



Removal of Methylene Blue from Aqueous Solution by Activated Carbon from Aloe Vera Wastes

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Abstract

Background: Many organic and inorganic pollutants are discharged by various industries into the water bodies. One of the most important pollutants is dye materials that have adverse effects on human health.

Objectives: In this study, Aloe Vera wastes-based activated carbon (AV-AC) was used as a low-cost sorbent for the removal of methylene blue (MB) as a textile dye from liquid medium.

Methods: The effect of different parameters such as contact time, pH, adsorbent dosage, and initial dye concentration was investigated on the sorption of MB. The experimental data were examined by various kinetic and isotherm models.

Results: The equilibrium was obtained at contact time of 40 minutes. Fitting the experimental data to different kinetics and isotherms models indicated that the experimental data were well fitted by the pseudo-second-order kinetic and Freundlich isotherm models.

Conclusions: The maximum adsorption capacity was 129.87 mg/g. Due to low cost and high uptake capacity, Aloe Vera wastes can be considered as one effective sorbent for removal of MB.

Keywords: Adsorption, Aloe Vera, Methylene Blue, Kinetic, Isotherm

1. Background

Water pollution due to discharging of colored wastewaters from the textile industries is an important environmental problem, especially in developing countries. Discharging these colored wastewaters into water bodies can cause adverse effects on aquatic life as well as human health. Most dyestuff components have toxic, carcinogenic and mutagenic properties (1, 2). Therefore, decolorization of these dyes are essential before their discharge into water bodies (3, 4). The treatment of the colored wastewater is very difficult, due to dyes recalcitrant organic molecules, resistance to biological degradation, and stability to heat and oxidizing agents (3). Dyes can be categorized to anionic, cationic and non-ionic dyes (5). Methylene Blue (MB) or basic blue 9 is one of the commonly cationic dyes used for various purposes in industries especially in the textile industry. Methylene Blue can cause harmful impacts on animals and humans. Several physical, chemical and microbial methods, such as mem-

brane technologies, coagulation-flocculation, chemical oxidation, advanced oxidation, biological degradation, electrochemical techniques and adsorption, have been developed for removal of the dye from wastewaters. Among these treatment methods, adsorption process owing to its high efficiency and non-toxicity has been widely applied for the removal of dye containing wastewaters (6, 7). Activated carbon due to simplicity, high surface area, and high sorption capacity has been used as a common adsorbent to remove dye pollutants, but it is expensive and inflexible to regenerate (3, 8). Jawad et al. used activated carbon developed from biomass and reported that it is an effective and low-cost adsorbent for the removal of MB from aqueous solution (9). In another study by Rahman et al., the sorption of MB was studied by activated carbon from rice husk and it was concluded that activated carbon of rice husk can be employed as a low-cost adsorbent for the treatment of dyeing water (10). Valliammai et al. illustrated that activated carbon from Vigna mungo can be used as a potential low-cost adsorbent for MB dye removal from aqueous solution

(11). In addition to activated carbon, other adsorbents such as montmorillonite nanocomposite (12), montmorillonite (4), spent tea (13), Moroccan clay (1), regenerated clay (14), diatomite (15), chitosan hydrogel (16), durian leaf powder (17), neem sawdust (18), and fly ash (19) have been employed for this purpose. Nowadays, more attention is paid to the use of low-cost sorbents for the removal of dye from aqueous solutions. Aloe Vera is a local plant that in addition to Iran, grows in other areas such as the United State, India, Australia, Africa, Mexico and South America (20, 21). The Aloe Vera leaves wastes are by-products of agricultural industries that were used for the creation latex and drug substances (20, 21).

2. Objectives

In this work, activated carbon was prepared from Aloe Vera wastes and used as a low-cost sorbent for the removal of methylene blue from synthesized wastewater. The influences of various parameters such as contact time, pH, sorbent dosage, and initial concentration of pollutants on adsorption were evaluated.

