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Low-Cost Activated Carbon for Methylene Blue Adsorption Using Inert Gas-Free Carbonization and Microwave Activation

Pornprapa Kongtragoul[1], Kanchalar Keeratirawee[2,*], and Orasa Choola-aied[3]

[1]Department of Agricultural Technology, King Mongkut's Institute of Technology Ladkrabang Prince of Chumphon Campus, Thailand
[2]Department of General Sciences, King Mongkut's Institute of Technology Ladkrabang Prince of Chumphon Campus, Thailand
[3]Laboratory Services, King Mongkut's Institute of Technology Ladkrabang Prince of Chumphon Campus, Thailand
Email: Pornprapa.ko@kmitl.ac.th (P.K.); Kanchalar.ke@kmitl.ac.th (K.K.); Orasa.ch@kmitl.ac.th (O.C.)
*Corresponding author

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***Abstract*—This study investigated the preparation of activated carbon from spent coffee grounds for methylene blue adsorption using inert gas-free carbonization and microwave activation. Spent coffee grounds were obtained from a local coffee shop in Pathio, Chumphon, Thailand. The coffee grounds were pretreated with 1M H_2SO_4 before undergoing carbonization at 600 ºC for 3 h in a limited-oxygen atmosphere. The obtained carbon materials were chemically activated with zinc chloride ($ZnCl_2$) and physically activated using a microwave-assisted method at 300 W for 180 seconds. The adsorption of methylene blue was evaluated using a batch method. Methylene blue adsorption contact time, effect of pH, and initial concentration were investigated. The maximum adsorption capacity of the activated carbon for methylene blue removal was found to be 29.24 mg/g. The prepared activated carbon exhibited a BET surface area of 70.08 m^2/g and reached equilibrium in 120 min at the optimum pH of 9. The adsorption isotherm was consistent with the Freundlich model, while the adsorption kinetics followed a pseudo-second-order model. The findings indicate that activated carbon is a cost-effective and efficient adsorbent for methylene blue removal from aqueous solutions.**

***Keywords*—methylene blue, coffee waste, activated carbon, microwave-assisted, zinc chloride**

I. INTRODUCTION

Methylene Blue (MB), a cationic dye, is widely utilized in various industries such as silk, paper, cotton, and wool, with its most extensively employed in textile dyeing [1, 2]. However, methylene blue is a non-degradable hazardous substance that can significantly impact living organisms, the environment, and aquatic ecosystems. Human exposure to methylene blue can result in health issues, including digestive problems, breathing difficulties, blindness, and neurological disorders. It also inhibits plant growth and lowers pigment and protein levels in microalgae [1, 3, 4]. While methylene blue is commonly used in aquaculture as a disinfectant and antifungal agent to control fish diseases, reduce mortality during transport, and maintain water quality, its use must be carefully regulated to prevent harm to aquatic life [5].

Various methods have been employed for the removal of methylene blue, including electrochemical methods [6], biological methods [7], photocatalytic degradation methods [8, 9], precipitation methods [10], and ozonolysis methods [11]. Among these, the adsorption method is recognized as a more efficient and cost-effective approach compared to other techniques for treating methylene blue due to its advantages in convenience, ease of optimization, and high suitability for liquid-phase applications in the environment [10, 12].

Many types of adsorbent materials have been utilized for methylene blue adsorption such as polymers [13], chitosan-nanocomposites [14], nanofibers [15], zeolite [16], and adsorption onto water hyacinth [17]. Within these materials, activated carbon is the most widely used material for dye adsorption due to its high surface area and excellent adsorption capacity for dye removal including methylene blue [18]. Many studies have investigated the conversion of low-cost and readily available agricultural wastes such as coconut shells, sugarcane bagasse, tea waste, and fruit peels into high-performance activated carbon. These activated carbons have been reported to have a high surface area and porosity that contribute to a strong adsorption capacity for organic and inorganic contaminants. This served as an alternative to commercial activated carbon as a renewable source [19].

