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Equilibrium and Kinetic Studies for Basic Yellow 11 Removal by *Sargassum binderi*

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Abstract: Dyes have been widely used in many industries such as textile, paint and also ink industry. Discharge of dyes from these industries need to undergo treatment prior of disposal to avoid ecosystem to be devastated. Upon this, *Sargassum binderi*, a species of brown seaweed was used as a low cost biosorbent to remove a basic dye, Basic Yellow 11 (BY11). Various parameters such as pH, initial concentration and sorbent dosage were conducted using batch sorption process. In this study, *S. binderi* exhibited good performance (almost 100% of BY11 removal) with 1 g of sorbent dosage in 100 mg L^{-1} of dye solution. pH of the solution, however, does not seem to have high influence on removal of BY11. Langmuir, Freundlich and Temkin isotherm models were used to study sorption behaviors of *S. binderi* at equilibrium stage. From the analysis, the sorption of BY11 onto the *S. binderi* was found obeyed Freundlich model with coefficient correlation (R^2) value of 0.9959 which indicates heterogeneous layers of sorption. The Freundlich constant, K_F and n values obtained were 32.46 mg g^{-1} (L $mg^{-1})^{1/n}$ and 1.968, respectively. Value of Freundlich component, n, was more than 1 indicates favourable adsorption process of this system. The kinetic sorption of BY11 using *S. binderi* were analyzed by using pseudo-first-order, pseudo-second-order and intraparticular diffusion model. The sorption process followed pseudo-second-order kinetic which involved chemisorption. Calculated q_e values from pseudo-second-order kinetic model were found fitted well with the experimental q_e values. Each R^2 values from linear regression line of pseudo-second-order kinetic model were more than 0.99. In this study, *S. binderi* was proven to have high sorption efficiency towards BY11 and thus, it can be categorized as one of the promising biosorbent to be applied in treating the effluent discharge from industries.

Key words: *Sargassum binderi*, basic yellow 11, sorption capacity, equilibrium isotherm, kinetic studies

INTRODUCTION

Dyes have been widely used in many industries. These industries will therefore release coloured effluent which requires treatment process to remove intense colour prior of disposal. Upon this, colour was the first pollutant to be identified by using naked eyes. Among all dyes, basic dyes or also known as aniline dyes were widely used in textile industries due to its strong tinctorial strength and brightness (http://www.jagson.com/basic_dyes.htm.). Basic dyes were not light sensitive and will not fade under direct exposure of light (http://www.jagson.com/basic_dyes.htm.). Thus, this characteristic will reduce the visibility of light into the water even it is in low concentration. Reduction of light penetration will then lead to low photosynthesis rate and thus, caused the whole ecosystem in the water to be severely impaired (Turabik, 2008).

Various researches have been carried out in search of effective method to remove colour compound from effluent. Several conventional methods were applied in removing dye component from dye solutions such as flocculation, chemical precipitation, filtration, oxidation, coagulation, adsorption and membrane separation (Aksu *et al*., 2008). Adsorption by conventional activated carbon was found to be the most effective method. In this adsorption process, dye compound was conveyed from solutions to solid form, reducing the volume of effluent to the minimum and subsequently saturated adsorbent can be either regenerated or avoided from direct contact with the environment (Lima *et al*., 2008). However, usage of activated carbon was expensive which limits its application to large scale of wastewater treatment (Lima *et al*., 2008). Hence, alternative adsorbent which is low in cost and with high removal capability is in great interest of researchers around the world. The ability of dye removal by low cost agricultural waste such as beer brewery waste (Tsai *et al*., 2008), *Hevea brasiliensis* seed coat (Hameed *et al*., 2008a), banana stalk waste (Hameed *et al*., 2008b), coconut bunch waste

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(Hameed *et al*., 2008c), coconut-husk (Jain and Shrivastava, 2008) and rice hull (Ong *et al*., 2007) were conducted by various researchers and it was proven that these agricultural wastes consist of promising potential in the removal of various dyes.

Sargassum binderi, a species of brown seaweed which was found to be abundant, especially from coastal area around Malaysia and with high regeneration rate was used as low cost biosorbent in this study. Brown seaweeds consist of high alginic acid content which contributes to the binding capability towards dye compound in solution (Davis *et al*., 2003).

