

Article

Adsorptive Removal of Methylene Blue (MB) Dye from Synthetic Wastewater by Biochar Prepared from Olive Mill Solid Wastes: Process Performance, Kinetics, Equilibrium, and Thermodynamics

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Abstract

This study evaluated the potential of biochar (BC) produced from olive mill solid waste (OMSW) for methylene blue (MB) removal from synthetic solutions. BC was prepared at pyrolysis temperatures of 400 °C, 500 °C, and 600 °C for residence times of 1, 3, and 5 h. The produced BCs were characterized using BET, SEM, FTIR, XRD, zeta potential, and elemental analysis, while adsorption performance was assessed through equilibrium, kinetic, and thermodynamic studies. The results demonstrated that increasing pyrolysis temperature improved the physicochemical properties of BC. The BET surface area increased from 1.34–1.9 m²/g at 400 °C to 135.23 m²/g at 600 °C, accompanied by enhanced pore volume and the development of a highly porous structure observed by SEM. FTIR analysis revealed progressive transformation of oxygen-containing functional groups, while XRD indicated increased structural ordering and carbonization at higher pyrolysis temperatures. BC produced at 600 °C exhibited the highest adsorption performance, achieving a maximum MB removal efficiency of 99.57% and an adsorption capacity of 17 mg/g. Adsorption kinetics were best described by the pseudo-second-order model ($R^2 > 0.99$), indicating strong adsorbate–adsorbent interactions. Equilibrium results were best fitted by the Langmuir isotherm with a maximum R^2 of 0.9963, suggesting predominantly monolayer adsorption, whereas the Freundlich model showed a maximum R^2 of 0.9788 under elevated temperature and prolonged contact conditions, indicating surface heterogeneity and multilayer adsorption. Thermodynamic analysis confirmed that MB adsorption was spontaneous ($\Delta G^\circ = -38.61$ to -47.57 kJ/mol), predominantly endothermic (ΔH° up to 4.11 kJ/mol), and associated with increased randomness at solid–solution interface ($\Delta S^\circ > 0$). Overall, BC produced at 600 °C for 3–5 h exhibited the most favorable structural characteristics and adsorption performance, highlighting its potential as a sustainable and low-cost adsorbent for wastewater treatment applications.



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Keywords: adsorption of dyes; biochar preparation; adsorption isotherm; adsorption kinetics; methylene blue wastewater; olive mill solid wastes; pyrolysis; wastewater treatment

1. Introduction

The olive oil industry is a key agricultural sector in Jordan, contributing significantly to the national economy and rural development [1,2]. However, it simultaneously generates large quantities of olive mill solid waste (OMSW), locally known as “Jift”, with annual production estimated at around 120,000 tons [3–5]. Improper management of this waste can lead to serious environmental problems, including soil degradation, groundwater contamination, and emission of unpleasant odors, which highlight the need for sustainable waste management and valorization strategies.

In recent years, the conversion of agricultural residues into biochar (BC) via thermochemical processes has gained increasing attention as an effective waste management and environmental remediation strategy [6,7]. BC is a carbon-rich, porous material produced under oxygen-limited pyrolysis conditions, typically between 300–700 °C, and its physicochemical properties are highly dependent on feedstock type and production parameters such as temperature, residence time, and heating rate [7–9]. These parameters directly influence surface area, pore structure, and surface functional groups, which ultimately govern adsorption performance [10,11].

Among various agricultural residues, OMSW is considered a promising precursor for BC production due to its rich lignocellulosic composition, mainly consisting of cellulose (38–50%), hemicellulose (23–32%), and lignin (15–25%) [12]. This composition enhances carbon yield and contributes to the development of functionalized carbon surfaces suitable for pollutant adsorption. Previous studies have confirmed that BC derived from olive-related wastes exhibits potential for removing organic and inorganic contaminants from aqueous solutions [12,13]. The physical and chemical properties of BC, such as its high surface area, high porosity, and the presence of functional groups, equip it with excellent adsorptive capabilities for a variety of contaminants, including hazardous heavy metals and synthetic dyes [12,13].

Methylene blue (MB) is a widely used cationic dye in textile and dyeing industries, and its discharge into aquatic environments represents a significant environmental risk due to its toxicity, persistence, and potential ecological impacts [13]. Consequently, the development of efficient, low-cost, and sustainable adsorbents for MB removal has become an important research focus [13–15]. A wide range of BC derived from different feedstocks has been investigated for dye adsorption, including agricultural residues such as rice husk, wheat straw, corn cob, sawdust, lychee seeds, and algal biomass [16–18]. Reported adsorption capacities vary significantly depending on precursor type and preparation conditions, for example, rice husk BC (315.4–359.7 mg/g) [19], lychee seeds BC (124.5 mg/g) [18], sawdust-based BC (568.16 mg/g) [20], and algae-derived carbon materials (>200 mg/g in some modified forms) [21,22]. These variations highlight the critical role of feedstock selection and pyrolysis conditions in determining adsorption efficiency.

The efficiency of BC's adsorption of MB is influenced by several critical factors, including the pH of the solution, the initial MB concentration, and the temperature [23]. For example, recent studies highlight that higher pH levels improve the adsorption capacity by reducing competition from HO⁻ ions [24], while increased initial concentrations of MB provide a greater driving force for adsorption [25]. Temperature also plays a significant role; elevated temperatures generally enhance molecular kinetic energy, promoting more effective interactions between the dye molecules and BC [26–28]. Despite the growing

number of studies on dye removal using BC, there remains a need for systematic evaluation of OMSW-derived BC produced under controlled pyrolysis conditions, particularly with respect to the combined effect of temperature and residence time. Most previous studies have focused on single-condition pyrolysis or chemically modified BCs, while limited attention has been given to optimizing production time as an additional controlling parameter affecting surface development and adsorption behavior.

In this study, biochar was produced from OMSW at different pyrolysis temperatures (400, 500, and 600 °C) and residence times (1, 3, and 5 h), and its performance for MB removal was evaluated using aqueous model solutions under controlled conditions. The influence of key operational parameters, including pH, initial dye concentration, and temperature, was also investigated to understand adsorption behavior and optimize removal efficiency. In addition, adsorption kinetics, equilibrium isotherms, and thermodynamic models were applied to describe the adsorption process. The novelty of this work lies in the systematic evaluation of residence time as a key parameter in OMSW BC production, as well as the comparative assessment of adsorption performance under different pyrolysis conditions. The study also establishes a relationship between production conditions and MB removal efficiency. Overall, this work contributes to the valorization of agricultural waste by converting OMSW into a functional adsorbent for dye removal. It also supports the optimization of BC production conditions and highlights its potential application in water treatment processes.

2. Materials and Methods

2.1. Collection and Preparation of Biochar from Agriculture Solid Waste

The OMSW, locally known as Jift or Zibar, was collected from an olive mill in Ajloun between October and December 2023. OMSW is a lignocellulosic agricultural by-product rich in organic matter, cellulose, hemicellulose, lignin, and residual oil content, which makes it a suitable precursor for BC production due to its high carbon availability and reactivity [12,29]. The samples were subsequently transported to the Chemistry Laboratory at the University of Jordan for BC preparation.

The preparation process involved several steps. Initially, the OMSW was thoroughly washed with distilled water to remove impurities. The cleaned materials were then dried in an oven (MATEST A005-08 KIT, Matest S.p.A., Arcore, Italy) at 50 °C for 24 h to reduce moisture content. After drying, the biomass was sieved through a 4.75 mm mesh to remove residual olive leaves and stones, ensuring uniform feedstock for pyrolysis.

For the pyrolysis process, 100-g samples of prepared OMSW were wrapped in aluminum foil and heated in a Nabertherm muffle furnace (Lilienthal, Germany) at temperatures of 400 °C, 500 °C, and 600 °C for 1, 3, and 5 h, respectively. After pyrolysis, the produced BC was ground using a Silver Crest SC-1880 grinder (Silver Crest, Hong Kong, China) to obtain a uniform particle size. The BC was then sieved using an electric shaker Electromagnet A059-02 KIT, (Matest S.p.A., Arcore, Italy) to obtain particles of 0.075 mm, which were stored in labeled containers for further use.

2.2. Characterisation of Biochar

The BC yield was calculated based on dry mass relative to the initial biomass. The pH was measured by suspending the BC in distilled water at a ratio of 3:50 (BC:water) and shaking for 2 h before measurement using a Hann pH meter “HI 2002-02, Hanna Instruments Ltd., Bedfordshire, UK” [30].

Electrical conductivity was assessed with “a conductivity meter” “CON 6-LaMotte, LaMotte Co., Washington, DC, USA”. The ash content was evaluated according to a modified ASTM standard, which involved measuring weight loss [30,31]. Approximately 5 g

of oven-dried BC, dried for 24 h at 105 °C, was weighed and combusted at 750 °C for 6 h [32]. After combustion, the samples were cooled to room temperature in a desiccator and reweighed to determine the ash content [32].

Additionally, “Fourier-transform infrared spectroscopy” (FTIR) and morphological analysis using “scanning electron microscopy” (SEM) were performed following protocols established in our previous study [32]. “X-ray diffraction” (XRD) analysis was conducted using an “AERIS X-ray diffractometer from Malvern Panalytical (Almelo, The Netherlands).”

2.3. Chemicals

All chemicals utilized in this study were of analytical grade. The chosen cationic dye for adsorption experiments was MB, also known as methylthioninium chloride. Its chemical structure is represented as 3,7-bis (Dimethylamino)-phenothiazine-5-ium chloride, with the molecular formula $C_{16}H_{18}N_3SCl$ and a molecular weight of 319.85 g/mol. MB was sourced from Sigma Aldrich (St. Louis, MO, USA) with a purity of 99.9%. A stock solution with a concentration of 1000 mg/L was prepared by dissolving 1000 mg of MB powder in a 1000 mL volumetric flask, subsequently filled with distilled water to reach the desired volume. Various experimental concentrations of the dye, specifically 10, 20, 30, 40, 50, 60, 70, 80, 90, and 100 mg/L, were obtained by diluting the stock solution with additional distilled water. To adjust the pH of these solutions, either 1 M HCl (Sigma-Aldrich, St. Louis, MO, USA) or 0.5 M NaOH (Sigma-Aldrich, St. Louis, MO, USA) was added, with pH levels monitored using a digital pH meter “Hanna HI 2002-02, Hanna Instruments Ltd., Bedfordshire, UK”. The concentration of MB in solution was quantified using a UV/Vis spectrophotometer. A calibration curve was constructed by plotting the absorbance against the concentrations of the dye solutions, facilitating the determination of unknown concentrations based on the established linear relationship.

2.4. Wastewater Treatment Application of BC

Adsorption experiments utilizing MB dye were conducted to assess the efficacy of BC in synthetic wastewater treatment. The study investigated the effects of various parameters including pH, temperature, and initial MB concentration using BC produced through pyrolysis of OMSW at temperatures of 400, 500, and 600 °C, with treatment durations of 1, 3, and 5 h. The effect of several operating parameters such as pH, temperature, initial dye concentration was investigated.

2.4.1. Effects of Operational Parameters

To evaluate the impact of pH on adsorption, 0.04 g of BC produced at 600 °C for both 3 and 5 h was placed in a glass cylindrical cell. This BC was mixed with 25 mL of MB solution at an initial concentration of 50 mg/L across a pH range of 2 to 12. The experiments were conducted in an incubator at room temperature for 24 h. The pH of the solutions was adjusted as necessary using 0.1 M HCl or 0.5 M NaOH solutions. In addition, the adsorption experiments were replicated under similar conditions as the pH experiments, maintaining an initial MB concentration of 50 mg/L at a pH of 12. Approximately 0.04 g of BC was mixed with 25 mL of the MB solution in a glass beaker. The mixture was agitated at 200 rpm in an incubator set to temperatures of 20, 30, and 40 °C for a duration of 24 h.

2.4.2. Adsorption Capacity of BC

The concentration of MB at equilibrium was determined using a calibration curve obtained at the wavelength of maximum absorbance (664 nm). The removal efficiency of the MB dye (R%) was calculated using the following equation [33]:

$$R(\%) = \frac{(C_0 - C_e)}{C_e} * 100 \quad (1)$$

where C_0 and C_e represent the initial and final MB concentrations (mg/L), respectively.

