Greener Cleaner: Liquid-Phase Adsorption of Dye Using Macroalgae Based Biosorbent Materials

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Abstract

Dyes are considered one of the most dangerous pollutants. Due to its toxicity to many life forms. Dying-wastewater also has a great impact on the ecosystem by putting the marine lives at high level of risk. In this study, treated macroalgae with potassium carbonate (K_2CO_3) has been used as a low cost and eco-friendly adsorbent materials for the removal of Crystal violet (CV) dye from water using batch mode system. The Influence of various important parameters such as contact time, initial dye concentrations, solution pH, amount of biomass, and solution temperature were investigated. The UV-visible spectrophotometer was applied to analyze the remaining dye concentrations in the treated solution. According to the data derived, Langmuir isotherm model was best fit the experimental results. The equilibrium adsorption time was reached within 90 min. Maximum adsorption capacity for CV adsorption onto macroalgae was found to be 38.4 mg/g. The kinetics analysis revealed that the pseudo-second-order model was a better fit for the experimental data than the pseudo-first order. The thermodynamic study demonstrates the spontaneous and endothermic nature of adsorption system. Finally, the presented study observed that macro alga-based biomass could be utilized in the removal of basic dyes from wastewater with high removal efficiency up to 98%. Also, algae biomass could be more economic and effective biosorbent for the control of dyes from industrial liquid wastes.

Keywords: Adsorption; Algae; Batch mode; Crystal violet; Equilibrium; Isotherm; Kinetic

Introduction

Water is used everywhere and plays such a vital role in our lives, but its supply is increasingly threatened by an increase in the generation of waste effluents [1]. Managing waste and preserving the quality of water are now two of the important tasks of human life [2]. Dyes are organic pollutants that pose a particular danger to the environment. Synthetic dyes (SD) are widely used in many areas of advanced technology such as dyestuff, paper, printing, carpet, plastic, food and cosmetic, paint manufacture and textiles to provide colour to their products [3,4], and as additives in petroleum products [5]. The textile industry uses large volumes of water in its manufacturing processes. As a result, they generate large amounts of colored wastewater [6]. It is reported that in the dyeing section of a textile industry, about 1 m³ of water is used for every ton clothes processed [5]. Thousand different types of dyes and pigments are available commercially and around 10-15% of such dyes are lost in the effluent during manufacturing and processing operations [5,7,8]. Chemically, synthetic dyes possess aromatic structures that are biologically non-degradable and whose efflu-

ents are difficult to treat [5,9,10]. A particularly difficult task for the wastewater treatment plants of textile industries is the elimination of the color of these compounds, as dyes and pigments are resistant to biodegradation. They typically remain in the environment for a long period of time. For example, the hydrolyzed dye reactive blue 19 has a half-life of around 46 years at room temperature and a pH of 7 [7]. A number of treatment techniques are available with different degree of sources to control and reduce pollution of water [11]. These technologies including chemical precipitation, ion exchange, membrane separation, electrochemical treatment, oxidation and coagulation/flocculation [12]. Nowadays, adsorption process is a very cost-effective and efficient treatment approach for the removal of pollutants from water and wastewater in comparison with the conventional methods [12]. In addition, adsorption process has offered flexibility in operation and design, and could be produce high-quality treated effluent [12]. Algae is a diverse group of photosynthetic organisms ranging from unicellular (microalgae) to multicellular (macroalgae) forms inhabiting freshwater and marine environments [13].

In recent years, there has been a growing interest in utilizing seaweed such as macroalgae to remove various water pollutants including heavy metals and dyes [11,13,14]. More recently, the possible application of algae biomass in nuclear waste management has been explored [14]. Because the structures of these biochemical substances are associated with a various functional group, such as carboxyl, hydroxyl, amine, carbonyl, and sulfate, which participated in the adsorption system [13,14,15]. However, there are several studies being conducted in this trend, particularly in the adsorption of heavy metals from liquid media [13]. Because algae biomass is abundance, economically attractive, and environmentally friendly adsorbents [14]. In the current study macro algae-based biomass has been utilized as a green and potential low-cost adsorbent for the adsorption of crystal violet from aqueous phase. Studies of equilibrium, isotherm, kinetics and thermodynamics are the main purposes of the study. This study indicates that algae biomass could be successfully applied at industrial effluents as adsorbent materials without or with a little bit of modification. Figure 1 shows the application of algae biomass.

