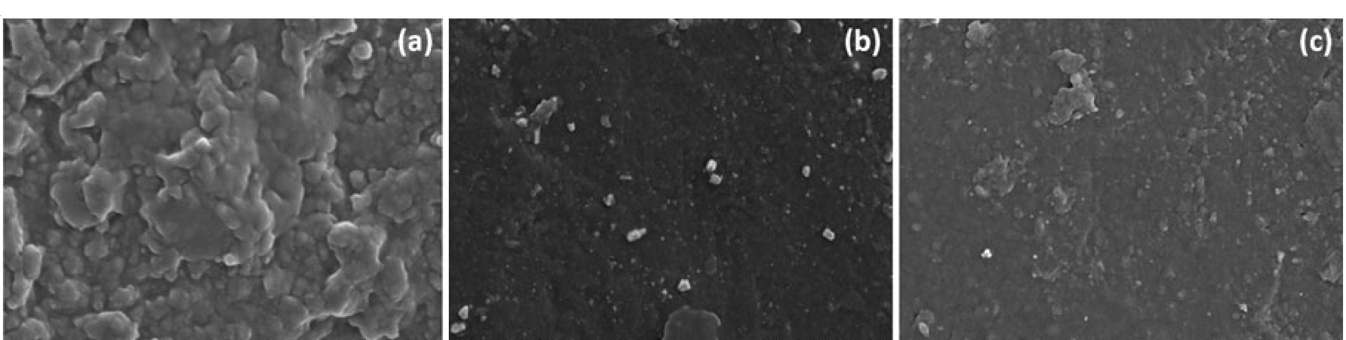

Fig. 1. SEM images of rSFB surface (a) before adsorption (b) after methylene blue adsorption and (c) after Lanasyn Navy M-DNL adsorption

bution of MMT can be observed in the polymer matrix as shown in Fig. 1b-e.

Effect of contact time: The contact time was studied in the range of 0 to 9 h with other experimental conditions being held at fixed values. As can be seen from Fig. 2, during the first 7 h of experiment, the concentration of methylene blue and Lanasyn Navy M-DNL dyes adsorbed on the rSFB increased with the prolonged time. This observation could be explained as at the very beginning of adsorption process, abundant binding sites are available on the surface of adsorbents, which make the adsorption process easier. From 7 to 9 h, no significant change of dyes concentration was observed. The results indicated that the adsorption equilibrium was reached only after 7 h. Thus, in the following experiments, 7 h were selected to ascertain the equilibrium of methylene blue and Lanasyn Navy M-DNL dyes adsorption to the rSFB.

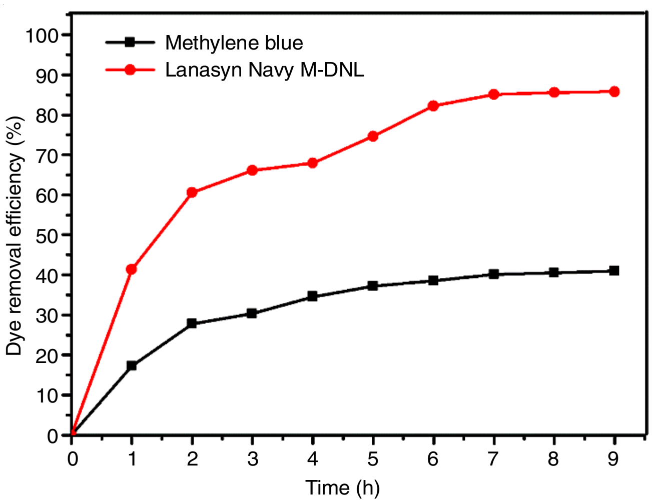

Fig. 2. Effects of contact time on dye removal efficiency (adsorbent concentration = 1 mg/L, dye concentration = 50 mg/L, volume = 50 mL, pH = 7)

Effect of pH: The pH of aqueous solution is another key parameter during the adsorption process, because it affects the surface properties of the adsorbents. In addition, the degree of ionization of dyes present in the aqueous solution is related to the pH solution, which in turn affects the degree of dye adsorption onto adsorbents. Fig. 3 shows the removal of methylene blue and Lanasyn Navy M-DNL dyes on the rSFB in solution having an initial pH varied between 4 and 10. Methylene blue dye removal efficiency of rSFB was increased with increasing pH. At pH 10, the removal efficiency for methylene blue was 72.95%, indicating that the adsorption of rSFB for cationic dye was favoured under alkaline conditions. In contrast, in the case of anionic dye (Lanasyn Navy M-DNL), the removal efficiency was high in acidic medium (90.73%) and decreased with the increase in pH of the solution.

