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Investigation on adsorptive removal of basic dye by seaweed-derived biosorbent: considering effects of sorbent dosage, ionic strength and agitation speed

Pei-Ling Tan^a, Ching-Lee Wong^b, Siew-Ling Hii^{a,*}

^aDepartment of Chemical Engineering, Faculty of Engineering and Science, Universiti Tunku Abdul Rahman, Jalan Genting Kelang, 53300 Setapak, Kuala Lumpur, Malaysia Tel. +60 3 41079802; Fax: +60 3 41079803; email: hiisl@utar.edu.my, yvonne_hii@ymail.com ^bSchool of BioSciences, Taylor's University, Taylor's Lakeside Campus, No. 1, Jalan Taylor's, 47500 Subang Jaya, Selangor Darul Ehsan, Malaysia

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ABSTRACT

Discharge of dyes from industries such as textile, cosmetic, paint and ink is one of the main concerns around the world nowadays. In the present study, locally available brown seaweed, *Sargassum binderi* was used to remove a basic dye – Basic Yellow 11 (BY11) – from aqueous solution. In batch sorption process, affecting parameters such as sorbent dosage, ionic strength and agitation rate were investigated for the optimum removal of BY11. The optimal sorption of BY11 (with more than 80% of uptake) was attained by contacting 1 g of dried *S. binderi* at 200 rpm in 100 mg/L of BY11 solution containing 500 mmol/L NaCl. By considering the effect of sorbent dosage, the adsorption mechanisms complied well with the Freundlich model ($R^2 = 0.94$) which suggested that the adsorption of BY11 could be attributed to the functional groups on the cell wall via the multiple layer adsorption process. Value of an important Freundlich constant (*n*) is 1.27, indicating a favourable adsorption process. The results of present study highlighted the practicability and potential of marine seaweeds as a low cost biosorbent in treating dyecontaminated wastewater.

Keywords: Seaweed; *Sargassum binderi*; Basic Yellow 11; Freundlich isotherm; Langmuir isotherm

1. Introduction

Colour, one of the prime pollutants which radically affect the ecosystem was getting great attention in wastewater treatment. Release of these untreated colours to the environment devastates the whole ecosystem and jeopardises human health as it consists of aromatic complexes which are resistant towards biodegradation. Conventional methods such as chemical precipitation, coagulation, filtration, oxidation and adsorption were used in wastewater mill in removal of colour compound prior to disposal [1]. Adsorption by activated carbon was found to be the best treatment method as compared to the others, however, due to economic constraint, its' applications on industrial scale are limited [2]. Various researches have been conducted on derivation of low cost biosorbent especially from agriculture waste to synthetic adsorbent media as a substitute of activated carbon in adsorption process [2–8].

Azo dyes, covers big range of dyes namely basic, acid, reactive, disperse, vat, metal, etc. was found to be the culprit towards environmental pollution [9]. Among

^{*}Corresponding author.

various types of dye available, basic dyes were found to be the brightest class of soluble dyes widely used by textile industry [10]. These dyes are stable due to the presence of aromatic and various functional groups which in turn render recalcitrant effect of the dyes [11]. Basic Yellow 11 (BY11) gives rise to high colour strength and was practically used in dyeing of polyacrylonitrile fibers [12]. Hence, textile waste containing of Basic Yellow 11 required pretreatment prior of discharge.

As reported by Hii et al. [13], seaweeds are potential biosorbent due to their special surface properties which enables them to adsorb different kinds of metallic and organic pollutants from solutions. Though the binding capability of seaweeds towards metal compounds has been reported extensively [14], a literature survey revealed that application of seaweeds in removal of dyes from solution is still remained scarce. There are few categories of seaweeds available: Phaeophyta (brown algae), Rhodophyta (red algae) and Chlorophyta (green algae). The cell wall of seaweeds possess some functional groups such as amino, carboxyl, sulphate, phosphate and imidazoles associated with polysaccharides, alginic acid and proteins which enable them to bind various pollutants [13]. In comparison, large amount of alginate compounds and followed by sulfated polysaccharides (fucoidons) dominated the components of Phaeophyta [14]. Alginic acid, an acidic polymer carboxylic compound contains ?-1,4 manuronic acid associated to ?-1,4 guluronic acid plays important role in removal of divalent cations followed by sulfonic acid of fucoidans which typically plays secondary role in metal binding process [15]. From our previous investigation, brown macroalgae (Sargassum binderi) was proven to have high sorption efficiency towards BY11 [16], with approximately 100% removal of BY11 recorded. The pH of dye solution, however, does not possess any significant influence on the removal of BY11 as the % uptake of BY11 and sorption capacity of *S*. binderi remained almost constant with the increasing of pH from 3 to 9 [16]. The present study aimed to further investigate the isotherm and kinetic of BY11 adsorption process, by taking into accounts the effect of using different dosage of S. binderi. Comparison of isotherm analysis among two different parameters (initial dye concentration and sorbent dosage) was reported in the present report. In addition, study was also conducted to evaluate the effects of agitation speed and ionic strength of dye solution towards the binding capabilities of *S. binderi*.

