Turkish J. Eng. Env. Sci.
36 (2012) , 161 – 169.
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doi:10.3906/muh-1108-6

Removal of methylene blue from aqueous solution using alkali-modified malted sorghum mash

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Received: 16.08.2011

Abstract

The removal of methylene blue (MB) from aqueous solution by NaOH-modified malted sorghum mash (NSM) at different contact times, initial solution pH, adsorbent dose, temperature, and initial dye concentration was investigated. The adsorption of MB by NSM increased with increasing contact time and the process attained equilibrium within 18 min. The extent of dye removal decreased with increasing adsorbent dosage but increased with increase in temperature. The adsorption isotherm was found to follow the Langmuir model. The maximum monolayer adsorption capacity obtained was 357.1 mg/g at 306 K. The estimated adsorption free energy (ΔG°), enthalpy change (ΔH°), and entropy change (ΔS°) for the adsorption process were –20.85 kJ/mol, 38.31 kJ/mol, and 68.16 J/mol K, respectively, at 306 K. The adsorption process was feasible, endothermic, and spontaneous. The low value of ΔG° suggests a physical adsorption process, while the positive value of ΔS° reflects the affinity of NSM for MB.

Key Words: Adsorption, methylene blue, malted sorghum mash, Langmuir isotherm

1. Introduction

The use of dyes by industries such as textile, paper, printing, leather tanning, food processing, and electroplating may lead to the production of coloured wastewater (McKay et al., 1998). Allen et al. (2004) estimated that about 84,000 t of dyes produced annually are discharged from the operation of manufacturing, textile, and associated industries. The presence of colour in wastewater, even in low concentrations, is highly visible and undesirable (Robinson et al., 2001).

The conventional methods for the treatment of coloured wastewater include chemical oxidation, coagulation and flocculation, ozonation, electrochemical techniques, fungal decolourisation, and adsorption. Adsorption is preferred to the other techniques in terms of initial cost, flexibility and simplicity of design, ease of operation, and insensitivity to toxic pollutants (Uddin et al., 2009; Santhi et al., 2010). Activated carbon is the most

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widely used adsorbent in water and wastewater treatment. However, activated carbon is expensive and problems of regeneration and difficulty in separation from the water after use promoted the research into low-cost adsorbents (Uddin et al., 2009).

The capability of low-cost adsorbents to remove dyes from aqueous solution has been investigated by researchers. Some materials that have been studied include jackfruit leaf powder (Uddin et al., 2009), water hyacinth roots (Low et al., 1995), rich husk (Jain et al., 2006), wheat shells (Buluk and Aydin (2006), pineapple stems (Hameed et al., 2009), teak tree bark powder (Patil et al., 2010), and borassus bark (Arivoli et al., 2009).

Malted sorghum grains are used to brew traditional opaque beers in many African countries including Ghana, Nigeria, Cameroon, Benin, Togo, Burkina Faso, and Mali (Sawadogo-Lingani et al., 2010). Malted sorghum is specifically used to produce popular traditional beer called pito in the 3 regions of northern Ghana. The mash left after brewing pito is usually fed to ruminants or disposed of indiscriminately. The main aim of this study was to investigate the feasibility of adding value to malted sorghum mash by using it as an adsorbent for the removal of methylene blue (MB) from aqueous solution. MB is not an extremely hazardous dye. However, acute exposure to the dye may cause increased heart beat, vomiting, shock, jaundice, cyanosis, quadriplegia, and tissue necrosis in humans (Vadivelan and Kumar, 2005).

2. Materials and methods

2.1. Materials

Fresh malted sorghum mash was obtained from a local malted sorghum beer (pito) brewer at Navrongo, Ghana. The mash was washed several times with tap water to completely remove dirt and colour, and rinsed twice with distilled water. The mash was air-dried for 2 h, dried to constant weight in a hot oven at 378 K, ground, and sieved through 0.6 mm sieve. The pH of the mash in aqueous phase was determined by mass titration technique and found to be 7.3 (Fiol and Villaescusa, 2009).