II. LITERATURE REVIEW

Activated carbon for methylene blue adsorption has been prepared with various methods. Thabede *et al.* [20] prepared activated carbon from black cumin seeds for methylene blue adsorption from aqueous solution by using sulfuric acid (H_2SO_4) as an activating agent. The cumin seeds material was carbonized at 300 ºC under a nitrogen atmosphere. The adsorption capacity of activated carbon for methylene blue adsorption varied from 11 to 16 mg/g of adsorbent, and the optimum pH condition was at pH 9. The removal efficiency was found to range from 73 to 95%. Yağmur *et al.* [21] utilized coconut shells as raw materials for producing activated magnetic carbon for methylene blue adsorption. The materials were chemically activated with $ZnCl_2$ and carbonized at 500 ºC under nitrogen gas flow for 1 h. The maximum adsorption capacity was determined to be 156 mg/g adsorbent. Coffee ground-based activated carbon has been studied for methylene blue removal using various approaches, including variations in carbonization temperature, activating agent, and physical activation processes. Mutalib *et al.* [22] investigated the preparation of coffee grounds for methylene blue adsorption by using microwave-assisted methods for the activation process. The microwave power was varied from 100 to 400 Watts. The adsorption capacity of activated carbon for methylene blue adsorption was found to be 35 mg/g adsorbent. In another study, microwave-assisted activation combined with conventional heating was used to prepare activated carbon

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from almond shells with phosphoric acid as the activating agent for methylene blue adsorption. The researchers applied microwave power ranging from 250 to 1000 W for 10 to 20 min, following 45 min of conventional heating at temperatures between 400 and 600 ºC, with phosphoric acid to sample ratio under various conditions. The prepared activated carbon achieved a high surface area of 1128 m²/g and exhibited a methylene blue adsorption capacity of 148 mg/g [23]. This approach demonstrated that the use of microwave-assisted activation is a promising method for methylene blue removal. In another study by Reffas *et al.* [24], they prepared the activated carbon from coffee grounds using phosphoric acid (H_3PO_4) as an activating agent for the adsorption of methylene blue and Nylosan Red N-2RBL. After acid treatment, the coffee grounds material was heated at 450 ºC in an air atmosphere. The surface area of obtained activated carbon was comparable to that of commercial activated carbon. This approach confirmed that the preparation of activated carbon from coffee grounds can be carried out without the need for inert gas flow during carbonization.

In this study, we demonstrated the use of coffee ground waste as a raw material for preparing activated carbon to adsorb methylene blue. We combined carbonization under a limited air atmosphere, microwave activation, and chemical activation with $ZnCl_2$. This method is effective in removing methylene blue from aqueous solutions.

III. MATERIALS AND METHODS

A. Materials and Instruments

Spent coffee grounds were obtained from a local coffee shop in Pathio, Chumphon, Thailand. Sulfuric acid (98%) and silver nitrate were purchased from QREC, New Zealand. Potassium hydroxide, sodium hydroxide, and zinc chloride were purchased from KEMAUS, Australia. All chemicals used in this study were of analytical grade. The instruments used in this study included a bench pH meter (model STARTER3100) from Ohaus, New Jersey, USA, an ultraviolet-visible spectrophotometer (model V-750) from Jasco, Japan, and a benchtop shaker (model innova®40) from New Brunswick Scientific, New Jersey, USA.

B. Preparation of Adsorbents

The coffee grounds were dried in a hot air oven at 105 ºC overnight, it was later washed with 1M H_2SO_4 (v/v) at a ratio of 1 g of coffee grounds to 1 mL of H_2SO_4. The mixture was soaked and shaken at an agitation speed of 150 rpm and a fixed temperature of 25 ºC for 24 h. The resulting materials were then washed with DI water and dried in the hot air oven at 105 ºC overnight. The coffee ground materials were carbonized at 600 ºC for 3 h. The obtained carbon material was chemically activated by soaking it in a $ZnCl_2$ solution for 24 h at a ratio of 1:1 (mixing 10 grams of carbon with 10 grams of $ZnCl_2$ and adding 50 mL of DI water). Carbon materials after activation were dried overnight and then physically activated using a microwave-assisted process at a power of 300 watts for 180 s, with a pause after 30 s to prevent potential overheating because the process was performed without the inert gas flow. The obtained activated carbon was washed several times with DI water until the rinsed water became clear and showed no presence of Cl^- ions when tested with a drop of $AgNO_3$. The resulting materials were then dried overnight at 105 ºC and stored in a desiccator before use.

