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Original Article

Adsorption of malachite green from synthetic wastewater using banana peel adsorbents

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Abstract

The adsorption of malachite green (MG) from synthetic wastewater on banana peel adsorbent was studied in batch experiments. Two species of banana peel adsorbents (*Musa x paradisiaca*, ABB and *Musa acuminate*, AAA) with two maturation stages (2 and 6) were studied. The best efficiency of MG removal (97.64%) was obtained using the powder of ripe ABB banana peel (stage 6) as the adsorbent under the optimal conditions of pH 5, 12 h equilibrium time, initial dye concentration of 25 mg/L, adsorbent particle size of 0.150-0.212 mm at the dose of 1 g/L, 30 °C, and shaken at the speed of 150 rpm. The experimental results were best described by the Freundlich isotherm model and the pseudo-second-order kinetic model. Thermodynamic results of the adsorption revealed that it was a random, endothermic, and spontaneous process under the investigated temperature range.

Keywords: malachite green, banana peel, equilibrium isotherms, adsorption kinetics, thermodynamics

1. Introduction

Approximately 10-15% of dyes, which is about 100 tons/year, are discharged worldwide from textile industries to water resources (Ferreira, Coutinho, Fernandes, & Freire, 2014; Yagub, Sen, Afroze, & Ang, 2014). This can cause water pollution and toxicity to some aquatic organisms. For example, water pollution interfered with light penetration which resulted in a decrease in photosynthetic activity and the amount of dissolved oxygen in water. Carcinogenic, mutagenic or teratogenic effects in various fish species, and potential damage to human health were also reported (Salleh, Mahmoud, Karim, & Idris, 2011; Yagub *et al.*, 2014).

Malachite green (MG) is a basic or cationic dye that is commonly used for dying of silk, nylon, and acrylics. The dye can be removed from wastewater by many treatment methods such as coagulation-flocculation, ion exchange, and adsorption (Rangabhashiyam, Anu, & Selvaraju, 2013; Seow & Lim, 2016). The interesting advantages of adsorption are high efficiency, less expense, less energy consumed, uncomplicated design, and can operate without difficulty (Seow & Lim, 2016). Therefore, many researchers have attempted to perform dye removal using inexpensive adsorbents, such as sludge, egg shells, chitosan, bottom ash, rich husk, and orange peel (Devi & Saroha, 2017; Fernandez, Nunell, Bonelli, & Cukierman, 2014; Gandhimathi, Remesh, Sindhu, & Nidheesh, 2013; Khalek, Rahman, & Francis, 2017; Kiakhani, Arami, & Gharanjig, 2013; Mane, Mall, & Srivastava, 2007).

Because of high nutrition and easy digestion, bananas are one of the most popular fruits in the world which were approximately 16% of global total fruit production and the second largest produced fruit in the year 2007. Approximately 107 million tons of bananas were produced in 135 countries and territories across the tropics and subtropics in the year 2013 and the highest production was found in Asia (BananaLink Office, 2017; Mohapatra, Mishra, & Sutar, 2010). In addition, banana peel accounts for about 30-40% of the total fruit weight and is composed of lignin (6-12%), pectin (10-21%), cellulose (7.6-9.6%), hemicellulose (6.4-9.4%), and functional groups, such as carboxyl, hydroxyl, and

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amide, which have high tendencies for dye adsorption in wastewater (Liu, Ngo, Guo, & Tung, 2012; Mohammed & Chong, 2014; Mohapatra *et al.*, 2010; Palma, Contreras, Urra, & Martinez, 2011; Tan, Hameed, & Ahmad, 2007). Therefore, this research was interested in the adsorption of MG from synthetic wastewater using banana peel as the adsorbent.

2. Materials and Methods

2.1 Adsorbate

MG has the chemical formula $C_{25}H_{25}ClN_2$ and the molar mass is 364.91 g/mol (Figure 1). MG is a basic dye that was purchased from a dye company in Bangkok, Thailand and was used as the adsorbate in the current study.

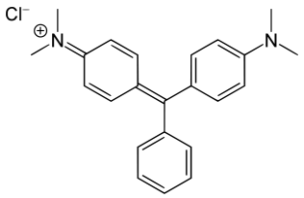

Figure 1. Chemical structure of malachite green.

2.2 Adsorbent

There are two species of banana peel according to the genome group, i.e. *Musa acuminata*, AAA (non–Cavendish) or Kluai Hom Thong and *Musa x paradisiaca*, ABB or Kluai Namwa, that were purchased from a local market and used as adsorbents in this study (Valmayor *et al.*, 2017). Two maturation stages of unripe (stage 2) and ripe (stage 6) banana peel were also compared for their adsorption capacities (Tapre & Jain, 2012). They were cut into small pieces (1-2 mm), washed and dried in a hot air oven at 60 °C until constant weight was achieved. Then, the pieces were ground and sieved to obtain a particle size in the range of 0.150-0.212 mm. The resultant banana peel powder (BPP) was stored in a desiccator for further experiments.

2.3 Adsorption experiment

The MG solubility under different pH (5-9) conditions was tested at the initial concentration of 25 mg/L and the required pH was adjusted by 0.1 M HCl or 0.1 M NaOH. Two species and the two maturation stages of BPP were evaluated as test adsorbents at the dose of 1 g/L, initial concentration of 25 mg/L, and shaken at 150 rpm at 30 °C under a pH range of 5-9. Then the BPP was separated by filtration and the residual dye concentration of filtrate was determined by absorbance measurement using a UV-Vis spectrophotometer (Analytical Lab Science Co., Ltd., Thailand, Model V-530) at 616 nm wavelength. The removal percentage at equilibrium condition was calculated by using the following equation:

$$\text{Dye removal (\%)} = \frac{(C_0-C_e)}{C_0} \text{ X } 100$$

where C_0 is the initial dye concentration (mg/L) and C_e is the final dye concentration (mg/L).

