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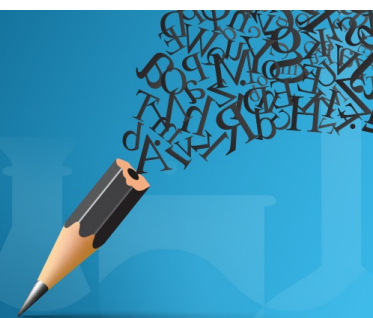


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***Leucaena leucocephala* Biomass Treated with Sodium Dodecylbenzene sulfonate (SDBS) as an Adsorbent for Malachite green uptake from Contaminated Water**

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Abstract. Dyes are significant pollutant of our clean water. Adsorption using activated carbon uses lots of energy in the production stage. Thus, it is an expensive choice for contaminant clearance from wastewater. Modification of biomass particles using suitable chemical can produce adsorbent with lower cost. In this study, the *Leucaena leucocephala* biomass was treated with sodium dodecylbenzene sulfonate (SDBS) as an adsorbent for Malachite green uptake from contaminated water. Batch adsorption studies were done to study the effect of various parameters on the adsorption percentage of the prepared adsorbent. The results showed that the optimum values evaluated for each adsorption parameters are 20 ppm for initial adsorbate concentration, 60 minutes contact time, 30 °C for solution temperature, pH of 7, adsorbent particle size of 0.425 – 0.5 mm and 0.5 g/L adsorbent dosage. The results showed that SDBS treated *Leucaena leucocephala* has the potential to be used as a low-cost adsorbent for Malachite green dye from contaminated water.

INTRODUCTION

Dyes are one of the largest polluters in the modern world and become common industrial environmental pollution. Dyes are widely used in industries such as textiles, paints, rubber, cosmetics, paper, leather, plastics, food and drug [1]. Dyes are used mostly in textiles industries during their synthesis and later during fibre dyeing [2]. Textiles industries consumed more than 60 % dyes of the total world production and it was estimated that 10-20 % of dye was lost during the dyeing process and released as effluent. There are various types of dyes used in manufacturing industries such as solvent dyes, azoic dyes, direct dyes, sulphur dyes, vat dyes, mordant dyes, ingrain dyes, metal complex dyes, acid dyes and disperse dyes [3]. The obvious problem with colored water is that it is not acceptable to the public. The public prefers clear, uncolored water [4]. The color is the first contaminant to be recognized in wastewater [5]. The presence of even very small amounts of dyes in water, less than 1ppm for some dyes is highly visible and undesirable [6]. Apart from the aesthetic point of view by public, dyes are undesirable because they can give bad effect to living organisms in the water discharged. It will decrease light penetration in stream due to synthetic dyes from industrial sewage. Dyes in water affect the aquatic flora for their photosynthetic activities and affecting the food source of aquatic organisms. The thin layers of discharged dyes that can form on the

surfaces of the waters also reduce the amount of dissolved oxygen [7]. Depletion of dissolved oxygen (DO) in water is the most serious effect of textile waste as dissolved oxygen is very essential for aquatic life. This effluent also will flow to the fields, and then it clogs the pores of the soil resulting in loss of soil productivity [8].

One of the techniques used in water treatment is adsorption. Adsorption on activated carbon is a well-known method for the treatment of water and wastewater [9]. However, the high cost of material is limits its use in wastewater treatment. Therefore, in recent years, innovative adsorption using locally low cost adsorbent for dyes removal has been extensively investigated. But, still many new economical, easily available and highly effective adsorbents are still needed.

Leucaena leucocephala is ideal as a low-cost adsorbent. On the other hand, the quick growing and short rotation of *Leucaena leucocephala* trees is some of the requirement for biomass. Currently, some researchers have studied on the effectiveness of *Leucaena leucocephala* seed pods as the removal of Congo red (acidic dye) from aqueous solutions at room temperature. It has been observed from experiments that adsorbent prepared from seed pods of *Leucaena leucocephala* plant was effective for the removal of Congo red from aqueous solutions at room temperature [10]. Research was also done on the studied of the effectiveness of *Leucaena leucocephala* seed pods as the removal crystal violet from aqueous solution. Adsorbent were successfully used to remove the crystal violet from an aqueous solution in a batch process. In this research, *Leucaena leucocephala* biomass were treated with Sodium Dodecyl benzene sulfonate (SDBS) to improve the effectiveness of the *Leucaena leucocephala* biomass as adsorbent for dye and increasing the number of bind particles on adsorbent. Batch adsorption studies were done to evaluate the effect of each parameters to the adsorption percentage.