C. Batch Experiment for Methylene Blue Adsorption

Adsorption experiments were performed using a batch method. In each trial, 20 mg of activated carbon was placed into an Erlenmeyer flask, followed by the addition of 50 mL of 10 mg/L of methylene blue solution (equivalent to 0.01 g/L). The mixture was stirred at 150 rpm for varying durations from 2–240 min to evaluate equilibrium conditions. Samples were periodically collected and filtered using Whatman No.1 filter paper. The concentration of methylene blue was determined using a UV spectrophotometer at a maximum absorbance wavelength of 662 nm.

The effect of pH on methylene blue adsorption was studied by adding 20 mg of activated carbon to an Erlenmeyer flask, followed by 50 mL of 10 mg/L methylene blue solution. The pH was adjusted between 3–11 using 0.05 M NaOH and HCl before the experiments. The mixtures were shaken at 150 rpm for 120 min.

The influence of initial methylene blue concentration was examined by adding 20 mg of activated carbon to an Erlenmeyer flask, followed by 50 mL of methylene blue solutions with concentrations ranging from 2–10 mg/L, the pH of each concentration was adjusted to 9. The mixtures were shaken at a constant rate of 150 rpm for 120 min.

Adsorption isotherms were investigated by adding Methylene blue solutions with concentrations ranging from 2–10 mg/L to 20 mg of activated carbon in an Erlenmeyer flask. The pH of all solutions was adjusted to 9 before the experiment. The mixtures were stirred at 150 rpm or 120 min to facilitate fitting to adsorption models.

Kinetics studies were conducted by adding 20 mg of activated carbon to an Erlenmeyer flask, followed by 50 mL of 10 mg/L of methylene blue solution. The pH of methylene blue solution was adjusted to pH 9, and the mixtures were agitated at 150 rpm. Samples were taken at various time intervals between 2–240 min for analysis.

IV. RESULT AND DISCUSSION

A. Effect of Acid Washing on Ash Formation

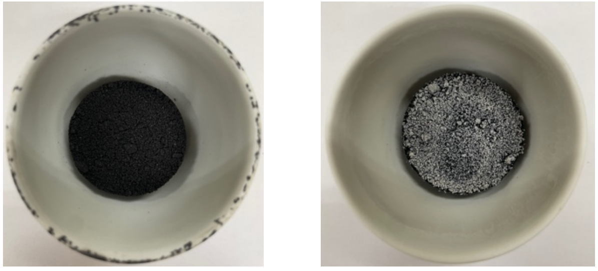
(a) (b)

Fig. 1. Effect of acid washing on ash formation (a) washed with 1M H_2SO_4 and (b) un-washed with 1M H_2SO_4.

The acid washing step was aimed at reducing ash formation during carbonization, as our study investigated carbonization and microwave-assisted activation without inert gas flow. The effect of acid washing was demonstrated by comparing ash formation in washed and unwashed coffee grounds with H_2SO_4 as illustrated in Fig. 1. The acid washing significantly affected as formation, with visible ash present in the unwashed materials but none observed in the washed materials. This demonstrates that acid washing effectively removes ash-forming impurities before carbonization. Our findings align with those of Hwang *et al.* [25] They reported that acid washing reduces ash formation and increases specific surface area. Therefore, the acid-washing step was employed in this study to prepare for the subsequent activation process in the experiment.

B. Characterization of Coffee-Based Activated Carbon

1) Scanning Electron Microscopy (SEM)

The surface morphology of prepared activated carbon was examined using SEM, and the resulting micrograph is shown in Fig. 2.

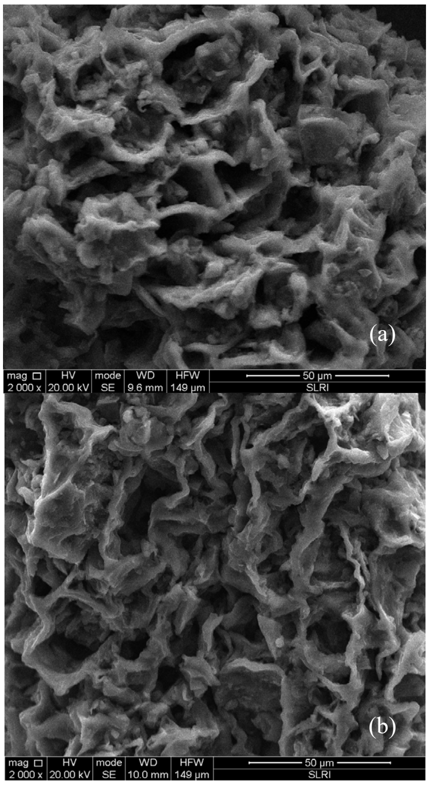

Fig. 2. The SEM image of (a) coffee grounds carbonized at 600 ºC for 3 h, (b) coffee grounds carbonized at 600 ºC for 3 h, activated with $ZnCl_2$ and using microwave-assisted method.

