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Marine brown macroalga *Sargassum wightii* as a novel biosorbent for removal of brilliant green dye from aqueous solution: kinetics, equilibrium isotherm modeling and phytotoxicity of treated and untreated dye

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ABSTRACT

In this study, the removal of brilliant green dye from aqueous solution using seaweed *Sargassum wightii* was carried out. The effects of different variables such as adsorbent dose, pH, initial dye concentrations and temperature were studied through batch experimental system. The adsorbent–adsorbate interaction was characterized by Fourier transform infrared spectroscopy and UV–Vis spectrophotometer and the surface structure of the sorbent was documented by scanning electron microscopy and chemical composition was determined by energy dispersive X-ray spectroscopy. The maximum biosorption of brilliant green dye was observed at the sorbent concentration of 0.1 g/L, 7 of pH at 35°C of temperature and initial dye concentration of 10 mg/L. Sorption interaction of dye on to algal biosorbents obeyed the pseudo-second-order rate (R^2 = 0.99). Experimental data showed good fit with the Langmuir adsorption isotherm (R^2 = 0.996) model with a maximum biosorption capacity of 43.48 mg/g. Furthermore, significant reduction in physicochemical parameters was obtained in the treated dye compared with the untreated dye. Phytotoxicity analyses suggested that the treated dye could be tuned as beneficial source for agricultural practices. Accordingly macroalga could be utilized as an efficient adsorbent for dye removal from aqueous solution; prompting to resulting subsequent reduction of toxic effects of the treated dye, alleviates environment damage.

Keywords: Brilliant green; Seaweed; Isotherm; Kinetics; Desorption; Phytotoxicity

1. Introduction

Water pollution is one of the major environmental problems that cause severe threat to living organisms. Increased population, industrialization and urbanization are responsible for environmental pollution. Industrialization of numerous sectors such as food, pharmaceutical, leather, textile, cosmetics, paper, printing, etc. utilize dye compounds to colour their end products [1]. Among these industries, textile industry is the principal contributor of wastewater effluent due to high utilization of water during dyeing, washing and finishing processes.

These wastewaters consist of high content of other products besides dye compounds such as dispersants, acids, bases, salts, detergents and oxidants [2]. Therefore, discharge from textile industries were usually high in colour content, biological oxygen demand (BOD), chemical oxygen demand (COD) and suspended solids [3]. The direct discharge of textile wastewater into the water stream is certainly impermissible

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and every one of us is being exposed to contamination from past and present industrial practices. In addition, these textile dyes reduce light penetration; affect the photosynthetic activity in aquatic life and it may also be toxic and carcinogenic to living organisms [4]. Thus, it is necessary to develop an effective and efficient method to remove the colour from wastewater before being discharged into natural water stream.

With the search of new dimension of treatment methods; biological techniques of dye decolourization are cheaper and easier to operate when compared with conventional treatments like physical and chemical methods. Removal of dye from aqueous solution using biosorbents is also a nonconventional technology and it mainly takes place on the biomass surface and the binding site at the surface activated and thus increasing the effective approach of enhancing the biosorption capacity [5]. Brilliant green (BG) dye (triphenylmethane) is a cationic dye used for decolourization study was selected on the basis of its extensive use as colouring agents (silk, wool) including soluble dyes and insoluble pigments, inks, paints and as indicators and reagents. It causes threats to human beings includes irritation in respiratory tracts and gastrointestinal tracts that leads to damage in organs with symptoms of nausea, diarrhoea and vomiting. It might form harmful products like nitrogen oxides and sulphur oxides during decomposition [6]. Due to its toxicity to the environment, in this research, brown marine macroalgae which are popularly known as seaweed was used as adsorbent to remove BG dye from aqueous solution. Seaweed was chosen as it is ubiquitous biological resource and most predominantly that contains alginate gel in their cell walls which offer a convenient basis for the production of biosorbent particles that are suitable for sorption process and also the presence of abundant dye uptake functional groups [7]. Keeping in view the significance of textile dyes and their environmental tribulations, the current study was undertaken to explore the biosorption potential of brown marine macroalga *Sargassum wightii* for the removal of dye from aqueous solution and the applicability of the use of treated dye for the growth of the selected plants (phytotoxicity). Decolourization kinetics and isotherms were also calculated using different models.

2. Materials and methods

2.1. Dye and reagents

BG dye was procured from Sigma-Aldrich, Mumbai, India, of 90% purity and used without further purification. All other chemicals used in the present study were of analytical grade. Deionized water was used for all dilutions. The properties of dye and the structural formula were shown in Table 1 and Fig. 1, respectively.

2.2. Collection and identification of algal sample

Brown marine macroalga (seaweed *S. wightii*) was collected from the coastal area of Kanyakumari, Tamil Nadu, India (latitude 0.0780°N and longitude 77.5410°S). Algal species was identified according to Chapman and Gellenbeck [8]; and Bold and Wynne [9]. Taxonomic classification of the algal species was made according to the system developed and modified by Papenfnss [10]. The identification was based on (a) morphological, using external and internal characteristics and (b) ecological distribution and habitat.

2.3. Preparation of adsorbent

The seaweed was washed with seawater and freshwater thoroughly to remove the contaminants. The alga was transported to the laboratory and air dried in shade (to evade thermal degradation of the metabolites), pulverized and sieved through a mesh to obtain fine particles of size 150 μm. The powdered sample was stored in an airtight container until use.

2.4. Preparation of dye solution

Dye stock solutions were prepared by dissolving accurately weighed dye in distilled water at a concentration of 1 g/L and left overnight to make the dye powder fully dissolved. The bottles were covered with aluminium foil in order the prevent decolourization caused by light and stored in dark environment at room temperature. All experimental solutions were prepared by diluting the stock

Table 1
Properties of BG dye

Chemical name	[4-[[4-(Diethylamino)phenyl]-phenylmethylidene]cyclohexa-2,5-dien-1-ylidene]-diethylazanium; hydrogen sulphate
Synonyms	Astradiamant green GX, Basic Green 1, Diamond Green, Emerald Green, Ethyl Green, Malachite Green G, Solid Green JO, Basic brilliant green
Molecular formula	$C_{27}H_{34}N_2O_4S$
Molecular weight	482.639 g/mol
CAS Reg. No.	633-03-4
EC number	211-190-1
Melting point	210°C (lit)
Product category	Organics
Hazard code	GHSO7
Industry uses	Dyes
Consumer uses	Silk and wool, spirit inks, wood stains, pigments, antiseptic

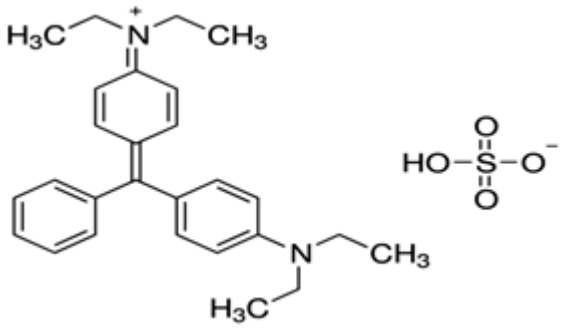

Fig. 1. Structure of brilliant green dye.

solution to the required concentration. Absorbance values were recorded at the corresponding maximum absorbance wavelength (λ_{max}) of 625 nm.