The adsorption capacity of the BC was calculated using the following equation [34]:

$$q \left(\frac{\text{mg}}{\text{g}} \right) = \frac{(C_0 - C_e) * V_{\text{solution}}}{m_{\text{ads}}} \quad (2)$$

where V_{sol} is the volume of the MB solution used in the adsorption (L), m_{ads} is the mass of BC used as the adsorption (g), and q is the adsorption capacity of the BC (mg/g).

2.5. Batch Adsorption Kinetic Experiments

Adsorption measurements were conducted through batch experiments involving a fixed amount of BC and 25 mL of aqueous MB dye solution at known concentrations. A series of 50 mL plastic bottles was prepared, each containing varying initial concentrations of MB ranging from 10 to 100 mg/L. A consistent amount of 0.04 g of BC was added to each flask, based on findings from a previous study. The mixture was agitated at a constant temperature using a Stuart Scientific Stirrer SS3 (Stuart Scientific Co., Ltd., Stone, Staffordshire, UK) set to 200 rpm and maintained at room temperature. Mixing time intervals varied from 5 to 1440 min, during which periodic samples were collected to monitor changes in MB concentrations. The data obtained from these kinetic experiments were critical in evaluating the efficiency of BC for removing MB from aqueous solutions. Further adsorption experiments were systematically designed by varying several parameters, including initial solution pH, contact time, adsorbent dosage, initial MB concentration, and temperature.

To analyze the adsorption kinetics, both pseudo-first-order and pseudo-second-order models were employed. The pseudo-first-order adsorption kinetic rate equation is generally represented as [35]:

$$\text{Log}(q_e - q_t) = \text{Log}(q_e) - \frac{K_1}{2.303} t \quad (3)$$

where q_e and q_t represent the adsorption capacities (mg/L) at equilibrium and at time t , respectively, and K_1 is the rate constant of pseudo-first-order adsorption (min^{-1}).

The kinetic equation for pseudo-second-order adsorption is given by [36]:

$$\frac{1}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \quad (4)$$

where K_2 denotes the rate constant of pseudo-second-order adsorption (g/mg/min).

2.6. Adsorption Isotherm Study

In the study of adsorption isotherm, ten samples of MB solution were prepared and placed into 25 mL flasks, each containing a fixed initial concentration (C_0) of 50 mg/L. To each flask, a predetermined amount of BC (0.04 g) was added. The samples were mixed for 24 h at varying temperatures of 30, 40, and 50 °C to achieve equilibrium. After this period, the mixtures were allowed to settle for 1 h. Samples were subsequently collected from the bottom of the flasks using appropriate pipette techniques and filtered before analysis.

Adsorption isotherms play a vital role in comprehensively understanding the adsorption dynamics. In this analysis, two models, the Freundlich and Langmuir isotherms, were applied to interpret the adsorption data effectively. These models are frequently utilized in literature to describe the nonlinear relationship between q_{eq} and C_{eq} under constant temperature conditions [18,37]. To calculate the adsorbate amount at any specified time, the following equation was used [38]:

$$q_t = \frac{(C_0 - C_t)V}{m_s} \quad (5)$$

where C_0 and C_t signify the concentrations of MB in mg/L at the beginning and at time t , respectively. V is the volume of the MB solution measured in L, and m_s represents the mass of biochar in grams.

The Freundlich isotherm can be expressed mathematically as [39]:

$$q_e = K_F C_e^{\frac{1}{n}} \quad (6)$$

where q_e corresponds to the amount of adsorbate adsorbed per unit mass of the adsorbent (mg/g), C_e indicates the concentration of the dissolved solute in the MB solution after equilibrium is reached (mg/L), and K_F and n are empirical constants that characterize the specific adsorbent–solute interaction, temperature, and material type.

The Langmuir isotherm is defined by the equation [40]:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{K_1 * q_m * C_e} \quad (7)$$

where C_e denotes the equilibrium concentration of MB dye (mg/L), q_m reflects the maximum adsorption capacity per unit weight, q_e pertains to the amount of adsorbate taken up by the adsorbent (mg/g), and K_1 represents the maximum adsorption coefficient (L/mg).

Recent advancements in research, notably by Ho [41], have highlighted the benefits of nonlinear chi-square analysis (X^2) for assessing how well experimental data fits isotherm models. The chi-square value is computed using this formula:

$$X^2 = \sum \frac{(q_e - q_{e,m})^2}{q_{e,m}} \quad (8)$$

where X^2 signifies the non-linear chi-square value (mg/g), q_e (mg/g) represents the experimentally observed equilibrium capacity (mg/g), and $q_{e,m}$ (mg/g) indicates the equilibrium capacity derived from the models. According to Ho's Ho [41] findings, a lower X^2 value indicates a superior fit of the model to the experimental data.

2.7. Thermodynamic Analysis of Adsorption

The thermodynamic parameters governing an adsorption process are essential for evaluating its feasibility and efficiency [42]. In this study, key thermodynamic factors, standard enthalpy change (ΔH°), standard entropy change (ΔS°), and Gibbs free energy change (ΔG°), were investigated to elucidate the temperature effects on the adsorption process. These parameters were calculated using the following equations [42]. The distribution coefficient (K_d) is defined as follows:

$$K_d = \frac{q_{eq}}{C_{eq}} \quad (9)$$

The relationship between the distribution coefficient and the thermodynamic parameters can be expressed by:

$$\ln K_d = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{R * T} \quad (10)$$

Additionally, ΔG° can be calculated using:

$$\Delta G^\circ = \Delta H^\circ - T * \Delta S^\circ \quad (11)$$

In these equations, T represents the adsorption temperature in Kelvin, R is the ideal gas constant (8.314 J/mol. K), and K_d is the distribution coefficient.

3. Results and Discussion

3.1. Characterization of BC

The production of BC from OMSW through pyrolysis at varying temperatures of 400, 500, and 600 °C for different periods of 1, 3, and 5 h results in BC with distinct physiochemical properties and structural characteristics. Understanding these properties is essential for their application in pollutant adsorption, especially for dyes like MB. The properties of BC produced under these conditions are summarized in Table 1. This analysis provided a comparison of the yield, moisture content, electrical conductivity, ash content, surface area, pore structure, elemental composition, and functional properties of BC produced at these pyrolysis temperatures.

Table 1. Physiochemical properties and structure characteristics of BC produced at 400, 500, and 600 °C pyrolysis temperature for durations of 1, 3, and 5 h.

Parameters	Unit	Pyrolysis Temperature								
		400 °C			500 °C			600 °C		
		1 h	3 h	5 h	1 h	3 h	5 h	1 h	3 h	5 h
Moisture Contents	%	−0.121	1.126	0.735	1.48	1.86	1.20	1.79	2.61	4.18
Electrical Conductivity	mS/cm	0.01125	0.0212	0.0162	0.082	0.179	0.078	0.018	0.029	0.077
Yield	%	32.08	29.81	27.73	25.18	27.23	28.11	24.62	24.27	18.18
Ash Contents	%	3.56	6.7	6.47	3.83	7.55	7.79	6.07	5.95	10.83
Zeta Potential	mV	−22.72	−25.15	−27.93	−32.21	−27.08	−26.39	−25.2	−23.53	−29.89
Average pore width	Å	27.73	35.54	24.53	31.59	23.11	23.99	22.06	21.74	21.95
Total pore volume	cm ³ /g	0.00097	0.0011	0.0012	0.006	0.025	0.026	0.075	0.065	0.0055
Langmuir Surface area	m ² /g	2.12	2.03	2.73	64.77	10.72	70.50	201.6	170.12	14.84
BET Surface Area	m ² /g	1.39	1.34	1.90	6.99	43.63	44.19	9.96	119.89	135.23
“Single point Surface area at P/Po = 0.297”	m ² /g	1.33	1.31	1.98	41.28	6.94	38.92	134.5	116.73	9.91
H	%	1.791	1.882	2.321	2.87	2.877	3.323	3.575	3.65	3.123
N	%	2.876	2.492	-	-	-	-	-	2.454	3.123
C	%	86.484	87.895	87.8	49.68	85.65	80.07	80.77	72.68	67.21
O	%	8.849	7.731	9.879	47.45	11.473	16.607	15.655	21.216	26.544
Atomic ratio H/C	%	0.0207	0.0214	0.0264	0.0578	0.0336	0.0415	0.0443	0.0502	0.047
Atomic ratio O/C	%	0.1029	0.0878	0.1128	0.9547	0.1336	0.207	0.1938	0.2912	0.395
Atomic ratio (O+N)/C	%	0.1356	0.1163	0.1125	0.9551	0.1336	0.207	0.1938	0.3257	0.441

The properties of BC produced from OMSW at varying pyrolysis temperatures (400 °C, 500 °C, and 600 °C) can be analyzed by comparing key characteristics associated with different thermal decomposition periods, as shown in Table 1. This analysis also examines the correlation between these properties and the capacity for MB dye adsorption.

At a relatively low temperature of 400 °C, the BC yield was notably high, ranging from 32.08% after 1 h to 27.73% after 5 h. However, surface area measurements were limited, showing values between 1.34 and 1.9 m²/g, indicating minimal porosity. The zeta potential values ranged from −22.72 mV to −27.93 mV, reflecting the presence of negatively charged functional groups capable of interacting with positively charged dyes. Despite these interactions, the limited surface area and pore structure adversely impacted the BC's adsorption capability. These findings align with existing literature, such as Ji et al. [43], which reported that BC with restricted surface area exhibits lower adsorption efficiency for MB.

Increasing the pyrolysis temperature to 500 °C resulted in significant enhancements in structural properties. The BET surface area increased substantially, ranging from 6.99 m²/g to 44.19 m²/g, and pore volume values ranged from 0.006 cm³/g to 0.026 cm³/g. Additionally, a higher zeta potential of −32.21 mV was recorded, suggesting the development of fine and mesoporous structures. The BC maintained a high carbon content of 85.65% at 3 h, alongside a reduced O/C ratio of 0.1336, which indicates a decrease in surface oxygen functionalities while preserving chemical reactivity. This configuration indicates that BC produced at 500 °C achieves a favorable balance of porosity, surface area, and carbonization, thus improving its ability to adsorb MB, consistent with findings by Dos Santos et al. [44].

At 600 °C, notable changes were observed as the BET surface area reached 135.23 m²/g after 5 h, and maximum pore volume was recorded at 0.075 cm³/g after 1 h. Despite a decrease in yield to 18.18%, the increased surface area and pore network enhanced the biochar's capacity to adsorb MB efficiently. The zeta potential ranged between −23.53 mV and −29.89 mV, coupled with suitable ratios of oxygen and nitrogen that facilitate oxidative functional groups. These conditions significantly promote hydrogen bonding and electrostatic interactions with the dye. This observation is consistent with Abid et al. [12], who demonstrated that BC from OMSW at higher temperatures exhibited substantial improvements in porosity and surface area, thus enhancing the adsorption of organic compounds.

Comparing the three pyrolysis temperatures (400 °C, 500 °C, and 600 °C) reveals that BC at 600 °C offers the highest adsorption efficiency due to its substantial surface area, developed pore volume, adequate zeta potential, and oxidative functional groups. Conversely, the 400 °C model exhibits a higher yield but is limited in porosity, while the 500 °C model achieves a reasonable balance yet does not match the exceptional surface area of the 600 °C variant. This trend is supported by other studies, including Dawood et al. [23] and Dos Santos et al. [44], which indicate that elevated pyrolysis temperatures enhance surface area and micro-porosity, key factors in improving the adsorption of MB and other organic pollutants. Overall, selecting a higher pyrolysis temperature (600 °C) combined with an appropriate duration (3 to 5 h) yields the most efficient BC for the removal of MB. The enhanced surface area, micro-porosity, and oxidative functionalities provide numerous active adsorption sites, surpassing the limitations of lower temperature models.