The upper SEM image represents the external structure of coffee grounds carbonized at 600 ºC for 3 h under limited oxygen, without any chemical or physical activation. The surface appears relatively compact, with fewer visible pores and less structural openness.

In contrast, the lower SEM image shows the coffee grounds treated under the same conditions but subjected to chemical activation with $ZnCl_2$ and physical activation using a microwave-assisted method. This carbon material reveals a more porous structure with larger pore openings. The improvement in porosity is attributed to chemical activation by $ZnCl_2$, which promotes pore development through decomposition processes, while the rapid energy input from microwave heating enhances structural expansion and the volatilization of organics during carbon formation [26].

2) Surface area analysis

The nitrogen adsorption-desorption isotherm of the prepared activated carbon is shown in Fig.3. According to the IUPAC classification, the curve follows a type IV isotherm with an H4-type hysteresis loop. This indicates the presence of mesopores and slit-like pores found in the carbon structure [27–29].

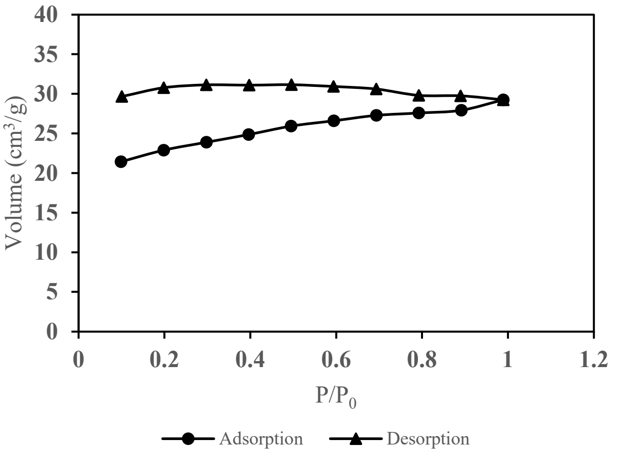

Fig. 3. The nitrogen adsorption-desorption isotherm.

The specific surface area and porosity properties of prepared activated carbon are shown in Table 1. BET analysis indicated a surface area of 70.08 m^2/g, and carbon materials contained micropores of 50.43 m^2/g. The total pore volume was 0.045 cm^3/g with an average pore diameter of approximately 2.6 nm.

Although the type IV isotherm indicates mesoporosity, the high micropore surface area and volume reveal a predominantly microporous structure with some contribution from mesopores. The H4-type hysteresis loop further supports the presence of slit-shaped pores commonly found in carbon-based materials.

Table 1. Specific surface area and porosity properties of prepared activated carbon

Parameter	Value
Surface area	70.08 m^2/g
Micropore area	50.43 m^2/g
Total pore volume	0.045 cm^3/g
Micropore volume	0.027 cm^3/g
Average pore diameter	2.58 nm

C. Effect of Contact Time

Adsorption equilibrium of methylene blue onto activated carbon was observed at different time intervals: 2, 10, 30, 60, 90, 120, 180, and 240 min. The quantity of methylene blue adsorbed per gram of activated carbon (q) at each interval was determined using Eq. (1) [29]

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

where q_e is the adsorption capacity at equilibrium (mg/g), C_o is the initial concentration (mg/L), C_e is the concentration at equilibrium (mg/L), V is the volume of methylene blue solution (L), and m is the mass of activated carbon used (g).

The effect of contact time on methylene blue adsorption is depicted in Fig. 4. It was observed that the adsorption rate increased rapidly within the first 2 min and stabilized after 120 min. The adsorption capacity was calculated to be 19 mg/g, using Eq. (1). This reveals that the most of active sites on the activated carbon were occupied with no further changes occurring thereafter. Therefore, a contact time of 120 min was chosen as the optimum condition for subsequent experiments.