3. Methods

3.1. Materials

In this study, Aloe Vera wastes were collected from suburban farms of Ahvaz (Khuzestan, Iran). Chemical substances including methylene blue, sulfuric acid and sodium hydroxide were purchased from Merck Co (Germany). Figure 1 shows the characteristics and chemical structure of MB. The pH of the solution was adjusted with diluted and concentrated sulfuric acid and sodium hydroxide solutions using a digital pH-meter (50-pp-sartorius model). The other chemicals used in this study were analytical-grade reagents. The stock solution of 1000 mg/L MB was prepared and the working concentrations were obtained with dilution of the stock solution. The solutions of the adsorbent and adsorbate were mixed using an orbital shaker (Behdad-Rotomix model, Iran) at 150 rpm.

3.2. Preparation of Adsorbent

After separation of the gel of Aloe Vera wastes, the remaining wastes were carefully washed with deionized water to remove impurities and surface adhered particles and then were dried in an electrical oven until constant weight at 150°C for 24 hours. The dried wastes were crushed by a laboratory mill to obtain a particle size in the range of 300 to 600 μm . Besides, particles were carbonized in a furnace at 550°C for 20 minutes. Finally, activated carbon was dried

in an electrical oven at 105°C for 12 hours and was crushed and sieved to obtain a uniform particle size of 40-mesh for sorption experiments.

3.3. Characterizations and Analysis

The surface morphology of activated carbon before and after the sorption process was characterized under a vacuum of a scanning electron microscope (SEM, Jeol Model Jsm-T330). Elemental analysis of Aloe Vera wastes-based activated carbon (AV-AC) was performed using a Heraeus elemental analyzer (Jobin-Yvon Ultima ICP-AES). The concentrations of MB in the solution phases were determined by a UV-Vis spectrophotometer (PG Instrument Limited Model, UK) at maximum wavelengths of 665 nm.

3.4. Determination of pH_{zcp}

The pH at the zero point charge (pH_{zcp}) for the activated carbon synthesized from Aloe Vera wastes was determined by preparation of 50 mL of 0.01 M NaCl solution in a series of 100-mL Erlenmeyer flasks. The initial pH values of the NaCl solution were adjusted, as initial pH (pH_i), between 2 to 12 by adding H_2SO_4 (0.1 M) or NaOH (0.1 M) solution. Then, sufficient amounts of adsorbent were poured into each flask and the suspensions were mixed by a mechanical shaker for 24 hours at 200 rpm. After this period, the solutions pH was measured as final pH (pH_f). The pH_{zcp} was determined by plotting the difference between pH_f and pH_i values ($\text{pH}_{\text{zcp}} = \text{pH}_f - \text{pH}_i$) versus pH_i (21). The contacting local of the resulting curve with abscissa given the pH_{zcp} , the point at which pH is equal to zero (22).

3.5. Batch Adsorption Study

The batch sorption system was applied to identify the influence of different parameters including contact time (0 - 80 minutes), pH (2 - 12), adsorbent dosage (1 - 10 g/L), and initial content of pollutants (30 - 110 mg/L) on the sorption of MB by AV-AC from synthesized wastewater. All of the adsorption experiments were carried out at room temperature (25°C) and agitated at 200 rpm with 100 mL pollutant solution in a 250 mL Erlenmeyer flask. After the adsorption process, the mixture of adsorbate and AV-AC was filtered using a fiberglass paper. The sorption experiments were carried out in duplicates and the average amounts were considered. The uptake capacity and percentage of MB was computed by Equations 1 and 2.

Equation 1.

$$q_e = \frac{(C_0 - C_e) \times V}{m} \quad (1)$$

Equation 2.

$$\text{Sorption (\%)} = \frac{C_0 - C_e}{C_0} \times 100 \quad (2)$$

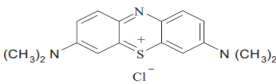
Generic Name	Scientific Name	Chemical Formula	Chemical Structure	Nature	λ_{\max} (nm)
Methylene Blue	Basic Blue 9	$C_{16}H_{18}N_3C_1S$		Cationic	665

Figure 1. The Physical and Structural Characteristics of Methylene Blue

Where q_e (mg/g) is the equilibrium uptake capacity per gram AV-AC. The parameters of C_0 and C_e (mg/L) are the initial and equilibrium concentrations, respectively (19). Also, V (L) is the volume of the solution and m (g) is the adsorbent mass (21).

4. Results

4.1. Characterization

Figure 2A - 2B are show the surface morphology of the sorbent before and after the MB sorption from liquid effluent. As indicated, the original activated carbon has cavities that are more irregular, have fine open pores, uneven structure and a relatively uniform pore size distribution.