3.1.1. FTIR Analysis

FTIR analysis of BC derived from OMSW at 400 °C, 500 °C, and 600 °C reveals key chemical and structural transformations during pyrolysis, as shown in Figure 1. These transformations are crucial for evaluating the adsorptive capacity of BC for organic pollutants, particularly MB.

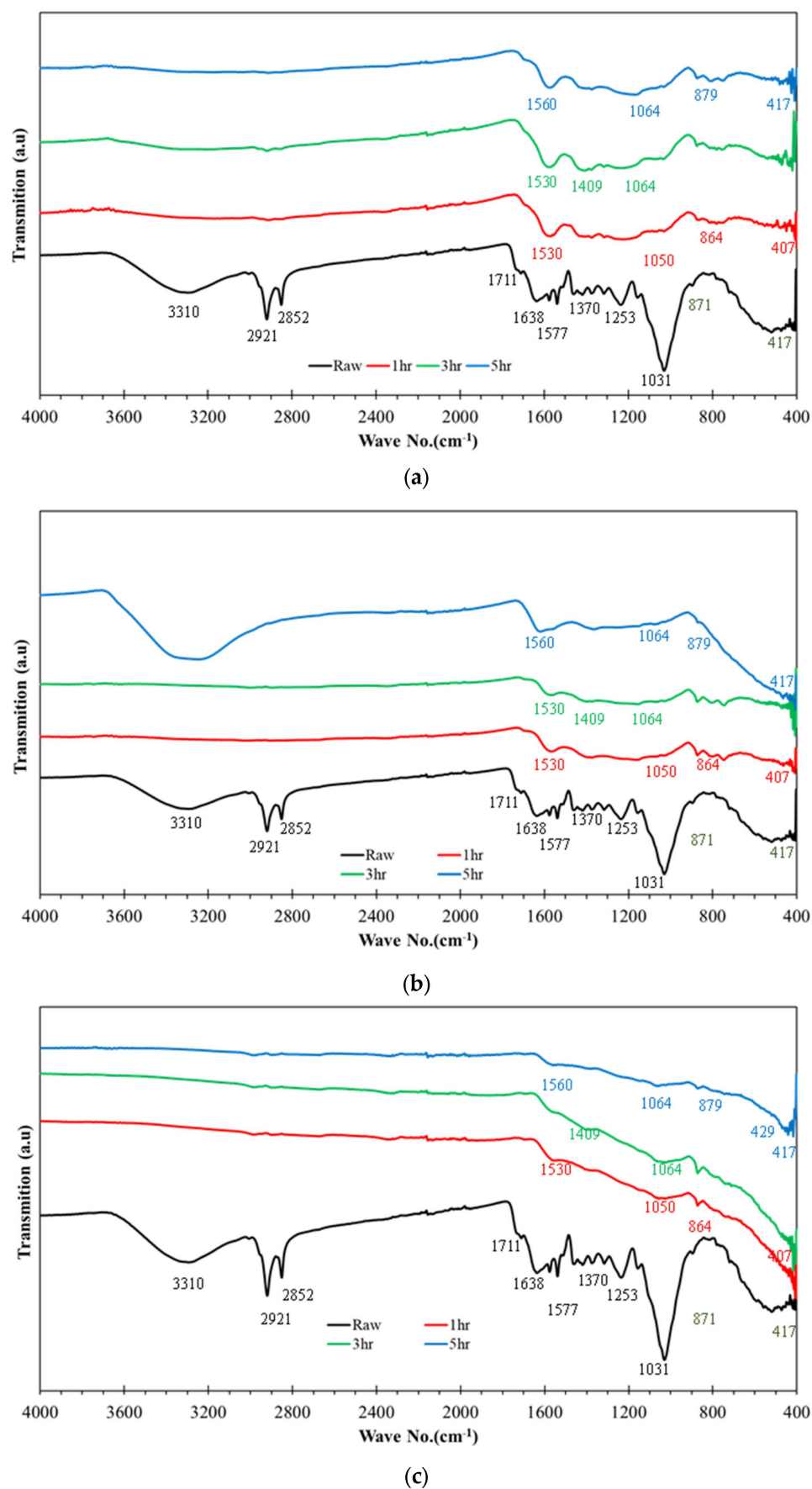


Figure 1. FTIR spectra of raw feedstock and BC produced at: (a) 400 °C, (b) 500 °C, and (c) 600 °C.

The FTIR spectrum of raw OMSW exhibits several distinct absorption bands indicative of its lignocellulosic composition. A broad band at 3310 cm^{-1} corresponds to the stretching vibrations of hydroxyl (-OH) groups. Absorption peaks at 2921 cm^{-1} and 2852 cm^{-1} are associated with aliphatic C-H stretching vibrations. The band at 1711 cm^{-1} is attributed to carbonyl (C=O) functional groups, while peaks at 1638 cm^{-1} and 1577 cm^{-1} correspond to C=C stretching within aromatic rings, primarily linked to lignin structures. Bands in the range of $1459\text{--}1376\text{ cm}^{-1}$ are assigned to CH_2 bending vibrations of cellulose and lignin. The absorption bands at 1234 , 1129 , and 1031 cm^{-1} are characteristic of C-O and C-O-C stretching vibrations in cellulose and hemicellulose backbone structures [45].

After heat treatment at $400\text{ }^{\circ}\text{C}$ for 1, 3, and 5 h, the intensity of bands at 3310 , 2921 , 2852 , 1711 , 1234 , 1129 , and 1031 cm^{-1} decreases significantly. This reduction results from dehydration and the progressive degradation of cellulose, hemicellulose, and lignin. Conversely, the aromatic C=C band in the $1580\text{--}1620\text{ cm}^{-1}$ region becomes more prominent, indicating the formation of aromatic structures during carbonization.

With additional thermal treatment at $500\text{ }^{\circ}\text{C}$ and $600\text{ }^{\circ}\text{C}$, the intensity of these bands continues to decrease and eventually disappears as pyrolysis temperature increases, indicating extensive thermal decomposition and the loss of oxygen-containing functional groups. New bands appearing in the $400\text{--}600\text{ cm}^{-1}$ range are attributed to the formation of metal-oxygen bonds from inorganic constituents (micronutrients) present in OMSW.

3.1.2. X-Ray Diffraction of BC

X-ray diffraction (XRD) is an effective technique for examining the structural organization and crystallinity of BC derived from the thermal decomposition of OMSW [46]. In this study, XRD was used to evaluate BC produced at different pyrolysis temperatures (400 , 500 , and $600\text{ }^{\circ}\text{C}$) and residence times (1–5 h), as shown in Figure 2, to assess the influence of these parameters on its suitability for methylene blue (MB) adsorption. The diffractogram of raw OMSW exhibits a broad peak within the range of $2\theta \approx 15\text{--}35^{\circ}$, indicating a largely amorphous and disordered structure, typical of untreated biomass with limited crystalline organization [47].

At $400\text{ }^{\circ}\text{C}$ (Figure 2a), BC retains a predominantly amorphous structure, as evidenced by broad diffraction peaks, suggesting incomplete carbonization and the presence of unconverted organic matter. Although increasing the residence time slightly improves peak intensity, the temperature remains insufficient to induce significant structural reorganization, thereby limiting the formation of active adsorption sites. At $500\text{ }^{\circ}\text{C}$ (Figure 2b), the BC shows notable structural enhancement, with sharper peaks around $2\theta \approx 23^{\circ}$ indicating increased aromatic condensation and improved carbon layer ordering, along with reduced background intensity due to the removal of volatile components. At $600\text{ }^{\circ}\text{C}$ (Figure 2c), the biochar exhibits the highest degree of structural development, as evidenced by the emergence of well-defined peaks at approximately $2\theta \approx 26.7^{\circ}$, 29.4° , 36° , 39.4° , 43° , 47.5° , 48.4° , 50° , and 51.3° . These features indicate enhanced carbonization, reduced volatile content, and the formation of partially ordered carbon domains, along with the development of minor crystalline phases associated with inorganic constituents (micronutrients) present in OMSW.

Overall, increasing pyrolysis temperature promotes the transition from an amorphous biomass structure to a more organized, carbon-rich framework. Consequently, BC produced at $600\text{ }^{\circ}\text{C}$, particularly with extended residence time, demonstrates the most favorable structural characteristics for MB adsorption due to improved stability and increased availability of active sites.

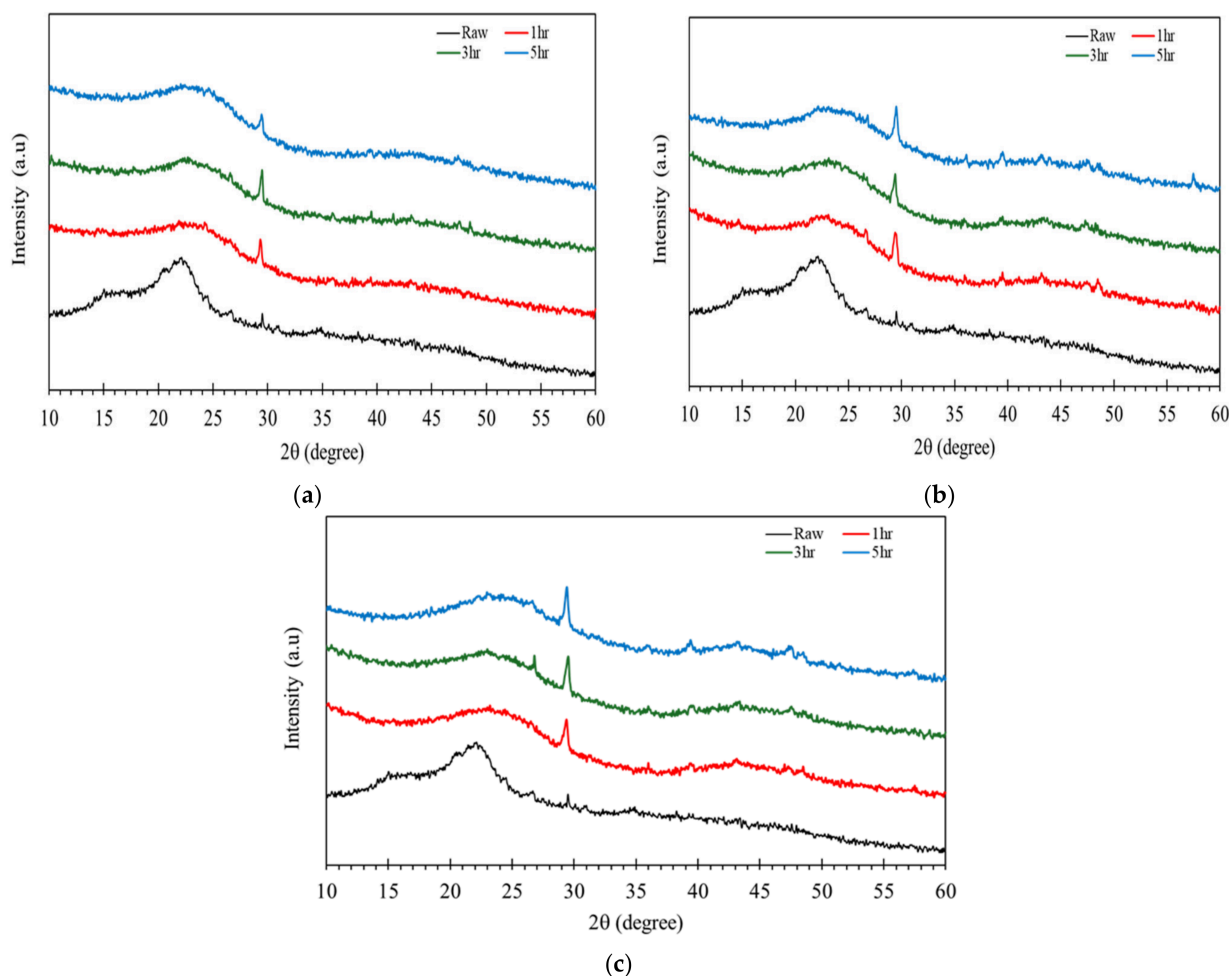


Figure 2. X-ray diffraction patterns of Raw feedstock and BC produced at: (a) 400 °C, (b) 500 °C, and (c) 600 °C.