Then 25 g of mash was put in a 1 L beaker and 250 mL of 0.1M NaOH added. The beaker with its contents was agitated at 100 rpm for 4 h on a mechanical shaker. The alkali-modified mash was filtered, washed thoroughly with distilled water until the pH of the wash was about 7, and dried to constant weight in a hot oven at 343 K. The dry mash was ground, kept in an air-tight container, and labelled NaOH-modified malted sorghum mash (NSM).

Ninety-eight percent pure analytical grade MB (C.I. = 52,015) produced by Merck, Darmstadt, Germany, was used for this study. The formula, molecular weight, and maximum wavelength of MB was $C_{16}H_{18}ClN_3S$, 319.85 g/mol, and 662 nm, respectively. The dye was used without further purification.

2.2. Experimental methods

MB stock solution containing 2000 mg/L was prepared and dilute solutions needed for the adsorption experiments were prepared from the stock solution. Distilled water was used for the preparation of all reagents. The pH of the solution was adjusted with 0.1 M HCl or 0.1 M NaOH and read using a Crison Basic 20 pH meter. The pH meter was standardised with buffer solution at pH 4, 7, and 12. All experiments, unless specified otherwise, were carried out at average room temperature of 306 $\pm$ 1 K. Except for the study of the effects of adsorbent dose and batch adsorption isotherm, the weight of NSM and volume of MB used for each experiment were 0.1 g and 25 mL, respectively. Erlenmeyer flasks of 250 mL capacity containing a mixture of NSM and

MB were shaken at 100 rpm for 25 min. At the end of the adsorption period, the content of each flask was centrifuged at 4000 rpm for 5 min. The absorbance of the residual dye left after the adsorption process was read at a wavelength of 662 nm using a Jenway 6305 UV/visible spectrophotometer. The amount of MB adsorbed by ACLP was calculated using the equation

$$Q_e = (C - C_e)V/M \tag{1}$$

where Q_e is the amount of MB adsorbed (mg/L) and C (mg/L) and C_e (mg/L) are the initial and equilibrium liquid-phase concentration of MB, respectively; V (L) is the volume of MB solution and M (g) is the weight of NSM used.

The effect of contact time on the amount of MB adsorbed was studied at the initial MB concentrations of 10 and 15 mg/L. The effect of initial solution pH on adsorption of MB by NSM was investigated from pH 2 to 12. The initial concentrations of MB used were 20 and 30 mg/L. The effect of adsorbent dosage on the adsorption of MB was studied with NSM dose ranging from 0.1 to 1.0 g. The volume of MB used was 25 mL in each case. To investigate the effect of temperature on adsorption, Erlenmeyer flasks containing a mixture of NSM and MB were put in a thermostatic water bath at 306 K and 323 K and shaken intermittently for 25 min. Study of the batch adsorption experiments at pH 7 was done with MB concentrations ranging from 200 to 1000 mg/L. Langmuir and Freundlich equations were used to model the adsorption data.

3. Results and discussion

3.1. Effect of contact time

The effect of contact time on the adsorption of MB from aqueous solution by NSM is presented in Figure 1. The amount of MB adsorbed (mg/g) was rapid for the first 5 min with more than 90% of dye in solution removed by NSM. Removal of dye by the adsorbent beyond this time proceeded at a slower rate, finally attaining saturation in about 18 min. Further agitation beyond this optimum time did not lead to an increase in the quantity of dye adsorbed. Adsorption density was directly proportional to the concentration of MB in solution. The rapid adsorption rate during the initial period (the first 5 min) may be due to an increased number of vacant sites available. This led to an increased concentration gradient between MB in solution and MB in adsorbent surface (Uddin et al., 2009). The short time for equilibrium, coupled with a high removal, indicates a high affinity of NSM for MB, indicating a probable chemical adsorption process (Hameed, 2009).