2. Materials and methods

2.1. Biosorbent and sorbate preparation

Sargassum binderi was collected from Cape Rachado, Port Dickson, Negeri Sembilan, Malaysia. The seaweed samples were washed and cleaned with distilled water to remove the salt particles, sand and epiphytes. Cleaned seaweeds were then dried in oven at 60 °C for 24 h. The dried seaweeds were pretreated with 0.2% v/v of formaldehyde in order to avoid organic leaching from seaweed during subsequent processes. Following that, treated seaweeds were rinsed with distilled water and dried in oven at 60 °C for 24 h before storage for further usage. In order to reduce the cost of operation, seaweed in intact form (without comminution) was used directly for adsorption after pretreatment and drying process.

Stock solution of BY11 (1000 mg/L) (Sigma Aldrich, C.I. No. 48055) was prepared by using distilled water. Working solution with various concentrations was prepared by diluting the stock solution with distilled water. Structure of BY11 is illustrated in Fig. 1.

2.2. Batch biosorption process

Biosorption process was carried out by agitating a fixed amount of treated seaweed with 150 mL synthetic solution of BY11 at 130 rpm 30 °C for 4 h, unless otherwise stated. A small volume of dye solutions were withdrawn at predetermined time interval and determination of dye solution concentration was conducted by using double beam UV/VIS spectrophotometer. Sorption capacity, q_e (mg/g) and percentage (%) of dye uptake were calculated by using equations as describe previously [16].

2.3. Effect of sorbent dosage, agitation rate and ionic strength

The effect of sorbent dosage was determined by contacting the dye solution with a series of biosorbent dosage in the range of 0.1–2.0 g at 130 rpm, 30 °C for 4 h. For the evaluation of the effect of agitation speed, seaweeds (0.5 g) were agitated at various agitation rates (50–250 rpm) at 30 °C for 4 h. In order to determine the effects of ionic strength, various concentrations of sodium chloride (NaCl) were added into dye solutions at 30 °C and the mixture were agitated for 4 h.

2.4. Isotherm and kinetic analysis

Both Langmuir (Eq. (1)) and Freundlich (Eq. (2)) models were employed for the analysis of the algal-dye



Fig. 1. Structure of BY11 (Sigma Aldrich, United States of America).

sorption process in the system with different sorbent dosage. Comparison of correlation coefficient values (R^2) derived from each respective models were used to examine the relationship between sorbed and aqueous concentration at equilibrium.

$$\frac{1}{q_e} = \frac{1}{Q_o} + \frac{1}{bQ_oC_e} \tag{1}$$

$$\ln q_e = \ln K_F + \left(\frac{1}{n}\right) \ln C_e \tag{2}$$

Linearized equation of pseudo-first-order kinetic model was presented in Eq. (3)

$$\log (q_e - q_t) = \log q_e - \frac{k_1}{2.303}t$$
(3)

Pseudo-second-order kinetic analysis was expressed as in Eq. (4)

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$$
(4)

2.5. Microscopy analysis

Scanning electron microscopy was conducted in analysing the surface structure of *S. binderi* before and after biosorption process at 20.0 KV and 500× magnification (Philips, XL30 ESEM).

3. Results and discussion

3.1. Characterization of biosorbent

By comparing with the surface structure of unmodified *S. binderi* (Fig. 2), occurrence of obvious protuberant on the surface of *S. binderi* after biosorption process (Fig. 3) indicated the possible sign of dye uptake by *S. binderi*.

3.2. Effect of sorbent dosage

Effect of sorbent dosage on removal of BY11 by *S. binderi* was conducted by varying sorbent dosage (0.1–0.2 g) while keeping other variables constant. The uptake of BY11 increased exponentially from 91.8% to 99.4%, with the increasing of sorbent dosage from 0.1 to 1.0 g (Fig. 4). This phenomenon might be due to the presence of more surface area in system with higher dosage of biosorbent, which contributed to the increment of available binding sites. Consequently, more dye compounds could be adsorbed [17–19]. However,



Fig. 2. SEM micrograph of unmodified *S. binderi* before the sorption process.



Fig. 3. SEM micrograph of S. binderi after dyed by BY11.