MATERIALS AND METHOD

Biosorbent preparation: *Sargassum binderi* was collected from coastal area in Port Dickson, Seremban, Malaysia. Harvested seaweed was then washed under running tap water to remove solid particles on the surface of the seaweed. Cleaned seaweed was then washed with deionized water several times before drying at 80°C for 24 h. Chemical modification of seaweed was then carried out by contacting 10 g L^{-1} of dried seaweed with 0.2% formaldehyde for 24 h at 30°C and the mixture were agitated at 150 rpm. The objective of this treatment process was to avoid organic leaching which is a common phenomenon observed in marine algae. Organic leaching will then lead to secondary pollution which might devastate the biosorption process in water and wastewater treatment (Chen and Yang, 2005). Treated seaweed was then washed with deionized water several times to remove the residues of formaldehyde. The cleaned seaweed was dried in oven at 60°C for 24 h.

Sorbate: Synthetic dye solution of Basic Yellow 11 (BY11) (Sigma Aldrich, United States of America) was used in this study. Stock solution with concentration of 1000 mg L^{-1} was prepared without further purification. Any of the solutions used in this adsorption experiment were prepared from this stock solution to the desired concentrations by successive dilution. The properties and structures of BY11 is listed and shown in Fig. 1. A calibration curve was constructed with concentration ranging from 1 to 1000 mg L^{-1}. The concentration of BY11 in the solution were measured by using double beam UV/VIS spectrophotometer (Perkin Elmer, United States of America) at the maximum wavelength of 413 nm.

Batch biosorption process: Batch biosorption process was conducted by contacting 1 g of treated sorbent (unless otherwise stated) with a fixed volume of sorbate (150 mL) and agitated at 130 rpm, 30°C for 4 h. The pH of solution and the concentration of dyes were varied according to experiments. A fix amount of sorbate was collected at predetermined time interval for determination of dye residual. Calculation of sorption capacity, q_e (mg g^{-1}) was conducted based on the Eq. 1:

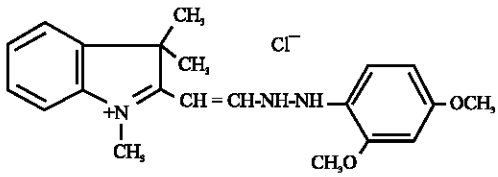

Fig. 1: Structure of BY11 (Sigma Aldrich, United States of America)

$$qe = \frac{(C_o - C_e)V}{W} \quad (1)$$

where, C_0 and C_e indicates initial concentration (mg L^{-1}) and concentration of sorbate at equilibrium (mg L^{-1}), respectively. V is the volume of sorbate (L) and W is the weight of sorbent (g).

Percentage of BY11 uptake (% uptake) by seaweed was calculated using Eq. 2:

$$\text{Uptake (\%)} = \frac{C_o - C_e}{C_o} \times 100\% \quad (2)$$

Effect of pH: A series of working solutions with different pH values were prepared from stock solution. pH of these working solutions were altered by using 1 M NaOH and 1 M HCl from the range of pH 2 to pH 9.

Effect of Initial dye concentration and contact time: Series of different dye concentrations range from 100 to 1000 mg L^{-1} were prepared and batch biosorption was conducted by contacting 1 g of seaweed with 150 mL of dye solution.

Effect of sorbent dosage: Effect of sorbent dosage was determined by agitate various amount of sorbent dosage (0.1, 0.5, 1.0, 1.5 and 2.0 g) with 150 mL of sorbate (100 mg L^{-1}) at 130 rpm, 30°C.

Equilibrium and kinetic studies: Equilibrium isotherms were determined by using initial concentration parameter. Langmuir, Freundlich and Temkin isotherm were applied for removal of BY11 by *S. binderi*. Determination of isotherm applied by *S. binderi* in this BY11 was conducted by comparing the correlation coefficient, R^2 value for each respective isotherm.