3.2. Effect of initial solution pH

The initial pH of solution affects the adsorption capacity. The effect of initial solution pH on adsorption is shown in Figure 2. The adsorption density of MB onto NSM increased from pH 2 to 4. The adsorption density was approximately constant between pH 4 and 10, and decreased slightly from pH 10 to 12. The decrease in MB adsorbed below pH 4 is probably because the NSM surface was positively charged, making H^+ ions compete effectively with MB cation leading to a decrease in the adsorption density (Hameed et al., 2009). The pH at the point of zero charge (pH_{pzc}) for the unmodified adsorbent was 7.3. The modification of the adsorbent to produce NSM depressed the pH_{pzc} and increased the pH range over which the adsorbent might take up the dye more effectively. At pH values higher than pH_{pzc}, the surface of NSM became negatively charged and the adsorption of positively charged MB was enhanced through electrostatic force attraction (Ncibi et al., 2007).

Electrostatic force attraction might not fully explain the approximately constant adsorption density from pH 4 to 10; it is possible that ion exchange was involved in the adsorption process (Bulut and Aydın, 2006).

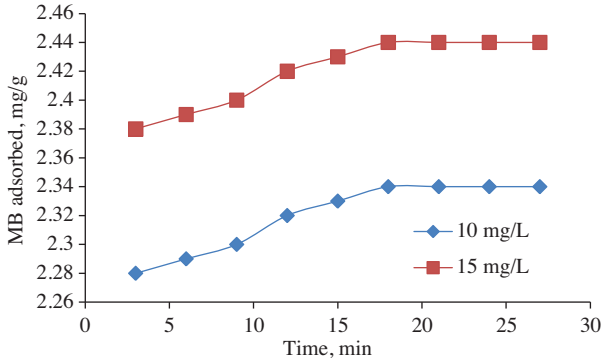

Figure 1. Effect of contact time on the adsorption of MB onto NSM (T = 306 K, V = 0.025 L, M = 0.1 g).

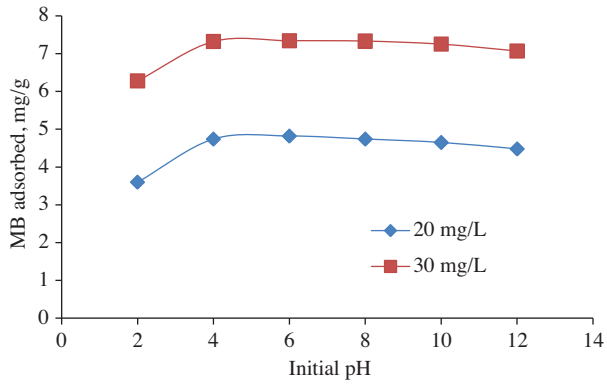

Figure 2. Effect of initial solution pH on the adsorption of MB onto NSM (T = 306 K, V = 0.025 L, M = 0.1 g).

3.3. Effect of NSM dosage

Figure 3 presents the equilibrium adsorption of MB against NSM dose. The adsorption of dye varied with varying adsorbent mass and it increased with increase in adsorbent dose. The amount of MB removed from aqueous solution by NSM increased rapidly with increase in adsorbent dose up to 0.5 g, then gradually increased with further increment in adsorbent dose up 0.7 g and thereafter remained unchanged. The dye adsorbed increased from 84.9% to 92.8% as the adsorbent dose increased from 0.1 to 1.0 g. The increase in amount of dye adsorbed was due to the increase in the available adsorption surface sites. However, the adsorption density decreased from 10.6 to 1.2 mg/g as the adsorbent dose increased from 0.1 to 1.0 g. At higher adsorbent to adsorbate concentration ratios, there was a very fast superficial adsorption of MB onto the surface of NSM that produced a lower solute concentration in the solution than when the biomass to solute concentration ratio was lower. The decrease in equilibrium adsorption density (mg/g) with increasing adsorbent mass was due to the concentration gradient between MB concentration in aqueous solution and MB concentration at the surface of the adsorbent (Uddin et al., 2009).