MATERIALS AND METHODS

Sample Preparation

Samples of *Leucaena leucocephala* trunk were collected in Kuala Kangsar, Perak, Malaysia. The collecting process was done randomly in the areas where the abundance growth of *Leucaena leucocephala* tree was founded. After collection, the samples were washed with distilled water several times to remove all the dirt particles. The trunk samples are needed to be making sure free from any impurities that might alter the accuracy of the result. Then, the portions were going through the process of chipping, drying and grinding. Samples were dried in an oven before used [11].

Adsorbent Preparation

The adsorbent of *Leucaena leucocephala* wood particles was prepared and done according to the method by Wan Ibrahim, Mohamad Amini [12]. Sodium Dodecylbenzene sulfonate ($\text{CH}_3(\text{CH}_2)_{11}\text{C}_6\text{H}_4\text{SO}_3\text{Na}$) (SDBS) was used as a modifier of the sawdust. Every ten grams of *Leucaena leucocephala* sawdust were mixed with 100 ml of 2% SDBS and were stirred using a magnetic stirrer for 24 hours. The sawdust was filtered to remove the remaining solution. Later it was rinsed with distilled water for several times to remove excessive surfactant. Treated sample was dried inside an oven at 70 °C. Treated particles were sieved into a different range of sizes at 0.075 – 0.15 mm, 0.15 – 0.425 mm, 0.425 – 0.5 mm and 0.5 > mm sieve sizes and stored for further use.

Adsorbate Preparation

Basic dye that was used in this research is Malachite green. Malachite green has the molecular formula $\text{C}_{23}\text{H}_{25}\text{ClN}_2$ (364.911 g/mol). In order to prepare the adsorbate, the dye stock solution was prepared by dissolving weighed dye accurately in distilled water to the initial concentration of 1000 ppm. Malachite green was diluted in respective volumetric flasks. The required experimental solutions were produced by diluting the 1000ppm dye stock solution in by serial dilution until needed initial concentrations was obtained [13].

Batch Adsorption Studies

Adsorption is an effective method by lowering the concentration of the dissolved dye in the effluent, in turn, removing colour [14]. Batch adsorption was made to determine the capacity of dye adsorption. Before conducting the studies, six parameters were determined. Malachite green stock solutions were diluted from 1000 ppm into the concentration of 10, 20, 30, 40 and 50 ppm, respectively. Dye stock solutions with dye concentration of 10, 20, 30, 40 and 50 ppm were prepared in a separated conical flask. A 50 ml solution for each concentrations were mixed with the adsorbent at 0.5 g/L. The samples were then shaken at 130 RPM at a temperature of 30 °C inside a computer controlled incubator shaker. At the end of predetermined time interval, mixtures was filtered and treated solutions were taken out by using a micropipette. Then, the filtrates solutions were analysed by using a spectrophotometer, DR500-UV-VIS for the final concentration of adsorbate. The wavelength corresponding to maximum absorbance is $\lambda_{\max} = 616 \text{ nm}$. A graph was plotted with adsorption percentage against initial adsorbate concentration. The adsorption percentage was expressed as;

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

Other parameters are initial adsorbate concentration, contact time, temperature, pH of adsorbate, size of adsorbent particles and dosage of adsorbent as shown in Table 1.