Pseudo-first-order, pseudo-second-order and intraparticular diffusion model were applied to the system by pH studies. R^2 values and comparison of calculated sorption capacity, q_e and experimental q_e verify the application kinetics in uptake of dye compound by *S. binderi*. Equations applied in determination of isotherm and kinetics studies were shown in discussion.

RESULTS AND DISCUSSION

Effect of pH: pH was found to be an important parameter which affect significantly on adsorption process especially for cationic dye adsorption (Wang *et al*., 2008). Figure 2 shows the removal of BY11 by *S. binderi* in pH range from 2 to 9. Sorption capacity, q_e of *S. binderi* increased from 29.14 mg g^{-1} in pH 2 to 29.71 mg g^{-1} in pH 3 and level off thereafter. Cationic dyes (basic dyes) possessed positively charge groups which play important role in binding of with surface charge of sorbent (Wang *et al*., 2008). These surface charges were greatly influenced by pH of the solution (Wang *et al*., 2008). This phenomenon can be explained by the presence of excessive H^+ ions which competes with positively charged dye compounds, hence, inhibits binding of dyes on the surface of biosorbent (Aravindhan *et al*., 2007). Similar outcome were reported by adsorption of methylene blue by pineapple stem (Hameed *et al*., 2009). The % uptake of dye and sorption capacity of seaweed remained almost constant with the increasing of pH from 3 to 9 (Fig. 2). Therefore, pH does not have significant influence towards removal of BY11 by *S. binderi*.

Brown seaweed consist high content of alginic acid, followed by sulfonic acid or fucoidon (Aravindhan *et al*., 2007). Carboxyl groups in alginic acid contribute to the binding of dye compounds, while sulfonic acid which usually plays secondary role bound effectively in adsorption at low pH (Davis *et al*., 2003). Although, removal of BY11 was the lowest at pH 2, the q_e value obtained from pH 2 was not significantly low as compared to q_e values of pH 3 to pH 9. Hence, the original pH of BY11 (approximate pH 5.41) was used for subsequent experiments.

Effect of initial concentration and contact time: As shown in Fig. 3, percentage of BY11 uptake decreased from 99.29 to 81.28, while the q_e value increased from 29.79 to 1219.17 mg g^{-1}. Higher chances of collision between dye compound and seaweed occurred with increasing of initial concentrations, thus enhance the dye absorbed by *S. binderi*. Similar phenomena was observed in removal of basic dye by apricot stone activate carbon (Demirbas *et al*., 2008), pumpkin seed hull