3.4. Effect of temperature and initial MB concentration

Temperature influences the adsorption of an adsorbate from aqueous solution. A study of the temperature dependence of adsorption reactions gives valuable information about important thermodynamic parameters such as enthalpy and entropy changes during adsorption. Figure 4 shows the effects of temperature and initial concentration on adsorption of MB by NSM. The adsorption of MB by NSM increased with increase in temperature. The dye adsorbed increased from 42.6 to 314.2 mg/g when the adsorption experiment was conducted at 306 K. However, the dye adsorbed rose from 45.8 to 329.1 mg/g when the experiment was conducted at 323 K. The fact that the adsorption of dye was in favour of temperature indicates that the mobility of the dye molecule increased with increase in temperature. This implies that the dye molecule should interact more effectively with the adsorbent surface with rise in temperature. The increase in adsorption capacity with increasing temperature suggests that the process of removal of MB by NSM is endothermic in nature.

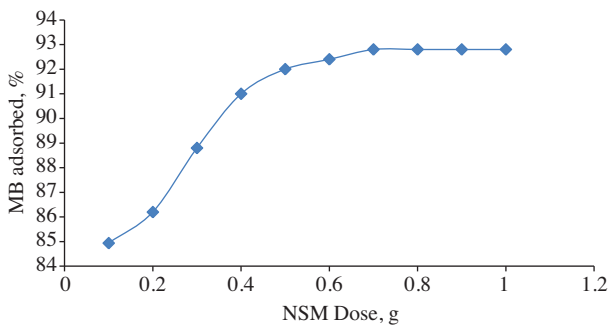

Figure 3. Effect of adsorbent dosage on the adsorption of MB onto NSM (T = 306 K, C_0 = 50 mg/L, V = 0.025 L, pH 7).

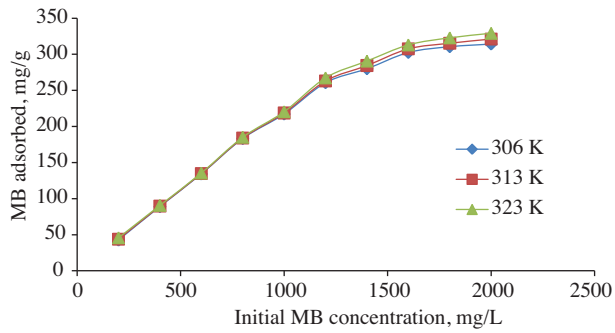

Figure 4. Effects of temperature and initial concentration on the adsorption of MB onto NSM (V = 0.025 L, pH 7).

Increase in initial MB concentration led to an increase in the adsorption of the dye by NSM and this trend was especially clear at high initial MB concentrations. The initial rapid removal of the dye from aqueous solution can be attributed to the concentration gradient created at the start of the adsorption process between MB in solution and that at the NSM surface. The concentration gradient decreased as the dye loading increased on the adsorbent and gave way to slower dye uptake (Oladoja et al., 2008).

3.5. Adsorption isotherms

Adsorption isotherms are often employed to characterise adsorption of solutes onto adsorbents from aqueous solutions. This is necessary to establish the most appropriate correlations for the equilibrium data in the design of adsorption systems (Hameed et al., 2009). The Langmuir and Freundlich isotherms were employed to analyse the results of the equilibrium isotherm based on Figure 4. The Langmuir model of adsorption assumes that the intermolecular forces decrease rapidly with distance, and, consequently, predicts the existence of monolayer coverage of adsorbate at the outer surface of the adsorbent, which is assumed to be homogeneous (Langmuir, 1918). The linear form of the Langmuir equation is:

$$\frac{C_e}{Q_e} = \frac{1}{bQ_m} + \left(\frac{1}{Q_m}\right) C_e \tag{2}$$

where Q_e (mg/g) is the amount of adsorbate adsorbed at equilibrium, C_e (mg/L) is the equilibrium concentration, Q_m (mg/g) is the mass of adsorbed solute required to saturate a unit mass of adsorbent, and b is the Langmuir constant related to rate of adsorption. The constants Q_m and b can be determined by plotting C_e/Q_e against C_e. This gives a straight line slope of $1/Q_{max}$ and intercept of $1/bQ_m$. The Langmuir adsorption isotherm plot is presented in Figure 5.