The suitable species and maturation stage of banana peel which provided the best removal percentage of MG was chosen as the adsorbent at the optimal pH for the next experiment. The following adsorption experiments were carried out in batch experiments.

Experiment 1: The effects of contact time and initial concentration of MG solution were evaluated. The adsorption experiments were performed as described in previous studies using initial concentrations in the range between 5-100 mg/L for contact time intervals that ranged from 0 to 24 h.

Experiment 2: The effect of adsorbent dose was evaluated. The adsorption experiments were conducted as in experiment 1 using a fixed amount of BPP (0.25-10 g/L) in the optimal initial concentration of MG at the equilibrium time obtained from experiment 1.

Experiment 3: The effect of particle size of adsorbent was evaluated. The experiments were operated as in experiment 2 using various particle sizes (0.063–1 mm) using the results from experiment 2 to obtain the optimal dose.

Experiment 4: The effect of MG solution temperature was evaluated. The adsorption experiments were done as in experiment 3 using the optimal particle size at MG solution temperatures that ranged 25-40 °C (298.15-313.15 K).

Adsorption isotherm: The experimental data at optimal conditions from experiment 1 were applied to the Freundlich and Langmuir models which can be described by linear equations 1 and 2, respectively (Amel, Hassena, & Kerroum, 2012; Baek, Ijagbemi, O, & Kim, 2010; Hazzaa & Hussien, 2015; Moubarak *et al.*, 2014).

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

where q_e (mg/g) is the amount of MG adsorbed per unit weight of the adsorbent at equilibrium and C_e (mg/L) is the equilibrium concentration of MG in solution. K_F and n are Freundlich constants indicating the adsorption capacity of the adsorbent and adsorption intensity or degree of favorability of adsorption, respectively. The values of K_F (mg/g) $(L/mg)^{1/n}$ and n were calculated from intercept and slope of the plot between log q_e versus log C_e.

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

where q_m (mg/g) is maximum monolayer adsorption capacity; K_L (L/mg) is Langmuir isotherm constant which is a direct measurement of the intensity of adsorption. The values of q_m and K_L were calculated from slope and intercept of the plot between C_e / q_e versus C_e

Adsorption kinetics: The experimental MG adsorbed rates onto the ripe BPP from experiment 1 were investigated by pseudo-first-order and pseudo-second-order kinetic models as expressed in equation 3 and 4, respectively (Ho, 2004; Ho & Mckay, 2000; Qiu *et al.*, 2009).

$$\log (q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (3)$$

where q_e, q_t (mg/g) is amount of MG adsorbed at a given time t and at equilibrium, respectively; k_1 (L/min) is the rate constant of pseudo-first-order adsorption. The values of k_1 and q_e were calculated from slope and intercept of the plot between log (q_e - q_t) versus t.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

where k_2 (g/mg.min) is pseudo-second-order rate constant. The values of k_2 and q_e were calculated from intercept and slope of the plot between t / q_t versus t.

Thermodynamic studies: The experimental results from experiment 4 on the effects of four temperatures (298.15, 303.15, 308.15, and 313.15 K (25, 30, 35, and 40 °C, respectively) on adsorption were investigated. The van't Hoff plot of ln K_c versus 1/T in equation 5 provided the values of enthalpy change (ΔH°) and entropy change (ΔS°) from the slope and intercept of the plot. The value of Gibb's free energy change (ΔG°) can be also determined by equation 6 (Tan *et al.*, 2007; Setiabudi, Juoh, Suhaimi, & Masrur, 2016).

$$\ln K_c = \frac{\triangle S^\circ}{R} - \frac{\triangle H^\circ}{RT} \quad (5)$$

$$\triangle G^\circ = - RT \ln K_c \quad (6)$$

where K_c (L/mol) is the equilibrium constant, R is the universal gas constant (8.314 J/mol K) , and T (K) is the absolute temperature (K).

2.4 Scanning electron microscopy and chemical oxygen demand measurement

The surface morphologies of the BPP samples were analyzed by scanning electron microscopy (SEM) . Chemical oxygen demand (COD) measurements of the filtrates of the adsorption experiments using 1 g/L of the suitable adsorbent (ripe BPP of ABB) in distilled water at 30 °C, pH 5 for five contact time intervals (0.5 h, 2 h, 6 h, 12 h, and 24 h) were also performed by the closed reflux, titrimetric method according to American Public Health Association, American Water Works Association and Water Pollution Control Federation (1985) Standard method 508 B.

3. Results and Discussion

3.1 Effect of initial pH of MG solution on its solubility

The allowable pH range of the textile effluent is 5-9 (Pollution Control Department [PCD], 2017). Therefore, the effects of pH that ranged between 5 and 9 on the solubility of MG were studied. The acceptable pH values of MG solution were pH 5 and pH 6 with the solubility of 98.37% and 91.80%, respectively (Figure S-1). The maximum solubility percentage of MG was at pH 5. But MG solubility decreased as the pH increased from 5.0 to 9.0. This was in good agreement with the results of Das, Pal, Saha, and Maji (2009) which reported that the ionic form of MG, i.e. the colored form, existed mostly in the range of pH 2.5-5.75. While at higher pH values (pH >7) MG changed to carbinol base which is the non-ionic form and colorless. Therefore, the optimal MG solution pH at pH 5 was chosen for the adsorption experiments of MG by BPP.