Fig. 4. Effect of sorbent dosage in removal of BY11 by *S. binderi*. Error bars indicate the mean ? standard deviation of triplicate. For data points without error bars, the errors were smaller than the size of the symbols.

as observed from Fig. 4, sorbent dosage of *S. binderi* above 1.0 g could only marginally raise the dye removal percentage. Thus, it is believed that the *S. binderi* had reached the saturation point of 1.0 g which considerably

satisfactory for sorption of BY11 with 99% of removal. The same behavior was also observed in removal of methylene blue by *Pyracantha coccinea* berries [20].

3.2.1. Isotherm analysis

Langmuir and Freundlich isotherm models were applied to investigate the overall sorption efficacy and to characterize the BY11 sorption process in the system using different dosage of *S. binderi*. Langmuir model assumes that adsorption occurs homogeneously and monolayer adsorption takes place within biosorbent [21], while Freundlich isotherm is an empirical model employed to describe the heterogeneous systems [22].

By comparing the parameter constants derived from both isotherm models with the values of constants reported previously (Table 1), the present study further proved that the adsorption of BY11 onto *S. binderi* could be a multilayer adsorption process. With higher R^2 value ($R^2 = 0.9370$) of Freundlich isotherm as compared with Langmuir isotherm ($R^2 = 0.7773$), the adsorption of BY11 onto *S. binderi* involves random distribution of sites which covered by more than one dye molecules. In addition, with the Freundlich adsorption indicator (*n* value) more than 1, the removal of BY11 by *S. binderi* could be confirmed to occur in a favourable sorption condition [23]. As shown in Table 1, discrepancy occurred between maximum adsorption capacity (Q_0) derived from the isotherm analysis of two different operating parameters (initial dye concentration and sorbent dosage), which might be due to the concentration of dye used. In the present study, the concentration of BY11 was fixed at 100 mg/L in the system using different sorbent dosage; while during previous study, the highest initial dye concentration applied was 5000 mg/L with 1 g of sorbent in the system [16].

3.2.2. Kinetic analysis

By considering the effect of sorbent dosage, the calculated q_e values $(q_{e,cal})$ of pseudo-second-order kinetic model compliance well with experimental q_e values $(q_{e'exp})$ which confirmed the suitability of this kinetic model to describe the uptake behaviour of BY11 by *S. binderi* (Table 2). Pseudo-second-order model assumed that BY11 uptake was controlled by chemical process (chemisorption) which involves valency forces through exchange or sharing of electrons [22].

3.3. Effect of agitation rate

From the present study, it is apparent that the increase of agitation speeds had no significant impact on the removal of BY11 by *S. binderi*. Regardless of

Table 1. Isotherm parameters for adsorption of BY11 onto *S. binderi*

1	1				
Models parameters	Langmuir		Freundlich		
	Dye concentration effect	Sorbent dosage effect	Dye concentration effect	Sorbent dosage effect	
Reference	Hii et al. [16]	This study	Hii et al. [16]	This study	
$Q_0 (mg/g)$	192.30	43.29	-	-	
<i>b</i> (L/mg)	0.2488	1.974	-	-	
$K_{\rm F} ({\rm mg/g.}({\rm L/mg})^{1/n})$	_	_	32.46	39.40	
N	_	_	1.968	1.268	
R^2	0.927	0.777	0.996	0.937	

Table 2

Comparison of pseudo-first-order and pseudo-second-order model in removal of BY11 by seaweed

Sorbent dosage	$\frac{q_{e'exp}}{(mg/g)}$	Pseudo-first order kinetic model		Pseudo-second order kinetic model			
(g)		$k_1 ({ m min}^{-1})$	$q_{e'cal}$ (mg/g)	R^2	k ₂ (10 ⁻⁵) (g/mg.min)	$q_{e'cal}$ (mg/g)	R^2
0.10	143.39	0.0311	104.59	0.9374	3.824×10^{-4}	156.25	0.9976
0.50	29.78	0.0212	2.67	0.4461	2.538×10^{-2}	30.03	0.9999
1.00	14.97	0.0207	2.71	0.3704	2.050×10^{-1}	14.99	1.0000
1.50	9.97	0.0018	6.32	0.0044	7.217×10^{-2}	9.64	0.9996
2.00	7.42	0.0083	2.62	0.1874	1.987×10^{-1}	7.30	0.9998

agitation speed applied, approximately 98% of dye uptake was achieved when all the systems were at their respective equilibrium stage (Table 3). The equilibrium stage was achieved when the adsorption rate of BY11 by S. dC_L *binderi* remained constant, i.e., $\frac{d}{dt} = 0$. In the present study, the systems with higher agitation speed required shorter period of time to reach equilibrium stage (Table 3). These phenomena is due to the fact that by increasing the agitation rate, film resistance to mass transfer which surrounds the sorbent particles could be reduced [24]. In addition, turbulence effect in the solution is induced by increasing agitation speed which consequently reduces the thickness of boundary layer [25]. Hence, both conditions enhance the mixing effect and lead to the increment in sorption of dye compounds by adsorbent.