2.5. Biosorption optimization study (decolourization)

The effect of various parameters for the removal of BG dye from aqueous solution onto the brown marine macroalga was studied in batch mode. The experiments were conducted in 250 mL Erlenmeyer flasks containing 100 mL dye solution with desired quantity of adsorbent. To evaluate the effect of different environmental factors on decolourization, the batch experiment was carried out with varying dye concentrations (10–110 mg/L), adsorbent dose (0.05–0.6 g) and pH (2–11) at different temperatures (20°C–60°C). The solution pH was adjusted by adding 0.1 N NaOH and 0.1 N HCl. Under the optimized conditions the samples (5 mL) were withdrawn at various time intervals (0, 5, 10, 15, 20, 30, 60, 120, 180 and 240 min) and evaluated the colour removal (%) and uptake capacity (q_{eq}). The absorbance of the supernatant solution was read in a colorimeter (μP Photo Colorimeter 1311) at 625 nm to estimate the final dye concentration. All the experiments were conducted in triplicates and conical flasks were capped in order to prevent evaporation at high temperature levels. The percentage of decolourization and dye uptake capacity were calculated using the following formula:

$$(\%)\ \text{Color removal} = \frac{\text{Initial concentration} - \text{Final concentration}}{\text{Initial concentration}} \times 100 \quad (1)$$

$$\text{Equilibrium uptake capacity (qe)} = \frac{(\text{Initial concentration} - \text{Final concentration})\text{V}}{\text{W}} \quad (2)$$

where V is total volume of the solution (L) and W is weight of adsorbent concentration (g).

2.6. Application of theories of adsorption

2.6.1. Sorption isotherm

The efficiency of adsorption process was determined by isotherm study. Langmuir [11] and Freundlich [12] models have been used to describe sorption isotherm.

2.6.2. Sorption kinetics

The linear form of pseudo-first-order and pseudo-second-order were applied in dye adsorption study. R^2 values and comparison of calculated sorption capacity, q_{eq} values and experimental q_{eq} values were used to verify the adsorption kinetics of dye by *S. wightii.*

2.7. Analytical studies (characterization of dye solution)

The dye solution before and after adsorption was characterized for neutralized pH, reduction in BOD and COD and further the adsorbent–adsorbate interaction was characterized by Fourier transform infrared spectroscopy (FTIR; Perkin Elmer PE 1600) and UV–Vis spectrophotometer (Shimadzu UV-1800). The surface morphology of the sorbent was documented by scanning electron microscopy (SEM; Model JSM-6100) and chemical composition was determined by energy dispersive X-ray spectroscopy (EDAX).

2.8. Dye desorption and regeneration study

After adsorption process, the dye loaded seaweed was filtered and dried overnight. Dye loaded algal biomass was placed in desorption medium (0.1 N HNO_3, 0.1 M HCl, 0.1 N NaOH and 10% CH_3COOH) and agitated at 150 rpm at 30°C for 30 min and were repeated for one cycle. Samples were taken to record for its pH and maximum absorbance was read in a colorimeter. Then seaweed was filtered and rinsed with double distilled water in order to remove remaining desorbing agent and left overnight to dry. After drying, the seaweed undergoes adsorption process and the regeneration cycle continues.

% dye desorption was calculated using the following formula:

$$\text{DE} = \frac{\text{Amount of dye desorbed}}{\text{Amount of dye adsorbed}} \times 100 \quad (3)$$

2.9. Physicochemical characterization of the dye solution

Treated and untreated textile dye were characterized for its pH, temperature, BOD, COD, total dissolved solids (TDS), total suspended solids (TSS), total solids (TS) and dissolved oxygen (DO) [13]. After analyzing the parameters, seaweed treated BG dye solution was assessed for its phytotoxicity (seed germination study).

2.10. Phytotoxicity study

Cow pea (*Vigna unguiculata* L.), green gram (*Vigna radiata* L.) and horse gram (*Macrotyloma uniflorum* L.) seeds were obtained from Tamil Nadu Agricultural University (TNAU), Coimbatore, Tamil Nadu, India. The healthy and even sized seeds were selected and surface sterilized with 0.1% $HgCl_2$ for 2 min. They were further washed with tap water to avoid surface contamination. A pot culture experiment was conducted with 20 seeds in each pot with respective treatments (T1 untreated dye and T2 treated dye) and tap water as control. Three replicates were maintained for each treatment.

Germination percentage, shoot length (cm), root length (cm), fresh weight (g), dry weight (g), vigour index, tolerance index and % phytotoxicity were measured on the seventh day. The fresh weight of seedlings was taken using an electrical single pan balance. The percentage of vigour index [14], tolerance index [15] and % phytotoxicity [16] were also calculated.

2.11. Statistical analysis

The phytotoxicity results obtained were subjected to one-way analysis of variance (ANOVA) and mean separation were accomplished by Duncan's multiple range test for significant differences ($p \leq 0.05$) using Statistica Software [17].

3. Results and discussion

3.1. Biosorption optimization study (decolourization)

3.1.1. Effect of adsorbent dosage

The effect of biosorbent dose (0.05–0.6 g) on BG dye biosorption on *S. wightii* was shown in Fig. 2. The maximum dye uptake capacity was observed at 0.1 g (25.1579 mg/g) adsorbent dose and minimum was observed at 0.05 g (19.1349 mg/g). When the adsorbent dose was further increased from 0.2 to 0.6 there was decrease in the sorption capacity of adsorbent. At the adsorption dose of 0.6 g, the dye uptake capacity was only 3.77193 mg/g (Table 2).

The results also revealed that dye uptake capacity tremendously increased at lower adsorbent dosage and in contrast decreased at higher doses. This is because; the active sites in the biosorbents could be efficiently utilized when the dosage is low. When the biosorbent dosages are higher, it is more likely that a significant portion of the available active sites remain uncovered, leading to lower specific uptake. Thus, adsorption efficiency decreased with increase in the biosorbent dose. The increase in adsorption might be due to increasing number of adsorption sites caused by breaking of some of the internal bonds near the edge of the active surface sites of the adsorbents [18].

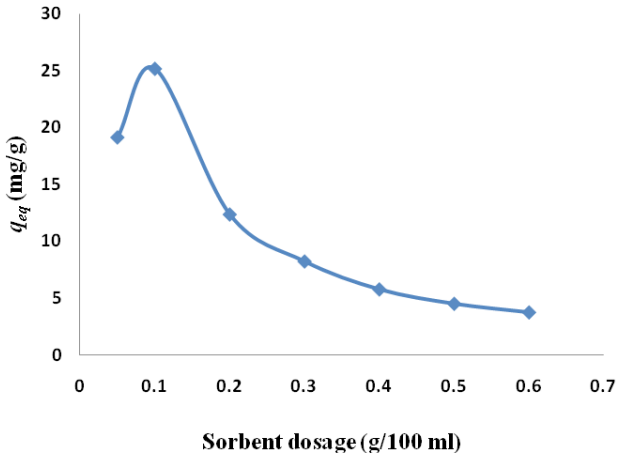

Fig. 2. Effect of biomass dosage on BG dye biosorption by *S. wightii* (pH: 7, dye concentration: 10 mg/L, contact time: 180 min, temperature: 35°C).

Our results also concur with Tan et al. [19] who studied the biosorption of basic yellow 11 using *Sargassum binderi* and stated that uptake capacity limit was found at 0.1 g adsorbent dose. The adsorption of BG dye using guava leaves and potato peels showed an increasing trend with increasing adsorbent dosage initially which then decreases and observed adsorbent dosage 0.6 g of guava leaves and 0.8 g of potato peels [20].

3.1.2. Effect of pH

The effect of pH on BG dye biosorption on *S. wightii* was showed in Fig. 3. Biological treatment process using marine macroalgae are highly pH dependent. The effect of pH was studied by varying the pH from 2 to 10. At pH 2 the minimum adsorption (uptake capacity) of 4.63 mg/g was observed. With further increase in pH, a gradual increase in adsorption was observed maximum of 14.73 mg/g at pH 7. Above pH 7 the adsorption was gradually decreased.

The above results indicated that the dye adsorption considerably increased when pH was raised from 3 to 6. At pH 2, uptake capacity was very low which increased as pH was raised to 7. pH 7 (neutral) was optimum and favours dye adsorption. Similar observations were reported by Tahir et al. [21] and they stated that optimum pH was 7 for methylene blue dye removal using *Ulva lactuca* and *Sargassum* sp.