The constants Q_m and b can be determined by plotting C_e/Q_e against C_e. This gives a straight line slope of $1/Q_m$ and intercept of $1/bQ_m$. The Langmuir adsorption isotherm plot is presented in Figure 5. The R^2 value (0.9979) implies that the Langmuir isotherm provides a good mathematical fit to the isotherm data. This may be due to the homogeneous distribution of active sites onto the NSM surface (Wang et al., 2005).

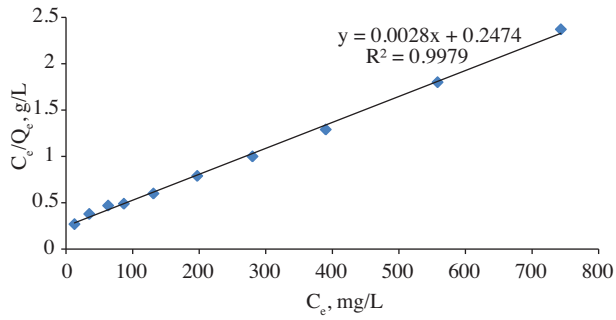

Figure 5. Langmuir isotherm for MB adsorption onto NSM (T = 306 K).

The essential features of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor, R_L (Hameed et al., 2009), which is defined as:

$$R_L = 1/(1 + bC_0) \tag{3}$$

where C_0 (mg/L) is the highest initial concentration of adsorbent and b (L/mg) is the Langmuir constant. The value of R_L is used to indicate the nature of shape of the isotherm as follows: $R_L > 1$ (unfavourable adsorption), $R_L = 1$ (linear adsorption), $1 > R_L > 0$ (favourable adsorption), and $R_L = 0$ (irreversible adsorption). The calculated values of Q_m, b, R^2, and R_L at different temperatures are presented in Table 1.

Table 1. Langmuir and Freundlich isotherm constants for adsorption of MB onto NSM at different temperatures.

	Langmuir isotherm constants						Freundlich constants		
T (K)	Q_m (mg/g)	b (L/mg)	b (L/mol)	R^2	R_L		K_F	n	R^2
306	357.1	0.01132	3621	0.9979	5.0×10^{-4}		17.98	2.12	0.9305
313	370.4	0.01103	3528	0.9981	5.0×10^{-4}		18.00	2.11	0.9329
326	384.6	0.01126	3602	0.9958	5.0×10^{-4}		17.62	2.07	0.9331

The Freundlich isotherm assumes heterogeneous surface energies in which energy varies as a function of the surface coverage. The linearised form of the Freundlich equation is:

$$\log Q_e = \log K_F + \left(\frac{1}{n}\right) \log C_e \tag{4}$$

A plot of $\log Q_e$ against $\log C_e$ (Figure 6) allows for the determination of K_F and n. K_F is a Freundlich constant that shows the adsorption capacity of an adsorbent, and 1/n is a constant that describes the adsorption intensity. The R^2 value (0.9305) for the Freundlich isotherm is lower than that obtained for the Langmuir isotherm. The calculated values of K_F, n and R_2 at different temperatures are presented in Table 1. The values of n ($n > 1$) indicated favourable adsorption.

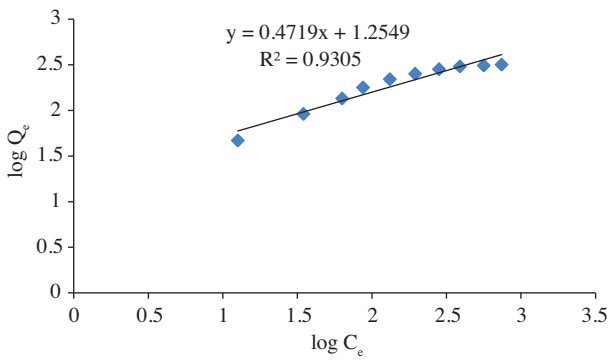

Figure 6. Freundlich isotherm for MB adsorption onto NSM (T = 306 K).