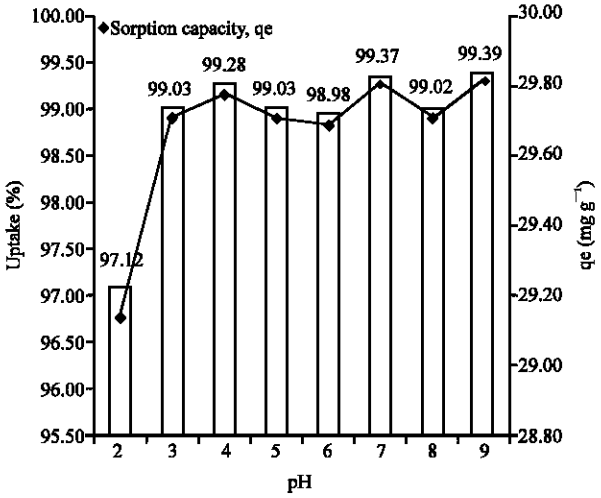

Fig. 2: Effect of pH in uptake of BY11 by *S. binderi*

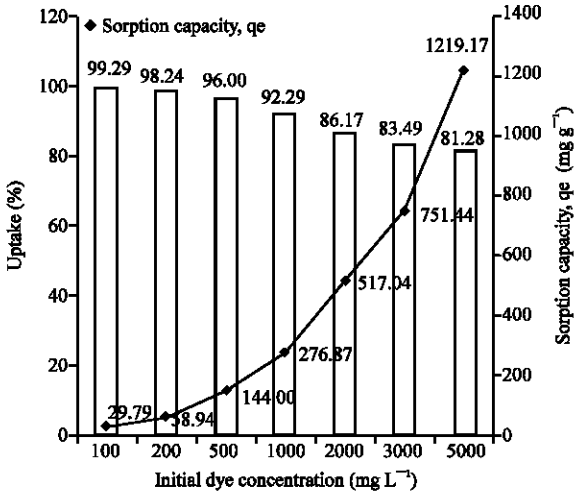

Fig. 3: Effect of initial concentration in removal of BY11 by *S. binderi*

(Hameed and El-Khaiary, 2008) and jute fiber carbon (Senthilkumaar *et al*., 2005). Figure 4 showed the time profile of sorption capacity of *S. binderi* in solution containing different dye concentration. Duration of time required for the system to achieve equilibrium increased with the increasing of initial concentrations from 60 to 180 min for initial concentration of 100 to 1000 mg L^{-1}, respectively. Uptake of BY11 by *S. binderi* was rapid in the beginning of adsorption process. The process were then gradually decreased and become constant when the system reaches equilibrium. The occurrence of this trend might be due to the rapid uptake of dye compound on the surface of the adsorbent until it reaches saturation level and lastly dye compounds will gradually diffuse into the porous structure of the adsorbent, thus contributes to

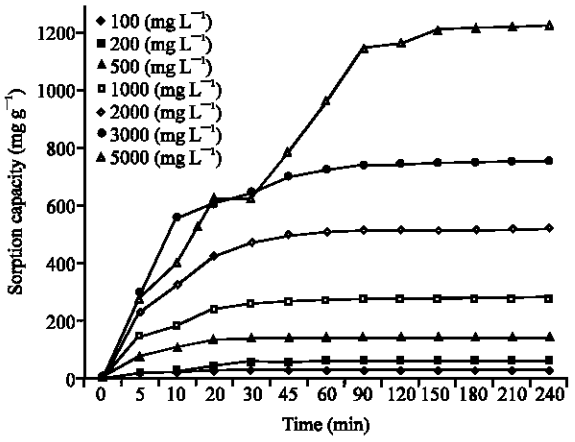

Fig. 4: Sorption capacity for removal of BY11 by *S. binderi* in various initial concentrations

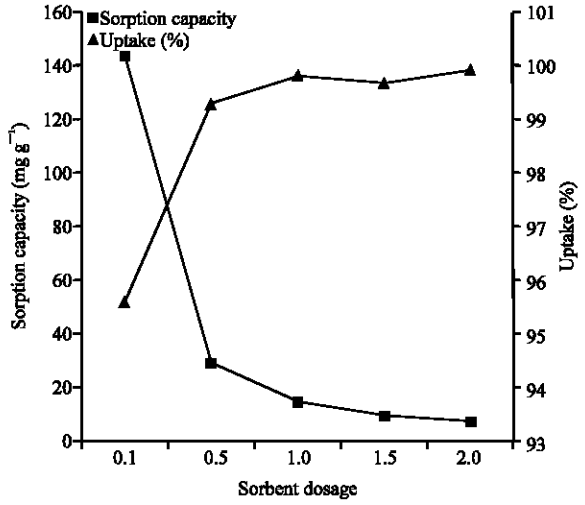

Fig. 5: Effect of sorbent dosage in removal of BY11 by *S. binderi*

longer contact time in order for the system to reach equilibrium (Senthilkumaar *et al*., 2005).

Effect of sorbent dosage: Effect of sorbent dosage on removal of BY11 by *S. binderi* was investigated by varying amount of sorbent dosage while other condition such as initial concentration (100 mg L^{-1}), temperature (30°C) and agitation speed (130 rpm) were kept constant. The uptake of BY11 removal by *S. binderi* increased from 91.78 to 99.37% with increasing of sorbent dosage. On the other hand, sorption capacity declined from 137.67 to 7.45 mg g^{-1}, as shown in Fig. 5. The increasing trend as observed in uptake of BY11 with the sorbent dosage might be attributed to the increased of surface area and presence of additional available binding sites (Ozer *et al*., 2007). However, this was inversely proportional to the sorption capacity of *S. binderi*. Increasing of sorbent dosage might lead to the overlapping of surface area due to the restricted area made available and thus, lead to reduction of effective surface area which plays important role in uptake of dyes (Garg *et al*., 2004).