The optimum pH for malachite green dye removal by *Ulva intestinalis* and *Ulva rigida* was 7 and showed the highest removal ability [22]. The pH of the solution may influence biosorption performance through different mechanisms and also biosorbent surface properties [23]. The pH of the solution affects surface charge of the adsorbent, the degree of

Table 2
Optimum conditions for maximum BG dye removal by *S. wightii*

Parameters	Adsorption condition	Uptake capacity (q_e)
Adsorbent dose	0.1 g/100 mL	25.1579
pH	7	14.73
Initial dye concentration and contact time	10 g/L and 180 min	43.7
Temperature	35°C	9.77

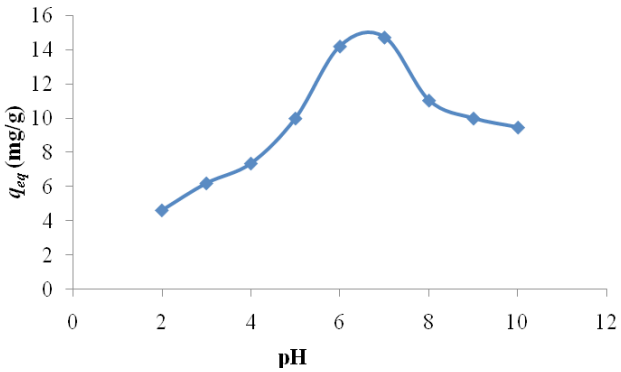

Fig. 3. Effect of pH on BG dye biosorption by *S. wightii* (adsorbent dose: 0.1 g, dye concentration: 10 mg/L, contact time: 180 min, temperature: 35°C).

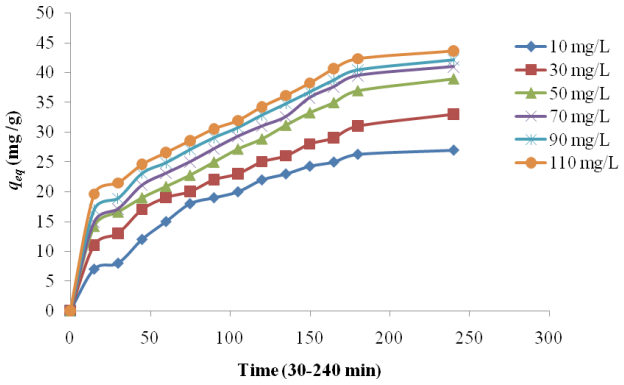

Fig. 4. Effect of initial concentration and contact time on BG dye biosorption by *S. wightii* (pH: 7, adsorbent dose: 0.1 g, temperature: 35°C).

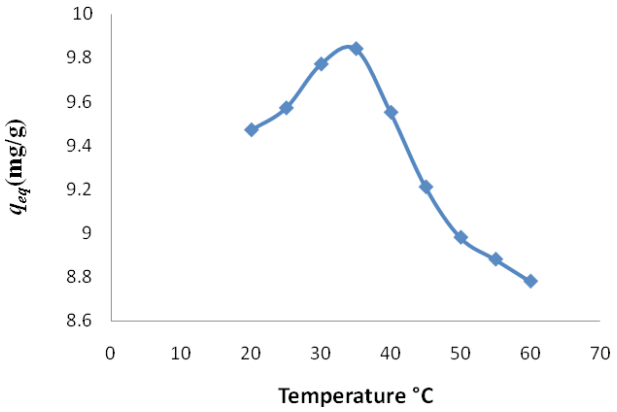

Fig. 5. Effect of temperature on BG dye biosorption by *S. wightii* (pH: 7, adsorbent dose: 0.1 g, dye concentration: 10 mg/L, contact time: 180 min).

ionization of adsorbate molecule, and extent of dissociation of functional groups on the active sites of the adsorbent [24]. The carbon atom of the BG dye acts as an electrophilic centre and the OH^- group favoured in adsorption, thus the uptake of the colour decreased with the increased in the pH [25]. pH 7 was considered as an optimum pH due to high sorption of BG dye onto red clay, as the surface of the adsorbent was negatively charged and the dye cations are positively charged [26].

3.1.3. Effect of initial dye concentration and contact time

The effect of initial dye concentration on BG dye biosorption capacity of *S. wightii* was investigated by changing concentrations from 10 to 110 mg/L. Fig. 4 shows the equilibrium sorption capacity of BG dye at various concentrations. The amount of BG dye achieved 94% colour removal in a very short time (10 mg/L; 43.7 mg/L in 180 min). For the first 30 min, the dye uptake capacity was rapid and thereafter it proceeds at a slower rate and finally attains saturation at different contact time for different initial concentrations of dye. The higher concentration solution of dyes employed, the longer equilibrium time was needed. Thus, the decolourization of dye was very rapid initially, followed by gradual decolourization until equilibrium. The plot also reveals that the uptake of dye adsorbed increases with increase in initial dye concentration. The curve on contact time was found to be smooth and continuous, leading to saturation indicating monolayer coverage of dye on adsorbent surface. It reveals that the rate of uptake capacity increased depending on the contact time.

Similar results have been reported in literature, the effect of initial dye concentration on the adsorption efficiency of biomass and their effects increased with increasing the initial dye concentration. Adsorption of BG dye using neem leaf powder showed that the adsorption rate was rapid during the first 5–15 min followed a gradual increase during 15–240 min with further increasing time there was no improvement in adsorption [27]. The equilibrium time reported for the adsorption of acid green 3 dye using dried *Azolla* was rapid for first 60–90 min and reached after 1.5 h [28]. Saturation of biomass occurred with 10 mg/L concentration of acid black 1 dye using *Nizimuddinia zanardini* and *Sargassum glaucescens* and 50 mg/L dye concentration for *Sargassum marginatum* [29]. The maximum percentage and q_{eq} were observed at 100 ppm for methylene blue dye using *U. lactuca*, *Caulerpa taxifolia*, *Chaetomorpha media* and *Enteromorpha intestinalis* [30].

3.1.4. Effect of temperature

The effect of temperature was studied in the range from 20°C to 60°C (Fig. 5). The dye uptake capacity was increased from 9.47 to 9.77 mg/L for the rise in temperature from 20°C to 35°C. Further increase in temperature from 40°C to 60°C could not increase dye removal, rather a decrease was observed from 9.55 to 8.78 mg/L. Therefore, the maximum adsorption (uptake capacity) of BG dye from aqueous solution was achieved at 35°C.

This was achieved because as temperature increased from 20°C to 35°C, a slight increase in surface area of the adsorbent could be possible but further increase in temperature could result in the loss of active surface area resulting from prolonged exposure to high temperatures. Hence, the adsorption was slow at higher temperature. The optimum temperature was 35°C at which the adsorption was very effective. The decrease of sorption capacity with increasing temperature suggested that the adsorption process was exothermic and the mechanism was mainly physical adsorption, dominant at lower temperature [31,32]. Hence, the present study considered as exothermic nature of adsorption. An elevation in the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores with temperature owing to the decrease in the viscosity of the solution [33]. An increased number of molecules may acquire sufficient energy to undergo an interaction with active sites on the surface. At high temperature, the decrease in biosorption of dyes might be due to the weakening of adsorptive forces responsible for the adsorption of dye molecules. This could be due to the deactivation of biosorbent active sites leads to decreased biosorption [34,35].

Under the above optimized conditions, the maximum adsorption capacity of BG dye from an aqueous solution by *S. wightii* was achieved and depicted in Table 1.

3.2. Biosorption isotherm

Two isotherm models were used to analyze the equilibrium curve at different initial dye concentration. The linear

correlation coefficient of determination (R^2) was used as an error function to evaluate the fitness of each isotherm equation obtained for the two isotherm models and their respective values are listed in Table 3.

3.2.1. Linearized Langmuir isotherm model

Linearized Langmuir isotherm model was expressed in Eq. (4) and separation factor was shown in Eq. (5):

$$1/q_{eq} = 1/Q^\circ + 1/Q^\circ b C_{eq} \quad (4)$$

where q_{eq} is the amount of dye adsorbed at equilibrium over unit mass of sorbent (mg/g), C_{eq} is the equilibrium concentration (mg/L), Q° is the maximum sorption capacity (mg/g) and b is the Langmuir constant (L/mg). Linear plot of $1/q_{eq}$ vs. $1/C_{eq}$ was plotted in order to determine the best fit isotherm data for the adsorption system.