Figure 1: Applications of macro algae based biomass.

Experimental work

Materials

In this study, macro algae has been selected as a potential low cost biosorbent material for the removal of basic dye from aqueous phase. The algae biomass was obtained from the saltwater ponds on farms in Libya. The raw materials were collected from the site and put into glass container and immediately transported to the laboratory. Then, the material was washed completely with water to remove salt, dust, and other impurities and undesirable materials. After that, the biomass was chemically treated by being soaked in potassium carbonate (K₂CO₃) solution and stirred in a water bath at 65°C for 4 hr. It was subsequently washed with distilled water and pure acetone respectively, and oven-dried at 105°C. Finally, the dried biomass was crushed and sieved and stored in plastic bags for further use.

Crystal violet (CV) {molecular weight =408g/mol; λ_{max} =584nm}, a cationic dye purchased from B.D.H Chemicals, Ltd., England was chosen as the model adsorbate. Working solutions were prepared from an aqueous stock solution (1000 mg/l) by diluting the dye stock solution with distilled water to give the needed initial concentrations ranging from 10 to 100 mg/L by using formula (1) [16]. To estimate the bulk density of dried biomass, a glass cylinder 25 ml was filled to a specified volume with powdered biomass and dried in an oven at 70°C for 24hr. The cylinder was tapped for few minutes to compact the material and the bulk density estimated and presented as g/ml using formula (2) [16,17], and was found to be 0.41g/ml.

C1 V1 = C2 V2 (1)

D = W/V (2)

where D is the bulk density of biomass (g/ml), W is the weight of dry biomass (g), and V is the final volume (ml)

Batch adsorption process

Adsorption experiments were carried out in 250 ml beakers containing fixed amount of biomass (0.3g) with 100 ml of dye solution (70 mg/l). The mixture was stirred at a temperature of 50oC, pH 8, for 30 min using normal mixer at stirring speed of 800rpm. The influence of different factors affecting adsorption process such as contact time, pH, adsorbent mass, initial dye concentrations, and temperature were investigated. The final concentration of dye was analyzed by UV-vis spectrophotometer after separation of solid phase from the mixture by filter paper. The adsorption capacity (q_e) was calculated from the following Equation [16]:

q_e = (C_i - C_e) V/M (3)

where C_i and C_e are the initial and equilibrium concentrations of dye (mg/l), respectively, V is the total volume of dye solution (l), and M is the mass of the adsorbent used (g). While, the percentage removal of CV was determined according to the equation (4) [16]. Matrix of the experimental work including, range and levels of independent process variables are given in Table 1.

% Removal = (C_i - C_e/C_i) x 100 (4)

Results and Discussion

Affecting parameters on the adsorption system

Influence of pH

The pH factor is very important and controlling parameter in the adsorption process for the treatment of water pollution [16]. The experiments were carried out as follow: 100 ml of 70 mg/l CV dye solution, were stirred with 0.25g of solid phase “biomass adsorbent” using 250 ml beakers at solution pH from 3 to 9 through a batch mode system at temperature of 50°C for 60 min. Stirring

Influential Factors	Performances				
	Adsorbent dosage	Initial conc.	pH	Contact time	Temperature
pH	0.25	70	3-9	60	323
Adsorbent mass(g)	0.15-0.5	70	6.5	60	323
Concentration (mg/l)	0.25	30-100	6.5	60	323
Temperature (K)	0.25	70	6.5	60	323
Contact time (min)	0.25	70	6.5	10-120	293-328

Table 1: The influence of factors on the rate of dye adsorption by macroalgae.

speed was fixed at 800 rpm for the whole adsorption process. The pH values of the solution were adjusted with 1M of HCl for the acid media and 1M of NaOH solution for the basic media respectively. Figure 2 shows the effect of pH on the adsorption efficiency for CV dye onto biomass. The results show that the percentage removal of CV dye rapidly increased with increasing pH values until reaches maximum at pH (6.5). A similar behavior was observed for the removal of basic dye from aqueous solution by orange peel powder [16], coffee waste [18], wasted Oolong tea [19], Gmelina Arborea biomass [20], olive-waste cake [21], marine macroalgae [22]. This trend may be described to electrostatic interaction between cationic dye (CV) and the negative surface of adsorbent. Lower removal of CV dye at the acid media may be due to the presence of excess H⁺ ions that compete with the basic dye for adsorption site [16]. On the other hand, as the pH value increased, the number of negatively charged surface sites on the biomass adsorbent increased, thus enhancing the removal efficiency of CV dye from aqueous solution. The maximum percentage removal CV dye was reached at pH 6.5 and found to be 98%.

