



Original Article

Batch adsorption and isothermic studies of malachite green dye adsorption using *Leucaena leucocephala* biomass as potential adsorbent in water treatment

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Abstract

The adsorption properties of *Leucaena leucocephala* biomass to clean malachite green contaminated water were investigated in this work. BET surface analysis showed a surface area of 0.5724 m²/g. Batch adsorption studies and isothermic studies determined that the best time and temperature for the adsorption process to become efficient were 60 min of contact time and 25 °C, respectively. The KF value of 0.3372 mg adsorbed per mg of adsorbent of the *L. leucocephala* biomass for malachite green was the highest at 50 °C. It was shown that the n values were n < 1 which suggested that malachite green dye adsorption on *L. leucocephala* biomass is favorable. Values of the correlation coefficient (R²) indicated that the Freundlich isotherm can be used for the adsorption of malachite green on *L. leucocephala*. The results also suggest that *L. leucocephala* biomass can be utilized as an adsorbent for malachite green dye from wastewater at a lower cost.

Keywords: adsorption, malachite green, batch adsorption, Langmuir, Freundlich, *Leucaena leucocephala*

1. Introduction

Wastewater comes mainly from the industrial sector. Examples of pollutant sources are textiles, paper, and other dyeing industries. Effluents containing dyes have become a major concern due to the adverse effects on living organisms. The discharge of dyes into the environment is a matter of concern for both toxicological and esthetical reasons. Dyes are used for coloring products such as plastics, paper, and textiles and at the same time consume significant volumes of water. As a consequence, industries generate a considerable amount of colored water. It is estimated that more than 100,000 commercially available dyes with over 7 x

10⁵ tons of dyestuff produced annually and from 40,000 to 50,000 tons of dye are discharged into the surface water every year (Parvathi & Maruthavanan, 2010).

There are many methods to reduce the color in textile effluent streams, involving physical and chemical treatment methods such as coagulation, precipitation, filtration, electrodialysis, membrane separation, and oxidation (Venckatesh *et al.*, 2010). However, these methods are very costly in terms of capital and the efficiency is low in treating waste water (Ho & Ofomaja, 2006). Adsorption using activated carbon, clay, chitin, and many others is the technique chosen most often since they can be used efficiently at a comparatively low cost (Weng & Pan, 2006). Adsorption is a mass transfer process in which substances are transferred from a liquid phase to a solid phase. It is a surface phenomenon where the process is bounded by physical or chemical forces. While adsorption using activated carbons or organic

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resins has been one of the procedures of choice, the cost of producing them is very high (Crini, 2006; Hai *et al.*, 2007). Therefore, it is a necessity to find alternative, cheaper adsorbent materials to replace activated carbon to clean contaminated water.

Research had been done on the use of *Hevea brasiliensis* sawdust as a bio-adsorbent for the removal of malachite green from aqueous solutions which had the adsorption capacity of 27.4 mg/g at 40 °C (Le Phan Linh *et al.*, 2012). Pavan *et al.* (2014) also did similar research by using *Carica papaya* L. seed powder to remove crystal violet from the aqueous phase. The Brunauer–Emmett–Teller (BET) surface area was found to be 1.38 m²/g which resulted in 85.99 mg/g of maximum adsorption capacity at 25 °C. Another example is the utilization of sulphuric acid treated fly ash (Lin *et al.*, 2008) and a powder from oil palm leaves (Sulaiman *et al.*, 2010) as low cost bio-sorbents with 2.2217 mol/g at 55 °C and 11.22 mg/g at 30 °C, respectively. A limited number of studies have reported the use of *Leucaena leucocephala* biomass particles as an adsorbent for dyes. *L. leucocephala* is a wild tree that grows very fast and can be found all over Malaysia. The rapid reproduction of *L. leucocephala* biomass makes it an ideal raw material as a low-cost adsorbent for dyes. Thus, this research was undertaken to investigate the effectiveness of *L. leucocephala* biomass as a low-cost adsorbent of dye from contaminated water.