3.1.3. SEM Analysis of BC

Figure 3 shows that raw BC exhibits a rough surface characterized by irregularly shaped particles. This non-uniform structure, typical of unprocessed biomass, reveals compact clusters of organic matter that retain a high degree of integrity. The pronounced lack of porosity significantly limits the surface area available for adsorption, making raw BC less effective in removing pollutants from solutions. Similar findings were noted by Abid et al. [12], who observed that the raw OMSW exhibited a rough surface without pores, underscoring inherent limitations in its applications for adsorption.

Figure 4 shows the feedstock produced at 400 °C, 500 °C, and 600 °C for varying durations of 1, 3, and 5 h. Figure 4a shows BC pyrolyzed at 400 °C for 1 h. The micrograph retains a rough texture similar to that of raw BC, but shows only minimal changes. It remains relatively compact with limited porosity. These slight alterations indicate that exposure to heat for this duration has not significantly enhanced its adsorptive capacity compared to the raw BC. Figure 4b depicts BC processed at 400 °C for 3 h. This micrograph reveals the emergence of small pores and a rougher surface texture, suggesting that prolonged exposure to heat facilitates structural modifications. This increase in porosity enhances the material's potential for adsorption, although it still lags behind the characteristics observed at higher temperatures. In Figure 4c, the BC produced at 400 °C for 5 h exhibits a marked increase in surface complexity and visible pore formation. This enhancement indicates that prolonged thermal treatment fosters sufficient degradation to improve adsorptive characteristics.

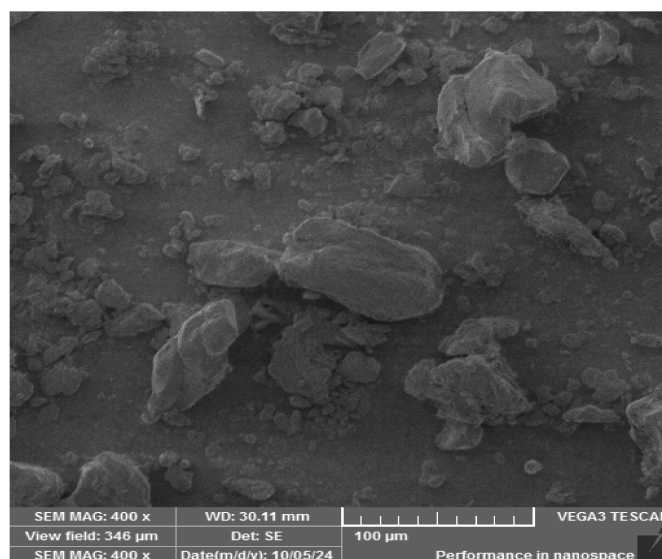


Figure 3. SEM micrographs of raw feedstock samples.

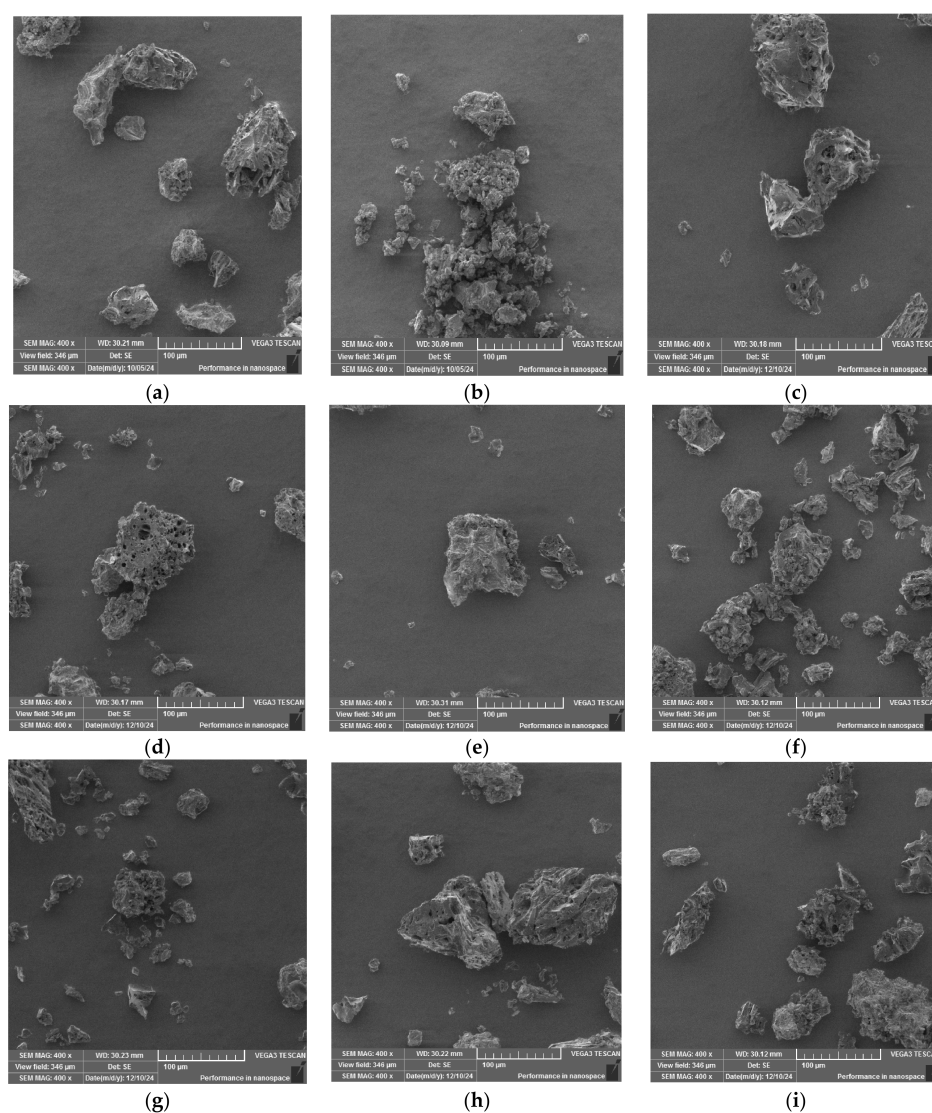


Figure 4. SEM micrographs of BC samples produced at (a) 400 °C at 1 h, (b) 400 °C at 3 h, (c) 400 °C at 5 h, (d) 500 °C at 1 h, (e) 500 °C at 3 h, (f) 500 °C at 5 h, (g) 600 °C at 1 h, (h) 600 °C at 3 h, and (i) 600 °C at 5 h.

Figure 4d presents BC processed at 500 °C for 1 h. The surface appears grainier, with initial signs of porosity becoming more evident. This transformation highlights how elevated temperature enhances structural characteristics even during shorter durations, positioning this biochar as more effective for adsorption compared to raw BC. Figure 4e shows BC produced at 500 °C for 3 h. The micrograph indicates significant increases in porosity and well-defined pore structures. These enhancements underscore the effectiveness of combining elevated temperature with extended dwell time, resulting in a material that exhibits marked improvements in adsorptive capacity, comparable to findings in Abid et al. [12], where BC at 450 °C demonstrated visible pores and grainy surfaces. In Figure 4f, the SEM analysis of BC pyrolyzed at 500 °C for 5 h reveals a highly porous structure. The abundant pore formations represent optimal conditions for adsorption, suggesting a substantial increase in surface area relative to raw BC and lower-temperature samples.

Figure 4g displays BC processed at 600 °C for 1 h. The micrograph showcases a significant increase in porosity, with numerous well-defined pores enhancing the surface area. This substantial structural improvement already marks a noticeable advantage over both the raw BC and samples processed at lower temperatures. In Figure 4h, the image illustrates BC treated at 600 °C for 3 h, with even greater pore development and uniform distribution observed across the surface. This advancement highlights the effectiveness of high-temperature pyrolysis in maximizing surface area, resulting in optimal adsorptive capability. Finally, Figure 4i features BC produced at 600 °C for 5 h. The SEM micrograph reveals extraordinary levels of porosity, with uniform pore sizes throughout the surface. This state, achieved after extended pyrolysis under high temperature, is critical for maximizing adsorption efficacy. The enhancements observed in this sample substantially outshine those of the raw and lower-temperature treatments, confirming that 600 °C is the most effective condition for producing high-quality BC suited for adsorption applications. This finding is consistent with the results of Abid et al. [12], who reported that BC produced at 650 °C exhibited improved porosity and more visible pores. Their study emphasizes that higher pyrolysis temperatures enhance the adsorptive properties of BC.

3.1.4. Effect of Surface Area and Pore Volume

The analysis of surface area and pore volumes for BC produced at different pyrolysis temperatures, as illustrated in Figure 5, underscores the significant influence of pyrolysis conditions on the structural properties of BC derived from OMSW. The trends indicate a consistent increase in both surface area and pore volume with rising temperatures and longer pyrolysis durations.

At a temperature of 400 °C, the surface area and pore volume are observed to be relatively moderate. Specifically, the surface area increases from 1.33 m²/g at 1 h to 1.9 m²/g after 5 h, while the pore volume experiences a corresponding rise from 0.97 to 1.2 × 10³ cm³/g. This suggests that even at lower pyrolysis temperatures, thermal treatment can enhance the structural characteristics of biochar, albeit modestly. Findings by Güleç et al. [24] and Varma et al. [48] support this observation, indicating that pyrolysis at lower temperatures leads to limited increases in surface area and porosity. At 500 °C, a significant enhancement is observed in both the surface area and pore volume. The surface area increases dramatically from 6.99 m²/g at 1 h to 44.19 m²/g at 5 h, while pore volume rises from 5.5 to 26 × 10³ cm³/g. This notable improvement highlights that higher temperatures promote enhanced carbonization and volatilization processes, which contribute to the development of more pronounced porous structures [24,49]. Increasing the temperature to 600 °C further amplifies these enhancements. At this level, the surface area reaches 135.23 m²/g, and the pore volume climbs to 75 × 10³ cm³/g at 5 h. Such

significant development can be attributed to the breakdown of organic materials and the formation of new, intricate pore structures.

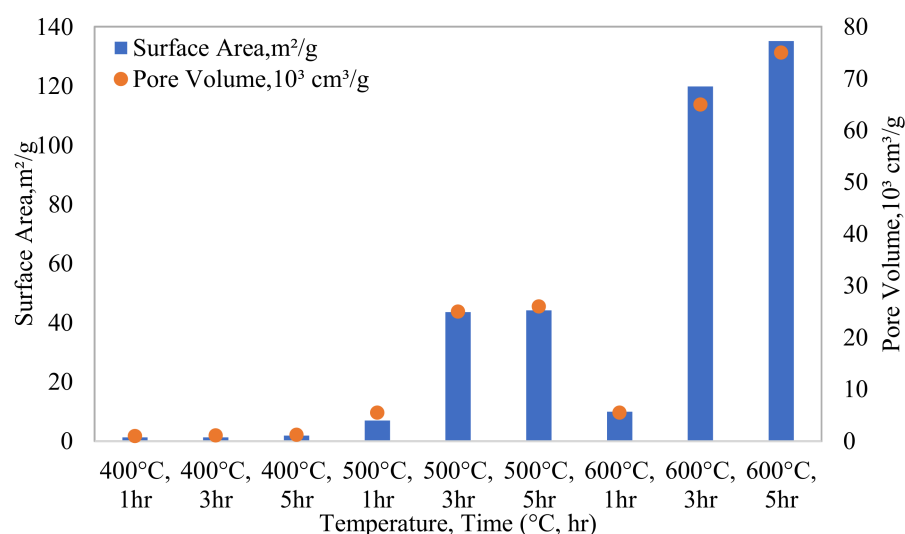


Figure 5. Surface area and pore volume for the BC produced at three pyrolysis temperatures for different durations.

3.2. Isotherm Adsorption Models

The equilibrium adsorption behavior of MB was evaluated using the Langmuir and Freundlich isotherm models, with parameters summarized in Table 2. These models elucidate the adsorption mechanisms, surface characteristics, and interactions between the adsorbate and BC produced from pyrolysis at varying temperatures (400, 500, and 600 °C) under different operational conditions and contact times.