Figure 2: Influence of pH on the removal of CV dye by macroalgae biomass.

Influence of adsorbent mass

The effect of adsorbent dose was performed by contacting different amounts of biomass in the range of (0.15g - 0.5g) using 100 ml of 70 mg/l of CV dye solution at pH of 6.5, reaction temperature of 50°C, at a fixed mixing speed of 800rpm for 60 min. Figure 3 shows the relation between the adsorption efficiency for CV removal and the adsorbent mass. From the figure it can be observed that with increase in adsorbent mass from 0.15 to 0.4g the percentage removal of CV dye rapidly increased from 86% to 97.7%. The adsorption equilibrium of dye was reached at adsorbent mass of 0.4g, and there is no change in the percentage removal of dye from 0.4 to 0.5g of adsorbent was observed. In other site, the adsorption capacity was decreased from 40.13 mg/g to 13.66 mg/g as the adsorbent mass increasing from 0.15 to 0.5g. This behavior can be explained as a result of overlapping or aggregation of adsorption sites resulting in a decrease in the total adsorption surface area available to the dye and an increase in the diffusion path length [23]. However, 0.25g was selected as optimum value of the adsorbent mass and has been used for the next stages. Similar trend for the influence of adsorbent amount on the adsorption efficiency for basic dyes was observed and reported in previous studies for other types of low-cost adsorbents [15-23].

Figure 3: Relationship between adsorbent mass and removal of dye.

Influence of initial dye concentration

To understand the influence of initial dye concentration on the removal efficiency, a series of different concentrations for the removal of dye were performed at pH of 6.5, solution temperature of 50°C, adsorbent dose 0.25g, adsorption time of 60 min, using 100ml of working solution and fixed agitation speed at 800rpm. The experimental result is shown in figure 4. In general, the adsorp-

tion efficiency decreases with increase in the initial concentration of dye, whereas the adsorption capacity increases with the initial concentration increase [18]. From the figure can be seen that the adsorption capacity for CV by macroalgae has been increased from 11.88 mg/g to 30.8 mg/g with increasing the initial concentration of CV dye from 30 mg/l to 100 mg/l, and then reached equilibrium. This is due to saturation of active sites on the adsorbent surface and exuthoin point takes place. This is agreement with several literatures that used low cost adsorbents for the removal of basic dyes from aqueous solution [16-22].

Figure 4: Influence of initial concentration on the adsorption capacity of CV.

Influenc of adsorption time

The effect of adsorption time for the removal of CV from its aqueous solution was investigated. The experiments was carried out by contacting 100 ml of 70 mg/l of CV initial dye concentration with 0.25 g of biomass at solution temperature of 50°C, agitation speed of 800 rpm, and pH 6.5 using 250 ml beakers, over time periods ranging from 10 to 120 min. The remaining concentration of CV dye as function of contact time is shown in figure 5. It can be concluded that the remaining concentration of dye in the solution after adsorption process was decreased from 28 mg/l to 1.12 mg/l with the increase of contact time from 10 min to 60 min and then reached equilibrium at contact time of 90 min. The percentage removal of CV at equilibrium was found to be 98.4%, while adsorption capacity for CV onto biomass was 27.6 mg/g. This results is in trend with several works have been done previously by many researchers [16-21].

Adsorption Isotherm

The equilibrium adsorption isotherm is important element in the design of adsorption process [18]. In order to describe the

Figure 5: The effect of contact time on the adsorption of CV.

equilibrium between the solid and the liquid phases in the adsorption system, two isotherm models (Langmuir and Ferundlich) are widely used for this purpose [16,18].