2. Materials and Methods

2.1 Preparation of the adsorbent

L. leucocephala wood samples were obtained from the environs of Prince of Songkla University, Pattani Campus, Thailand. The samples were then washed with distilled water several times to remove the dirt and unwanted particles. The washed materials were chipped into smaller pieces and dried in an oven at 70 °C for 24 h. The material was ground and sieved into 4 different ranges of particle sizes: <0.30; 0.30–0.71; 0.71–0.85; and 0.85–2.0 mm. The particles were then dried again at 70 °C in an oven for 2 h until constant moisture content was obtained before use.

2.2 Preparation of the adsorbate

In this study, the malachite green was obtained from Sigma-Aldrich. Malachite green stock solutions were prepared by dissolving 1.0 g of the dye into 1 liter of distilled water to a concentration of 1000 mg/l. The experimental solutions were obtained by diluting the dye in correct proportions to get concentrations of 10 mg/l, 20 mg/l, 30 mg/l, 40 mg/l, and 50 mg/l.

2.3 Characterization of adsorbent properties

Surface area characterization was done using Micromeritics 3Flex surface analyzer. Nitrogen adsorption isotherm measurement was done at -195 °C. The sample was first degassed at 100 °C for 2 h, and the surface areas were calculated by the BET method. FT-IR analysis of the solid phase was performed using a Fourier Transform Infrared Spectrometer, FT-IR (Thermo Scientific Nicolet iS10). Samples were scanned 16 times between 4000 and 400 cm⁻¹ at a resolution of 4 cm⁻¹ (Cui *et al.*, 2011).

2.4 Batch adsorption studies

Batch adsorption studies were done by measuring 50 ml of each dye solution with concentrations of 10, 20, 30, 40, and 50 mg/l in conical flasks and mixing with an adsorbent at a dose of 0.5 g/l. The samples were then put inside an incubator shaker and shaken at 130 rpm for pre-determined time intervals. The temperature of the mixtures was maintained at 30 °C. The final dye concentrations were measured after 30, 60, 90, and 120 min until equilibrium was reached. The adsorption percentages of the samples were estimated using a spectrophotometer (HACH DR5000 UV-VIS) at the wavelength corresponding to maximum absorbance, ($\lambda_{\text{max}} = 616$ nm). A graph was plotted with adsorption percentage against time. The adsorption percentage is expressed by Equation 1.

$$\text{Adsorption, \%} = \frac{\text{Initial conc. of dye ion} - \text{final conc. of dye ion}}{\text{Initial conc. of dye ion}} \times 100 \quad (1)$$

The effects of the initial concentration of adsorbate, solution temperature, pH of solution, adsorbent size, and adsorbent dose were determined by varying the parameters which were the initial concentration (10, 20, 30, 40, and 50 ppm), pH of the solution (2, 4, 6, 7, 8, 10, and 12), adsorbent size (<0.30, 0.30–0.71, 0.71–0.85, and 0.85–2.00 mm), and adsorbent dose (0.5, 1.0, 1.5, 2.0, and 2.5 g/l solution).

2.5 Isothermic studies

Isothermic studies were done to determine how the adsorption molecules were dispersed in the solid and liquid phase when the process of adsorption reached the equilibrium state. An analysis of the isotherm data by fitting the data to different isotherm models is a vital step to indicate the most suitable model to be used for design purposes. It is important to describe the interactions between the adsorbate and adsorbent and it is critical in optimizing the use of an adsorbent. To identify the most appropriate correlations for the equilibrium data in the design of the adsorption system, isothermic studies were carried out based on three common isotherm models: the Langmuir, Freundlich, and Temkin models. The applicability of the isotherm equations to the adsorption study was compared by judging the correlation coefficients, R² (Hameed, 2009).

In this study, 15 ml of adsorbate solutions of different concentrations (10, 15, 20, 25, 30, 35, 40, 45, and 50 ppm) were mixed with 0.5 g of adsorbent in separate conical flasks. Conical flasks were shaken at 130 rpm for 1 h which is the best time for adsorption to reach equilibrium in batch adsorption studies. Experiments were carried out at 30 °C in triplicates and were repeated at temperatures of 40 °C and 50 °C. Mixtures were filtered with filter paper and filtered solutions were analyzed using a spectrophotometer.