As shown in Table 2, the Langmuir model provided a superior fit for BC produced at 600 °C, particularly at a 1-h contact time. The R^2 of 0.9447 and the low X^2 of 0.2209 suggested monolayer adsorption on a relatively homogeneous surface, consistent with Langmuir's assumptions [50]. While the maximum adsorption capacity (q_{max}) remained stable (~5.6–6 mg/g), a gradual decline in R^2 and an increase in X^2 with longer contact times indicated potential surface heterogeneity and the onset of site saturation. At higher operational temperatures, specifically 40 °C, the Langmuir model significantly improved its fit, particularly at 5 h, achieving $R^2 = 0.9963$ and a low X^2 value (0.533). This finding suggests enhanced accessibility to uniform adsorption sites and improved interactions between the adsorbate and BC. The Freundlich model was less effective in this context, reinforcing the predominance of monolayer adsorption. When the temperature increased further to 50 °C, a transition in adsorption behavior occurred. While the Langmuir model showed moderate to low R^2 values at longer contact times, the Freundlich model exhibited an excellent fit at 1 h ($R^2 = 0.9788$, $n = 4.53$), indicating a shift towards heterogeneous adsorption and potential multilayer formation, in line with existing literature on thermally activated adsorption processes [51].

For BC produced at 500 °C, the Langmuir model initially performed poorly at 1 h due to a negative K_L value, indicating non-physical adsorption behavior. However, at 3 h, it exhibited a good fit with $R^2 = 0.8999$ and a low X^2 , suggesting better establishment of equilibrium conditions. The Freundlich model underperformed overall, as indicated by lower correlation coefficients. At this temperature, the Langmuir model continued to improve, particularly at 40 °C after 3 h, where R^2 reached 0.9189. The Freundlich parameters displayed low R^2 values despite high n , indicating that surface heterogeneity alone could not fully explain the adsorption behavior. At 50 °C, the Langmuir model

demonstrated excellent alignment with experimental data at 3 h ($R^2 = 0.9663$), yielding the highest recorded adsorption capacity for this BC ($q_{max} = 6.1$ mg/g). This result indicates optimal monolayer adsorption performance, suggesting that the BC produced at 500 °C is effective at elevated temperatures when given sufficient contact time.

Table 2. Langmuir and Freundlich constants for adsorption at different temperatures.

Pyrolysis Temperature (°C)	$T, ^\circ\text{C}$	Contact Time, h	Langmuir Model				Freundlich Model			
			K_L (L/mg)	q_{max} (mg/g)	R^2	X^2	K_F (mg/L)	N	R^2	X^2
600	30	1	0.641	5.903	0.9447	0.2209	0.703	6.716	0.8667	34.57
		3	1.391	5.56713	0.8538	0.5554	0.7984	10.3	0.5713	36.87
		5	1.152	5.9838	0.8144	4.887	1.0338	5.0223	0.7949	100.3
	40	1	1.036	5.37	0.9423	0.1838	0.7568	8.762	0.7679	33.15
		3	1.146	5.56134	0.8726	0.766	0.81226	9.3133	0.54	41.94
		5	0.829	5.9838	0.9963	0.533	0.99452	4.1973	0.8674	97.2
	50	1	0.586	5.38773	0.8995	35.358	0.70688	4.53	0.9788	28.6
		3	1.907	5.382	0.7758	0.2478	0.78512	20	0.3373	43.91
		5	1.195	6.059	0.7806	5.035	1.11838	3.7322	0.8483	154.9
500	30	1	−1.723	4.433	0.4164	0.5347	0.7143	12.66	0.232	12.33
		3	2.091	5.8912	0.8999	0.6391	0.8277	7.9587	0.8408	42.86
		5	0.624	4.7	0.8397	0.3365	0.633	7.1432	0.8322	20.05
	40	1	2.242	4.815	0.4672	0.1988	0.6732	20.891	0.4331	31.38
		3	2.775	5.99	0.9189	0.6614	0.8847	10.074	0.6001	54.45
		5	1.543	5.1505	0.7126	0.3107	0.7462	17.46	0.3543	37.77
	50	1	0.391	4.855	0.7487	2.4333	0.56545	3.877	0.9054	17.47
		3	4.765	6.1	0.9663	0.271	0.89904	10.076	0.7784	55.48
		5	0.802	4.92	0.8074	0.3237	0.6547	7.6148	0.8683	21.77
400	30	1	0.403	4.635	0.7882	0.7882	0.6855	6.697	0.5742	26.91
		3	0.156	4.178	0.9751	0.4503	0.4957	3.1509	0.9497	18.34
		5	0.218	4.491	0.9745	0.3939	0.6537	4.4336	0.7613	26.74
	40	1	0.249	4.5602	0.924	1.004	0.5737	3.7833	0.8901	20.26
		3	0.412	5.0521	0.9046	1.603	0.6892	4.7643	0.7793	29.1
		5	4.847	6.094	0.9295	0.4828	0.87862	9.4427	0.8537	51.4
	50	1	0.253	4.6586	0.9654	0.361	0.6166	3.9843	0.8991	23.22
		3	0.399	4.4618	0.8589	0.7205	0.5988	5.6431	0.7696	18.84
		5	2.795	6.475	0.8713	1.747	0.8593	6.2603	0.9769	46.94

The Langmuir model for BC produced at 400 °C improved significantly with increased contact time, achieving high R^2 values (>0.97) at 3 and 5 h, which indicates stable monolayer adsorption. The Freundlich model also showed reasonable agreement at shorter contact times, suggesting partial surface heterogeneity. At 30 °C, the Langmuir model provided a good fit, particularly with extended contact time, achieving high R^2 values. In the context of 40 °C, the Langmuir model consistently performed well across all contact times, with the highest adsorption capacity observed at 5 h ($q_{max} = 6.094$ mg/g). Comparatively, the Freundlich model was weaker, indicating that adsorption was primarily governed by uniform surface sites. When evaluating at 50 °C, the Langmuir fitting was notably strong at 1 h ($R^2 = 0.9654$), while the Freundlich model became more suitable at 5 h, achieving a very high R^2 value (0.9769). This behavior implies a combined adsorption mechanism, where initial monolayer adsorption transitions to multilayer adsorption as contact time and operational conditions increase.

Overall, the Langmuir isotherm generally provided the best description of adsorption behavior at low to moderate operational conditions, particularly for BCs produced at 400

and 500 °C. This confirms that adsorption predominantly occurs as monolayer coverage on relatively homogeneous surfaces. Conversely, the Freundlich model became more relevant under higher operational conditions and extended contact times, particularly for BC produced at 600 °C, indicating increased surface heterogeneity and multilayer adsorption. These findings align with existing literature, which demonstrates that elevated temperatures enhance the complexity of adsorption mechanisms and promote pore-filling effects [51].

3.3. Kinetics Adsorption Analysis

This section presents an analysis of the kinetics of MB adsorption on BCs produced at varying pyrolysis temperatures: 400 °C, 500 °C, and 600 °C. The performance of both the Pseudo-first-order and Pseudo-second-order kinetic models in characterizing the adsorption behavior of MB on these BCs is compared. The kinetic parameters for MB adsorption onto BCs produced at different pyrolysis temperatures are summarized in Table 3.

Table 3. Kinetic parameters for MB adsorption onto BC produced at different pyrolysis temperature (400, 500, and 600 °C).

Pyrolysis Temperature, °C	Time, h	q_e (exp.) (mg/g)	Pseudo-First Order Kinetic Model			Pseudo-Second Order Kinetic Model		
			q_e (cal.) (mg/g)	K_1 (1/h)	R^2	q_e (cal.) (mg/g)	K_2 (g/mg.h)	R^2
600	1	5.6985	5.0163	−0.000622	0.4539	5.828	0.00503	0.9992
	3	4.3689	4.31	−0.000666	0.3227	4.49	0.00588	0.9961
	5	17.004	16.9398	−0.000558	0.8831	17.1673	0.00326	0.9994
500	1	4.5956	5.3371	−0.000639	0.3469	5.12089	0.00093	0.2876
	3	8.4436	8.052	−0.000118	0.4522	8.6356	0.00344	0.999
	5	2.8431	3.02	−0.001119	0.6772	2.30879	−0.00201	0.8463
400	1	2.4265	1.42	−0.001664	0.4086	2.1309	−0.00465	0.3679
	3	4.4608	4.4982	−0.000764	0.3170	4.5056	0.01083	0.9995
	5	5.2083	5.6758	−0.000563	0.6373	5.29	0.00749	0.9994

The pseudo-first-order kinetic model shows limited capability to describe the adsorption behavior of MB for BC produced at 400 °C, as illustrated in Figure 6a. The calculated adsorption capacities (q_e (cal.)) diverge significantly from experimental values (q_e (exp.)), particularly during 1 h, where q_e (cal.) (1.42 mg/g) is notably lower than q_e (exp.) (2.43 mg/g), as illustrated in Table 2. The R^2 values range from 0.32 to 0.64, indicating poor linearity and inadequate model fitting. The presence of negative and very small K_1 values further suggests that the adsorption process does not conform to first-order kinetics at this temperature. Consequently, the pseudo-first-order model is deemed unsuitable for accurately describing MB adsorption on biochar produced at 400 °C, supporting findings from previous studies [52]. In contrast, the pseudo-second-order model provides a significantly improved fit for the adsorption data at 400 °C. At 3 and 5 h, the calculated adsorption capacities (4.51 and 5.29 mg/g) closely match the experimental values (4.46 and 5.21 mg/g), as illustrated in Table 3. This close alignment is reinforced by very high R^2 approximately equal 0.999, as shown in Figure 6b. Although the fitting at 1 h is relatively poor ($R^2 = 0.37$), the excellent agreement at longer contact times indicates that the pseudo-second-order model accurately reflects the adsorption behavior as equilibrium is approached. This suggests that adsorption at this temperature is primarily influenced by surface-related interactions rather than first-order mass transfer, consistent with findings from earlier studies [53]. For BC produced at 500 °C, the pseudo-first-order model exhibits limited applicability, as shown in Figure 6c. While q_e (cal) values appear moderately close to q_e (exp) at 3 and 5 h, the overall fitting quality remains poor, as evidenced by low R^2 val-

ues (0.35–0.68). Negative K_1 values indicate non-physical adsorption kinetics, confirming that the pseudo-first-order model inadequately represents the adsorption mechanism at this temperature. Moreover, inconsistent fitting across different contact times further implies that adsorption does not follow first-order kinetics. The pseudo-second-order model demonstrates mixed fitting behavior at 500 °C, as shown in Figure 6d. At 3 h, an excellent agreement is observed between q_e (exp) (8.44 mg/g) and q_e (cal) (8.64 mg/g), with a very high R^2 value (0.999), indicating that the adsorption kinetics at this stage are well characterized by the Pseudo-second-order model. However, lower R^2 values at 1 and 5 h (0.29 and 0.85, respectively) suggest incomplete establishment of equilibrium conditions or the potential occurrence of competing adsorption–desorption processes. Despite this variability, the pseudo-second-order model performs better overall than the pseudo-first-order model, particularly near equilibrium.

For BC produced at 600 °C, the pseudo-first-order model continues to display poor performance at shorter contact times, with low R^2 values at 1 and 3 h (0.45 and 0.32), as shown in Figure 6e and Table 3. Although a higher R^2 value (0.88) is noted at 5 h, the presence of negative K_1 values and overall inconsistencies across time intervals indicate that this model does not reliably depict the kinetics of MB adsorption. The pseudo-second-order model provides an excellent description of MB adsorption kinetics for BC produced at 600 °C across all contact times. The calculated adsorption capacities closely align with experimental values, and R^2 values consistently exceed 0.99, as shown in Figure 6f. Positive K_2 values and strong linearity suggest rapid initial adsorption followed by a controlled approach to equilibrium. This fitting confirms that adsorption at elevated pyrolysis temperatures is dominated by surface-controlled processes with strong interactions between MB molecules and the BC surface, which aligns with findings from previous research [24,37].