Crucial characteristic of Langmuir adsorption model was expressed as dimensionless separation factor, R_L which can be derived from Eq. (4) (Table 3):

$$R_{L=\frac{1}{1+bC_L}} \quad (5)$$

where b is the Langmuir adsorption constant (L/mg) and C_L is the highest concentration of solution. R_L value indicates the favourable condition of adsorption process.

3.2.2. Linearized Freundlich isotherm model

Freundlich isotherm was expressed as in Eq. (6), and linearized equation was shown in Eq. (7):

$$q_{eq} = K_f(C_{eq})\ 1/n \quad (6)$$

$$\ln(q_{eq}) = \ln(K_f) + (1/n)\ln(C_{eq}) \quad (7)$$

where K_f (mg/g(L/mg)1/n) and n were Freundlich constants derived from the linear slope of $\ln q_{eq}$ vs. $\ln C_{eq}$.

Table 3 also revealed that R^2 value of Langmuir (Fig. 6) and Freundlich isotherm (Fig. 7) are 0.996 and 0.959, respectively. Based on the correlation coefficients, the applicability of the isotherms was compared and it might assume that Langmuir model was best fit to the adsorption data of BG dye than Freundlich model. In addition, the R_L of Langmuir isotherm was obtained from 3.86 (10 mg/L) to 0.08 (130 mg/L) for biosorption of BG dye (Table 3) indicating that biosorption process was favourable. From Table 4, Langmuir constant b and q_m were 2.00 and 43.48 mg/g, respectively, while Freundlich constant K_f and 1/n were 2.641 and 1.41 mg/g, respectively. Both two sets of the isotherms constants are valid and both supported the applicability of the isotherms. Similar observation was reported for removal of malachite green dye from aqueous solution with adsorption technique using *Limonia acidissima* (wood apple) shell [36]. These experimental results and values of R^2 indicated that the sorption of BG dye onto *S. wightii* followed Langmuir model. The sorption process was described by Esan et al. [39] who observed that the Langmuir isotherm with maximum monolayer adsorption capacity of BG dye onto luffa cylindrical sponge.

From the result, the fitness of isotherm models describing the type of adsorption based on high correlation coefficient value (R^2) and Langmuir constant, accordingly the Langmuir isotherm model considered favourable towards adsorption process and described the characteristic of biosorption of BG dye onto *S. wightii*. The Langmuir model assumes monolayer coverage and constant adsorption energy while the Freundlich equation deals with heterogeneous surface adsorption. The applicability of Langmuir isotherm to the studied system implies the monolayer sorption

Table 3
R_L values from Langmuir isotherm model from various initial dye concentrations in removal of BG dye by *S. wightii*

Initial dye concentrations	R_L (10^{-2})
(mg/L)	BG dye
10	3.86
30	1.97
50	0.80
70	0.40
90	0.20
110	0.13
130	0.08

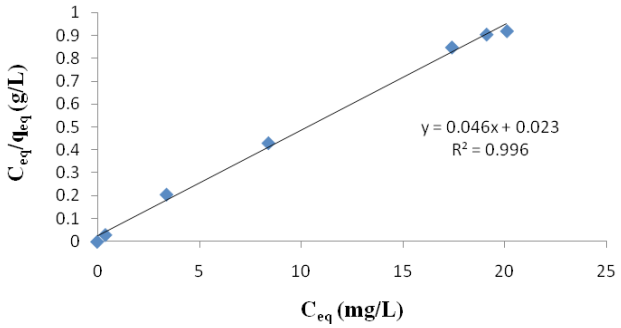

Fig. 6. Linearized Langmuir isotherm curve of BG dye biosorption by *S. wightii.*

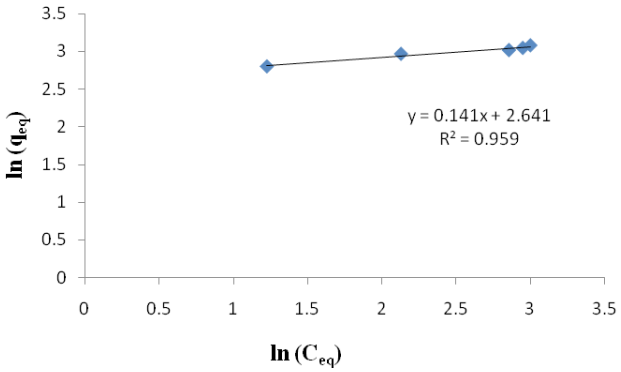

Fig. 7. Linearized Freundlich isotherm curve of BG dye biosorption by *S. wightii.*

and homogeneous surface conditions exist under the used experimental conditions.

3.3. Sorption kinetics

3.3.1. Pseudo-first-order kinetic model

Pseudo-first-order kinetic model (Fig. 8) was theorized by Lagergran [37] as shown in Eq. (8) which was further integrated as linearized form Eq. (9):

$$\frac{dq_t}{d_t} = k_{1(q_{eq}-q_t)} \tag{8}$$

$$\log(q_{eq} - q_t) = \log q_{eq} - (k_1 ad/2.303)*t \tag{9}$$

where q_t indicates the sorption capacity at time t (mg/g), q_{eq} is the sorption capacity at equilibrium (mg/g) and k_1 is the constant. Linear plot of $\log(q_{eq} - q_t)$ vs. t was used in determination of k_1 constant (min^{-1}) and R^2 values. The values of pseudo-first-order kinetic model are tabulated in Table 5.

3.3.2. Pseudo-second-order kinetic model

Pseudo-second-order kinetic model (Fig. 9) was expressed [38] in Eq. (10):

$$t/q_t = (1/k_2 ad\ q_{eq}^2) + (1/q_{eq})t \tag{10}$$

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

From Table 5, it could be observed that pseudo-second-order kinetic model was best fit rather than pseudo-first-order. The linearity of the plot showed the applicability of the pseudo-second-order kinetic model, which has regression coefficients (R^2) in the range from 0.96 to 0.99. These values are very close to 1 and thus it can be suggested that the present adsorption system are more favourably controlled by the pseudo-second-order kinetic model while that of pseudo-first-order varied from 0.92 to 0.98 randomly. The calculated q_{eq}(cal) based on pseudo-first-order model increased from 29.3 to 34.79 (10–90 mg/g) and with increase of initial concentration (90–110 mg/L) decreased in q_{eq}(cal) was observed while that of pseudo-second-order model increased from 101.69 to 252.39 mg/g with the increase of initial dye concentration from 10 to 110 mg/L. Also, the theoretical values of calculated q_{eq} for pseudo-second-order kinetic model agreed with the experimental data. In contrast, q_{eq}(cal) values of pseudo-first-order kinetic model do not match the experimental values. This expresses the chemisorptive behaviour of the biosorption process [6]. It thus showed that the system of study was more appropriately described by the pseudo-second-order model and were in good agreement with Esan et al. [39] and Naveen et al. [40].

Table 4
Comparison of Langmuir and Freundlich isotherm constants for removal of BG dye by *S. wightii*

Isotherms	Parameters	Values
Langmuir	q_m (mg/g)	43.48
	b (L/mg)	2.00
	R^2	0.996
Freundlich	$1/n$	1.41
	K_f (mg/g)	2.641
	R^2	0.959

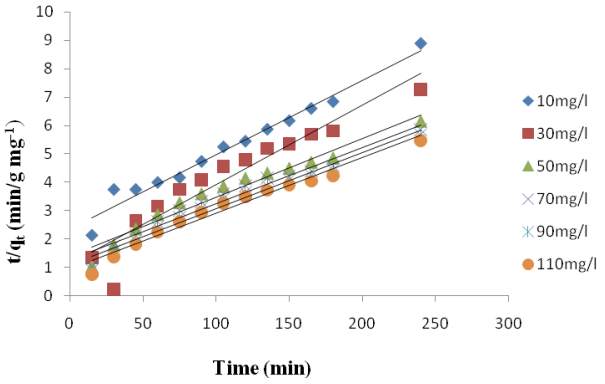

Fig. 8. Pseudo-first-order kinetic model at various initial dye concentrations for the removal of BG dye biosorption by *S. wightii*.