4.2. Effect of Contact Time

The effect of different contact time on the sorption capacity of MB is presented in Figure 3A. It is obvious that the adsorption capacity (q_e) of MB was quickly increased over the first 40 minutes and then slowly reached a stable state at 80 minutes. The sorption capacity of MB in equilibrium time was obtained at 19.89 mg/g. Therefore, 40 minutes was selected as the optimum contact time for the subsequent experiments.

4.3. Kinetic Study

The establishment of adsorption kinetic models is valuable for the evaluation the mechanisms of the sorption process (23). Two kinetic models including pseudo-first-order and pseudo-second-order kinetics were applied to analyse the adsorption data of MB by AV-AC. The pseudo-first-order kinetic can be depicted by Equation 3:

Equation 3.

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (3)$$

Where q_e (mg/g) and q_t (mg/g) are the quantity of MB adsorbed onto the AV-AC at the equilibrium and at time t (minute), respectively. k_1 (1/minute, the rate constant of the pseudo-first-order model) and q_e were gained from the slope and intercept of the liner plot of $\ln(q_e - q_t)$ versus t , respectively. The experimental data were also explored by

pseudo-second-order model (2, 24). This kinetic model is represented by the following formula:

Equation 4.

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

Where k_2 (g/mg. min, the rate constant of the pseudo-second-order model) and q_e were acquired from the intercept and gradient of t/q_t against t , respectively (13). Figure 3B and Table 1 show the plot of pseudo-second-order kinetic and quantities of kinetic parameters of MB adsorption by adsorbent, respectively. As indicated, the pseudo-first-order kinetic model has a correlation coefficient ($R^2 > 0.85$) value of lower than of the pseudo-second-order kinetic model ($R^2 > 0.98$). Thus, the sorption process of MB dye by AV-AC followed the pseudo-second-order kinetic model.

4.4. Effect of Solution pH and Determination of pH_{pzc}

The influence of solution pH on the sorption capacity of MB onto AV-AC surface was investigated over the pH range of 2 to 12. The maximum sorption rate occurred at pH of 12 equal to 22.98 mg/g. Thus, it appears that the sorption process is dependent on the pH of the solution. Methylene Blue produces molecular cations in aqueous. Figure 4B shows the pH_{pzc} of the Aloe Vera wastes-based activated carbon. As it is obvious, the pH_{pzc} of adsorbent was found to be equal to 11.3.

4.5. Effect of Adsorbent Dosage and Dye Concentration

The effect of adsorbent dosages (1 - 10 g/L) on the sorption capacity of MB was investigated at an initial concentration of 50 mg/L at pH of 12 and the results are presented in Figure 5A. As shown, with increase in the sorbent dosage from 1 to 10 g/L, the uptake capacity was reduced. Thus, dose of 1 g/L was chosen as the optimum dosage for the next stages of the experiments.

The effect of various initial concentrations of MB (30 - 110 mg/L) was investigated on the sorption capacity during contact time of 40 minutes, adsorbent dosage of 1 g/L, and pH of 12 at room temperature (25°C) and the results are presented in Figure 5B. As shown, with increasing the initial concentration of pollutant from 30 to 110 mg/L, the sorbent capacity of MB onto the sorbent was gradually increased.