The variation in sorption behaviour with different solution pH values can be explained on the basis of surface charge and property changes in the adsorbents. It is known that silk fibroin protein contains many hydrophobic amino acid residues and some charged amino acids (positively or negatively charged) that are produced by the ionization of carboxyl or amino groups

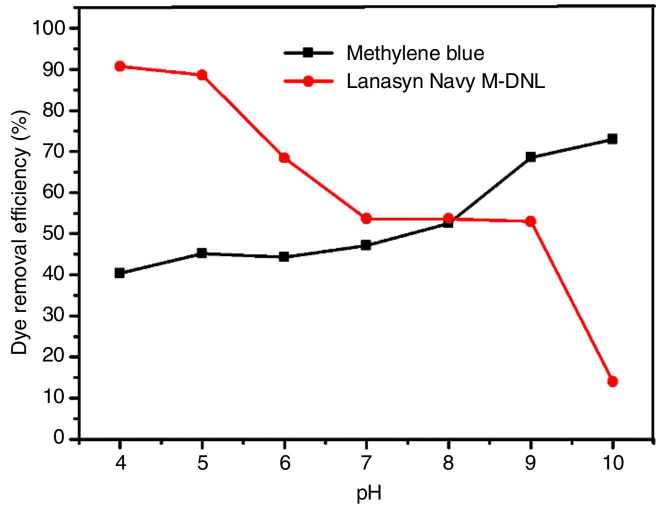

Fig. 3. Effects of pH on dye removal efficiency (adsorbent concentration = 1 mg/L, dye concentration = 50 mg/L, volume = 50 mL, contact time = 7 h)

in aqueous solution, which is dependent on the solution pH [14,15]. As a zwitterionic molecular, the ionization equilibrium of an amino acids in silk fibroin could occur with the pH variations [16].

When the solution pH was in acidic condition, the surface of rSFB exhibited positive charged, which would reveal the approach of anionic dye molecular (Lanasyn Navy M-DNL) to the surface of rSFB due to electrostatic attraction between adsorbent surfaces and dye molecular. On the other hand, when the solution pH was alkaline condition, more carboxyl groups of amino acid protonated with the pH increasing, resulting in a negatively charged surface of rSFB, thus more adsorbed cationic dye molecules (methylene blue) through electrostatic interaction. Therefore, the next batch experiments were controlled at pH 4 and 10 for Lanasyn Navy M-DNL and methylene blue, respectively.

Effect of adsorbent concentration: The effect of regenerated silk fibroin beads (rSFB) concentration in adsorption is presented in Fig. 4. The initial adsorptions for the dyes were rapid with the increasing adsorbent concentration after which change in contact time yielded no significant change in the adsorption. The initial rate of adsorption of the dyes may be explained by the fact that initially for adsorption, a large surface area and more adsorption sites were available, which slowed down later due to exhaustion of remaining surface sites [17] and the sorption process could be said to have attained equilibrium. A further increase in contact time did not increase the removal efficiency. Thus, 1 h (for Lanasyn Navy M-DNL) and 4 h (for methylene blue) with 5 g/mL rSFB were enough for the removal of 50 mg/L dye solutions.

Isotherm studies: The interaction between the adsorbent and adsorbate of any system can be described by adsorption isotherm parameters [18]. This is because these parameters present significant information on the surface properties, the adsorption mechanisms and efficiencies of the adsorbent [18]. Langmuir and Freundlich′s equations are the most suitable surface adsorption equations in a single solute system (Fig. 5).