3.2 Effect of initial pH of MG solution on its removal by various adsorbents derived from banana peel powder

The effects of the initial pH of MG solution on its removal by banana peel powder are shown in Figure 2. Ripe banana peel powder acted as a better adsorbent than unripe banana peel powder for both species of banana peel. The maximal MG removal percentage was 97.35% at pH 5. At this pH, the surface of the adsorbent was negatively charged which resulted in an increase in the adsorption of the positively charged MG cationic dye due to electrostatic forces of attraction (Sartape *et al.*, 2017). But MG removal decreased with an increase in pH from 5 to 9 because of the reasons explained in section 3.1. Therefore, the optimal adsorption condition using ripe banana peel powder (ABB) at pH 5 was used for further experiments.

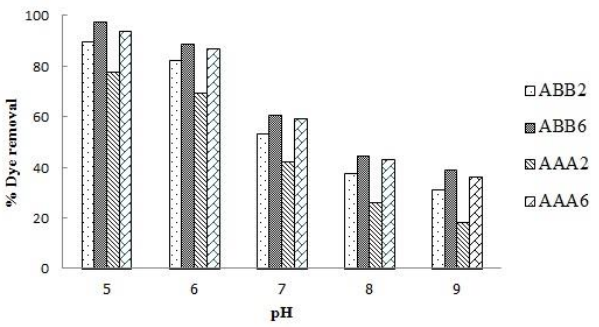

Figure 2. Effect of initial pH of solution on removal percentage of MG by various adsorbents derived from two species and two maturation stages of banana peel powder (initial concentration of MG: 25 mg/L, adsorbent dose: 1 g/L, particle size of adsorbent:0.150-0.212 mm, temperature: 30°C, the agitation speed:150 rpm, contact time: 24 h).ABB2 and ABB6 stand for unripe (stage 2) and ripe (stage 6) banana peel powder of ABB, and AAA2 and AAA6 stand for unripe (stage 2) and ripe (stage 6) banana peel powder of AAA.

3.3. Effects of contact time and initial dye concentration on MG removal

The effects of contact time (0-24 h) and initial dye concentration (5-100 mg/L) on MG removal by ripe banana peel powder (ABB) are presented in Figure 3. MG was adsorbed rapidly at the early range of contact time (0-0.15 h). Then a slight increase of dye adsorption was observed. However, the results showed that the equilibrium time for all of the test initial concentrations of MG solution was achieved within approximately 12 h. This possibly resulted from the availability of a large number of vacant adsorption sites on the

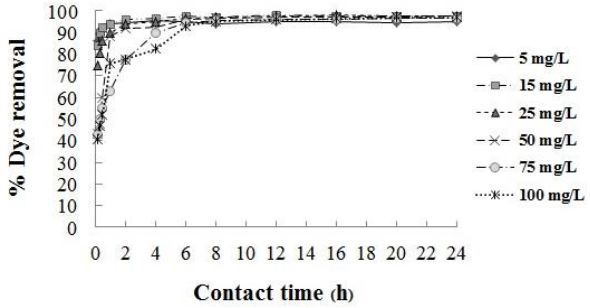

Figure 3. Effects of contact time and initial concentration on MG removal by banana peel powder at pH 5, adsorbent dose of 1 g/L, particle size of 0.150-0.212 mm, at 30 °C, and at the agitation speed of 150 rpm.

BPP surface which was higher at the beginning. The adsorption sites were then gradually occupied by MG and beyond the equilibrium time the adsorption was nearly stable (Seow & Lim, 2016; Kumar *et al.*, 2011; Kyzas, Lazaridis, & Mitropoulos, 2012;). The maximum removal percentage of MG was 97.76% at 12 h equilibrium time in 25 mg/L of initial MG concentration.

3.4 Effect of adsorbent dose on MG removal

The percentage of MG removal by BPP at different doses is shown in Figure 4. MG removal increased from 56.05 to 97.56% as the adsorbent dose increased from 0.25 g/L to 1 g/L. This can be explained by the increased number of sorption sites (Yagub *et al.,* 2014) . At an adsorbent dose greater than 1 g/L, MG removal did not increase significantly. Therefore, the adsorbent dose of 1 g/L was chosen for further studies.

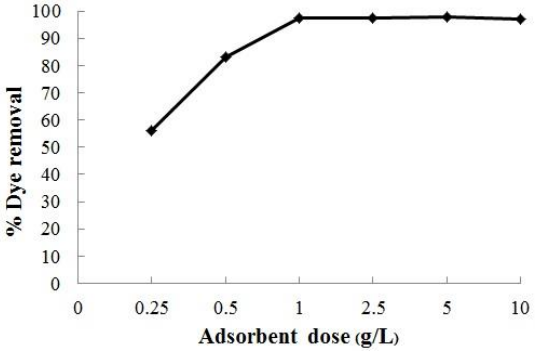

Figure 4. Effect of adsorbent dose on MG removal by banana peel powder at pH5, contact time of 12 h, initial dye concentration of 25 mg/L, adsorbent particle size of 0.150-0.212 mm, at 30 °C, and at the agitation speed of 150 rpm.

3.5 Effect of particle size of adsorbent on MG removal

Figure 5 exhibits the influence of various particle sizes of adsorbent on MG removal. Higher MG adsorption was found with smaller adsorbent particle size because of an increase in the total surface area for adsorption per unit mass of adsorbent (Krishan & Swamy, 2012; Liu *et al.*, 2012).