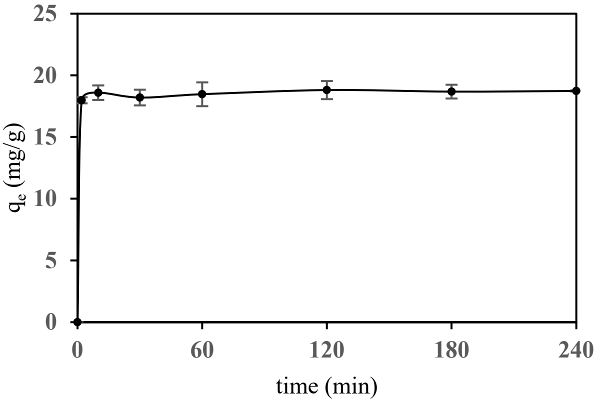

Fig. 4. Effect of contact time.

D. *Effect of pH*

The effect of pH on methylene blue adsorption was examined at pH 3, 5, 7, 9, and 11 with a contact time of 120 min. The effect of pH is shown in Fig. 5. In this study, adsorption strongly depends on pH due to changes in surface charge and electrostatic interactions. In acidic conditions (pH 3–5), adsorption capacity and removal efficiency increased rapidly as pH increased. This is because, at lower pH, the surface of activated carbon was not strongly negative charge, this reduced its attraction to methylene blue, a cationic dye. Under basic conditions, the surface of activated carbon became more negatively charged, this enhanced adsorption through stronger electrostatic force between the positively charged methylene blue and the negatively charged activated carbon. The maximum adsorption capacity and removal efficiency were observed at pH 9. These results were consistent with the findings of Tang *et al.* [30] and Larakeb *et al.* [31], who also reported that the adsorption capacity is higher in basic solutions. Based on these results, pH 9 was chosen for further experiments for further experiments to optimize adsorption performance.

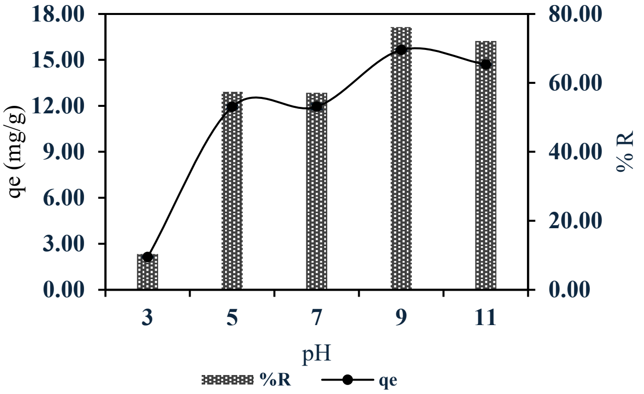

Fig. 5. Effect of pH.

E. *Effect of Initial Methylene Blue Concentration*

The effect of initial concentration on adsorption is the factor used to define the efficiency of adsorbent materials. In this study, the influence of initial methylene blue concentration on adsorption capacity was evaluated by varying the concentration at 2, 4, 6, 8, and 10 mg/L. The pH was adjusted to 9 for all methylene blue concentration tests. The adsorption capacity at equilibrium was calculated using Eq. (1). The effect of initial methylene blue concentration is shown in Fig. 6.

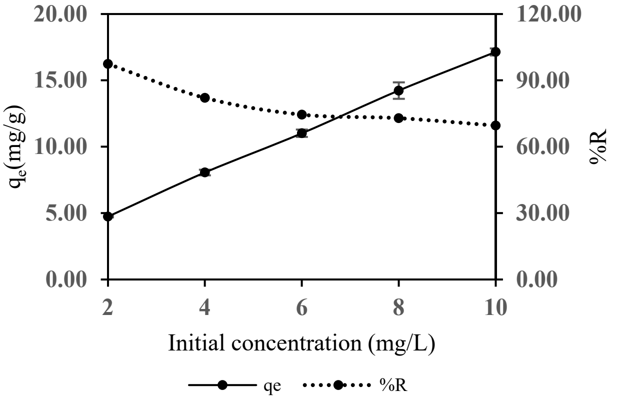

Fig. 6. Effect of initial methylene blue concentration.

The results indicated that the initial dye concentration significantly affected the removal efficiency. As the initial methylene blue concentration increased from 2 to 10 mg/L, the removal efficiency decreased from 97% to 69%. This is due to the reduction of available active sites on the activated carbon as the concentration of adsorbate increased [32]. In contrast, the adsorption capacity of activated carbon for methylene blue adsorption increased from 4 mg/g to 17 mg/g adsorbent. This is because there are more methylene blue molecules in the solution at higher concentrations, and it leads to more mass transfer being available to occupy the active site on the adsorbent [33].