2.5.1 Langmuir model

The Langmuir isotherm model assumes ideal monolayer adsorption onto a homogenous surface. The data of the equilibrium studies for the adsorption of dye at 30, 40, and 50 °C onto *L. leucocephala* biomass may follow the Langmuir model based on Equation 2 (Hameed *et al.*, 2009a):

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{K_a q_m C_e} \quad (2)$$

where q_e is the amount of adsorbate adsorbed at equilibrium in mg/g, q_m is the saturated amount of adsorbate adsorbed in mg/g, C_e is the equilibrium concentration of adsorbate in mg/l, and K_a is the Langmuir adsorption constant in l/mg. Hence, a plot of $1/q_e$ versus $1/C_e$ should be a straight line with a slope of $(1/K_a q_m)$ and an intercept as $1/q_m$. The value of constant K_a was calculated (Sulaiman *et al.*, 2010).

2.5.2 Freundlich model

The Freundlich isotherm is suitable for non-ideal adsorption on heterogeneous surfaces. This heterogeneity arises from the presence of different functional groups on the surface, and different adsorbent-adsorbate interactions. The Freundlich adsorption isotherm is expressed by Equation 3 (Rafatullah *et al.*, 2011):

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (3)$$

where K_F is the Freundlich adsorption constant and $1/n$ is a measure of the adsorption study. All other terms have the usual significance and n is an empirical constant. Thus, a plot of $\log q_e$ vs. $\log C_e$ should be a straight line with a slope $1/n$ and an intercept $\log K_F$. All related parameters were calculated.

2.5.3 Temkin model

The Temkin isotherm explicitly takes into account the absorbing species-adsorbate interactions. The isotherm uses two assumptions which are (i) the heat of adsorption of all the molecules in the layer decreases linearly with coverage due to adsorbate-adsorbate interaction and (ii) adsorption is characterized by a uniform distribution of binding energies to some maximum binding energy (Hameed *et al.*, 2009b). A plot of Q_e versus $\ln C_e$ was plotted using linear Equation (4):

$$Q_e = a + b \ln C_e \quad (4)$$

where Q_e is the amount adsorbed at equilibrium (mg/g) and C_e is the equilibrium concentration (mg/l) of metal ions in the solution.

3. Results and Discussion

3.1 Characterization of adsorbent properties

The result of the BET surface area measurement was $0.5724 \text{ m}^2/\text{g}$. This is considered very low compared to other adsorbents such as *Avena sativa* (oat) hull which showed $63.0 \text{ m}^2/\text{g}$ of surface area. This is confirmed by an isothermic analysis which showed low adsorption capacity of *L. leucocephala* biomass. Increasing the adsorption capacity can be achieved using surface modification or thermal activation means. Figure 1 shows the FT-IR spectra of *L. leucocephala* biomass. The peak at 3336.95 cm^{-1} indicates the presence of hydroxyl groups which are abundant in woody biomass. The peak at 2889.26 cm^{-1} showed methyne C-H stretch, while the peak at 2099.51 cm^{-1} showed cyclohexane ring vibrations. Meanwhile, peaks at 1025.35 cm^{-1} and 526.77 cm^{-1} determine groups of primary amine (CN stretch) and aliphatic iodo compounds (C-I stretch), respectively.

3.2 Effect of contact time

From the results, the adsorption equilibrium was attained after 60 min for the experiment with five different initial concentrations. Therefore, all equilibrium adsorption data in this study are collected after 60 min. The effect of contact time can be seen from Figure 2 for the adsorption of dyes. It is clear that the extent of adsorption for all five concentrations of dye (10, 20, 30, 40, and 50 ppm) was rapid in the initial stages and became slow in the later stages until saturation. The final dye concentrations did not vary significantly after 60 min from the start of the adsorption process. This shows that equilibrium can be assumed to be achieved after 60 min. It was observed that for every initial concentration, the highest dye uptake was recorded at 60 min adsorption with 75.36% maximum adsorption for an initial concentration of 10 mg/l. The higher removal percentage for the lower initial concentration of dye was due to the huge availability of adsorption sites compared to the amount of dye to be absorbed as well as the calculation was based on the percentage of dye removed from the initial concentration, not based on the mass of the dye.