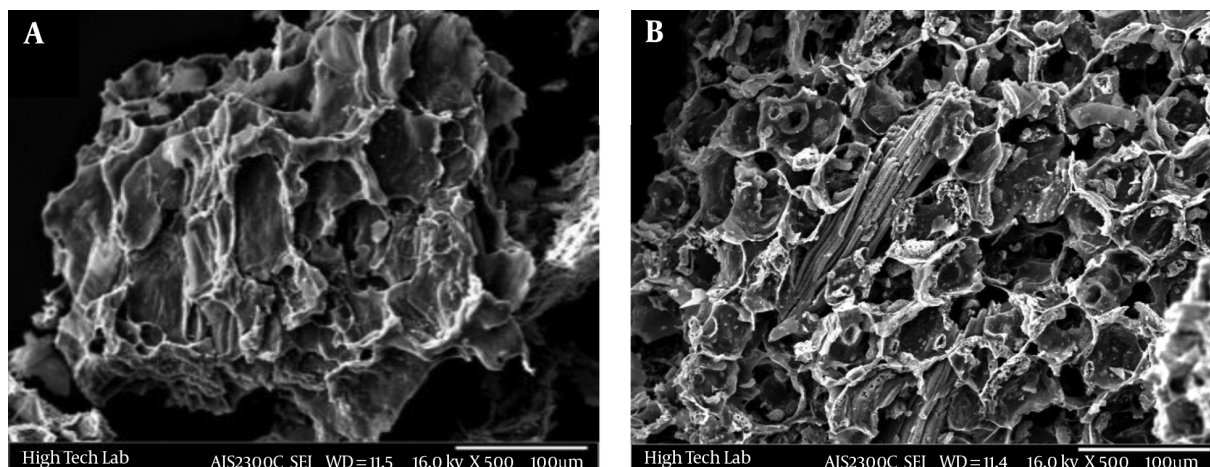


Figure 2. Scanning Electron Microscopy Image of Adsorbent (A) Before and (B) After the Sorption

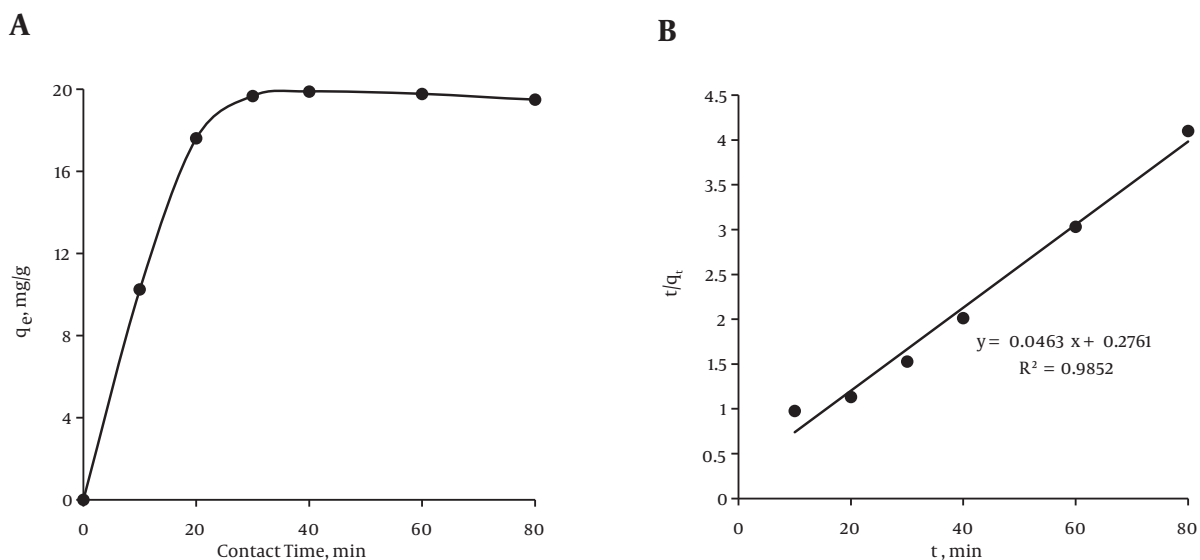


Figure 3. A, Effect of contact time on sorption capacity (dye concentration = 50 mg/L, adsorbent dose = 2 g/L, and ph = 7) and; B, pseudo-second-order kinetic

Table 1. Parameters of Pseudo-First-Order and Pseudo-Second-Order Kinetic Models

Adsorbate	Pseudo-First-Order			Pseudo-Second-Order			
	$q_e, \text{calculated, mg/g}$	$K_1, 1/\text{min}$	R^2	$q_e, \text{experimental, mg/g}$	$q_e, \text{calculated, mg/g}$	$K_2, \text{g/mg.min}$	R^2
MB	2.39	0.029	0.856	19.89	21.73	0.0093	0.985

4.6. Isotherm Study

The sorption isotherms are helpful to find out the adsorbate distribution onto the sorbent under equilibrium conditions (25, 26). The results of MB adsorption were used to determine adsorption isotherm. Therefore, two isotherms including Langmuir and Freundlich models

were employed for this purpose. The Langmuir isotherm approximates the greatest monolayer sorption on the uniform surface of the adsorbent (1, 3, 27). The linearized form of Langmuir isotherm is presented by the following Equation 5:

Equation 5.