3.4. Analytical studies (characterization of dye solution)

3.4.1. FTIR analysis

To better understand the nature of the functional groups responsible for the biosorption of BG dye by

Table 5
Adsorption kinetic model constants at various initial dye concentrations for the removal of BG dye by *S. wightii*

Initial dye concentration	Pseudo-first-order				Pseudo-second-order		
(mg/L)	q_{eq}(exp) (mg/g)	$k_1$ad (1/min)	q_{eq}(cal) (mg/g)	R^2	$k_2$ad (g/mg/min)	q_{eq}(cal) (mg/g)	R^2
10	27	–0.0015	29.3	0.98	38.17	101.69	0.99
30	33	–0.0108	27.57	0.96	35.71	216.22	0.98
50	39	–0.0115	34.39	0.95	48.31	169.33	0.96
70	41	–0.0124	36.96	0.93	50.5	188.74	0.96
90	42.2	–0.0122	35.33	0.95	50.25	221.28	0.97
110	43.7	–0.0122	34.79	0.92	51.02	252.39	0.97

S. wightii before and after adsorption, the FTIR spectra were obtained within the range of 4,000–800 cm^{-1} (Figs. 10(a) and (b)).

The major difference after seaweed treated BG dye, a new IR band was observed at 2,736 cm^{-1}. However, the following assignments can be done, the band at 1,635 cm^{-1} may be assigned to C=N bond in dye molecule. The shoulder peak at 1,573 cm^{-1} is due to the aromatic phenyl groups in BG dye. The band at 2,890 and 2,978 cm^{-1} are due to the symmetric and asymmetric stretching vibration of alkyl groups. The broad band centred at 3,360 cm^{-1} is due to the moisture content in sample. The broad and strong vibration around 3,000–3,500 cm^{-1} is an indicative of existence of –OH groups and –NH groups on the surface of *S. wightii*. The peaks at 2,823.79 and 2,893.22 cm^{-1} were assigned to the stretching vibration and bending vibration of C–H of the aliphatic groups. The absorption band at 1,242 and 1,249 cm^{-1} were attributed to stretching vibration of S=O group. The peak at 1,350.17 and 1,342.46 cm^{-1} were correspond to –N=O stretching vibrations for the biomass. The bands observed at 1,049.28 and 1,157.29 cm^{-1} were assigned to C–O stretching vibration of alcohols and carboxylic acids.

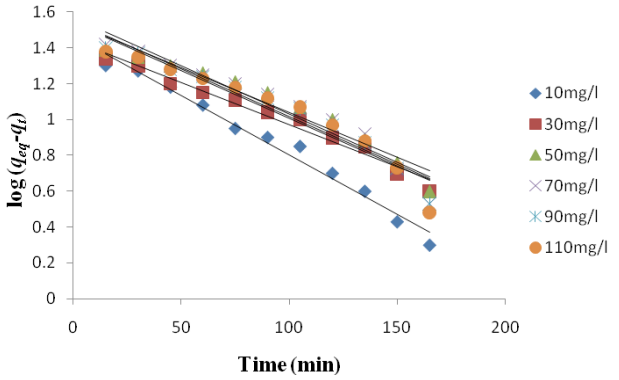

Fig. 9. Pseudo-second-order kinetic model at various initial dye concentrations for the removal of BG dye biosorption by *S. wightii*.

These observations indicated that several functional groups (hydroxyl, carboxyl and amine) on the surface of the biosorbents might be responsible for the binding of BG dye. Moreover, each adsorbent has different binding capacity for each dye molecules. Adsorption capacity is not only affected by the textural

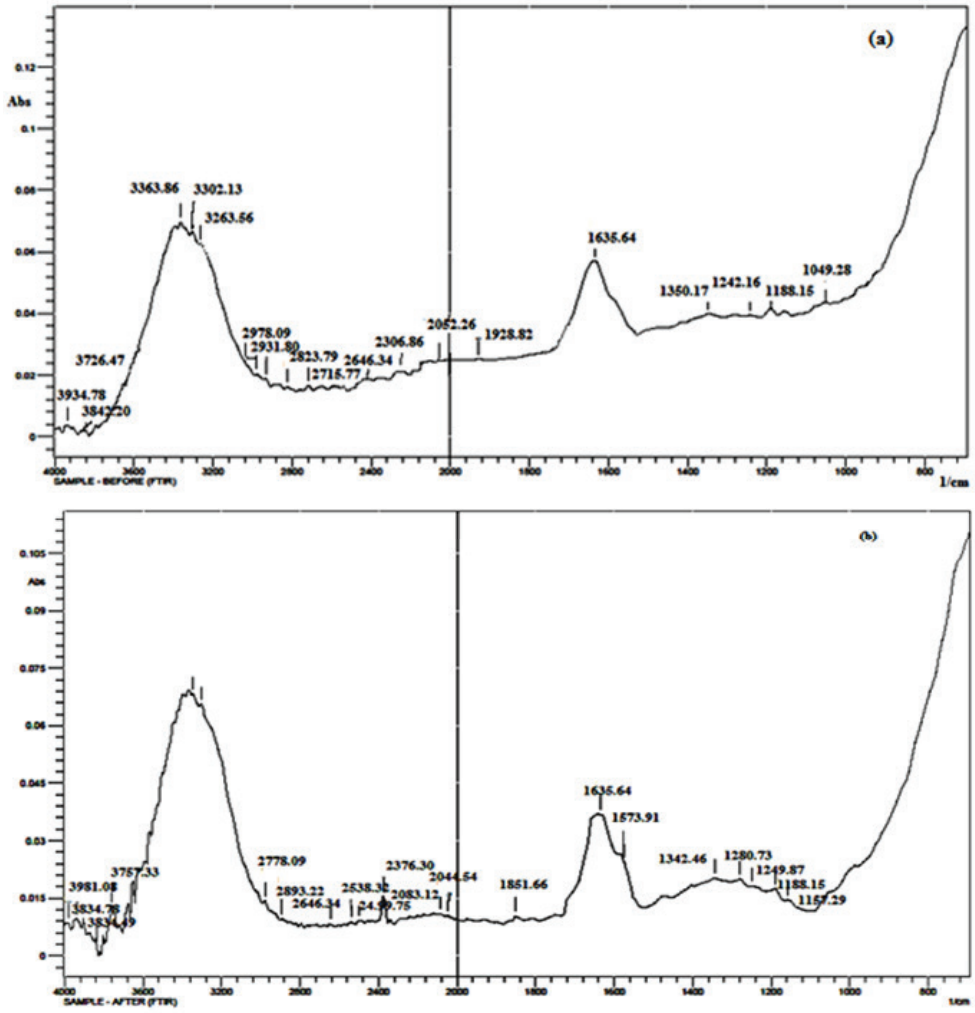

Fig. 10. (a) FTIR spectra of the BG dye before adsorption and (b) FTIR spectra of the BG dye after adsorption.

or porous structure of adsorbents but also strongly influenced by the chemical functionalities at the surface.

3.4.2. SEM analysis

The surface feature and morphological characteristics of the biosorbent were studied using SEM (Fig. 11). The study was useful in determining the particle shape and porous structure of the seaweed. From the micrograph, it was evident that before adsorption (Fig. 11(B)), pores within the seaweed particles are assorted, the surface was rough and irregular in shape which might be indicated that there is an adequate space for dye adsorption. Whereas after BG dye was adsorbed on seaweed, a significant change was observed in surface of the adsorbent (Fig. 11(A)), which clearly indicated the porous and fibrous texture of the biosorbent with homogeneity that could contribute to the biosorption of the dye. Also, it could be seen that the surface is smooth and linkage. Since they are regular in shape, there is high possibility that the dye might be trapped by adsorbent and causes the higher percentage removal of dye from the aqueous solution.