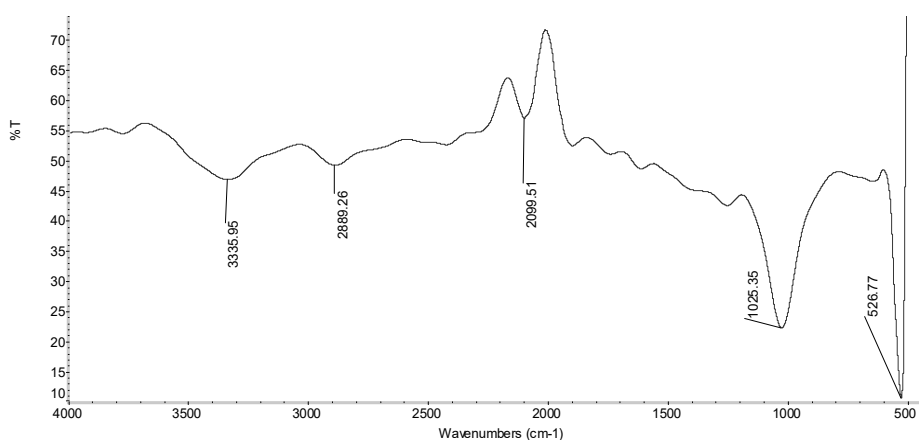


Figure 1. FT-IR spectra of *Leucaena leucocephala* biomass.

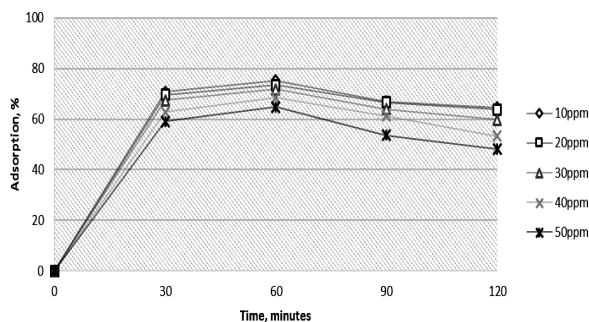


Figure 2. Percentage of adsorption against time.

According to Ho and Chiang (2001), fast diffusion on the external surface was followed by fast pore diffusion into the intraparticle matrix to attain rapid equilibrium. At equilibrium, due to the saturation of the active site, it does not allow further adsorption to take place. This means that, at initial stages, the adsorbate cations occupied many of the active sites resulting in the rapid uptake of ions. The active sites were then blocked at the later stage which caused the rate to decline. For the maximum removal of malachite green by the adsorbent, the solutions should be equilibrated for 60 min regardless of the initial concentration.

3.3 Effect of temperature

In this study, batch adsorption experiments were performed in the range of 15 °C to 60 °C using 0.5 g of 0.30–0.71 mm size adsorbent in 50 ml of dye solutions with five different concentrations as stated previously. Figure 3 shows the percentage of adsorption against the temperature. It indicates that as the temperature increased from 15 °C to 25 °C, the adsorption percentage increased. However, further increments of temperature above 25 °C resulted in a decrement of the adsorption percentage. The best temperature for adsorption of malachite green dye was carried out at 25 °C. Thus, it is recommended that malachite green be cooled to room temperature before any adsorption process takes place as waste water from the industries tends to have a higher temperature. Aksu and Tezer (2005) stated that the adsorption rate decrease with a further increase in temperature was possibly due to a decrease in surface activity. It was suggested that the adsorption process between malachite green and *L. leucocephala* was an exothermic process and the mechanism was mainly physical adsorption enhanced by a chemical effect.