Isotherm analysis: Langmuir isotherm model was the first isotherm model derived. Langmuir isotherm model assume that only homogenous adsorbent surface exist which is probably reasonable for the assumption in gas adsorption process (Droste, 1997). At equilibrium, saturation point has been achieved with no further adsorption process to be taken place, therefore, it is assumed that only one molecule will occupied one single site. Linearised form of Langmuir equation was as follow (Eq. 3):

$$\frac{1}{q_e} = \frac{1}{Q_o} + \frac{1}{bQ_oC_e} \quad (3)$$

where, Q_o and b indicates Langmuir constant which can determined from the linear plot of $1/q_e$ versus $1/C_e$. The q_e is the sorption capacity of *S. binderi* (mg g^{-1}) while C_e is the equilibrium concentration (mg L^{-1}).

Crucial characteristic of Langmuir adsorption model was expressed as dimensionless separation factor, R_L which can be derived from Eq. 4:

$$R_L = \frac{1}{(1+bC_o)} \quad (4)$$

where, b is the Langmuir adsorption constant (L mg^{-1}) and C_o is the highest concentration of solution. R_L value indicates the favourable condition of adsorption process with the status of adsorption to be favourable at $0 < R_L < 1$, unfavourable if $R_L > 1$ and lastly linear or in other words irreversible if $R_L = 1$.

Another popular isotherm model applied to the adsorption process was Freundlich adsorption isotherm. Freundlich model suggested that sorption process involved heterogenous sorption with different classes of adsorption sites (Aravindhan *et al*., 2007). Linearized Freundlich equation was shown in Eq. 5:

$$\ln q_e = \ln K_F + \left(\frac{1}{n}\right)\ln C_e \quad (5)$$

where, K_F (mg g^{-1} (L $mg^{-1})^{1/n}$) and n were Freundlich constants derived from the linear slope of ln q_e versus ln C_e. Freundlich magnitude of exponent, 1/n indicates favourable condition of adsorption with value of $n > 1$.

Temkin isotherm equation was based on the assumption where sorption energy decreased linearly as

Table 1: Comparison of Langmuir, Freundlich and Temkin isotherm constants for removal of BY11 by *S. binderi*

Models	Parameter values
Langmuir isotherm	
Q_o (mg g^{-1})	192.3077
b (L mg^{-1})	0.2488
R^2	0.9266
Freundlich isotherm	
K_F (mg g^{-1} (L $mg^{-1})^{1/n}$	32.46
n	1.968
R^2	0.9959
Temkin isotherm equation	
A (L g^{-1})	0.4385
B	141.70
R^2	0.7579

Table 2: R_L values from Langmuir isotherm model in different initial dye concentrations

Initial dye concentrations (mg L^{-1})	R_L (10^{-3})
100	38.60
200	19.70
500	7.97
1000	4.00
2000	2.01
3000	1.34
5000	0.80

the coverage of adsorbent decreased (Hameed and Duad, 2008). Temkin model assumed that adsorption take place through the characterization of uniform distribution in binding energies up to a certain level. Equation of Temkin isotherm was as Eq. 6:

$$q_e = B \ln A + B \ln C_e \quad (6)$$

Intercept and slope of linear plot from q_e versus C_e was used to calculate the value of A and b constant whereby B = RT/b. b, indicates Temkin constant which is related to the heat of sorption (J mol^{-1}), while A is the Temkin isotherm constant (L g^{-1}), R was the gas constant with the value of 8.314 (J mol^{-1} K) and lastly T was the absolute temperature with the unit of Kelvin.

Isotherm constants obtained from each equation were presented in Table 1. From the correlation coefficient obtained (R^2), the sorption process was found fitted well to Freundlich isotherm with R^2 values of 0.9959. Freundlich constant, K_F and n was 32.46 (mg g^{-1} (L $mg^{-1})^{1/n}$) and 1.968, respectively. The R^2 value of Langmuir isotherm was 0.9266 and it might indicate that homogeneous surface adsorption occurred but in a minor way. R_L values obtained from each respective initial dye concentrations were tabulated in Table 2. Each respective concentrations show favourable occurrence of monolayer adsorption with R_L values in between 0 and 1 (Hameed, 2008). On the other hand, this system does not obey Temkin isotherm equation at all time with the least value of R^2 (0.7579). Therefore, adsorption of BY11 by *S. binderi* occurred heterogeneously in majority, while homogenous process happened to be in minor.