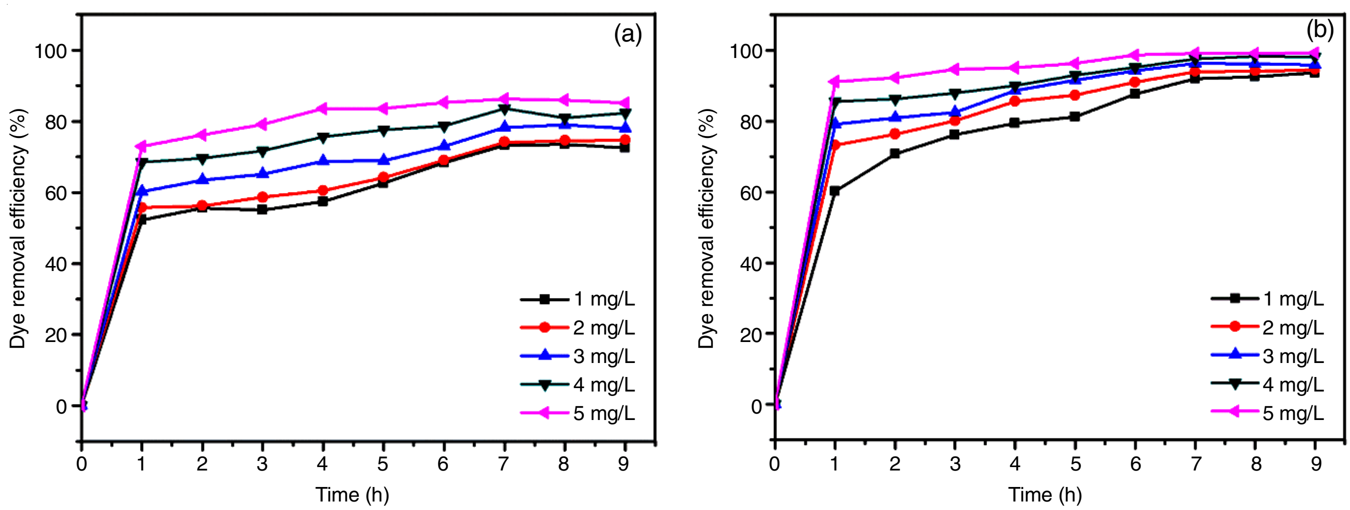

Fig. 4. Effects of adsorbent concentration on (a) methylene blue and (b) Lanasyn Navy M-DNL dye removal efficiency (dye concentration = 50 mg/L, volume = 50 mL, contact time = 7 h, pH = 10 (methylene blue) and pH = 4 (Lanasyn Navy M-DNL))