3.4 Effect of pH

In an aqueous solution, malachite green is a cationic dye that exists in the form of positively charged ions. As a charged species, the rate of adsorption onto the adsorbent surface is primarily influenced by the surface charge on the adsorbent, which in turn is affected by the solution pH (Ramakrishna & Viraraghavan, 1998). Maximum dye removal was noted at pH 4. Figure 4 shows the effect of pH on the percent removal of malachite green. It was observed that the adsorption capacity increased with an increase in the initial pH in the range of 2–4. The amount of adsorption, however,

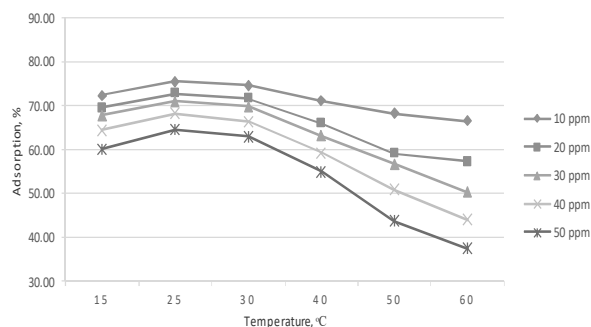


Figure 3. Percentage of adsorption against temperature at 60 min.

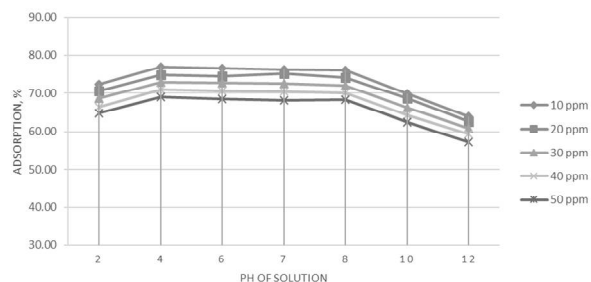


Figure 4. Percentage of adsorption against pH at 60 min.

remained almost constant in the range of pH 4–8. Somehow, dye removal decreased as the solution pH increased from 8 to 12. At a lower pH, the amine and carboxylic groups may take $-\text{NH}_3^+$ and $-\text{COOH}$, respectively. In another words, at a pH of 2, the surface of adsorbent is positively charged, which makes H^+ compete with dye cations causing a decrease in dye removal. While at a higher pH, the amine and carboxylic groups may take $-\text{NH}_2$ and $-\text{COO}^-$, respectively. The influence of solution pH on the change in the surface charge may be attributed to the adsorption behavior observed in this study.

3.5 Effect of size of adsorbent

The effect of adsorbent size on adsorption behavior was investigated using four different sizes of adsorbent: <0.30, 0.30–0.71, 0.71–0.85, and 0.85–2.00. Figure 5 shows the amount of adsorption of malachite green at 60 min as a function of the adsorbent size. The size <0.30 mm was found to have the greatest effect in dye removal from the solution. This occurred because the smallest adsorbent particles are free from the mass transfer effect, while the larger particles underwent the effect. The intake by smaller particles was also due to the greater accessibility to the pores and the greater surface area for bulk adsorption per unit mass of the adsorbent (Krishna & Swamy, 2012).

3.6 Effect of adsorbent dose

The effect of adsorbent dose on adsorption behavior was investigated using five different doses of adsorbent: 0.5, 1.0, 1.5, 2.0, 2.5, 3.5, and 5.5 g. Figure 6 shows the adsorption percentage of malachite green against the dose of adsorbent at 60 min. The amount of 2.5 g was found to be the most effi-

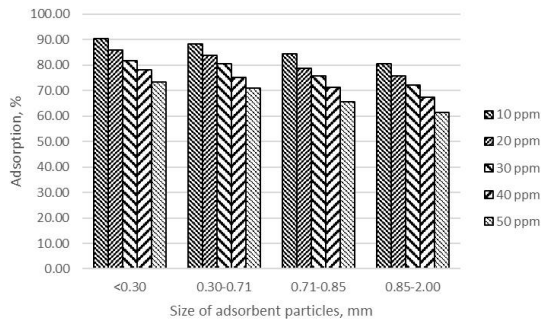


Figure 5. Percentage of adsorption against size of adsorbent.