3.6. Thermodynamic parameters

The free energy of adsorption (ΔG°) can be related with the equilibrium constant K (L/mol) corresponding to the reciprocal of the Langmuir constant, b, by the following equation:

$$\Delta G^\circ = -RT \ln b, \tag{5}$$

where R is the gas universal constant (8.314 J/mol K), T is the absolute temperature, and b (L/mol) is the Langmuir constant. The enthalpy change (ΔH°) and entropy change (ΔS°) can be estimated using the following equation:

$$\ln b = \Delta S^\circ / R - \Delta H^\circ / RT \tag{6}$$

ΔH° and ΔS° can be estimated from the slope and intercept of the straight line plot of ln b against 1/T. The estimated values of ΔG°, ΔH°, and ΔS° are presented in Table 2. The values of ΔG° were negative at all temperatures studied, which confirms the feasibility of the process and the spontaneous nature of MB adsorption onto NSM. The values of ΔG° were low, suggesting a physical adsorption process (Silva et al., 2005). The estimated enthalpy change (ΔH°) and entropy change (ΔS°) of adsorption were 38.31 kJ/mol and 68.16 J/mol K, respectively. The positive value of ΔH° (38.31 kJ/mol) indicates that the adsorption of MB onto NSM was an endothermic process. The positive value ΔS° (68.16 J/mol K) reflects the affinity of the adsorbent for the dye and increased randomness at the solid-solution interface during adsorption (Chowdhury et al., 2011).

Table 2. Values of thermodynamic parameters for the adsorption of MB onto NSM.

T (K)	ΔG° (kJ/mol)	ΔH° (kJ/mol)	ΔS° (J/mol K)
306	–20.85		
313	–21.26	38.31	68.16
326	–22.00		

3.7. Adsorption capacities of adsorbents for MB

Researchers have earlier investigated the feasibility of using other low-cost materials for removing MB from aqueous solution. The maximum monolayer adsorption capacities of MB onto various adsorbents of plant origin are listed in Table 3. The adsorption capacity of NSM (357 mg/g) is very large, implying that it could be a promising low-cost adsorbent for the removal of MB from aqueous solution.

Table 3. Adsorption capacities of various adsorbents for MB.

Adsorbent	Q_m (mg/g)	Reference
Neem leaf powder	8.76	Bhattacharyya and Sarma (2003)
Wheat shells	16.56	Bulut and Aydın (2006)
Orange peel	18.6	Annadurai et al. (2002)
Banana peel	20.80	Annadurai et al. (2002)
Rice husk	40.58	Vadivelan (2005)
Pineapple stem	119.05	Hameed et al. (2009)
Dehydrated peanut hull	123.5	Ozer et al. (2007)
Cedar sawdust	142.36	Pengthamkeerati et al. (2008)
Jackfruit leaf powder	326.32	Uddin et al. (2009)
Teak tree bark powder	333.33	Patil et. al. (2011)
Grass waste	457.64	Hameed (2009)
NaOH-modified malted sorghum mash	357.1	This study

4. Conclusion

The present study shows that NSM could be an effective adsorbent for removing cationic dyes like MB from aqueous solution. The equilibrium data were analysed using the Langmuir and Freundlich isotherms and the former model provided a better mathematical fit. The maximum monolayer adsorption capacity obtained was 357.1 mg MB/g NSM at 306 K. This value is high if compared to the results of other workers who investigated the use of plant-based materials to remove MB from aqueous solution. The adsorption process was feasible, endothermic and spontaneous with free energy (ΔG°) of 17.23 kJ/mol at 306 K. The estimated enthalpy change (ΔH°) and entropy change (ΔS°) for the adsorption process were 38.31 kJ/mol and 68.16 J/mol K. The low value of ΔG° suggests a physical adsorption process, while the positive value of ΔS° reflects the affinity of NSM for MB and increased randomness at the solid-solution interface during adsorption.

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