Langmuir isotherm model is applied to describe the adsorption system where the occupancy occurs at a single adsorption site at an energetically homogeneous range of adsorption sites [18]. The adsorption isotherm studies were performed by varying the initial concentration of dye from 30 to 150 mg/l, keeping all other conditions constant as follow: (pH 6.5, mixing speed 800 rpm, 100 ml of working solution, adsorbent mass of 0.25g). A plot of C_e/q_e versus C_e , indicates a straight line of slope $(1/q_{max})$ and an intercept of $(1/b q_{max})$, where C_e is the concentration of dye at equilibrium (mg/l), q_e is the amount of dye adsorbed at equilibrium (mg/g), and q_{max} is the maximum adsorption capacity for dye onto biomass (mg/g); b is the Langmuir constant (l/mg). Equation 5 shows the Langmuir isotherm expressions that can be linearized to Eq. 6 [24].

$$q_e = (q_{max} b C_e) / (1 + b C_e) \quad (5)$$

$$C_e/q_e = (1/q_{max} b) + (C_e/q_{max}) \quad (6)$$

A plot of C_e/q_e against C_e , gave a fitted curve, and the Langmuir constants were calculated regarding to figure 6 and data are presented in Table 2. The Langmuir isotherm can also be expressed in terms of a dimensionless constant separation factor R_L that is given by Eq. (7) [16,18].

$$R_L = 1 / (1 + b C_e) \quad (7)$$

The values of the factor R_L obtained are shown in figure 7 indicat that the adsorption process is favorable, since $(0 < R_L < 1)$. There were found to be in the range of (0.048 - 0.01). Similar results were reported in previous works [16,18,22].

Figure 6: Langmuir isotherm plot for the adsorption of CV by macroalgae.

Figure 8: Freundlich isotherm plot for the adsorption of CV by macroalgae.

Figure 7: R_L as function of initial CV dye concentration.

Freundlich isotherm model is mainly used to describe characteristics for the heterogeneous surface. The model assumes that adsorption takes place on a heterogeneous surface, and can be used for non-ideal adsorption system [16]. The linear form of Freundlich isotherm is given as: [25].

$$\ln q_e = 1/n \ln C_e + \ln K_f \quad (8)$$

where q_e is the amount of CV adsorbed onto biomass in equilibrium (mg/g), C_e is the equilibrium dye solution concentration (mg/l), K_f and n are the Freundlich equilibrium coefficients. A plot ($\ln q_e$) against ($\ln C_e$) gave lower fitted curve than the Langmuir isotherm model. The values obtained of the Freundlich equilibrium coefficients K_f and n from figure 8 are presented in Table 2.

Equilibrium isotherm constants					
Langmuir isotherm			Freundlich isotherm		
q_{max}	b	R^2	K_f	n	R^2
38.4	0.645	0.987	19.24	5.4	0.842

Table 2: Equilibrium isotherm modeling of CV dye adsorption onto macroalgae biomass.

Kintetic experiments

There are two types of kinetics models including pseudo-first order [26], and pseudo-second order [27] which have been widely used to investigate the mechanism of bioadsorption and potential rate controlling steps such as mass transfer and chemical reaction process [28].The linear pseudo-first order Eq. is given as [18]:

$$\log (q_e - q_t) = (\log q_e) - (k_1/2.303) t \quad (9)$$

where q_e and q_t are the amount of adsorbed CV dye onto biomass (mg/g) at equilibrium and at time t (min) respectively; k_1 is the rate constant of pseudo-first-order model. The pseudo-second order equation is given as follows [18]:

$$t/q_t = 1/(k_2 q_e^2) + (1/q_e) t \quad (10)$$

where k_2 (g/mg min) is the rate constant of pseudo-second-order equation.

Kinetic adsorption experiments were performed by stirring 100 ml of a constant CV concentration with 0.25 mg of adsorbent mass at pH value of 6.5, stirring speed of 800 rpm, for 90 min. The samples were then filtered by filter paper and the remanaing

concentration of CV in the final solution was analyzed using UV-vis spectrophotometer. The procedure was similar to that used by Aljeboree., *et al.* [29]. Figs 9 and 10 showed the plots of pseudo-first order kinetic model and the pseudo-second-order model, respectively, for the removal of CV dye from aqueous solution onto macroalgae biomass. According to the data obtained, the results suggested that the pseudo-frist-order model is less suitable to describe the adsorption system compared to the pseudo-second-order model. The correlation coefficient of second-order-kinetic model (0.9963) is greater than for pseudo-first-order model (0.8452). This confirmed that the rate limiting step is chemisorption, involving valence forces through sharing or exchange of electrons between adsorbent and adsorbate [18]. The kinetic parameters of both models are reported in Table 3. Works [18,28,30] were also observed a similar behaviour.