TABLE 1. Parameters for batch adsorption studies

Parameter	Values						
Initial adsorbate concentration (ppm)	10	20	30	40	50		
Time (minutes)	30	60	90	120			
Temperature (°C)	30	40	50	60			
pH of adsorbate	2	4	6	7	8	10	12
Size of adsorbent particles (mm)	0.075 – 0.15		0.15 – 0.425		0.425 – 0.5		0.5 >
Dosage of adsorbent (g/L)	0.5	1.0	1.5	2.0	2.5		

RESULTS AND DISCUSSION

Standard Calibration Curve

Dye stock solutions of Malachite green were prepared in 1000 ppm by dissolving 1.0 g of dyes into 1000 mL of distilled water. Then, Malachite green stock solutions were diluted from 1000 ppm into a concentration of 10, 20, 30, 40 and 50 ppm, respectively. Their optical densities for each concentration were then found out by using DR 500 UV-Vis spectrophotometer ($\lambda_{616\text{nm}}$). All the values were obtained, and a standard calibration curve was plotted, as shown in Fig. 1.

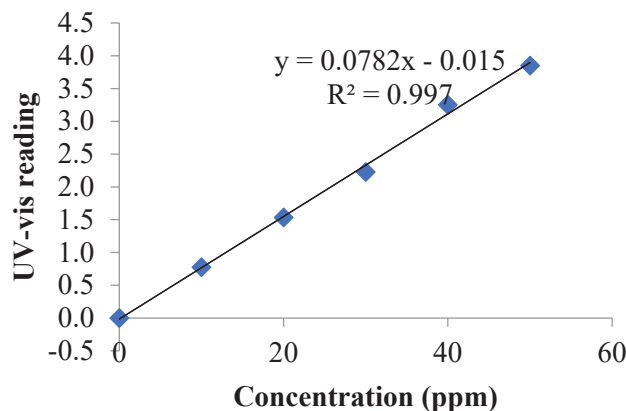


FIGURE 1. The standard calibration curve of Malachite green

Calibration is the key to accurate data. Calibration curves were made based on a linear relationship that can be expressed using the $y = mx + c$, which is an equation for a straight line. In this equation, y is the optical densities by using DR 500 UV-Vis spectrophotometer (λ_{616nm}), x is the concentration, m represents the slope of the line, and c is the y -intercept. The slope and intercept of that line provide a relationship between absorbance and concentration. From the graph obtained, the equation coefficient value of the calibration curve is 0.997. The calibrated results show the value is approximate to 1.0, which are very useful in identifying the capability of removing Malachite green dye.

Batch Adsorption Studies

Impact of Initial Adsorbate Concentration

In this study, the results were obtained at five different initial adsorbate concentrations: 10, 20, 30, 40 and 50 ppm. Figure 2 illustrates the effect of different initial adsorbate concentration on the adsorption of Malachite green. From the result obtained, it is clear that the adsorption percentage is increasing rapidly in the initial stage and become a slower decrease in later stages. Maximum percentages removal, 72.63% is observed at around 20 ppm initial adsorbate concentration. Further increase in initial adsorbate concentration at 30, 40, and 50 ppm does not show big change the adsorption yield. This is mainly due to almost all adsorbate ions had bind to the adsorbent surface. Equilibrium between the adsorbate on the adsorbent and in the solution also can be assumed to be achieved after 20 ppm. The removal efficiency of the adsorbents decreased with increasing initial dye concentration [15]. Therefore, in the following experiment, the initial adsorbate concentration was fixed at 20 ppm.

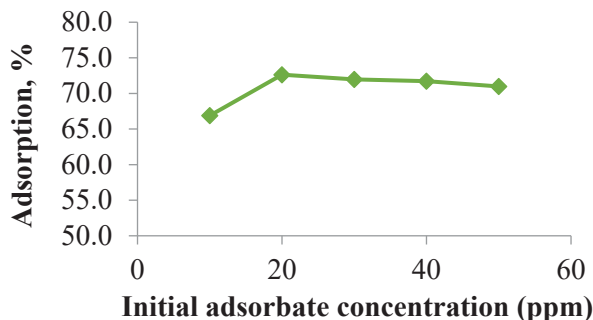


FIGURE 2. The effect of adsorption against the different initial concentration of adsorbate.