3.4.3. EDAX

In this study various elements were identified from biomass before and after the biosorption by EDAX spectrum. The variation in elemental quantity in brown seaweed was presented in Fig. 12. The amount of K, C, Ca, Cl, O were increased while that of Mg, Na, Si, Al, S were found decreased and Al and Si are found as new after biosorption. It could be due to the chemical interaction mainly employed in the adsorption process. Thus, the study determined possible interaction of seaweed in dye adsorption.

3.4.4. UV–Vis spectroscopic analysis

The initial and final absorbance values of untreated and treated peaks of BG dye were used to determine the intensity of dye decolourization (Fig. 13). The spectrum was recorded using at a range of 200–800 nm. The dye UV spectrum before treatment exhibited absorption band at 538 nm. The main absorption features of BG dye, absorption maximum (the alpha band) near 630 nm and a shoulder band (the beta band) near 590 nm are diminished in the sample treated with seaweed. The significant reduction of the absorption peak at 630 nm indicates the significant adsorption of the dye molecule. The appearance of new peak at 250 nm indicated the maximum adsorption of dye from aqueous solution. From the result, it could be confirmed that the spectrum displays the shift of the maximum absorbance towards shorter wavelengths upon seaweed treatment which indicates considerable amount of dye was adsorbed on sorbent.

3.5. Dye desorption and regeneration study

Desorption study has been carried out to recover the biosorbent for reuse and regeneration in order to reduce the

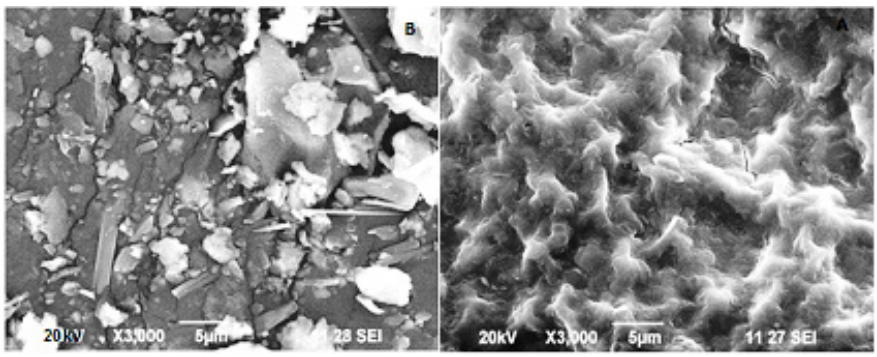

Fig. 11. SEM micrographs before (B) and after adsorption (A).

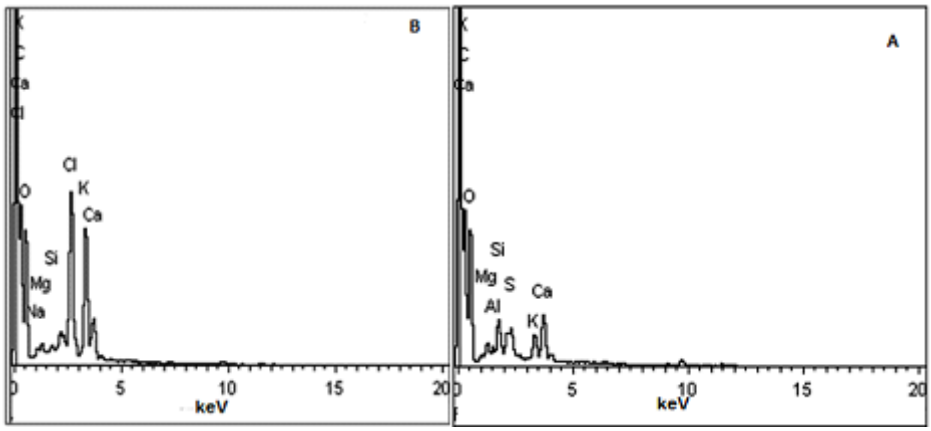

Fig. 12. EDAX spectrum before (B) and after adsorption (A).

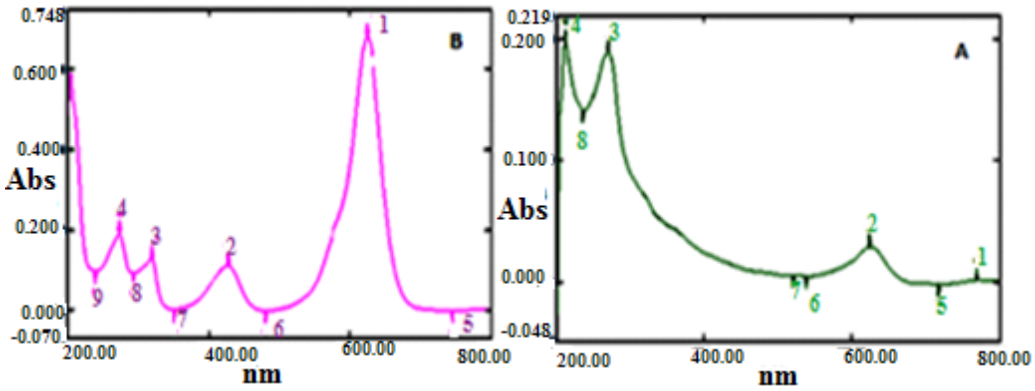

Fig. 13. UV–Vis peak values before (B) and after adsorption (A).

process costs and to determine the desirability of the biosorbent material to desorb the sorbed dye for another cycle. The study was conducted with different desorbing agent's like 0.1 M HCl, 10% CH_3COOH, 0.1 M NaOH and 0.1 M HNO_3. Prior to each experiment, seaweed after adsorption was washed repeatedly with tap water followed by distilled water to remove the unbound dye particles. For the desorption study, dye loaded seaweed was filtered from results obtained in adsorption study under optimized conditions of 7 of pH at 35°C was mixed with 100 mL of the desorbing medium and it was agitated at 150 rpm at 30°C for 30 min.

From the results (Table 6), HCl was found as the most powerful desorbing agent. Although HNO_3 and CH_3COOH showed minimum rate in desorption compared with HCl, they too performed as powerful desorbing agents. NaOH is a strong base that has been widely used as a desorbing agent in desorption process but it might have effective as precipitated (Fig. 14).

After desorption process, regeneration step (adsorption; Table 7) was applied (HCl washed biosorbent in 100 mL of dye) to prevent biomass deterioration or loss of biosorption capacity. The selection of desorbing (regenerating) agent depends on the type of biosorbent used and also the materials being adsorbed, as it determines the type of ion interaction with the biosorbent material [41]. If a protonated biomass like *Sargassum* alga was used, desorption with acid could be more useful, provided that the release of dye and regeneration process also could be achieved and also reducing the process cost [42]. From the study, we observed that biosorbent like *S. wightii* could be regenerated by a simple acidic wash that would quickly release the loaded dye. The main purpose of regeneration is to have a better overall process economy and also to recover the materials. Thus, recovery and recycle of adsorbent in adsorbate might be possible.

3.6. Physicochemical analyses of treated and untreated dye

Physicochemical characterization of the treated dye showed a pH of 7.6 at 30°C. DO content, BOD, COD, TSS, TDS and TS were reduced to the level of 2.0, 21, 240, 90, 2,000 and 2,090 mg/L, respectively. Whereas untreated dye has pH of 9.0 at temperature of 40°C. DO content, BOD, COD, TSS, TDS and TS were found to be 0.5, 90, 471, 240, 5,000 and 5,240 mg/L, respectively.

Table 6
Percentage of dye desorption by various solvents

Time (min)	Desorption (%)		
	HCl (0.1 M)	CH_3COOH (10%)	HNO_3 (0.1 M)
0	0.00	0.00	0.00
10	16.77	13.63	12.53
20	28.63	21.77	23.67
40	35.77	27.16	28.34
60	43.78	33.53	35.45
80	51.42	42.06	41.07
100	59.23	49.42	47.78
120	64.42	43.34	45.03
140	71.34	41.12	43.21
160	77.27	39.11	41.45
180	82.92	37.34	38.97
200	79.15	34.12	35.73

Fig. 14. Desorption study under different desorbing medium.