Figure 9: Pseudo-first-order reaction of adsorption (CV onto biomass).

Figure 10: Pseudo-second-order reaction of adsorption (CV onto biomass).

Kinetic constants					
First-pseudo-order			Second-pseudo-order		
q _e	K ₁	(r ₁) ²	q _e	K ₂	(r ₂) ²
29	0.0776	0.996	34	0.0044	0.845

Table 3: Kinetic constants for the adsorption of CV onto biomass algae

Thermodynamic studies

Temperature effect on the adsorption process is another significant parameter that shows whether the adsorption nature is an exothermic or endothermic system and changes the adsorption capacity of the adsorbent [3,31-33]. A temperature study also helps in understanding thermodynamic parameters such as free energy (ΔG_o), enthalpy (ΔH_o), and entropy (ΔS_o) [32]. These parameters were calculated using the following equations [30,31].

ΔG_o = -RTln Kc (11)

Kc = q_e/C_e (12)

Also,

ΔG_o = ΔH_o - TΔS_o (13)

lnKc = (ΔS_o/R) - (ΔH_o/RT) (14)

where q_e represents the amount of CV adsorbed onto biomass algae at equilibrium (mg/l), R is the gas constant (8.314 J/mol k), and C_e is the equilibrium concentration of CV in the solution (mg/l).

The effect of temperature on the adsorption efficiency were performed in the range of (293K -328K). Besides, the initail concentration of CV was maintained at 70 mg/l and all other conditions such as pH, adsorption time, adsorbent mass, speed of stirring, working solution were kept constant. Results of temperature study is showed in figure 11. From the figure it can be seen that the percentage removal of CV dye increassses rapidly with an increase in temperature from 293 to 328 K. This may be because of a rise in the mobility of adsorbate and a rise in the number of active sites for the adsorption with rising temperature [31,32]. In addition, it might be because of the rise in chemical interaction between the adsorbate and surface functionalities of the adsorbent [32,34,35]. Further rise in temperature reduces the swelling effect thus enabling the lage dye molecule to penetrate further [31]. This improves the diffusion rate of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent as a result of the viscosity decreases of the solution [32,34].

On the other hand, thermodynamic parameters for CV adsorption were evaluated using equations (11-14). The values of enthalpy and entropy were determined from the slope and entercept of the plot of (ln Kc) versus (1/T) as shown in figure 12. Table 4 illustrates the thermodynamic parameters for the removal of CV by magroalgae biomass. The obtained value of ΔH° (53.12 KJ/mol)

indicates that the adsorption process is an endothermic reaction. The spontaneous nature of adsorption appears due to negative value of ΔG° (-7.98 KJ/mol). This confirms affinity of macro algae adsorbent for the adsorbate CV dye [30,31]. Similar trend was observed by Singh and Kaur, (2013) when they used rice straw for the removal of malachite green (MG) from its aqueous solutions [36].

T, K	ΔG° (kJ/mol)	ΔH° (kJ/mol)	ΔS° (J/mol K)
293	-2.38	53.12	188.1
303	-3.52		
313	-5.26		
323	-7.98		
328	-8.73		

Table 4: Thermodynamic parameters of macro algae biomass.

Figure 11: Effect of temperature on the percentage removal of CV onto biomass.

Figure 12: Van't Hoff plot for the removal of CV onto biomass.

Conclusion

In this study, macro algae biomass shows a good example of natural biosorbents for the removal of crystal violet from aqueous solution. The optimum adsorption parameters were found to be (0.25 g of adsorbent mass, contact time of 90 min, and solution temperature of 323 K). Adsorption of CV dye was pH dependent and the maximum percentag removal was attained at pH value of 6.5. Equilibrium data were fitted well with the Langmuir isotherm model combarred to the Freundlich model. The kinetics analysis revealed that the pseudo-second-order model was a better fit for the experimental data than the pseudo-first order. The thermodynamic study demonstrates the spontaneous and endothermic nature of adsorption system. This study Also observed that the algae biomass could be widely used in the agriculture and environment cleaning purpose of toxic dyes from the industrial wastes polluting water in lakes and rivers. Further investigation about characterization of the adsorbent (including physical and chemical properties) are still necessary and recommended to support this study.

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