Impact of Contact Time on the Adsorption

The effect of contact time on the amount of Malachite green adsorbed was investigated at four different contact times, 30, 60, 90 and 120 minutes by using 20 ppm determined initial concentration of Malachite green. Data obtained from experiments are presented in Fig. 3. From the result obtained, it is revealed that the percentage removal for 20 ppm concentration is 72.34% at 60 minutes compared to 64.35 % at only 30 minutes. The graph showed the adsorption for 20 ppm dye concentration is very fast in the initial stages and later slows down until saturation is achieved. The percentage of dye removal did not vary significantly at 60 minutes onwards which could be assumed as an equilibrium point. Fast diffusion of metal ions on the external surface and later into the intra-particle matrix, results in rapid equilibrium. After equilibrium point, the active site was saturated, thus no further adsorption can take place [16]. For the optimum removal of Malachite green, the solutions should be treated for 60 minutes regardless of their initial concentrations. Therefore, in the following experiment, the time was fixed at 60 minutes.

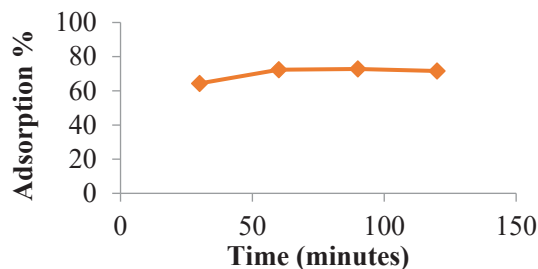


FIGURE 3. The effect of adsorption against different contact time

Impact of Temperature

Figure 4 illustrates the effect of temperature on the adsorption of Malachite green. The temperature was a crucial parameter that affects the adsorption process. The graph showed decrement in removal efficiencies with the increment of reaction temperature. The weakening of the bonds between the adsorbate molecules and the binding sites of the adsorbent could be the main factor resulting this phenomenon. This observation also suggests that the adsorption process of Malachite green by the adsorbent is kinetically controlled by an exothermic process. Similar finding have been reported in the previous literature for adsorption of Malachite green by tamarind fruit shell [17]. Therefore, in the following experiment, the temperature was fixed at 30°C.

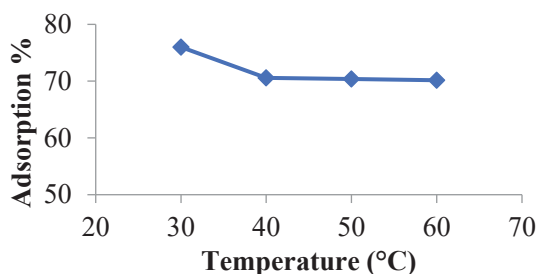


FIGURE 4. The effect of adsorption against temperature

Effect of pH

The solution pH is also a vital parameter to be considered in an adsorption process. The pH value influences the surface charge of the adsorbent, besides affecting the degree of ionization of the adsorbate species present in the solution. In an aqueous solution, Malachite green is a cationic dye that exists in the form of positively charged ions. Because of it is a charged species, the rate of adsorption onto the adsorbent surface is mainly influenced by the surface charge on the adsorbent, which also influenced by the solution pH [18]. Therefore, in the present investigation, batch experiments as described above were carried out in the pH range 2–12. Figure 5 showed the results of the adsorption at different pH. The adsorption percentage was observed to be rapidly increasing from pH 2 to pH 7. At pH 7 to pH 10, no significant change in adsorption percentage was observed. The maximum Malachite green removal takes places at pH 7. At lower pH, amine and carboxylic groups may take $-\text{NH}_3^+$ and $-\text{COOH}$, respectively. At pH 2, the surface of adsorbent is positively charged, therefore the H^+ ions was competing with dye cations, hindering them to attached to the surface, thus causing a decrease in dye removal. Meanwhile, amine and carboxylic groups may take $-\text{NH}_2$ and $-\text{COO}^-$ at higher pH. Generally, at highly acidic pHs, binding of positively charged adsorbate ions is reduced due to electrostatic repulsions. The positive nature of the adsorbent and the binding increases with the increase in pH, as the adsorbent becomes negatively charged. The influenced of solution pH on changes in the surface charge may be attributed to adsorption behaviour observed in this study. Therefore, in the following experiment, the pH was fixed at pH 7.