3.4. Effect of ionic strength

Generally, wastewater from textile industries is with high concentrations of salt compounds which contribute to high ionic strength [23]. The ionic strength of solution affects both electrostatic and non-electrostatic interactions between sorbent and sorbate, thus, it play an important role in adsorption of contaminants from solution [23]. Fig. 5 shows the sorption capacity of *S. binderi* in removal of BY11 with the presence of series concentrations of NaCl. Sorption capacity of S. binderi reduced from 29.1 to 25.4 mg/g with increasing of NaCl concentration from 10 to 500 mmol/L. The undesirable results indicated that ionic compounds of NaCl were adsorbed by S. binderi, in preference to molecules of BY11. These phenomena might be attributed by competition of available binding sites between positively charged dye compounds and ionic compounds [26,27]. Same phenomena were observed in removal of malachite green and methylene blue by rice bran and wheat bran [27] as well as removal of yellow RL by Rhizopus arrhizus [28].

Table 4

Comparison of adsorption capacities of various adsorbents for basic yellow dyes

Table 3		
Effect of agitation rate on removal of BY11 by	, S.	binderi

Agitation rate (rpm)	Time to reach equilibrium stage ^a (min)	% of removal (± s.d.) ^b
50	90	98.1 ± 0.03
100	60	98.5 ± 0.03
150	45	99.0 ± 0.02
200	45	98.5 ± 0.04
250	30	98.8 ± 0.02

^aEquilibrium stage is achieved when $\frac{dC_L}{dL} = 0$.

^bs.d. = standard deviation of triplicate. $\frac{dt}{dt}$



Fig. 5. Sorption capacity of *S. binderi* in solution of BY11 mixed with various concentration of NaCl. Error bars indicate the mean ? standard deviation of triplicate. For data points without error bars, the errors were smaller than the size of the symbols.

3.5. Evaluation of S. binderi as an adsorbent

Table 4 lists the maximum adsorption capacity (Q_0) values of different adsorbents used for basic yellow dye removal. By comparison of the results obtained in this study with those reported previously, biomass of

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Adsorbent	$Q_o (\mathrm{mg/g})$	References	
Clinoptilolite	52.9	Yener et al. [29]	
Amberlite	11.		
Caulerpa scalpelliformis			
27.0	Aravindhan et al. [30]		
Bentonite	333.3	Turabik [31]	
Boron waste	75	Olgun and Atar [32]	
Mesoporous adsorbents	169–203	Wu et al. [33]	
Sargassum binderi	43.3	This study	
	Adsorbent Clinoptilolite Amberlite <i>Caulerpa scalpelliformis</i> 27.0 Bentonite Boron waste Mesoporous adsorbents <i>Sargassum binderi</i>	Adsorbent Q_o (mg/g)Clinoptilolite52.9Amberlite11.Caulerpa scalpelliformis27.0Bentonite333.3Boron waste75Mesoporous adsorbents169–203Sargassum binderi43.3	

S. binderi presented a comparable adsorption capacity with most adsorbent used for removal of basic yellow dye. In view of the availability, the low cost and the interesting adsorption capacity of *S. binderi*, its utilisation for removal of basic yellow dye from aqueous solution is very promising.

4. Conclusion

Present study shows that % uptake of BY11 increased with increasing of *S. binderi* dosage and decreased with increasing of ionic strength of dye solution. The present results also indicated that with higher agitation speed applied to the system, shorter equilibrium time is sufficient for comparable % uptake of BY11 by *S. binderi*. Isotherm analysis predicted that the maximum adsorption of BY11 by *S. binderi* occurs when saturated multiple layers of BY11 molecules are present on the seaweed surface. Based on the current results, it is expected that the *S. binderi* could be an alternative for wastewater management of basic dyes in reducing the concentration of dye wastes to an acceptable levels prior to discharging into the river body.

Symbols

- C_{L} concentration of BY11 in the liquid (mg/L)
- $\overline{C_e}$ concentration at equilibrium (mg/L)
- W weight of biosorbent used (g)
- V volume of working solution (L)
- q_e equilibrium sorption capacity (mg/g)
- q_t sorption capacity at time *t* (mg/g)
- Q_0 maximum amount of dye sorbed per unit weight of biomass (mg/g)
- B energy of adsorption (L/mg)
- $K_{\rm F}$ Freundlich constants indicating sorption capacity 1 (mg/g (L/mg)^{1/n}
- \overline{n} Freundlich constants indicating intensity
- k_i pseudo-first-order constant
- $k_2 \text{pseudo-second-order model constant, (g/mg min)}$

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