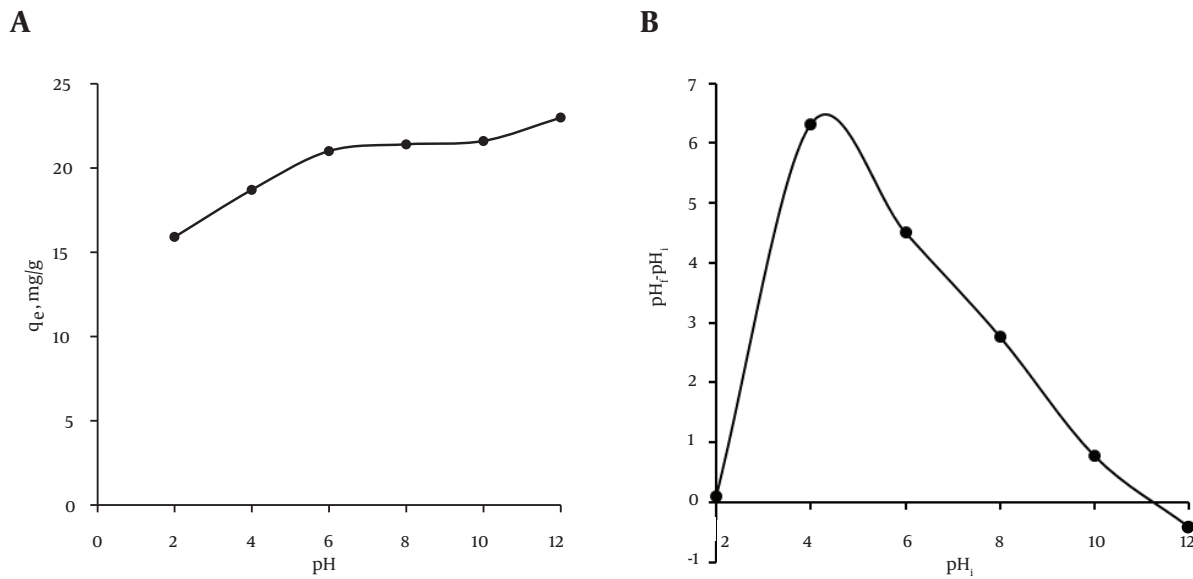


Figure 4. A, Effect of pH on sorption capacity (contact time = 40 minutes, dye concentration = 50 mg/L, and adsorbent dosage = 2g/L) and B, pH_{pzc} of adsorbent.