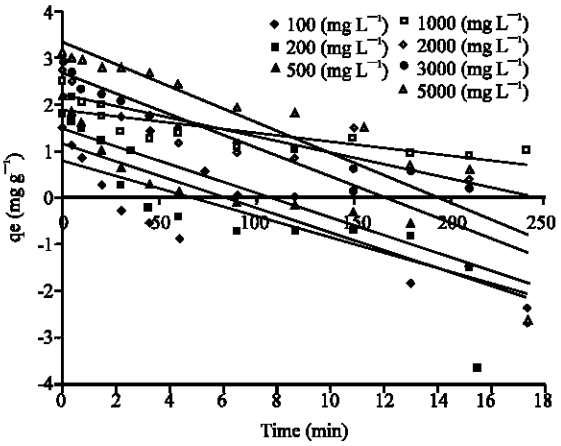

Fig. 6: Pseudo-first-order kinetics for removal of BY11 by *S. binderi*

Kinetic studies: The behavior of adsorption process were studied by using three kinetic models which comprises of pseudo-first-order, pseudo-second-order and intraparticular model. The correlation coefficient, R^2 values was used to determine the appropriateness of kinetic models that best fitted to the adsorption system.

Pseudo-first-order was theorized by Lagergran (Liu and Liu, 2008) with Eq. 7:

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (7)$$

which was further integrated to:

$$\log (q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (8)$$

where, q_t indicates the sorption capacity at time t (mg g^{-1}), q_e is the sorption capacity at equilibrium (mg g^{-1}) and k_1 is the constant. Linear plot of log (q_e-q_t) versus t (Fig. 6) was used in determination of k_1 constant (min^{-1}) and R^2 values which indicates the best fitted kinetic model.

Pseudo-second-order kinetics which was found to be the famous kinetic model as compared to the others was expressed in Eq. 9:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (9)$$

where, k_2 (g mg.min) indicates pseudo-second-order kinetic constant which was derived from linear plot of t/q_t versus t (Fig. 7).

R^2 and calculated q_e values obtained from both kinetic models were tabulated in Table 3. From the results, calculated q_e values derived from pseudo-second-order kinetic models was found to be compliance well to

Table 3: Comparison of R^2 values and constant of Pseudo-first and Pseudo-second-order in removal of BY11 by *S. binderi*

		Pseudo-first order kinetic model			Pseudo-second order kinetic model		
C_o (mg L^{-1})	q_e, exp (mg g^{-1})	k_1 (min^{-1})	q_e, cal (mg g^{-1})	R^2	k_2 (10^{-5}) (g mg^{-1} min)	q_e,cal (mg g^{-1})	R^2
100	29.79	0.0269	5.80	0.6522	1331.00	30.03	0.9992
200	58.94	0.0313	14.23	0.7741	252.80	61.73	0.9981
500	144.00	0.0313	28.37	0.8493	304.20	147.06	0.9999
1000	276.87	0.0113	73.43	0.6336	83.33	285.71	0.9996
2000	517.04	0.0207	155.38	0.7536	39.67	526.32	0.9990
3000	751.44	0.0368	458.78	0.8367	24.85	769.23	0.9998
5000	1219.17	0.0394	2138.45	0.8178	26.49	1428.57	0.9956