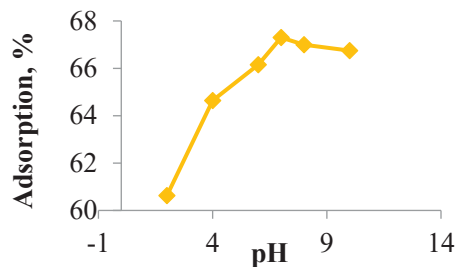


FIGURE 5. The impact of adsorption at different pH

Effect of Adsorbent Particle Size

Figure 6 illustrates the effect of adsorbent size on adsorption behaviour by using four different range size of adsorbent: 0.075 – 0.15 mm, 0.15 – 0.425 mm, 0.425 – 0.5 mm and more than 0.5 mm. As shown in Fig. 6, the size of 0.425 – 0.5 mm was found to be very useful in removal Malachite green from the solution at 89.19%. The amount of adsorbed ions increase as the size of adsorbent became larger. However, these studies show that optimum adsorption can be done using 0.425 – 0.5 mm sizes of the adsorbent. At 0.5 mm sizes and above, the adsorbent was observed as slowly remain constant, and the effectiveness as colour removal slowly decreases. Therefore, in the following experiment, the adsorbent size was fixed at 0.425 – 0.5 mm.

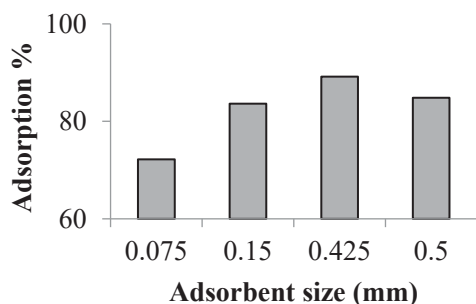


FIGURE 6. The effect of adsorption against adsorbent size.

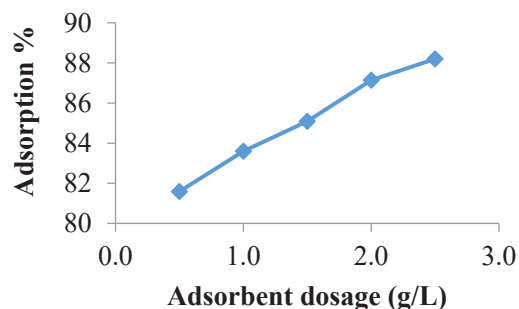


FIGURE 7. The effect of adsorption against adsorbent dosage

Effect of Adsorbent Dosage (g/L)

The adsorbent dosage is also an important parameter to be analyzed. It evaluates the capacity of an adsorbent for a certain initial concentration of adsorbent. The effect of adsorbent dose was studied with keeping all the experimental conditions predetermined based on previous steps. The removal of Malachite green at different adsorbent dosages from 0.5 to 2.5 g/l at initial dye concentration of 20 ppm and pH seven was studied. Figure 7 showed that as the adsorbent dosage increases from 0.5 to 2.5 g/l, the percentage of Malachite green removal was increased from 81.59 to 88.20 %. The percentage of Malachite green removal increase as the adsorbent dosage was increase due to an increase in the total number of binding sites. According to Bhattacharyya and Gupta [19], this is due to the greater availability of the surface area. The 2.5g/l of adsorbent dosage have higher surface area compared to 0.5g/l. The reaction between adsorbate and adsorbent increase, due to the greater surface area of adsorbent react with Malachite green.

CONCLUSION

The *Leucaena leucocephala* biomass was treated with sodium dodecylbenzene sulfonate (SDBS) as an adsorbent for Malachite green uptake from contaminated water in this study. The optimum values evaluated for each adsorption parameters are 20 ppm for initial adsorbate concentration, 60 minutes contact time, 30 °C for solution temperature, pH of 7, adsorbent particle size of 0.425 – 0.5 mm and 0.5 g/L adsorbent dosage. The results showed

that SDBS treated *Leucaena leucocephala* has the potential to be used as a low-cost adsorbent for Malachite green dye from contaminated water.

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