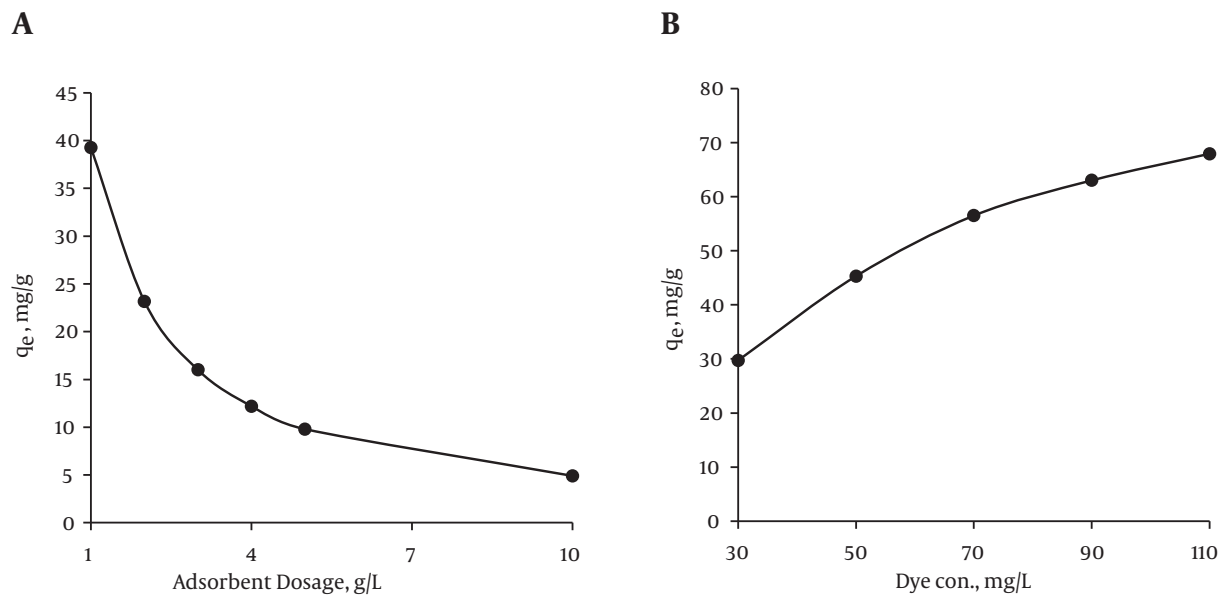


Figure 5. A, Effect of adsorbent dosage on sorption capacity (contact time = 40 minutes, dye concentration = 50, and $pH = 12$) and B, effect of initial dye concentration on Sorption (contact time = 50 minutes, adsorbent dosage = 1 g/L, and $pH = 12$).

$$\frac{C_e}{q_e} = \frac{C_e}{Q_m} + \frac{1}{bQ_m} \quad (5)$$

Where C_e (mg/L) and q_e (mg/g) are the initial MB concentration and AV-AC uptake capacity in the equilibrium time, respectively (18). As shown in Figure 6A, Q_m (maximum sorption amount, mg/g) and b (the rate constant of Langmuir, l/mg) were acquired from the slope and inter-

cept of linear plotting of C_e/q_e versus C_e , respectively. The Langmuir isotherm can be depicted via a dimensionless constant separation factor (R_L). This factor is also called the equilibrium parameter that is obtained from the following formula (20, 21):

Equation 6.

$$R_L = \frac{1}{1 + bC_0} \quad (6)$$

Where; C_0 is the maximum initial MB concentration. The value of separation factor (R_L) shows the adsorption condition as unfavorable (R_L more than 1), linear (R_L equals to 1), irreversible (R_L equals to 0) and favorable (R_L between 0 and 1). Based on the value of R_L in this study ($R_L = 0.58$), the adsorption process of MB using AV-AC was favorable (20, 21).

The Freundlich isotherm is often expressed for multi-layer sorption onto a heterogeneous surface of adsorbent (1, 28). This isotherm is illustrated by Equation 7.

Equation 7.

$$\ln q_e = \ln k_f + \frac{1}{n} \ln C_e \quad (7)$$

Where K_f (l/g) and n are the rate constants of Freundlich and depict the capability and strength of the sorption process, respectively. As illustrated in Figure 6B, K_f and n were achieved using the cut off and gradient of plotting $\ln q_e$ versus $\ln C_e$, respectively (29). The n value of more than one confirms that the adsorption bonds between MB and AV-AC are appropriately strong. Table 2 corresponds with the calculated values of isotherms parameters of MB sorption by AV-AC. As indicated, the value of n computed by Freundlich model was 1.56. Furthermore, the Freundlich isotherm demonstrated a higher R^2 value than Langmuir isotherm. Therefore, it was shown that the experimental sorption data of MB onto the AV-AC was best fitted by the Freundlich isotherm model.

4.7. Comparison With Other Studies

The maximum uptake capacity of MB dye by AV-AC in the present study was 129.87 mg/g. This value has been compared via Q_m obtained from other studies (Table 3). It can be concluded from Table 3 that AV-AC can be considered as a very effective adsorbent for the removal of MB from aqueous solution.

5. Discussion

In this work, Aloe Vera wastes-based activated carbon (AV-AC) as an inexpensive adsorbent was used for the removal of Methylene Blue (MB) from aqueous solutions. The development of pores in Figure 2B is due to the sorption of MB molecules that filled up pores of the sorbent surface and increased the surface area of the AV-AC. Chemical composition analysis of AV-AC showed that the main elements included O, Ca, K and Mg (92.7% of the total weight (wt. %)). Other minor elements were Na (5.92%) and Cl (1.35%) (20, 21). The fast adsorption at initial contact time can be