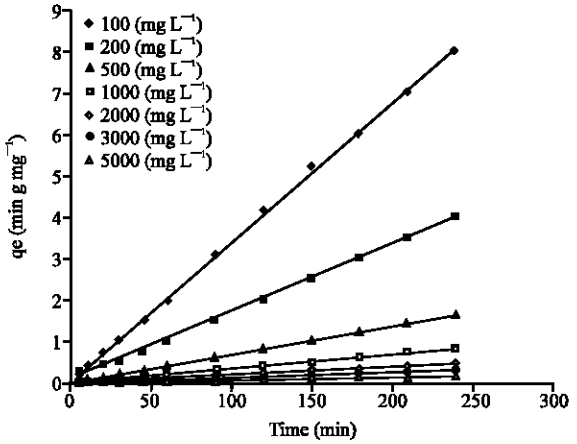

Fig. 7: Pseudo-second-order kinetics for removal of BY11 by *S. binderi*

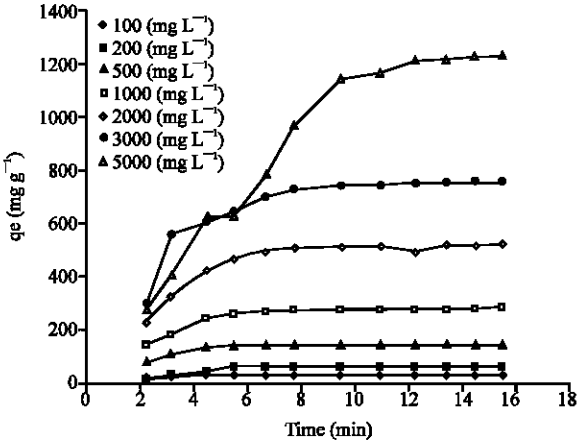

Fig. 8: Intraparticular diffusion model in adsorption of BY11 by *S. binderi*

experimental q_e values as compared to pseudo-first-order kinetic model. R^2 values obtained from pseudo-second-order kinetic model were more than 0.99 for all concentrations. Kinetic constants decreased with the increasing of initial concentrations. Pseudo-second-order kinetic model predicts the overall adsorption mechanism that occurs during the adsorption process (Fan *et al*., 2008). Since, pseudo-second-order kinetics model was best fitted with this system, it can be suggested that the adsorption process was controlled by chemical process or in other words, chemisorption (Hameed, 2008).

Diffusion mechanism might occur in adsorption process, therefore, intraparticular diffusion model was applied to the kinetic data and the equation applied is Eq. 10:

$$q_t = k_{id}^{1/2} + C \quad (10)$$

where, k_{id} was the intraparticular diffusion rate constant (mg/(gmin$^{1/2}$) and *C* is the intercept which was derived from the linear plot of q_t versus $t^{1/2}$ as shown in Fig. 8. Theoretically, diffusion was said to occur in adsorption process if linear plot was obtained and intraparticular diffusion was the rate-controlling step if these lines passed through origin (Fan *et al*., 2008). In removal of BY11 by *S. binderi*, diffusion occurred in adsorption process but these lines deviates from the origin which indicates that diffusion is not the only rate-controlling step.

From intraparticular plot obtained, the first sharp region indicates surface adsorption followed by intraparticular diffusion in the second region (Hameed, 2009). The first region illustrates boundary layer diffusion through mesopore diffusion while the second region indicates intraparticular diffusion by micropore diffusion (Lakshmi, 2009). From Fig. 8, it was proven that uptake of dye occurs initially through boundary layer diffusion as shown in the first sharp region and gradually transform to intraparticular diffusion in the second region.

CONCLUSION

The ability of *S. binderi* in removal for basic dye (BY11) was determined in this study. Various affecting parameters have been conducted in this study to determine the capability of BY11 uptake in batch sorption process. Effect of pH does not possess significant influence towards binding of BY11 compound by *S. binderi*. Uptake of BY11 increased with the increasing

of sorbent dosage. On the other hand, it was found that the removal of BY11 decline with the increasing of initial dye concentration. Removal of BY11 by *S. binderi* was found to obey Fruendlich model in which was heterogenous sorption with sorption capacity of 32.46 (mg g^{-1} (L $mg^{-1})^{1/n}$) and n value of 1.968 which indicates favourable adsorption process. It was also found that the adsorption process followed pseudo-second-order kinetic with the occurrence of chemisorption process as. *S. binderi* was found to be abundance along the coastal area in Malaysia and with high regeneration rate and thus, it might be a potential low cost biosorbent with high adsorption capability.

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