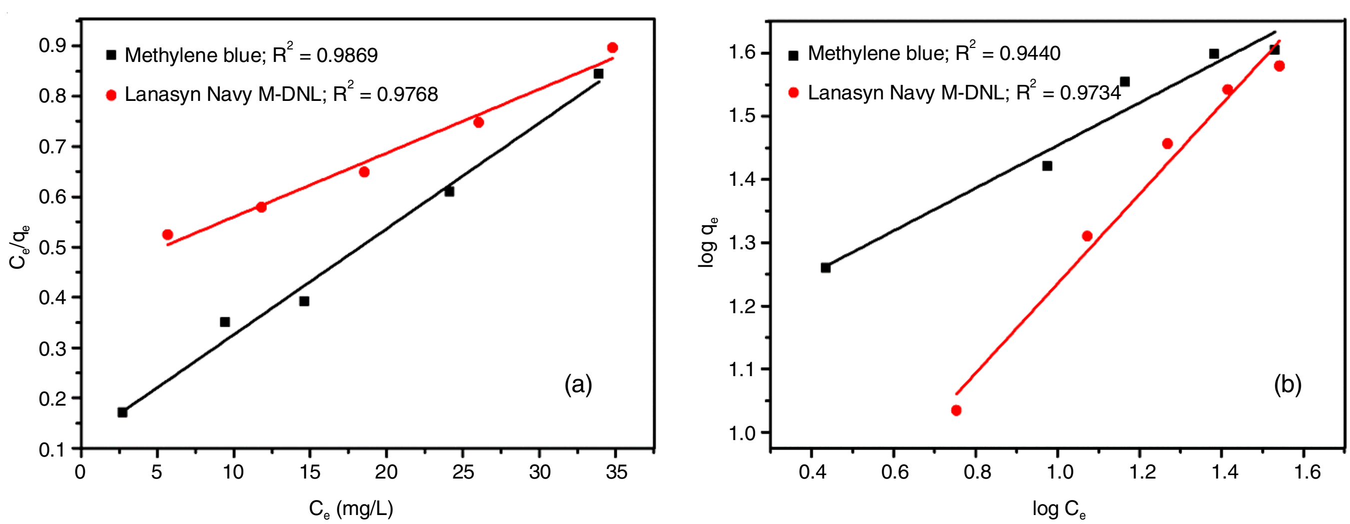

Fig. 5. (a) Langmuir isotherm and (b) Freundlich isotherm plots for adsorption of dye onto rSFB

The validity of the isotherm models can be determined by their linearized plots. The regression coefficient (R^2) is used as a criterion to compare the fitness of each model.

The linear form of the Langmuir isotherm can be expressed as follows:

$$\frac{C_e}{q_e} = \left(\frac{1}{bq_m}\right) + \left(\frac{C_e}{q_m}\right)$$

where q_e (mg/g) is the amount of dye adsorbed per unit mass of adsorbent and C_e (mg/L) is the equilibrium dye concentration, q_m (mg/g) is the maximum monolayer coverage capacity of adsorbent; and b (L/mg) is the Langmuir adsorption constant. The favourability of the adsorption process can be estimated [19,20] by a dimensionless constant, R_L, which is expressed as:

$$R_L = \frac{1}{1 + bC_o}$$

For the initial concentration of dye solution ranging from 10 mg/L to 50 mg/L, the values of R_L varied from 0.0991 to 0.7734. This range is between 0-1, which indicate that the adsorption process is favourable.

The Freundlich isotherm is given by the following equation:

$$\log q_e = \log K_f + \frac{1}{n}\log C_e$$

where K_f is a constant to describe the sorption capacity, and n is a constant representing the favourability of the sorption system [19,21]. In this work, the value of n is larger than 1 (n = 2.9568 and 1.4100 for methylene blue and Lanasyn Navy M-DNL, respectively), which indicate favourable adsorption of dyes onto rSFB at optimized conditions [22].

Various isotherm constants are summarized in Table-1. According to the linear regression coefficients (R^2), Langmuir isotherm model best described the adsorption data, which implied that monolayer adsorption with uniform adsorption energies may occur in the dye-rSFB system and can be described in terms of chemisorption as the formation of ionic bonds between adsorbent and adsorbate.

TABLE-1
ISOTHERM PARAMETERS FOR THE ADSORPTION OF METHYLENE BLUE AND LANASYN NAVY M-DNL ONTO rSFB

Dye	Langmuir			Freundlich		
	q_m (mg/g)	b (L/mg)	R^2	K_f	1/n	R^2
Methylene blue	47.55	0.1818	0.9869	13.0557	0.3382	0.9440
Lanasyn Navy M-DNL	78.74	0.0293	0.9768	3.3574	0.7092	0.9734

The calculated maximum monolayer coverage capacities from Langmuir isotherm were 47.55 and 78.74 mg/g for methylene blue and Lanasyn Navy M-DNL, respectively. A comparison of maximum adsorption capacities of dyes for some other reported adsorbents is shown in Table-2. It can be seen that rSFB has comparable adsorption capacity and could be used as an effective adsorbent for dye removal from waste-water.

Adsorption kinetics: In order to study the adsorption mechanism and possible rate limiting steps, the pseudo-first-order and pseudo-second-order models were used to fit the experimental data with a different initial dye concentration at 50 mg/L. The fitness of the two kinetic models can be checked by the correlation coefficient (R^2).

The pseudo-first-order model is described as follows [30]:

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

where K_1 (1/min) is the rate constant of pseudo-first-order adsorption, q_t and q_e (mg/g) are the adsorbed amount of dye at any time and equilibrium state. This model, the rate of adsorption was assumed to be dependent on the difference between the q_t and q_e, which relates to the driving force for mass transfer of adsorbate to reach at surface of adsorbent.