attributed to the accessibility of large numbers of the unfilled sites on the sorbent surface at the early step of the adsorption process and the adsorption sites were gradually occupied by the pollutant molecules as the contact time was increased (35, 36). This finding is consistent with the results of Hussin *et al.* for the removal of MB by durian leaf powder (17). Jourvand *et al.* reported that the equilibrium time was obtained in contact time of 60 minutes for removal of MB by HDTMA-Mt from aqueous solutions (4). Kazembigi *et al.* also presented that the uptake capacity for removal of MB by modified-RH was equal to 9.66 mg/g in an equilibrium time of 90 minutes (5). In another study, Utomo *et al.* obtained the equilibrium time of MB removal system by modified sugarcane bagasse equal to 30 minutes (37). The pseudo-second-kinetic model was fitted by the results of the present study. Similar kinetic results were expressed for the uptake of MB onto other adsorbents such as HDTMA-Mt (4), modified-RH (5), fly-ash (38), durian leaf powder (17), and *Scolymus hispanicus* L. (39). Valliammai *et al.* showed that R^2 value obtained for second-order-kinetic ($R^2 > 0.99$) is greater than that of first-order-kinetic ($R^2 > 0.98$); therefore, the second-order-kinetic is a perfect fit for the MB removal system (11). The solution pH influences the surface nature of adsorbent and the level of ionization of adsorbate (40, 41). Moreover, the sorption of adsorbate is affected by the solution pH, due to the sorption of H^+ and OH^- ions in the solution. The removal rate of MB was increased with increase of the solution pH from 2 to 12 due to the increase in the magnitude of the electrostatic attractions between MB and positive charge adsorption sites. The adsorption of MB on the AV-AC surface is primarily influenced by the surface charge on the adsorbent. This finding is in agreement with the results of the study of Barka *et al.* for the biosorption of MB. They showed that the sorption of MB is weak in acidic medium (39). Silanol groups on this surface become increasingly deprotonated as the pH of the sorption system rises, thereby increasing the number of negatively charged adsorbent sites. Reduced adsorption of MB at acidic pH reflects the presence of excess H^+ ions that compete with dye cations for the sorption sites (42). Valliammai *et al.* illustrated that higher sorption of MB at basic medium is due to the electrostatic attraction between the negatively charged sites of the adsorbent and the positively charged dye molecules. At higher pH, negatively charged adsorbent sites increased, which enhanced the adsorption of positively charged dye cations through electrostatic forces of attraction (11). Almeida *et al.* reported that the removal efficiency of MB by MC at pH of about 11 was about 97.38%, whereas at lower pH the removal efficiency was increased (42). To understand the sorption mechanism, it is required to determine the point zero charge (pH_{pzc}) of the adsorbent (43). On the other hand, pH_{pzc} is