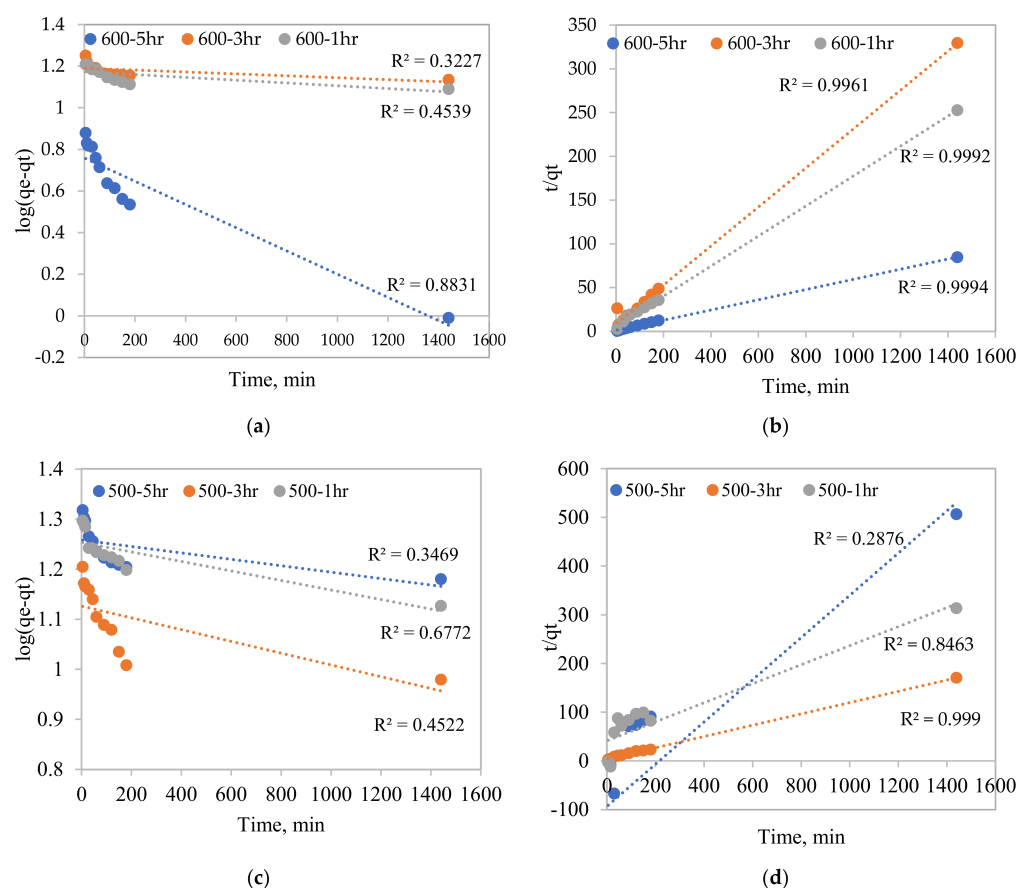


Figure 6. Cont.

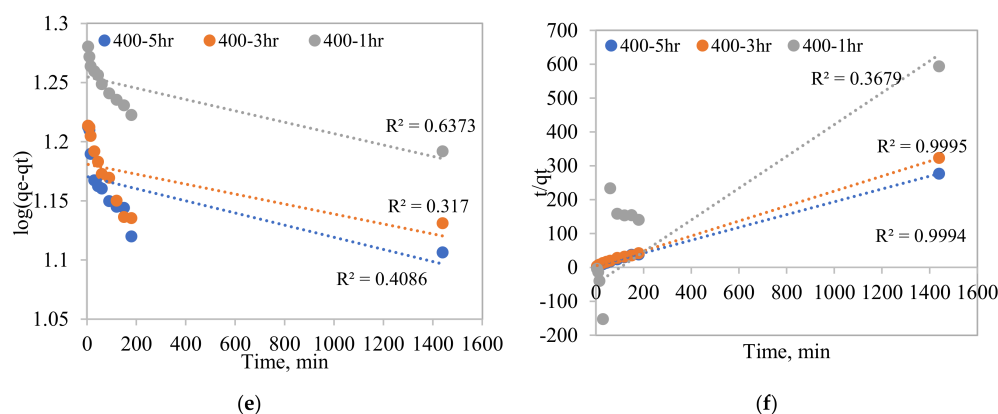


Figure 6. Kinetic Models of MB Adsorption on BC at 400, 500, and 600 °C: (a) Pseudo-1st-Order (400 °C), (b) Pseudo-2nd-Order (400 °C), (c) Pseudo-1st-Order (500 °C), (d) Pseudo-2nd-Order (500 °C), (e) Pseudo-1st-Order (600 °C), and (f) Pseudo-2nd-Order (600 °C).

The kinetic analyses indicate that the pseudo-first-order model is inadequate for describing the kinetics of MB adsorption across all investigated pyrolysis temperatures. In contrast, the pseudo-second-order model demonstrates superior fitting, particularly under equilibrium conditions and at higher pyrolysis temperatures. The most optimal kinetic performance is observed for BC produced at 600 °C, which suggests an enhanced adsorption efficiency due to improved surface characteristics.

3.4. Adsorption Thermodynamics

The thermodynamic parameters associated with the sorption of MB onto BC are essential for understanding the adsorption behavior and efficiency of the process. Table 4 presents the Gibbs free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) calculated at three different temperatures: 30 °C, 40 °C, and 50 °C.

Table 4. Thermodynamic parameters for MB sorption onto BCs at (30, 40, and 50 °C).

BC	ΔG° (30)	ΔG° (40)	ΔG° (50)	ΔH° (Kj/mol)	ΔS° (j/k.mol)	Conclusion
400 °C, 1 h	−38.609	−39.841	−42.407	18.737	188.478	spontaneous, endothermic, random
400 °C, 3 h	−37.390	−41.152	−42.380	38.665	252.188	spontaneous, endothermic, random
400 °C, 5 h	−44.663	−47.570	−40.756	−102.27	−185.023	spontaneous, exothermic, random
500 °C, 1 h	−39.706	−45.562	−46.310	61.248	335.646	spontaneous, endothermic, random
500 °C, 3 h	−46.008	−46.118	−46.830	−33.642	40.48	spontaneous, exothermic, random
500 °C, 5 h	−43.581	−44.590	−41.516	−9.540	107.581	spontaneous, exothermic, random
600 °C, 1 h	−40.726	−43.553	−43.653	4.107	149.293	spontaneous, endothermic, random
600 °C, 3 h	−43.700	−43.815	−45.735	−13.149	99.849	spontaneous, exothermic, random
600 °C, 5 h	−42.522	−42.972	−45.228	−1.803	133.389	spontaneous, exothermic, random

The ΔG° serves as an indicator of the spontaneity of the adsorption process. The negative ΔG° values observed across all BCs imply that MB sorption is a thermodynamically favorable process [54]. The values range from approximately −38.609 kJ/mol to −47.57 kJ/mol, suggesting that as temperature increases, the adsorption process becomes more spontaneous. This trend is consistent with findings in the literature that highlight the role of temperature in enhancing adsorption efficiency [42,55]. For instance, the BC produced at 400 °C for 5 h exhibits the most negative ΔG° value of −47.57 kJ/mol at 40 °C, indicating strong adsorption potential. In contrast, the higher ΔG° values for the BC produced at 400 °C for 1 h suggest less effective MB uptake, emphasizing the importance of pyrolysis duration for adsorption performance.

The ΔH° provides insight into the nature of the adsorption process. The varying ΔH° values indicate both endothermic and exothermic adsorption mechanisms. Most BCs dis-

play positive ΔH° values, suggesting that the adsorption process is primarily endothermic. For example, the BC produced at 600 °C for 1 h has a ΔH° of 4.107 kJ/mol, reinforcing the idea that higher temperatures facilitate enhanced adsorption [25]. Conversely, the BC produced at 600 °C for 5 h presents a ΔH° of −1.803 kJ/mol, indicating an exothermic process, which may suggest a transition from physical to chemical adsorption.

Entropy changes, represented by ΔS° values in Table 4, provide further understanding of the adsorption process. The positive ΔS° values across all samples indicate an increase in randomness at the adsorbent–adsorbate interface during the adsorption of MB. This reflects favorable conditions for sorption, as increased disorder often leads to improved adsorption efficiency [27]. For instance, the BC produced at 500 °C for 1 h exhibits a ΔS° value of 335.646 J/(K.mol), suggesting significant molecular rearrangements that enhance MB adsorption stability.

The thermodynamic analysis illustrates that MB sorption onto BCs is a spontaneous and thermodynamically favorable process. The combination of negative ΔG° , positive ΔH° , and positive ΔS° values collectively indicates that higher temperatures improve BC's adsorption capabilities. This aligns well with findings reported in the literature regarding various biochar sources [25,42]. The results presented in Table 4 emphasize the complex interplay between temperature, pyrolysis conditions, and thermodynamic parameters, suggesting that optimizing these factors can yield BCs with superior adsorptive properties for effective dye remediation applications.

3.5. Effect of Various Factors on Adsorption

3.5.1. Effect of Initial MB Dye Concentration

The impact of initial MB dye concentration on the adsorption performance of BC produced at varying pyrolysis temperatures (400, 500, and 600 °C) was systematically investigated through batch adsorption experiments. Additionally, the role of contact time (1 to 5 h) was evaluated across initial MB concentrations of 10, 50, and 100 mg/L. The results are illustrated in Figure 7, which highlights the interplay between initial dye concentration, contact time, and the temperature of BC production.

Figure 7a–c indicates that both the initial concentration of MB and the pyrolysis temperature significantly affect removal efficiency. At 400 °C (Figure 7a), the data reveals a remarkable removal efficiency of approximately 97.2% for the lowest concentration of 10 mg/L. However, as the initial concentration increases to 50 mg/L and 100 mg/L, the removal efficiency declines markedly, reaching only 68.4% and 18%, respectively, after 1 h of contact. This trend suggests that BC produced at lower pyrolysis temperatures has limited pore development, resulting in fewer active sites available for dye adsorption. The phenomenon of early surface saturation at higher dye concentrations restricts additional MB uptake. This observation aligns with findings by Abd El-Latif et al. [37], who described how increased initial dye concentrations intensify the driving force for mass transfer but concurrently diminish overall percentage removal due to adsorption effects [56–58].

In contrast, at 500 °C (Figure 7b), there is a noticeable enhancement in MB removal efficiency across all concentrations compared to the lower pyrolysis temperature. The removal efficiency reached about 97.52% for 10 mg/L, while it dropped to only 5.88% for 100 mg/L. This improved performance is likely attributed to the enhanced carbonization process and the more developed pore structure of the BC, which enables greater accessibility to internal adsorption sites. However, the reduction in percentage removal with increasing dye concentration indicates that saturation of adsorption sites remains a critical constraint at elevated concentrations, consistent with established adsorption equilibrium principles [57,58].