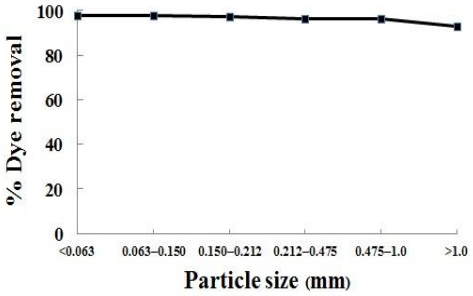

Figure 5. Effect of adsorbent particle size on MG removal by banana peel powder at pH 5, contact time of 12 h, initial dye concentration of 25 mg/L, adsorbent dose of 1 g/L, at 30 °C with the agitation speed of 150 rpm.

However, the mean data for MG removal percentage on BPP at the particle size 0.150–0.212 mm and <0.063 mm were 97.26% and 97.96%, respectively, which were not much different. Nevertheless, considering the greater convenience of preparation and the larger amount of BPP obtained, the particle size of BPP at the range 0.150–0.212 mm was more suitable and was chosen for further experiments.

3.6 Effect of temperature on MG removal

The mean values of MG removal percentage at 25 °C (298.15 K) was 97.48% and the value increased to 98.01% at 40 °C (313.15 K) (Figure 6). This was due to a decrease in its viscosity and an increase in MG diffusion across the external surface area to the internal pores of the adsorbent (Hazzaa & Hussien, 2015) . However, considering the slight differences in the percentage of MG removal using BPP among various temperatures and the optimal external environment temperature in Thailand, the temperature at 30 °C (303.15 K) was chosen for further experiments.

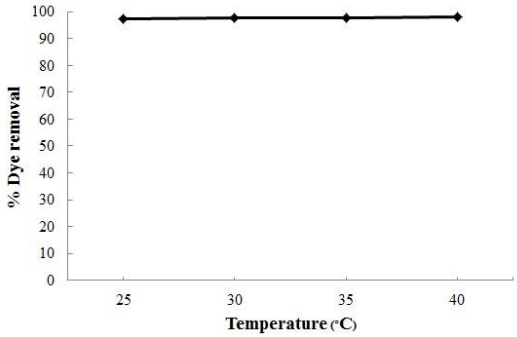

Figure 6. Effect of temperature on MG removal by banana peel powder at pH 5, contact time of 12 h, initial dye concentration of 25 mg/L, adsorbent dose of 1 g/L, adsorbent particle size of 0.150-0.212 mm and at the agitation speed of 150 rpm.

3.7 SEM

Figure 7 exhibits the surface morphologies of the banana peels analyzed by SEM. At a lower magnification (100x) both species of test banana peel showed stuck fibers of lignin, pectin, and other viscous compounds which were

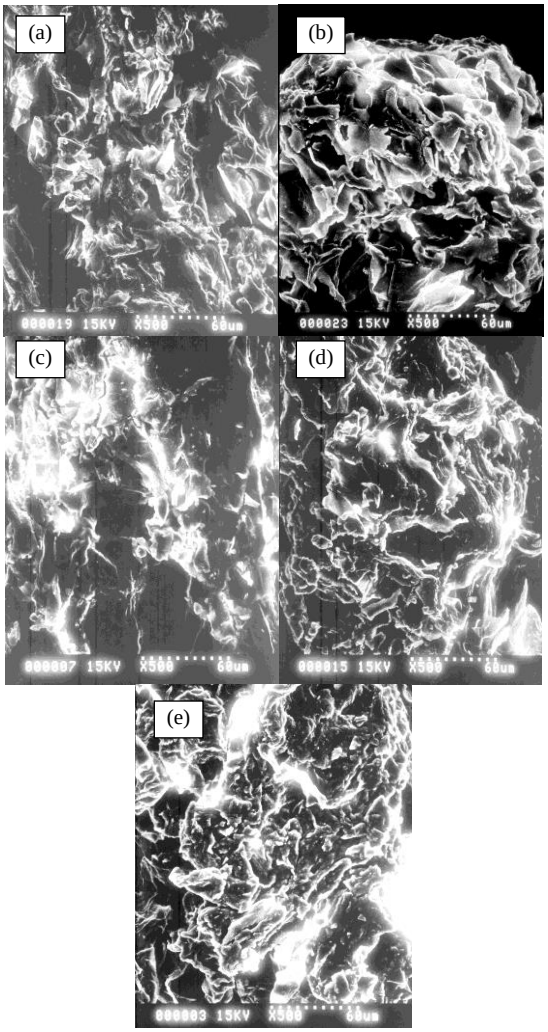

Figure 7. SEM image of banana peel powder of (a) unripe *Musa x paradisiaca* (ABB), (b) ripe ABB, (c) unripe *Musa acuminata* (AAA), (d) ripe AAA, and (e) ripe ABB after adsorption of MG. All images are at magnification of 500x (scales indicated in each image).

similar to the results reported by Ali, Saeed, and Mabood (2016). At higher magnifications (≥1,000x) the surface morphology revealed an irregular shape with a heterogeneous, rough surface with crater-like pores which was in accordance with previous studies (Mohammed & Chong, 2014; Pathak & Mandavgane, 2015). In addition, at the middle magnification (500x), a lumpy denseness was seen inside which was quite similar to SEM images of *Zalacca edulis* peel (Sirilamduan, Umpuch, & Kaewsarn, 2011). It is also important to note that the unripe banana peel was more loosely packed and had a more heterogeneous surface than the ripe banana peel. Therefore, ripe banana peel revealed an interior with a continuous lumpy denseness with irregular pores more than the unripe banana peel. However, ripe banana peel of *Musa x paradisiaca* (ABB) showed this effect was more pronounced than *Musa acuminata* (AAA). Therefore, the structure of ripe banana peel of *Musa x paradisiaca* (ABB) can support the adsorption of MG more than the *Musa acuminata* (AAA) and those of unripe banana peel of both species. Furthermore, no differences were observed between the SEM images of the ripe banana peel of *Musa x paradisiaca* (ABB) that were loaded or unloaded with MG (Figures 7a and 7e). These results were in good agreement with the results of *Zalacca edulis* peel samples that were either loaded or unloaded with Cu^{2+} (Sirilamduan *et al.,* 2011).