The pseudo-second-order model is expressed as follows [31]:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e}$$

where K_2 (g/mg min) is the rate constant of pseudo-second-order adsorption. This model is based on the assumption that the rate-limiting step may be chemical sorption.

Figs. 6 and 7 show the plots of a pseudo first-order model and second-order model and the kinetic parameters are listed in Table-3. As shown in the figures, linear regression correlation coefficient (R2) for the pseudo second-order was much higher than that of the pseudo first-order, indicating better fitness using the pseudo second-order model. This implied that the extremal film diffusion may not be the rate-limiting step for methylene blue and Lanasyn Navy M-DNL dyes adsorption onto rSFB [32,33]. Moreover, the calculated adsorption capacity ($q_{e(cal)}$) obtained from the pseudo-second-order model was also closer to the experimental adsorption capacity ($q_{e(exp)}$). These results demonstrated that the kinetics experimental data were better fitted to the pseudo-second-order model, indicated that the adsorption process was likely to be a chemisorption process.

Desorption and regeneration of adsorbent: In practical applications, reuse of dyes and adsorbents can be achieved through desorption. Fig. 8 shows the desorption efficiency of methylene blue and Lanasyn Navy M-DNL within 7 h. The desorption efficiency of rSFB for methylene blue were decreased with increasing pH. Whereas, in the case of Lanasyn Navy M-DNL the desorption efficiency was low in acidic medium and increased with the increase in pH of the solution. Therefore, the regeneration experiments were controlled at pH 4 and 10 for methylene blue and Lanasyn Navy M-DNL, respectively.

As shown in Fig. 9, initial removal of rSFB for methylene blue and Lanasyn Navy M-DNL was 72.96 and 90.73%, respectively. With increased cyclic adsorption-desorption times, the removal was slightly decreased. The removal efficiency of rSFB for dyes remained above 50% after five consecutive adsorption/desorption cycles. It can be concluded that rSFB has a good

TABLE-2
DYE ADSORPTION CAPACITIES OF SEVERAL ADSORBENTS FROM AQUEOUS SOLUTIONS

Dyes	Adsorbents	Adsorption capacity (mg/g)	Ref.
Cationic dyes			
Basic Yellow 11	Alginate/sericin composite	5.89	[23]
Toluidine blue	Montmorillonite clay	33.00	[24]
Methylene blue	Regenerated silk fibroin beads	47.55	This study
Maxilon blue GRL	Coconut shell activated carbon	62.06	[25]
Basic Yellow 28	Smectite rich natural clay	76.92	[26]
Methylene blue	Potato peel	107.20	[27]
Rhodamine B	Modified bentonite	173.50	[28]
Methylene blue	Polyacid doped polyaniline	466.50	[29]
Anionic dyes			
Acid Red 336	Alginate/sericin composite	7.60	[23]
Acid Brown 75	Smectite rich natural clay	8.33	[26]
Congo red	Montmorillonite clay	9.00	[24]
Direct Yellow 12	Coconut shell activated carbon	13.78	[25]
Orange G	Potato peel	23.60	[27]
Lanasyn Navy M-DNL	Regenerated silk fibroin beads	78.74	This study
Acid red 1	Modified bentonite	157.40	[28]
Rose Bengal	Polyacid doped polyaniline	440.00	[29]