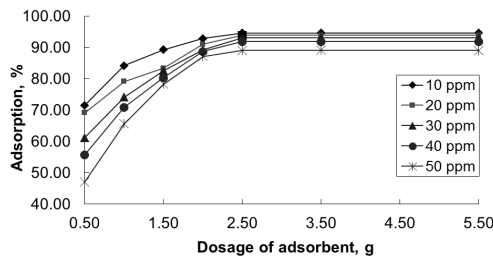


Figure 6. Percentage of adsorption against dose of adsorbent.

cient in dye removal from the solution with the adsorption percentage between 89.03% and 94.58% for the initial concentrations of 50 ppm and 10 ppm, respectively. Additional amounts of adsorbent at 3.5 g and 5.5 g made the adsorption percentage about 0.03% higher or lower. These small changes were considered insignificant and indicated the saturation point for the adsorption sites. The percentage of adsorption of dye increased with an increase in the amount of adsorbent. This is due to the greater availability of the adsorption sites at the higher adsorbent dose (Erdem *et al.*, 2005).

3.7 Isothermic studies

3.7.1 Langmuir model

All of the constants of the Langmuir equation were obtained from the intercept and slope of a linear plot of the experimental data of $1/Q_e$ versus $1/C_e$ (Figure 7). Table 1 shows the values of q_m and K_a which were calculated from the intercept and slope of the linear plot. The maximum adsorption capacities for malachite green onto the *L. leucocephala* biomass particles at 30 °C, 40 °C, and 50 °C were found to be 2.389, 2.310, and 2.383 mg/g, respectively. The high values of the correlation coefficient, shown as R^2 , indicated good agreement between the parameters. However, negative values for the intercept showed that there was some desorption in some parts of the adsorbent as reported by Boekhold *et al.* (1993) and the inadequacy of the isotherm model to explain the adsorption process.

3.7.2 Freundlich model

A plot of $\log q_e$ vs. $\log C_e$ gave a straight line, the slope of $1/n$, and the intercept $\log K_F$ (Figure 8). All the related parameters were calculated and are listed in Table 1. The value of K_F of *L. leucocephala* biomass for malachite

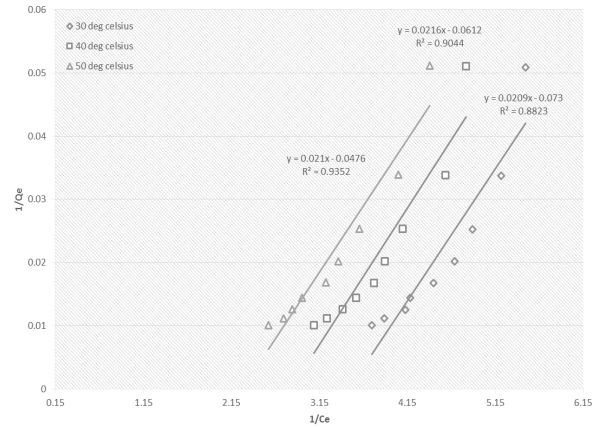


Figure 7. Langmuir adsorption isotherm plots for the adsorption of dye at 30 °C, 40 °C, and 50 °C.

Table 1. Adsorption isotherm model constants and correlation coefficients for the adsorption of dye on *Leucaena leucocephala* at 30 °C, 40 °C, and 50 °C.

Adsorption isotherms	Isotherm constants	Temperature (°C)		
		30	40	50
Langmuir	q_m (mg/g)	2.389	2.310	2.383
	K_a (l/mg)	-0.287	-0.354	-0.441
	R^2	0.9352	0.9044	0.8823
Freundlich	K_F (mg/mg) (l/mg) ^{1/n}	1.1741	0.3000	0.0878
	n	0.2464	0.2819	0.3372
	R^2	0.9071	0.9760	0.9879
Temkin	a	18.554	14.95	11.739
	b	10.376	8.9842	7.4255
	R^2	0.992	0.9825	0.9752