3.8 Leaching of COD from the adsorbent

The COD values leached from the adsorbent under the same conditions of adsorption experiments for five contact time intervals (0.5 h, 2 h, 6h, 12 h, and 24 h) were analyzed (Table S-1). It was found that the COD values increased from 16 mg/L to 32 mg/L with the increase of contact time from 0.5 h to 24 h. However, the COD values leached from the adsorbent were acceptable and much less than the maximum allowable standard value for the textile effluents in Thailand (≤400 mg/L) (PCD, 2017).

3.9 Adsorption isotherm

The results of equilibrium adsorption experiments were applied to the Freundlich and Langmuir models and their constants and correlation values are summarized in Table 1. Regarding the R^2 values, the results show that the Freundlich isotherm was more appropriate than the Langmuir isotherm for MG adsorption. This can imply that the adsorption was performed on heterogeneous surfaces in multiple layers. In addition, the n value obtained from this study was 1.1403 (n>1), which indicated that physical adsorption was the predominant process for MG removal by ripe ABB banana peel (Kumar *et al.,* 2011), which was consistent with the results of the SEM study (section 3.7). Ali (2017) also reported that the pores and gaps between cellulose fibers of untreated agricultural adsorbents were covered by viscous compounds such as lignin and pectin surrounding the available active functional groups and could not interact well with the adsorbates. Due to the limitation of time and budget, the functional groups of the adsorbents used in this study could not be studied. Nevertheless, a study of the functional groups is suggested in order to obtain clearer results. The capacities of various fruit peels to adsorb MG were reported as 483.63 mg/g by orange peel and 51.42 mg/g by lemon peel, while it was 243.90 mg/g by ripe ABB banana peel in the current study. Therefore, these adsorption capacities can be arranged in this order orange peel > ripe ABB banana peel > lemon peel (Table S-2). The percentage of MG removal in this

Table 1. Langmuir and Freundlich isotherm parameters and correlations for the adsorption of MG by banana peel powder.

Isotherms models	Isothermparameters	
Langmuir	q_m (mg/g)	243.90
	K_L (L/mg)	0.1126
	R^2	0.9540
Freundlich	1/n	0.8770
	N	1.1403
	K_F (mg/g) $(L/mg)^{1/n}$	23.0781
	R^2	0.9918

study was relatively high (97.35%). Therefore, modification of the adsorbent does not seem to be necessary. However, for other adsorbents with relatively low adsorbate removal percentages, modification of adsorbents may be carried out in different ways, such as chemical modification, activation by heat, and ultrasound-assistance (Asfaram, Ghaedi, Hajati, Goudarzi, & Dil, 2017; Hokkanen, Bhatnagar, & Sillanpää, 2016; Suhas *et al.*, 2016;).

3.10 Adsorption kinetics

The adsorption kinetic data from the experimental results are shown in Table 2. Due to the high values of the correlation coefficients and the close calculated values ($q_{e,cal}$) to the actual values ($q_{e,exp}$), these indicate that the pseudo-second-order model provided a more satisfactorily description of the results than the pseudo-first-order model. This indicated that chemisorption might be the rate limiting step as reported by Kyzas *et al.* (2012); Sirilamduan *et al.* (2011).

3.11 Adsorption thermodynamic

The adsorption thermodynamic parameters are presented in Table 3. The negative values of Gibbs free energy change (ΔG°) indicated that the adsorption of MG by banana peel powder was spontaneous. In addition, an increase in the magnitude of negative values of $\triangle G^{\circ}$ with an increase in temperature confirmed that the adsorption became more spontaneous at higher temperatures. The ΔG° values ranged from -9.06 to -10.15 kJ/mol, which were related to the spontaneous physical processes reported by Seki & Yurdakoç (2006). The high positive value of entropy change (ΔS°) indicates the enhancement in the degree of freedom of the adsorbed species. A positive value of enthalpy change (ΔH°) suggests that the adsorption is an endothermic process and a large amount of heat is consumed for the transfer of MG from an aqueous solution to a solid phase of the adsorbent.

Table 3. Thermodynamic parameters for the adsorption of MG by banana peel powder.

Temperature °C (K)	K_c (L/mol)	ΔH° (kJ/mol)	ΔS° (J/mol K)	ΔG° (kJ/mol)
25 (298.15)	38.92	12.6207	72.7442	-9.0571
30 (303.15)	41.67			-9.4208
35 (308.15)	45.70			-9.7845
40 (313.15)	49.51			-10.1483

4. Conclusions

The banana peel powder of *Musa x paradisiaca* (ABB) at maturation stage 6 performed as a good adsorbent for MG adsorption from synthetic wastewater with the removal efficiency of 97.64% under optimal conditions in batch experiments at pH 5, 12 h equilibrium time, initial dye concentration of 25 mg/L, adsorbent dose of 1 g/L, adsorbent particle size of 0.150-0.212 mm, 30 °C, and at an agitation speed of 150 rpm. The equilibrium adsorption isotherm and kinetics were satisfactorily described by Freundlich isotherm (R^2=0.9918) and pseudo-second-order kinetic model (R^2>0.99), respectively. The results from adsorption thermodynamic parameters indicated that it was a random, endothermic, and spontaneous process.