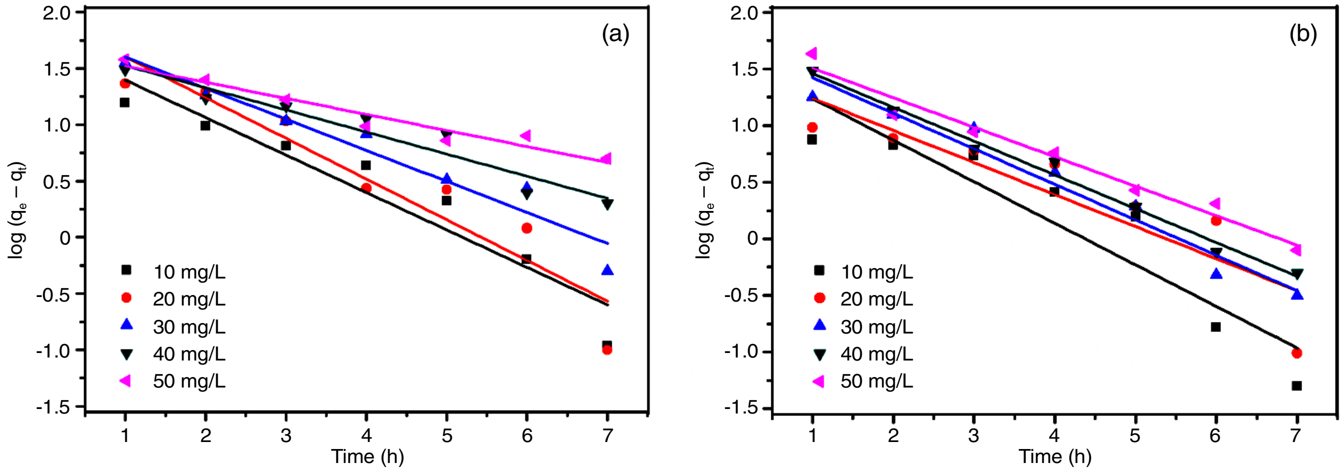

Fig. 6. Pseudo-first-order plots for adsorption of (a) methylene blue and (b) Lanasyn Navy M-DNL onto rSFB

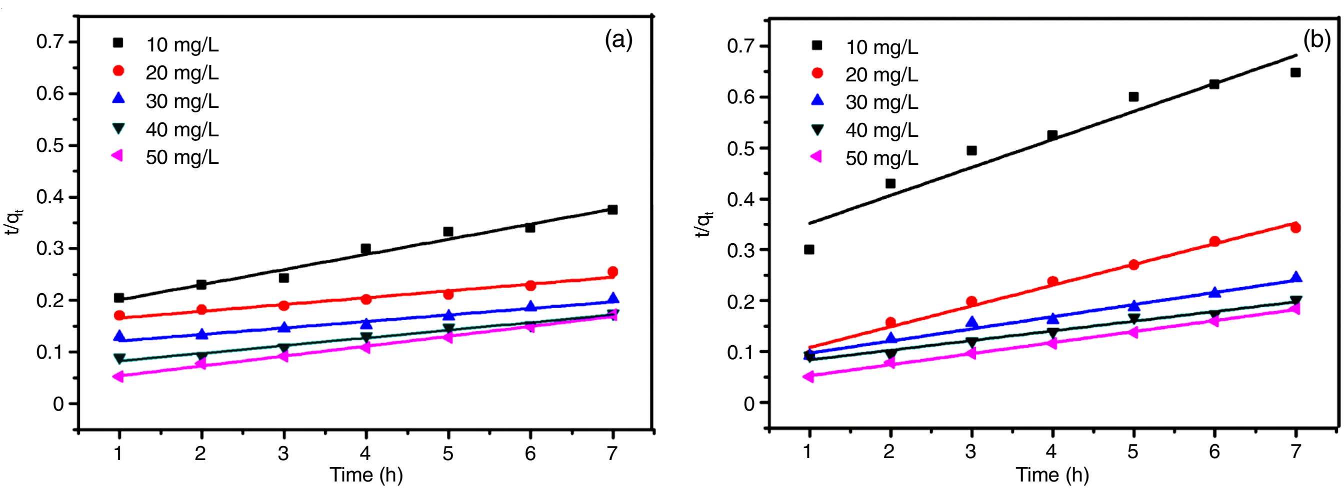

Fig. 7. Pseudo-second-order plots for adsorption of (a) methylene blue and (b) Lanasyn Navy M-DNL onto rSFB