F. *Adsorption Isotherms*

The adsorption isotherms of methylene blue on activated carbon derived from coffee grounds were studied by adjusting the methylene blue concentration to 2, 4, 6, 8, and 10 mg/L. The data were evaluated using the Langmuir and Freundlich model, which were calculated using Eq. (2) and Eq. (3) [29], respectively. The results are shown in Table 2. The Langmuir and Freundlich isotherms plots are shown in Fig. 7.

$$\frac{C_e}{q_e} = \frac{1}{K_L \times q_{max}} + \frac{C_e}{q_{max}} \tag{2}$$

where C_e is the concentration at equilibrium (mg/L), q_e is the adsorption capacity at equilibrium (mg/g), K_L is the Langmuir constant (L/mg), and q_{max} is the adsorption capacity of the monolayer (mg/g).

$$\log q_e = \log K_F + \frac{1}{n} \times \log C_e \tag{3}$$

where K_F is Freundlich constant (mg/g(L/mg)$^{1/n}$), and $1/n$ is the adsorbate for the adsorbent.

The results from the adsorption study indicated that the Freundlich isotherm model provides a better fit for the adsorption of methylene blue onto coffee grounds-based activated carbon, as indicated by a slightly higher correlation coefficient value of 0.9315 compared to the Langmuir isotherm model, which had a value of 0.8936. Therefore, the adsorption of methylene blue onto activated carbon in this study follows a heterogeneous surface with a non-uniform distribution of adsorption sites. The low adsorption capacity of methylene blue is likely due to the moderate surface area measured by BET, which limits available active sites. Additionally, carbonization and microwave activation without an inert gas atmosphere may have resulted in a less developed porous structure.

Table 2. Adsorption isotherms for methylene blue adsorption onto activated carbon

Isotherm Models					
Langmuir			**Freundlich**		
q_m (mg/g)	K_L (L/mg)	R^2	K_f (mg/g(L/mg)$^{1/n}$)	$1/n$	R^2
29.24	1.55	0.8936	10.66	3.34	0.9315

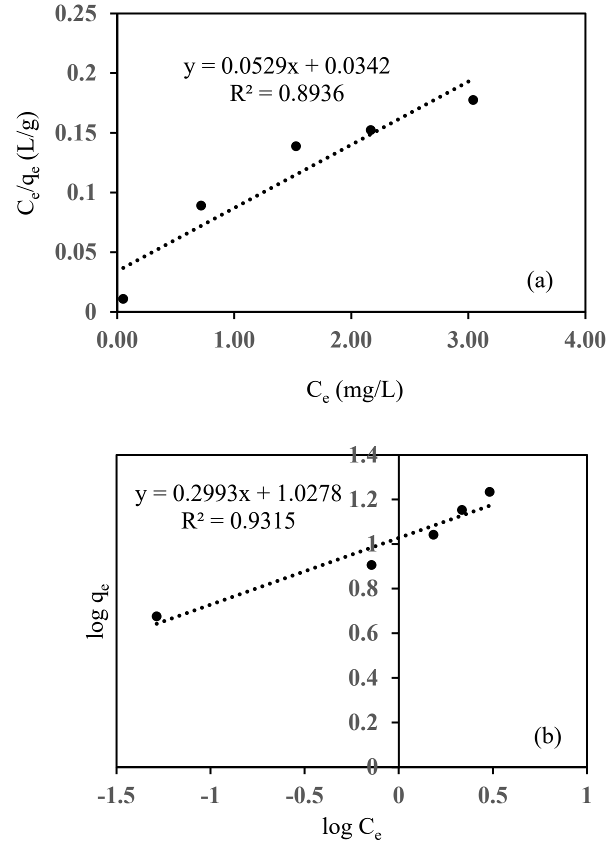

Fig. 7. Adsorption isotherm of methylene blue activated carbon (a) the Langmuir model and (b) the Freundlich model.