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References

Abbasi, Z., Alikarami, M., Nezhad, E. R., Moradi, F., & Moradi, V. (2013). Adsorptiveremoval of Co^{2+} and Ni^{2+} by peels of banana from aqueous solution. *Universal Journal of Chemistry*, *1*(3), 90-95. doi:10.13189/uje.2013.010303

Ali, A. (2017). Removal of Mn (II) from water using chemically modified banana peels as efficient adsorbent. *Environmental Nanotechnology, Monitoring and Management, 7*, 57-63. doi:10.1016/j.enmm.2016.12.004

Ali, A., Saeed, K., & Mabood, F. (2016). Removal of chromium (VI) from aqueous medium using chemically modified banana peels as efficient low-cost adsorbent. *Alexandria Engineering Journal, 55*, 2933-2942. doi:10.1016/j.aej.2016.05.011

Amel, K., Hassena, M. A., & Kerroum, D. (2012). Isotherm and kinetics study of biosorption of cationic dye onto banana peel. *Energy Procedia, 19*, 286-295. doi:10.1016/j.egypro.2012.05.208

American Public Health Association, American Water Works Association and Water Pollution Control Federation. (1985). 508 B. Closed reflux, titrimetric method. In M. A. H. Franson (Ed.), *Standard methods, for the examination of water and wastewater* (16th ed.) (pp. 535-537). Washington, DC: Author.

Table 2. Pseudo-first-order and pseudo-second-order kinetic parameters for adsorption of MG by banana peel powder.

Initial dye concentration (mg/L)	$q_{e,exp}$ (mg/g)	Pseudo-first-order			Pseudo-second-order		
		$q_{e,cal}$ (mg/g)	k_1 (L/min)	R^2	$q_{e,cal}$ (mg/g)	k_2 (g/mg.min)	R^2
5	5.74	0.73	0.0032	0.5377	5.53	0.0523	0.9990
15	18.95	1.79	0.0035	0.3349	18.62	0.0142	0.9997
25	40.82	12.59	0.0064	0.7474	40.82	0.0023	0.9984
50	61.48	23.05	0.0106	0.9312	63.69	0.0009	0.9986
75	94.41	66.56	0.0078	0.9679	101.01	0.0002	0.9941
100	131.44	75.84	0.0048	0.9114	131.58	0.0002	0.9931

Annadurai, G., Juang, R. S., & Lee, D. J. (2002). Use of cellulose-based wastes for adsorption of dyes from aqueous solutions. *Journal of Hazardous Materials, 92*, 263-274. PII: S0304-3894(02)00017-1

Asfaram, A., Ghaedi, M., Hajati, S., Goudarzi, A., & Dil, E. A. (2017). Screening and optimization of highly effective ultrasound-assisted simultaneous adsorption of cationic dyes onto Mn-doped Fe_3O_4-nanoparticle-loaded activated carbon. *Ultrasonics Sonochemistry, 34*, 1-12. doi:10.1016/j.ultsonch.2016.05.011

Baek, M. H., Ijagbemi, C. O., O, S. J., & Kim, D. S. (2010). Removal of Malachite Green from aqueous solution using degreased coffee bean. *Journal of Hazardous Material, 176*, 820-828. doi:10.1016/j.jhazmat.2009.11.110

BananaLink Office. (2017, March 27). All about bananas. Retrieved form http://www.banana.org.uk/all-about-banas/

Betiku, E., Akintunde, A. M., & Ojumu, T. V. (2016). Banana peels as a biobase catalyst for fatty acid methyl esters production using Napoleon's plume (*Bauhinia monandra*) seed oil: A process parameters optimization study. *Energy, 103*, 797-806. doi:10.1016/j.energy.2016.02.138

Das, A., Pal, A., Saha, S., & Maji, S. K. (2009). Behaviour of fixed-bed column for the adsorption of malachite green on surfactant-modified alumina. *Journal of Environmental Science and Health Part A, 44*, 265-272. doi:10.1080/10934520802597929

Devi, P., & Saroha, A. K. (2017). Utilization of sludge based adsorbents for the removal of various pollutants: A review. *Science of the Total Environment, 578*, 16-33. doi:10.1016/j.scitotenv.2016.10.220

Fernandez, M. E., Nunell, G. V., Bonelli, P. R., & Cukierman, A. L. (2014). Activated carbon developed from orange peels: Batch and dynamic competitive adsorption of basic dyes. *Industrial Crops and Products, 62*, 437-445. doi:10.1016/j.indcrop.2014.09.015

Ferreira, A. M., Coutinho, A. P., Fernandes, A. M., & Freire, M. G. (2014). Complete removal of textile dyes from aqueous media using ionic-liquid-based aqueous two-phase systems. *Separation and Purification Technology, 128*, 58-66. doi:10.1016/j.seppur.2014.02.036

Gandhimathi, R., Remesh, S. T., Sindhu, V., & Nidheesh, P. V. (2013). Bottom ash adsorption of basic dyes from their binary aqueous solutions. *Songklanakarin Journal of Science and Technology, 35*(3), 339-347. Retrieved from http://rdo.psu. ac.th/sjstweb/journal/35-3/35-3-339.pdf/

Hazzaa, R., & Hussien, M. (2015). Adsorption of cationic basic dye from aqueous solution onto activated carbon derived from banana peel. *Proceedings of the 14th International Conference on Environmental Science and Technology*, Rhodes, Greece. Retrieved from http://cest2015.gnest.org/papers/cest2015_000 31 _oral_paper.pdf/

Hokkanen, S., Bhatnagar, A., & Sillanpää, M. (2016). A review on modification methods to cellulose-based adsorbents to improve adsorption capacity. *Water Research, 91*, 156-173. doi:10.1016/j.watres.2016.01.008