TABLE-3
KINETIC PARAMETERS FOR THE ADSORPTION OF METHYLENE BLUE AND LANASYN NAVY M-DNL ONTO rSFB

Dye	C_o (mg/L)	q_e (exp) (mg/g)	Pseudo first-order			Pseudo second-order		
			K_1 (1/min)	q_e (cal) (mg/g)	R^2	K_2 (g/mg min)	q_e (cal) (mg/g)	R^2
Methylene blue	10	35.82	0.3282	45.98	0.8889	0.0050	34.12	0.9678
	20	52.53	0.4523	52.65	0.8741	0.0106	52.19	0.9442
	30	66.11	0.7664	53.62	0.9291	0.0033	66.98	0.9557
	40	76.25	0.6354	75.20	0.9191	0.0011	75.99	0.9684
	50	80.36	0.8330	92.17	0.9325	0.0015	79.37	0.9939
Lanasyn Navy M-DNL	10	16.81	0.6513	33.23	0.8381	0.0102	18.20	0.9159
	20	21.43	0.8450	40.20	0.7465	0.0246	24.54	0.9857
	30	38.85	0.7218	54.27	0.9510	0.0078	41.96	0.9803
	40	47.68	0.5999	58.40	0.9866	0.0151	46.19	0.9712
	50	48.99	0.6870	57.10	0.9659	0.0056	52.63	0.9971

reusability for removing cationic and anionic dyes in aqueous solution.

Conclusion

In this work, a new kind of natural biomaterial bead based on regenerated silk fibroin was prepared. The regenerated silk fibroin beads (rSFB) could be used to remove both cationic and anionic dyes from aqueous solutions. The results indicated that several factors such as contact time, pH and adsorbent dosage affect the adsorption process. Under optimal conditions, the adsorption capacities for methylene blue and Lanasyn Navy M-DNL dyes were 47.55 and 78.74 mg/g, respectively. In addition, the regeneration experiment revealed that rSFB might be feasible for practical applications. The rSFB as bioadsorbent has potential applications for the efficient adsorption and separation of dyes from wastewater.

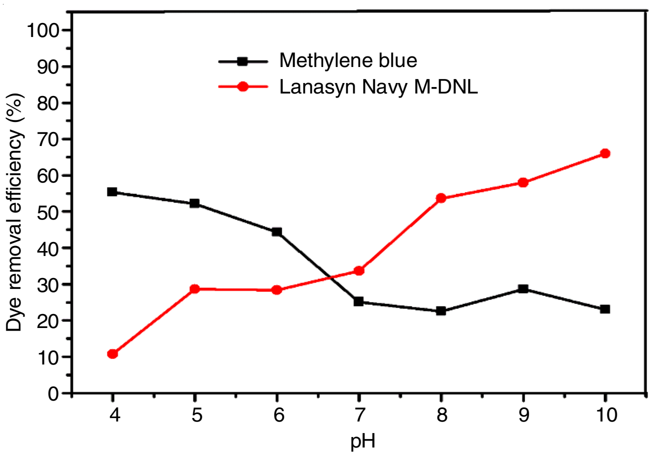

Fig. 8. Desorption efficiency at different pH (adsorbent concentration = 1 mg/L, dye concentration = 50 mg/L, volume = 50 mL, contact time = 7 h)

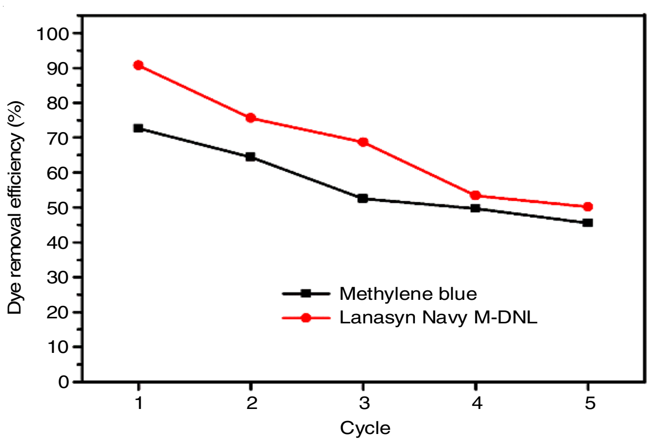

Fig. 9. Dye removal efficiency at different cycle times (adsorbent concentration = 1 mg/L, dye concentration = 50 mg/L, volume = 50 mL, contact time = 7 h, pH = 10 (methylene blue) and pH = 4 (Lanasyn Navy M-DNL))

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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