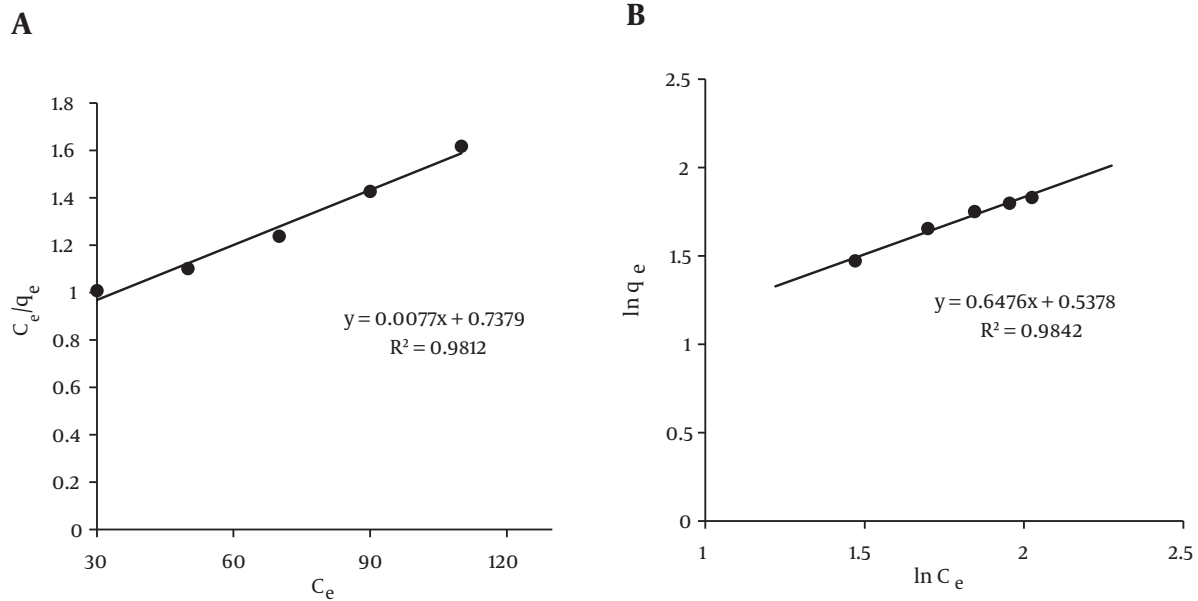


Figure 6. Plots of (A) Langmuir and (B) Freundlich Isotherm Models

Table 2. Parameters of Langmuir and Freundlich Isotherms Obtained From the Present Study

Adsorbate	Langmuir Isotherm			Freundlich Isotherm			
	Q_m , mg/g	B , l/mg	R^2	R_L	K_f , l/g	n	R^2
MB	129.87	0.014	0.981	0.58	3.16	1.56	0.984

Table 3. Comparison of Maximum Uptake Capacity of Methylene Blue by Aloe Vera Wastes-Based Activated Carbon and Other Adsorbents

Adsorbent	Q_{max}	Ref.
Neem saw dust	3.62	(30)
Fly ash	5.71	(31)
Coir pith carbon	5.87	(32)
Silk fibroin powder	20.8	(29)
Modified-RH	66.66	(5)
Cetylpyridinium-modified montmorillonite	80.67	(33)
Carbonaceous adsorbent	92	(34)
AV-AC	129.87	This study

a very important parameter to identify the interaction of sorbent surface with adsorbates (44). It has been identified that at any pH lower than pH_{pzc} , the surface charge is positive, whereas at pH higher than pH_{pzc} the surface charge is negative (22). Regarding pH_{pzc} , it can be stated that the surface of activated carbon will be de-protonated at pH values lower than 11.3. Hence, the sorption of negatively charged MB is hindered at pH values above pH_{pzc} . The pH_{pzc} of ad-

sorbent was more than 7.3 in the study of Hussin et al. (17). Decrease in the sorption capacity at higher dosage of AV-AC can be due to the unavailability of the MB molecules that cannot cover all the surface active sites of the adsorbent. In other words, a large number of active sites of the sorbent surface cannot reach saturation state at higher dosages of AV-AC. Barka et al. showed that with an increase of adsorbent dosage, the uptake capacity of the MB in solution was decreased (39). The sorption capacity of AV-AC was increased with increasing of MB concentrations. This is probably due to the increase in driving force of MB molecules including Vander Waal's force to the surface active sites of the adsorbent that occurs at higher concentrations of adsorbate. The experimental data of MB by AV-AC were well fitted by Freundlich isotherm model. Similar results were presented for the sorption of MB by several adsorbent such as ash of walnut shell (45), HDTMA-Mt (4) and modified-RH (5). The results of the study of Kazembigi et al. (2014) showed that the uptake of MB by rice husk was well fitted by the Freundlich isotherm model (5), which is consistent with the results of our study. Chang et al. (12) indicated that the adsorption data of MB using montmorillonite followed

by Freundlich isotherm ($R^2 > 0.99$) than that of Langmuir ($R^2 > 0.98$) and Tempkin ($R^2 > 0.93$) isotherm models.

5.1. Conclusion

The effect of various parameters such as contact time, pH, adsorbent dosage, and initial dye concentration was assessed on sorption. The optimum contact time in the sorption process was achieved after 40 minutes. Furthermore, the optimum pH was also obtained at pH 12. The experimental data were well fitted by pseudo-second-order kinetic and Freundlich isotherm models. The maximum monolayer adsorption capacity of MB was 129.87. The results illustrated that AV-AC as a low cost, eco-friendly, non-toxicity, and high capacity sorbent towards other sorbents can be used as an effective material for the removal of MB from aqueous solution.

Footnotes

Authors' Contribution: The manuscript was written by Yusef Omid, and other authors were involved in other tasks of the research.

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