Ho, Y. S. (2004). Citation review of Lagergren kinetic rate equation on adsorption reactions. *Scientometrics, 59*(1), 171-177. doi:10.1023/B:SCIE.0000013305.99473.cf

Ho, Y. S., & Mckay, G. (2000). The kinetics of sorption of divalent metal ions onto sphagnum moss peat. *Water Resource Journal, 34*(3), 735-742. doi:10.1016/S0043-1354(99)00232-8

Khalek, M. A. A., Rahman, M. K. A., & Francis, A. A. (2017). Exploring the adsorption behavior of cationic and anionic dyes on industrial waste shells of egg. *Journal of Environmental Chemical Engineering, 5*, 319-327. doi:10.1016/j.jece.2016.11.043

Kiakhani, M. S., Arami, M., & Gharanjig, K. (2013). Preparation of chitosan-ethyl acrylate as a biopolymer adsorbent for basic dyes removal from colored solutions. *Journal of Environmental Chemical Engineering, 1*, 406-415. doi:10.1016/j.jee.2013.06.001

Krishan, R. H., & Swamy, A. V. V. S. (2012). Physico-chemical key parameters, Langmuir and Freundlich isotherm and Lagergren rate constant studies on the removal of divalent nickel from the aqueous solutions onto powder of calcined brick. *International Journal of Engineering Research and Development, 4*(1), 29-38. Retrieved from https://www.idc-online.com/technical_references/pdfs/chemical_engineering/Physico%20Chemical.pdf/

Kumar, K. V. (2007). Optimum sorption isotherm by linear and non-linear methods for malachite green onto lemon peel. *Dyes and Pigments, 74*, 595-597. doi:10.1016/j.dyepig.2006.03.026

Kumar, K. V., & Porkodi, K. (2007). Batch adsorber design for different solution volume/adsorbent mass ratios using the experimental equilibrium data with fixed solution volume/adsorbent mass ratio of malachite green onto orange peel. *Dyes and Pigments, 74*, 590-594. doi:10.1016/j.dyepig.2006.03.024

Kumar, P. S., Abhinaya, R. V., Lashmi, K. G., Arthi, V., Pavithra, R., Sathyaselvabala, V., & Sivanesan, S. (2011). Adsorption of methylene blue dye from aqueous solution by agricultural waste: Equilibrium, thermodynamics, kinetics, mechanism and process design. *Colloid Journal, 73*(5), 651-661. doi:10.1134/S1061933X11050061

Kyzas, G. Z., Lazaridis, N. K., & Mitropoulos, A. C. (2012). Removal of dyes from aqueous solutions with untreated coffee residues as potential low-cost adsorbents: Equilibrium, reuse and thermodynamic approach. *Chemical Engineering Journal, 189-190*, 148-159. doi: 10.1016/j.cej.2012.02.045

Liu, C., Ngo, H. H., Guo, W., & Tung, K. L. (2012). Optimal conditions for preparation of banana peels, sugarcane bagasse and watermelon rind in removing copper from water. *Bioresource Technology, 119*, 349-354. doi:10.1016/j.biortech.2012.06.004

Mane, V. S., Mall, I. D., & Srivastava, V. C. (2007). Kinetic and equilibrium isotherm studies for the adsorptive removal of brilliant green dye from aqueous solution by rice husk ash. *Journal of Environmental Management, 84*, 390-400. doi:10.1016/j.jenvman.2006.06.024

Mohammed, R. R., & Chong, M. F. (2014). Treatment and decolorization of biologically treated palm oil mill effluent (POME) using banana peel as novel biosorbent. *Journal of Environmental Management, 132*, 237-249. doi:10.1016/j.jenvman.2013.11.031

Mohapatra, D., Mishra, S. M., & Sutar, N. (2010). Banana and its by-product utilisation: an overview. *Journal of Scientific and Industrial Research, 69*, 323-329. Retrieved from http://sa.indiaenvironmentportal.org.in/files/Banana%20and%20its%20by%20product%20utilisation.pdf/

Moubarak, F., Atmani, R., Maghi, I., Elkouali, M., Talbi, M., Bouamrani, M. L., & Kenz, A. (2014). Elimination of Methylene Blue dye with natural adsorbent (banana peels powder). *Global journal of Science Frontier Research: B Chemistry, 14*(1), 38-44. Retrieved from https://globaljournals.org/GJSFR_Volume14/5-Elimination-of-Methylene-Blue.pdf/

Palma, C., Contreras, E., Urra, J., & Martinez, M. J. (2011). Eco-friendly technologies based on banana peel use for the decolourization of the dyeing process wastewater. *Waste Biomass Valor, 2*, 77-86. doi:10.1007/s12649-010-9052-4

Pathak, P. D., & Mandavgane, S. A. (2015). Preparation and characterization of raw and carbon from banana peel by microwave activation: Application in citric acid adsorption. *Journal of Environmental Chemical Engineering, 3*, 2435-2447. doi:10.1016/j.jece.2015.08.023

Pollution Control Department. (2017, March 3). Water quality standards. Retrieved from http://www.pcd.go.th/info_serv/en_reg_std_water04.html/

Qiu, H., LV, L., Pan, B. C., Zhang, Q. J., Zhang, W. M., & Zhang, Q. X. (2009). Critical review in adsorption kinetic models. *Journal of Zhejiang University Science A, 10*(5), 710-724. doi:10.1631/jzus.A0820524

Rangabhashiyam, S., Anu, N., & Selvaraju, N. (2013). Sequestration of dye from textile industry wastewater using agricultural waste products as adsorbents. *Journal of Environmental Chemical Engineering, 1*, 629-641. doi:10.1016/j.jece.2013.07.014