G. Kinetic Studies

The kinetic studies for methylene blue onto activated carbon were evaluated using pseudo-first-order and pseudo-second-order models, which can be calculated using Eq. 4 and Eq. (5) respectively [29].

$$ln\,(q_e - q_t) = lnq_e - k_1 t \tag{4}$$

where q_e is the amount of methylene blue adsorbed at equilibrium (mg/g), q_t *is* the amount of methylene blue adsorbed at time *t* (mg/g), *k* is the rate constant for the pseudo-first-order model (1/min), and t is the time (min)

$$\frac{t}{q_t} = \frac{1}{K_2 \times q_e^2} + \frac{t}{q_e} \tag{5}$$

where K_2 is the rate constant for the pseudo-second-order model (g/mg·min)

The adsorption kinetics were evaluated using both pseudo-first-order and pseudo-second-order models. A summary of adsorption kinetics for methylene blue adsorption is shown in Table 3, and the adsorption kinetics of methylene blue on activated carbon are illustrated in Fig. 8.

Table 3. Adsorption kinetics for methylene blue on activated carbon

Kinetic Model	k_1 (1/min)/k_2 (g/mg·min)	q_e (mg/g)	R^2
Pseudo-first order	k_1 = 0.0064	1.137	0.8492
Pseudo-second order	k_2 = 0.0526	36.23	0.9999

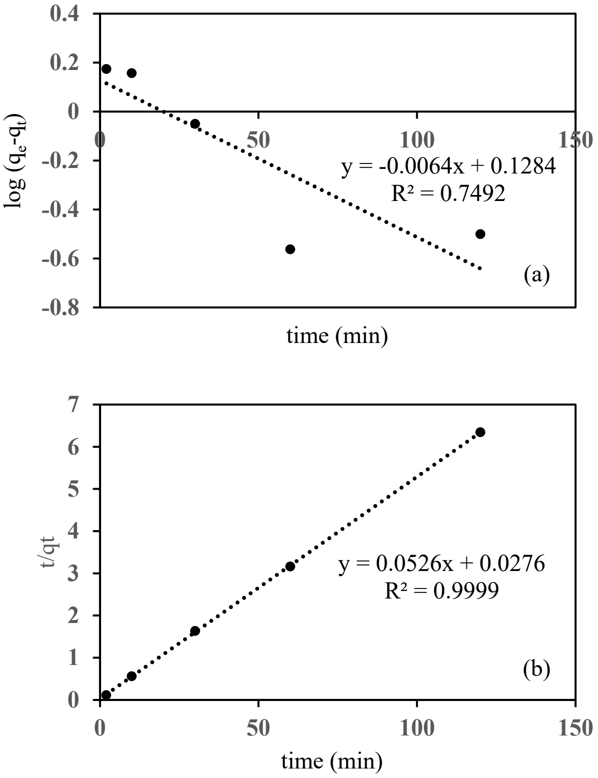

Fig. 8. Adsorption kinetics of methylene blue on activated carbon (a) pseudo-first-order and (b) pseudo-second-order.

The pseudo-first-order provided a correlation coefficient (R^2) value of 0.7492 and the rate constant (k_1) was 0.0064 (1/min), revealing a poor fit. On the other hand, the pseudo-second-order demonstrated a significantly better fit with the correlation value, R^2 of 0.9999, and the rate constant of 0.0526 g/mg·min. The results suggested that the adsorption of methylene blue onto activated carbon follows a chemical adsorption mechanism. The results obtained from this study aligned with the investigation of Paluri et al. and Jawad *et al.* [34, 35], they reported that the adsorption of methylene blue fit better with pseudo-second-order.

V. CONCLUSION

This study successfully developed low-cost activated carbon from coffee grounds for the adsorption of methylene blue. The activation process was carried out without the need for an inert gas flow during both the carbonization and microwave-assisted activation processes, which makes it both cost-effective and practical. The adsorption results followed the Freundlich isotherm model, which indicated that the adsorption process involves multilayer adsorption on a heterogeneous surface. The pseudo-second-order kinetic model fits the data better than the pseudo-first-order, indicating that the adsorption process is influenced by chemical adsorption. This approach could be a promising method for further development in various adsorption applications.

CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

AUTHOR CONTRIBUTIONS

Pornprapa Kongtragoul conducted the experiments, provided sources, and assisted with the data analysis. Kanchalar Keeratirawee conceptualized and designed the study, mainly conducted the experiments, analyzed the data, and wrote the manuscript. Orasa Choolaaied prepared glassware for the experiments.; all authors had approved the final version.

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