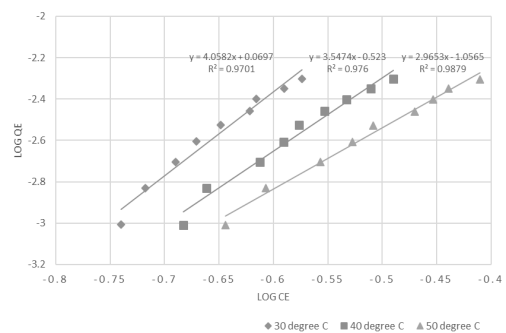


Figure 8. Freundlich adsorption isotherm plots for the adsorption of dye at 30 °C, 40 °C, and 50 °C.

green at 30 °C, 40 °C, 50 °C were on the order of 0.2464, 0.2819, and 0.3372 mg/mg, respectively. It was shown that the n values were $n < 1$, which suggested that malachite green dye adsorption on *L. leucocephala* biomass was favorable. Higher values of the correlation coefficient (R^2) indicated that the Freundlich isotherm can be applied for the adsorption of malachite green on *L. leucocephala* (Table 2). This suggested multilayer adsorption of the adsorbate on the adsorbent particles.

Table 2. Adsorption capacity of malachite green with different types of adsorbents.

No.	Adsorbent	Adsorption capacity, (mg/g)	Equilibrium time, min	Temperature, °C	Dose of adsorbent, g/l	BET surface area analysis, m ² /g	Reference
1.	<i>Avena sativa</i> (oat) hull	83.0	80	40	0.25	63.0	Banerjee <i>et al.</i> (2016)
2.	Waste pea shells	7.04	40	35	0.24	-	Khan <i>et al.</i> (2014)
3.	Sugarcane dust	4.88	14	25	2.5	-	Khatti and Singh (1999)
4.	Neem sawdust	4.35	16	25	0.25	-	Khatti and Singh (2009)
5.	<i>Leucaena leuco-cephala</i>	2.389	60	30	0.5	0.5724	This work
6.	Tamarind fruit shell	1.951	60	30	30	-	Saha <i>et al.</i> (2010)

3.7.3 Temkin model

A plot of Q_e vs. $\ln C_e$ gave a straight line, slope of b , and intercept c (Figure 9). All of the related parameters were calculated (Table 1). The b (J/mol) is the Temkin constant related to the heat of sorption, whereas a (l/g) is the equilibrium binding constant corresponding to the maximum binding energy. The best fit of the adsorption to the Temkin isotherm was at 30 °C with R^2 value of 0.992.

3.8 Comparisons

Table 2 shows the comparison of adsorption capacities between *L. leucocephala* biomass and other types of adsorbents for the sequestration of malachite green dye from contaminated water. Maximum adsorption capacities differ from as low as 1.951 mg up to 83.0 mg per gram of adsorbent. The data clearly showed the capability of *L. leucocephala* biomass as an adsorbent for malachite green dye to be considerably low compared to the others (Table 2). Its adsorption capacity was 2.389 mg/g which was only slightly higher from the tamarind fruit shell based adsorbent which was placed at the bottom. Chemical modification needs to be done to the adsorbent surface to increase the adsorption site for the dyes.

4. Conclusions

The use of *L. leucocephala* biomass as an adsorbent for malachite green dye has been experimentally evaluated. The results showed that the adsorption of dye depends on the contact time, temperature, pH, adsorbent size, and adsorbent dose. Adsorption percentage tends to increase with contact time with the equilibrium between the adsorbate and adsorbent was practically achieved in 60 min. The Langmuir isotherm model, Freundlich isotherm model, and Temkin isotherm model were found to fit well with the adsorption system as their R^2 values were quite near to 1. However, the Temkin isotherm had the best fit, followed by the Freundlich isotherm. The negative value of the Langmuir isotherm constant indicated that the adsorption process did not fit well. Therefore, this study concludes that the *L. leucocephala* biomass has the potential to be a low cost adsorbent for removing cationic dye from aqueous solutions. It can be employed as an alternative adsorbent to the current expensive methods of removing dyes from textile effluents.

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