3.7. Phytotoxicity study

The phytotoxicity study of untreated and *S. wightii* treated BG dye solutions compared with control (tap water) were carried out in three different nutritive valued seeds viz., cow pea (*Vigna unguiculata* L.), green gram

Table 7
Percentage of reuse efficiency (adsorption cycle 1)

Time (min)	Decolourization (%)
0	9
10	21
20	27
40	33
60	39
80	43
100	49
120	51
140	55
160	58
180	61
200	57

(*Vigna radiata* L.) and horse gram (*Macrotyloma uniflorum* L.) and the results were tabulated in Table 8. From the result, ANOVA test showed that the germination percentage, seedling growth and their fresh weight, dry weight were maximum in control followed by seaweed treated BG dye and minimum was observed in untreated dye. Untreated dye influences their toxic effect whereas treated dye after decolourization showed better seed germination and spontaneous growth of plants. The results of the present investigation suggested that the significant increasing growth in treated dye solutions might be due to the prevented accumulation of dye in the sample there by reducing its toxicity significantly. Our results also coherence with Phugare et al. [43] reported the toxic and nontoxic effects of untreated and treated effluent on the seed germination of *Triticum aestivum* and *Phaseolus mungo*. Phytotoxicity study showed higher germination rate of *T. aestivum* and *Ervum lens* plants grown after decolourization when compared with the plants grown in dye [44]. *Triticum* sp. grown in treated dye showed better growth when compared with untreated dye [45].

Table 8
Phytotoxicity study of control, untreated and seaweed treated BG dye (n = 20), and its effect on seeds germination (%), shoot length (cm), root length (cm) weight (g), vigour index and tolerance index

Germination seeds	Observations	Control (tap water)	Untreated dye	Seaweed treated dye
Cow pea 	Germination (%)	93	42	80
	Shoot length (cm)	13.43 ± 0.208^{c}	3.10 ± 0.020^{a}	12.60 ± 0.057^{b}
	Root length (cm)	6.43 ± 0.152^{c}	2.26 ± 0.020^{a}	5.40 ± 0.0570^{b}
	Fresh weight (g)	0.27 ± 0.020^{c}	0.10 ± 0.005^{a}	0.24 ± 0.005^{b}
	Dry weight (g)	0.23 ± 0.010^{c}	0.09 ± 0.005^{a}	0.18 ± 0.005^{b}
	Vigour index	558	84	400
	Tolerance index	–	0.333	0.833
	% of phytotoxicity	–	–7.42	–78.85
Green gram 	Germination (%)	95	40	83
	Shoot length (cm)	13.40 ± 0.050^{c}	3.10 ± 0.026^{a}	13.60 ± 0.057^{b}
	Root length (cm)	6.40 ± 0.010^{c}	2.16 ± 0.005^{a}	5.40 ± 0.005^{b}
	Fresh weight (g)	0.28 ± 0.005^{c}	0.10 ± 0.005^{a}	0.27 ± 0.010^{b}
	Dry weight (g)	0.20 ± 0.0025	0.09 ± 0.005^{a}	0.18 ± 0.005^{b}
	Vigour index	1,235	80	1,079
	Tolerance index	–	0.333	0.833
	% of phytotoxicity	–	–10.07	–94.69
Horse gram 	Germination (%)	93	41	87
	Shoot length (cm)	17.40 ± 0.351^{c}	3.10 ± 0.010^{a}	15.6 ± 0.809^{b}
	Root length (cm)	4.40 ± 0.665^{c}	2.26 ± 0.010^{a}	4.73 ± 0.665^{b}
	Fresh weight (g)	0.18 ± 0.005^{c}	0.10 ± 0.005^{a}	0.27 ± 0.005^{b}
	Dry weight (g)	0.25 ± 0.010^{c}	0.10 ± 0.005^{a}	0.19 ± 0.005^{b}
	Vigour index	1,581	123	1,218
	Tolerance index	–	0.5	1.5
	% of phytotoxicity	–	–0.64	–65.35

Note: Results are the mean value of triplicates ± standard deviation followed by different superscripts with significant difference at $p < 0.05$.

4. Conclusions

The brown seaweed, *S. wightii* proved its potency for BG dye biosorption from aqueous solution. The effect of different parameters such as adsorbent dosage, pH, initial dye concentration and temperature were studied. Optimization study showed that maximum biosorption occurred at lower biomass dosage (0.1 g), 7 of pH at 35°C. BG dye adsorption decreased with increase in initial dye concentration may be due to the loss of the active binding sites on the biosorbent and maximum removal percentage obtained was 94%. The consequence of equilibrium adsorption was successfully described through Langmuir isotherm for biomass. Kinetic models were also examined and the pseudo-second-order was found to be the applicable kinetic model in the present study. FTIR analysis confirmed the role of different surface functional groups for the sorption of BG dye. SEM photographs clearly indicated the porous and fibrous texture of the biosorbent with high heterogeneity that could contribute to the biosorption of the dye. EDAX analysis concluded that chemical composition mainly employed in the adsorption process. UV–Vis spectroscopy indicated the intensity of absorption spectra was reduced considerably after absorption. Phytotoxicity study reveals the treated BG dye could be valorized as an excellent nutrient rich source for agricultural practices. The results of the present study concluded that utilization of brown marine macroalga could be a good alternative and could drive the current expensive methods for the removal of dyes from aqueous solution.

Acknowledgement

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Symbols

b	—	Equilibrium constant of biosorption
C_0	—	Initial concentration, mg/L
C_{eq}	—	Equilibrium concentration, mg/L
$HgCl_2$	—	Mercurous chloride
$k_1$ad	—	Pseudo-first-order rate constant, min^{-1}
$k_2$ad	—	Pseudo-second-order rate constant, g/mg min
$K_f v$	—	Freundlich isotherm constant, L/mg
N^f	—	Freundlich isotherm constant
$Q_0 b$	—	Langmuir isotherm constant, L/mg
q_{eq}	—	Equilibrium uptake capacity, mg/g
q_t	—	Sorption capacity at time t, mg/g
R^2	—	Condition factor
R_L	—	Dimensionless constant separation factor
V	—	Volume of the solution, L
W	—	Weight of adsorbent, g