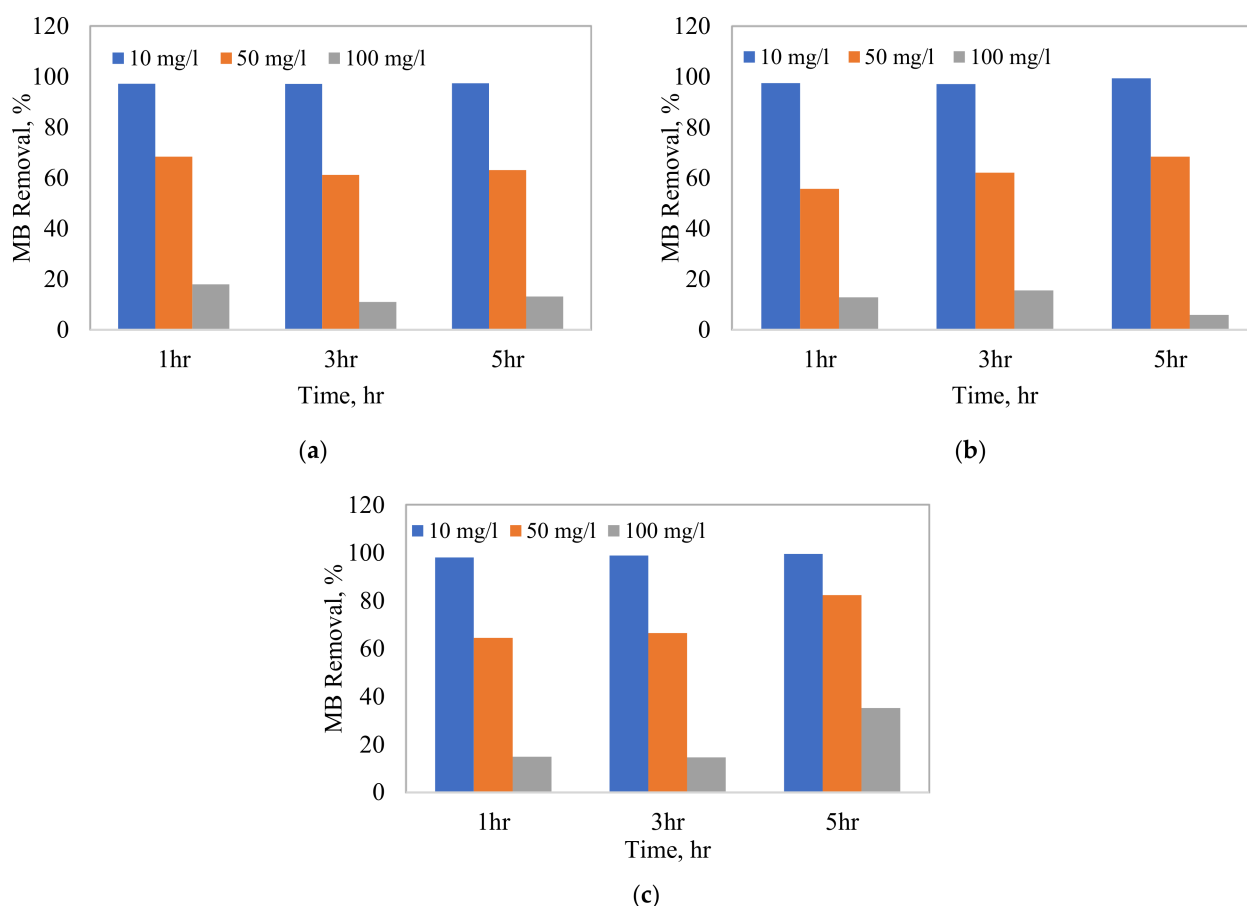


Figure 7. Effect of initial MB concentration (10–100 mg/L) on removal efficiency of BC produced at: (a) 400 °C, (b) 500 °C, and (c) 600 °C.

The biochar produced at 600 °C (Figure 7c) exhibited the highest MB removal efficiencies, particularly at 50 and 100 mg/L, with peak efficiencies observed at 99.57% for 10 mg/L. The superior adsorption performance at this temperature can be attributed to the significantly enhanced pore connectivity and surface area, which facilitate intraparticle diffusion processes and promote the adsorption of additional MB molecules even after the initial rapid surface uptake phase. Research has consistently noted similar enhancements in adsorption dynamics when utilizing BC subjected to higher thermal processing [25,59].

Across all pyrolysis temperatures studied, a common trend emerged: increasing the initial MB concentration led to decreased percentage removal, despite extended contact times. This trend can be accounted for by the saturation of available adsorption sites and increased competitive interactions among MB molecules as their concentration rises. Dawood et al. [23] emphasized that as initial dye concentrations rise, the accumulation of dye on the adsorbent's surface results in the saturation of active sites, which adversely affects removal efficiency. Similarly, Güleç et al. [24] noted that while higher solution concentrations resulted in lower percentage uptakes, they correspondingly increased overall adsorption capacity due to heightened mass transfer driving forces [25].

3.5.2. Adsorption Capacity and Effect of Contact Time

Despite the lower percentage uptakes observed for higher solution concentrations, Figure 8 illustrates that the overall absorption capacity significantly increases with higher concentrations of MB. This can be linked to the more pronounced mass transfer driving forces that facilitate effective adsorption mechanisms at elevated concentrations, as ref-

erenced in prior literature [24,25]. A consistent solution concentration of 50 mg/L was employed for all subsequent tests to maintain experimental uniformity.

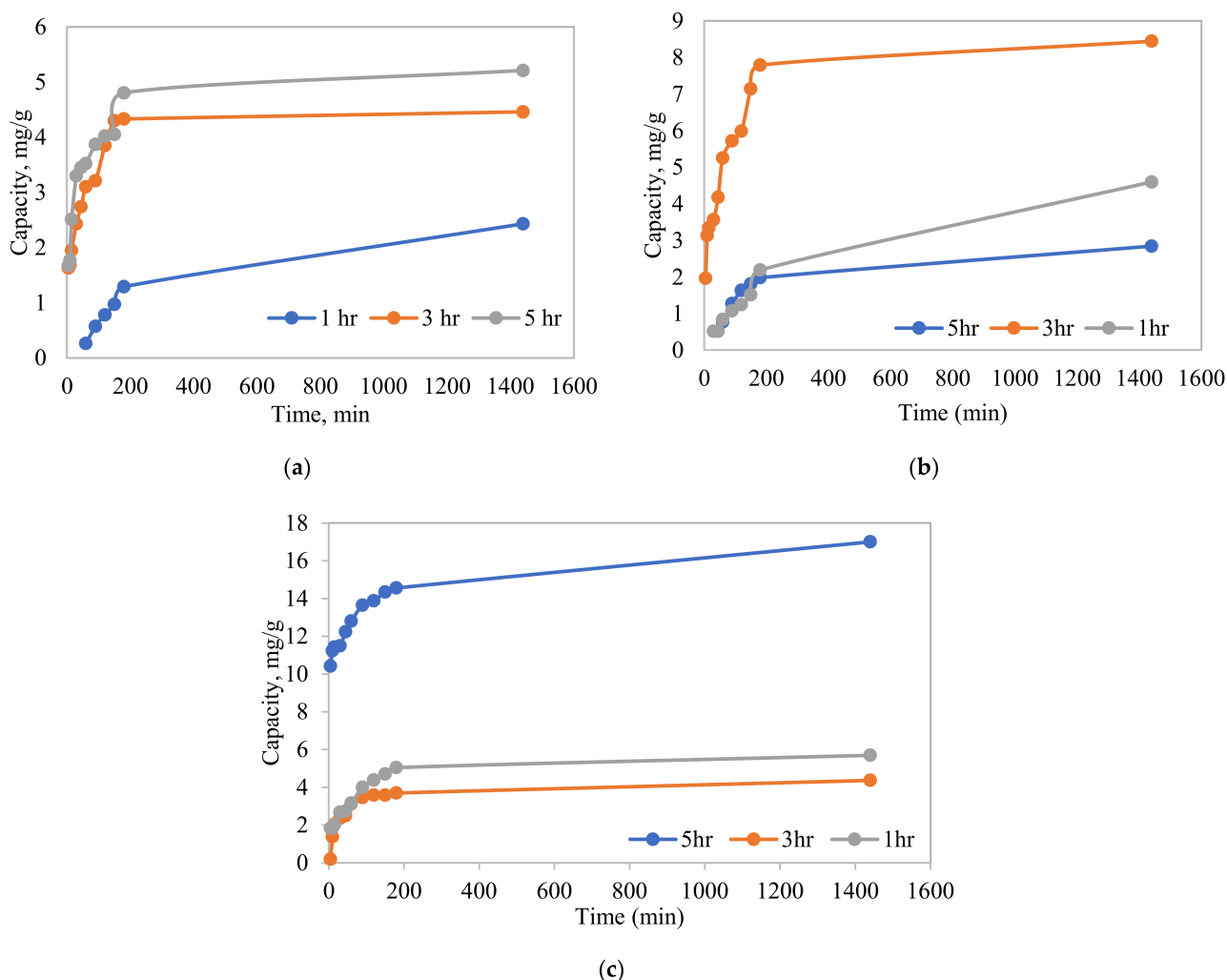


Figure 8. Adsorption capacity of BC at different pyrolysis temperatures (a) 400 °C, (b) 500 °C, and (c) 600 °C.

Figure 8a illustrates a progressive increase in adsorption capacity as contact time increases for all pyrolysis durations (1, 3, and 5 h) at 400 °C. At shorter pyrolysis times (1 h), the adsorption values are relatively low, ranging from 0.26 to 1.29 mg/g during the first 60 to 180 min. After 1440 min, the capacity gradually increases to 2.43 mg/g, indicating a slow adsorption kinetics and a limited number of available active sites. In contrast, extending the pyrolysis duration to 3 and 5 h results in noticeable improvements in adsorption capacity, reaching values of 4.46 mg/g and 5.21 mg/g, respectively, after 24 h. This behavior can be attributed to the gradual development of the porous structure and the active surface area of the biochar at 400 °C. However, achieving equilibrium requires a relatively long contact time, exemplifying that this temperature may be insufficient for generating high-activity adsorption sites in the early stages, as suggested by prior research.

At a pyrolysis temperature of 500 °C, Figure 8b demonstrates a marked improvement in adsorption capacity compared to 400 °C, particularly at the 3-h pyrolysis duration. The adsorption capacity steadily increases over time, peaking at 8.44 mg/g after 1440 min, which is significantly higher than the 4.46 mg/g recorded at 400 °C for the same duration. However, at the 1-h pyrolysis duration, the capacities remain relatively low, registering at 4.6 mg/g after 24 h. This observation indicates that merely raising the temperature is

insufficient without an appropriately extended pyrolysis duration. The values observed at the 5-h pyrolysis duration exhibit irregular behavior in the early stages, stabilizing below those at the 3-h pyrolysis duration. This irregular behavior may suggest the formation of blocked pores or partial structural agglomeration due to excessive pyrolysis time at this temperature.

Figure 8c displays the highest adsorption capacities among all samples, particularly at the 5-h pyrolysis duration. Initial capacities are already elevated from the early stages, exceeding 10 mg/g within the first 5 to 15 min, and further rising to 17 mg/g after 1440 min. This behavior demonstrates rapid adsorption kinetics with high-energy active sites. In contrast, the samples subjected to 1 and 3 h of pyrolysis display more gradual increases, reaching 5.7 mg/g and 4.37 mg/g after 24 h, respectively. This finding reinforces that maximizing the benefits of higher temperatures requires sufficient pyrolysis time to develop an open and stable porous network. The results suggest that adsorption at 600 °C initially undergoes a rapid surface mass transport mechanism, followed by pore filling as contact time extends, aligning with the structural dynamics discussed in existing literature [24,25].

Figure 9 illustrates the relationship between BET surface area and the maximum adsorption capacity of the prepared biochars under different pyrolysis conditions. A positive correlation can be observed, where an increase in surface area is accompanied by an enhancement in adsorption performance. This trend confirms that pore development plays a key role in governing the adsorption behavior of the prepared materials.

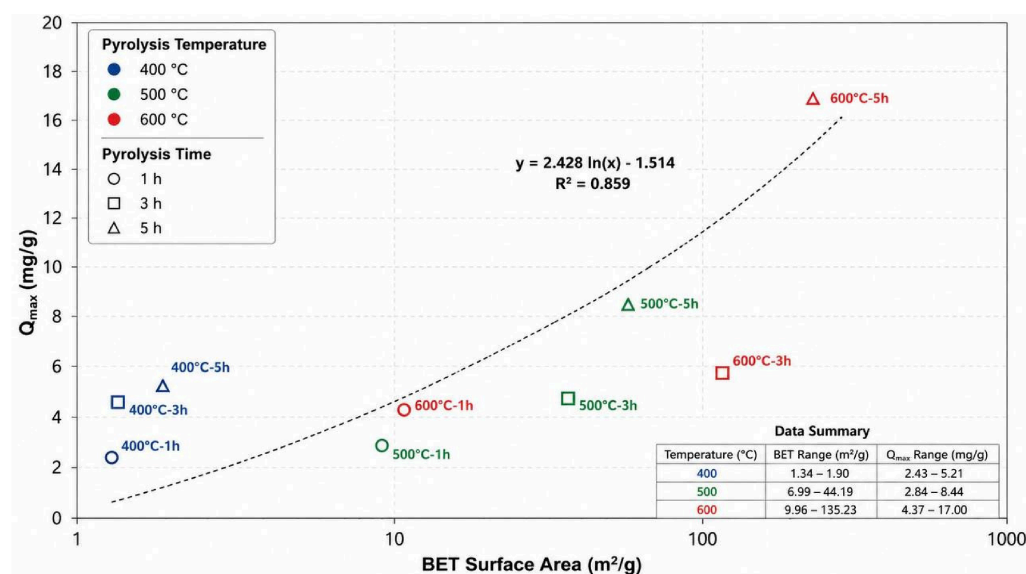


Figure 9. Correlation between BET surface area and maximum adsorption capacity of BC samples.