Saikaew, W., & Kaewsarn, P. (2009). Cadmium ion removal using biosorbents derived from fruit peel wastes. *Songklanakarin Journal of Science and Technology, 31*(5), 547-554. Retrieved from http://rdo.psu.ac.th/sjstweb/journal/31-5/0125-3395-31-5-547-554.pdf/

Salleh, M. A. M., Mahmoud, D. K., Karim, W. A. W. A., & Idris, A. (2011). Cationic and anionic dye adsorption by agricultural solid wastes: A comprehensive review. *Desalination, 280*, 1-13. doi:10.1016/j.desal.2011.07.019

Sartape, A. S., Mandhare, A. M., Jadhav, V. V., Raut, P. D., Anuse, M. A., & Kolekar, S. S. (2017). Removal of malachite green dye from aqueous solution with adsorption technique using *Limonia acidissima* (wood apple) shell as low cost adsorbent. *Arabian Journal of Chemistry, 10*, S3229-S3238. doi:10.1016/j.arabjc.2013.12.019

Seki, Y., & Yurdakoç, K. (2006). Adsorption of Pronmethazine hydrochloride with KSF motmorillonite. *Adsorption, 12*(1), 89-100. doi:10.1007/s10450-006-0141-4

Seow, T. W., & Lim, C. K. (2016). Removal of dye by adsorption: A review. *International Journal of Applied Engineering Research, 11*, 2675-2679. Retrieved from http://www.ripublication.com/ijaer16/ ijaerv11n4_86.pdf/

Setiabudi, H. D., Juoh, R., Suhaimi, S. F. R. M., & Masrur, S. F. (2016). Adsorption of methylene blue onto oil palm (*Elaeis guineensis*): Process optimization isotherm, kinetics and thermodynamic studies. *Journal of the Taiwan Institute of Chemical Engineers, 63*, 363-370. doi:10.1016/j.jtice.2016.03.035

Sirilamduan, C., Umpuch, C., & Kaewsarn, P. (2011). Removal of copper from aqueous solutions by adsorption using modify *Zalacca edulis* peel modify. *Songklanakrin Journal of Science and Technology, 33*(6), 725-733. Retrieved from http://rdo.psu.ac.th/sjstweb/journal/33-6/0125-3395-33-6-725-732.pdf/

Suhas, Gupta, V. K., Carrott, P. J. M., Singh, R., Chaudhary, M., & Kushwaha, S. (2016). Cellulose: A review as natural, modified and activated carbon adsorbent. *Bioresource Technology, 216*, 1066-1076. doi:10.1016/j.biortech.2016.05.106

Tan, I. A. W., Hameed, B. H., & Ahmad, A. L. (2007). Equilibrium and kinetic studies on basic dye adsorption by oil palm fibre activated carbon. *Chemical Engineering Journal, 127*, 111-119. doi:10.1016/j.cej.2006.09.010

Tapre, A. R., & Jain, R. K. (2012). Study of advanced maturity stages of banana. *International Journal of Advanced Engineering Research and Studies, 1*(3), 272-274. Retrieved from http://www.technicaljournalsonline.com/ijaers/VOL%20I/IJAERS%20VOL%20I%20ISSUE%20III%20APRIL%20JUNE%202012/178.pdf

Valmayor, R. V., Jamaluddin, S. H., Silayoi, B., Kusumo, S., Danh, L. D., Pascua, O. C., & Espono, R. R. C. (2017, January 2). Banana cultivar names and synonyms in Southeast Asia. Retrieved from http://www.bioversityinternational.org/uploads/tx_news/Banana_cultivar_names_and_synonyms_in_Southeast_Asia_713.pdf/

Wikimedia Foundation, Inc. (2017, January 16). Malachite green. Retrieved from https://en.wikipedia.org/wiki/Malachitegreen/

Yagub, M. T., Sen, T. K., Afroze, S., & Ang, H. M. (2014). Dye and its removal from aqueous solution by adsorption: A review. *Advances in Colloid and Interface Science, 209*, 172-184. doi:10.1016/j.cis.2014.04.002

Appendix

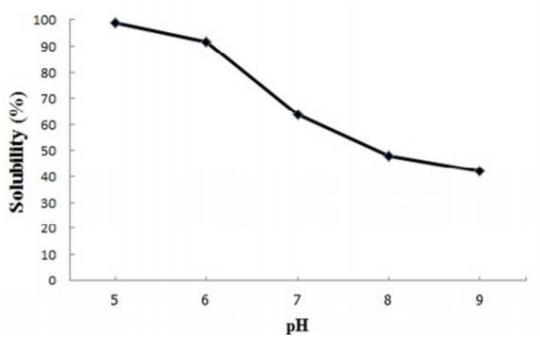

Figure S-1. Effect of initial pH of solution on the solubility of MG (initial MG concentration of 25 mg/L, contact time for 24 h, at 30 ℃ with the agitation speed: 150 rpm).

Table S-1. Leaching of COD from banana peel adsorbent in distilled water at pH 5, adsorbent dose of 1 g/L, particle size of 0.150-0.212 mm, at 30 ℃ with the agitation speed of 150 rpm.

Mean ±SD values of leached COD (mg/L) from banana peel adsorbent at various contact time (h)				
0.5	2	6	12	24
16±4	19±4	23±4	30±4	32±4

Table S-2. Comparison of MG adsorption capacity values between previous studies and the current study.

Adsorbent	Adsorption Capacity (mg / g)	Reference
Orange peel powder (0.180-1 mm)	483.63	Kumar & Porkodi (2007)
Ripe Banana peel powder (0.150-0.212)	243.90	Current study
Lemon peel powder (0.165-0.193 mm)	51.42	Kumar (2007)