References

[1] V.K. Garg, M. Amita, R. Kumar, R. Gupta, Basic dye (methylene blue) removal from simulated wastewater by adsorption using Indian Rosewood sawdust: a timber industry waste, Dyes Pigm., 63 (2004) 243–250.
[2] S.G. de Souza, L.C. Peruzzo, A.U. de Souza, Numerical study of the adsorption of dyes from textile effluents, Appl. Math. Modell., 32 (2008) 1711–1718.
[3] V.K. Garg, R. Kumar, R. Gupta, Removal of malachite green dye from aqueous solution by adsorption using agro-industry waste: a case study of *Prosopis cineraria,* Dyes Pigm., 62 (2004) 1–10.
[4] D.D. Asouhidou, K.S. Triantafyllidis, N.K. Lazaridis, K.A. Matis, S.S. Kim, T.J. Pinnavia, Sorption of reactive dyes from aqueous solutions by ordered hexagonal and disordered mesoporous carbons, Microporous Mesoporous Mater., 117 (2009) 257–267.
[5] J. Mao, S.W. Won, J. Min, Y.S. Yun, Removal of Basic Blue 3 from aqueous solution by *Corynebacterium glutamicum* biomass: biosorption and precipitation mechanisms, Korean J. Chem. Eng., 25 (2008) 1060–1064.
[6] A. Mittal, D. Kaur, I. Mittal, Applicability of waste materials—bottom ash and deoiled soya—as adsorbents for the removal and recovery of a hazardous dye, brilliant green, J. Colloid Interface Sci., 326 (2008) 8–17.
[7] R. Vieira, B. Volesky, Biosorption: a solution to pollution?, Int. Microbiol., 3 (2000) 17–24.
[8] D.J. Chapman, K.W. Gellenbeck, An Historical Prospective of Algal Biotechnology, Algae and Cyanobacterial Biotechnology, Longman Group, UK, 1983, pp. 1–27.
[9] H.C. Bold, M.J. Wynne, Introduction to the Algae: Structure and Reproduction, Prentice Hall, Inc., Englewood Cliffs, New Jersey, 1978, pp. 706.
[10] G.F. Papenfnss, Classification of the Algae, A Century of Progress in the Natural Sciences, California Academy Science, San Francisco, 1955, pp. 115–224.
[11] I. Langmuir, The constitution and fundamental properties of solids and liquids, J. Am. Chem. Soc., 38 (1916) 2221–2295.
[12] H.M.F. Freundlich, Over the adsorption in solution, J. Phys. Chem., 57 (1906) 385–470.
[13] APHA, AWWA, WEF, Standard Methods for the Examination of Water and Wastewater, 21st ed., Washington, 2005.
[14] A.A. Abdul-Baki, J.D. Anderson, Vigour determination in soybean seed by multiple criteria, Crop Sci., 3 (1973) 630–633.
[15] R.G. Turner, C. Marshall, Accumulation of zinc by subcellular fractions of roots of *Agrostis tenuis Sibth* in relation of zinc tolerance, New Phytol., 71 (1972) 671–676.
[16] C.H. Chou, C. Chaiang, C.I. Kao, Impacts of water pollution on crop growth in Taiwan, Bot. Bull. Acad. Sinica, 19 (1978) 107–124.
[17] Statistica for Windows [Computer Program Manual], StatSoft, Inc., Tulsa, OK, 1999.
[18] S.M. Kanawade, R.W. Gaikwad, Removal of dyes from dye effluent by using sugarcane bagasse ash as an adsorbent, Int. J. Chem. Eng. Appl., 2 (2011) 203.
[19] P. Tan, C. Wong, S. Ong, S. Hii, Equilibrium and kinetic studies for basic yellow 11 removal by *Sargassum binderi*, J. Appl. Sci., 9 (2009) 3005–3012.
[20] R. Rehman, T. Mahmud, M. Irum, Brilliant green dye elimination from water using *Psidium guajava* leaves and *Solanum tuberosum* peels as adsorbents in environmentally benign way, J. Chem., 2015 (2015) 8.
[21] H. Tahir, M. Sultan, Q. Jahanzeb, Removal of basic dye methylene blue by using bioabsorbents *Ulva lactuca* and *Sargassum*, Afr. J. Biotechnol., 7 (2008) 2649–2655.
[22] K. Pansamrit, S. Ruangsomboon, Effect of pH on Biosorption of Basic Dye Malachite Green by Algae, The 8th International Symposium on Biocontrol and Biotechnology, 2010, pp. 132–136.
[23] N. Daneshvar, M. Ayazloo, A.R. Khataee, M. Pourhassan, Biological decolorization of dye solution containing Malachite Green by microalgae *Cosmarium* sp., Bioresour. Technol., 98 (2007) 1176–1182.
[24] B.K. Nandi, A. Goswami, M.K. Purkait, Adsorption characteristics of brilliant green dye on kaolin, J. Hazard. Mater., 161 (2009) 387–395.
[25] C.V. Rao, A.S. Giri, V.V. Goud, A.K. Golder, Studies on pH dependent colour variation and decomposition mechanism of Brilliant Green dye in Fenton reaction, Int. J. Ind. Chem., 7 (2016) 71–80.

[26] M.S.U. Rehman, M. Munir, M. Ashfaq, N. Rashid, M.F. Nazar, M. Danish, J. Han, Adsorption of Brilliant Green dye from aqueous solution onto red clay, Chem. Eng. J., 228 (2013) 54–62.
[27] K.G. Bhattacharyya, A. Sarma, Adsorption characteristics of the dye, Brilliant green, on Neem leaf powder, Dyes Pigm., 57 (2003) 211–222.
[28] D. Balarak, E. Bazrafshan, F.K. Mostafapour, Equilibrium, kinetic studies on the adsorption of Acid Green 3 (Ag_3) dye onto *Azolla filiculoides* as adsorbent, Am. Chem. Sci. J., 11 (2016) 1–10.
[29] M. Kousha, E. Daneshvar, H. Dopeikar, D. Taghavi, A. Bhatnagar, Box–Behnken design optimization of Acid Black 1 dye biosorption by different brown macroalgae, Chem. Eng. J., 179 (2012) 158–168.
[30] R. Deokar, A. Sabale, Biosorption of methylene blue from aqueous solution onto green seaweeds, Int. J. Recent Sci. Res., 5 (2014) 347–351.
[31] F. Deniz, S.D. Saygideger, Equilibrium, kinetic and thermodynamic studies of Acid Orange 52 dye biosorption by *Paulownia tomentosa* Steud. leaf powder as a low-cost natural biosorbent, Bioresour. Technol., 101 (2010) 5137–5143.
[32] A. Abbas, S. Murtaza, K. Shahid, M. Munir, R. Ayub, S. Akber, Comparative study of adsorptive removal of congo red and brilliant green dyes from water using peanut shell, Middle-East J. Sci. Res., 11 (2012) 828–832.
[33] S. Durairaj, S. Durairaj, Colour removal from textile industry wastewater using low cost adsorbents, Int. J. Chem. Environ. Pharm. Res., 3 (2012) 52–57.
[34] M. Asgher, H.N. Bhatti, Removal of reactive blue 19 and reactive blue 49 textile dyes by citrus waste biomass from aqueous solution: equilibrium and kinetic study, Can. Chem. Eng. J., 90 (2012) 412–419.
[35] D. Vigneshpriya, E. Shanthi, Biosorption of textile effluent using sugarcane bagasse, Int. J. Curr. Res., 8 (2016) 31324–31331.
[36] A.S. Sartape, A.M. Mandhare, V. Jadhav, P.D. Raut, M.A. Anuse, S.S. Kolekar, Removal of malachite green dye from aqueous solution with adsorption technique using *Limonia acidissima* (wood apple) shell as low cost adsorbent, Arabian J. Chem., 7 (2013). http://dx.doi.org/10.1016/j.arabjc.2013.12.019.
[37] S. Lagergran, About the theory of so-called adsorption of soluble substances, K. Sven. Vetensk.akad. Handl., 24 (1898) 1.
[38] G. Mckay, Y.S. Ho, Pseudo second order model for sorption process, Process Biochem., 34 (1999) 451.
[39] O.S. Esan, O.N. Abiola, O. Owoyomi, C.O. Aboluwoye, M.O. Osundiya, Adsorption of brilliant green onto luffa cylindrical sponge: equilibrium, kinetics, and thermodynamic studies, ISRN Phys. Chem., 2014 (2014) 1–12.
[40] N. Naveen, P. Saravanan, G. Baskar, S. Renganathan, Equilibrium and Kinetic modeling on the removal of Reactive Red 120 using positively charged *Hydrilla verticillata*, J. Taiwan Inst. Chem. Eng., 42 (2011) 463–469.
[41] Y. Mata, M. Blazquez, A. Ballester, F. Gonzalez, J. Munoz, Studies on sorption, desorption, regeneration and reuse of sugar-beet pectin gels for heavy metal removal, J. Hazard. Mater., 178 (2010) 243–248.
[42] P. Lodeiro, R. Herrero, M.E. Sastre De Vicente, Batch desorption studies and multiple sorption–regeneration cycles in a fixed-bed column for Cd(II) elimination by protonated *Sargassum muticum*, J. Hazard. Mater., 137 (2006) 1649–1655.
[43] S. Phugare, P. Patil, S. Govindwar, J. Jadhav, Exploitation of yeast biomass generated as a waste product of distillery industry for remediation of textile industry effluent, Int. Biodeterior. Biodegrad., 64 (2010) 716–726.
[44] U. Shedbalkar, J.P. Jadhav, Detoxification of malachite green and textile industrial effluent by *Penicillium ochrochloron*, Biotechnol. Bioprocess Eng., 16 (2011) 196–204.
[45] N.H. Brahmbhatt, R.T. Jasrai, The role of algae in bioremediation of textile effluent, Int. J. Eng. Res. Gen. Sci., 4 (2016) 21–28.