As shown in Figure 9, at a lower pyrolysis temperature (400 °C), the BCs exhibit very limited surface development (1.34–1.90 m²/g), which corresponds to low adsorption capacities (2.43–5.21 mg/g), indicating insufficient pore formation. With increasing temperature to 500 °C, a significant improvement in BET surface area is observed (up to ~44 m²/g), leading to a moderate increase in adsorption capacity.

The most pronounced improvement is obtained at 600 °C, where extensive pore development is achieved, reaching a maximum BET surface area of 135.23 m²/g. This enhancement results in a substantial increase in adsorption capacity, reaching 17 mg/g. This confirms that higher pyrolysis temperature and longer residence time promote the formation of a more porous structure, which directly enhances adsorption performance. However, minor deviations from perfect linearity suggest that adsorption is not solely

controlled by surface area, but may also be influenced by surface chemistry and functional groups present on the BC surface.

3.5.3. pH Effect

The adsorption behavior of MB on BC is influenced by solution pH, which governs the surface charge of the adsorbent and the ionization state of functional groups. Since MB is a cationic dye, its adsorption is primarily controlled by electrostatic interactions with negatively charged sites on the BC surface, as well as additional mechanisms such as π - π interactions and hydrogen bonding.

As illustrated in Figure 10, the adsorption capacity of BC produced at 600 °C (3 and 5 h) varied significantly with initial pH, confirming the strong dependence of MB uptake on surface chemistry. For BC produced for 3 h, the adsorption capacity decreased from 0.5117 mg/g at pH 2 to 0.0718 mg/g at pH 12, while the removal efficiency increased up to 86.7%. A similar trend was observed for BC produced for 5 h, where adsorption capacity decreased from 0.2591 mg/g to 0.0435 mg/g, while removal efficiency reached 91.94% at pH 12.

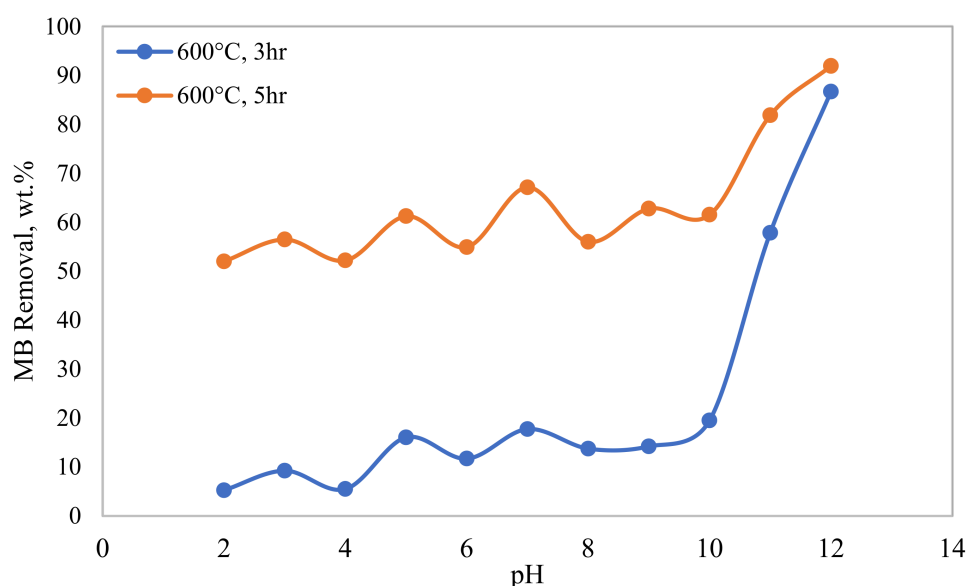


Figure 10. Effect of initial pH on the adsorption of MB dye onto BC produced at 600 °C for both 3 and 5 h.

Under acidic conditions, the surface functional groups of BCs become protonated, resulting in a positively charged surface. Since MB is a cationic dye, electrostatic repulsion may occur between the adsorbent surface and dye molecules, limiting adsorption. As the pH increases, surface functional groups gradually deprotonate, generating a more negatively charged surface that enhances the electrostatic attraction of positively charged MB molecules and promotes adsorption.

The observed behavior in Figure 10 can be interpreted in terms of the point of zero charge (pHpzc) of BC. At pH values below pHpzc, the BC surface becomes protonated, leading to a positively charged surface that electrostatically repels the cationic MB molecules, thereby reducing adsorption capacity. At higher pH values, deprotonation of oxygen-containing functional groups (such as $-\text{OH}$ and $-\text{COOH}$) occurs, generating negatively charged surface sites that strongly attract MB molecules, enhancing adsorption via electrostatic attraction [23,25,37].

In addition, under alkaline conditions, the abundance of OH^- reduces competition with dye molecules for active adsorption sites and promotes greater accessibility of MB to

porous structures. Similar pH-dependent behavior has been widely reported for MB adsorption onto lignocellulosic and biochar-based adsorbents, where increased pH enhances dye uptake due to stronger electrostatic attraction and reduced proton competition [37,60].

The slight difference in adsorption capacity between BCs produced for 3 h and 5 h can be attributed to differences in surface development and functional group density, which affect the density of active adsorption sites available for MB interaction. Overall, the results confirm that alkaline conditions favor MB adsorption onto BC due to enhanced negative surface charge development and stronger electrostatic attraction between BC and MB molecules.

3.5.4. Effect of Adsorption Temperature

Three different temperatures (30, 40 and 50 °C) were investigated using an MB dye solution to assess the effect of temperature on the MB uptake of the BC produced at three different pyrolysis temperatures. Figure 11 presents the MB removal efficiency (%) for BC produced at different pyrolysis temperatures. Specifically, Figure 11a depicts the BC produced at 400 °C, Figure 11b presents the BC produced at 500 °C, and Figure 11c illustrates the BC produced at 600 °C.

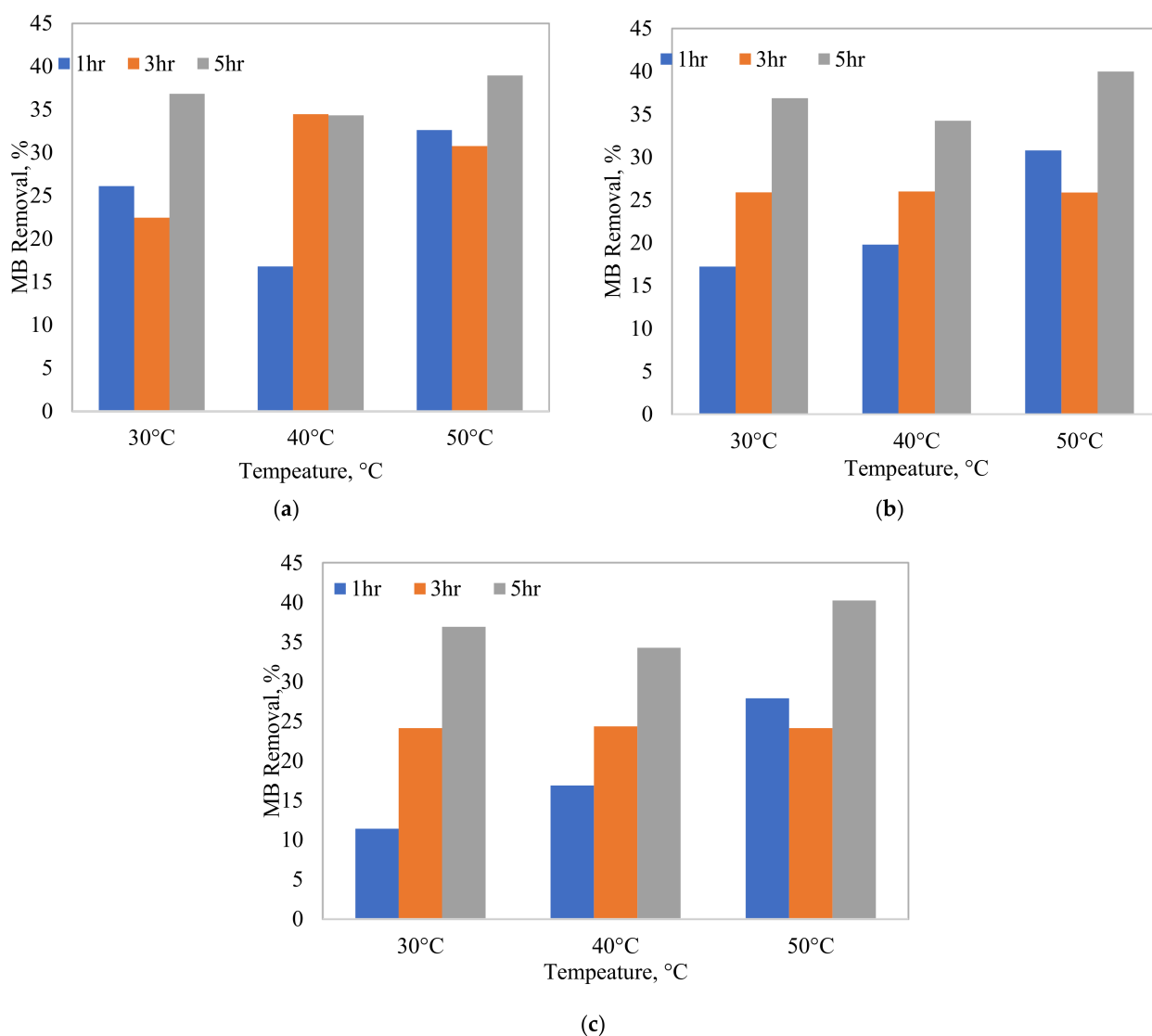


Figure 11. Effects of temperature on MB removal efficiencies of BC produced (a) 400 °C, (b) 500 °C, and (c) 600 °C.

The results indicate a clear temperature dependence in the removal efficiencies across all BC samples, with higher pyrolysis temperatures leading to more effective MB adsorption. For BC produced at 600 °C, the removal rates show a marked increase with rising temperature. After 1 h of treatment, the removal efficiency escalates from 11.41% at 30 °C to 27.87% at 50 °C. Over time, the adsorption efficiency stabilizes around 24% after 3 h but demonstrates a significant improvement at the 5-h mark, where removal increases to 40.24% at 50 °C. This pattern confirms the enhancement of adsorption capabilities at higher temperatures, aligning with findings in previous studies that emphasized the positive correlation between temperature and adsorption capacity [26,27].

In the case of BC produced at 500 °C, the results are somewhat lower but still exhibit a similar trend. The initial 1-h treatment shows an increase from 17.23% at 30 °C to 30.78% at 50 °C. Although the removal efficiencies stabilize around 25% after 3 h, the 5-h treatment indicates a notable improvement, peaking at 40% at the highest temperature. This suggests that while lower than the effectiveness observed at 600 °C, the BC from 500 °C still benefits from elevated temperatures.

Conversely, the BC produced at 400 °C shows a more varied performance. After 1 h, the removal efficiency ranges from 26.1% at 30 °C to 32.6% at 50 °C. Notably, the 3-h results peak at 34.5% at 40 °C, demonstrating that even at lower pyrolysis temperatures, effective removal is achievable. The 5-h results show that removal efficiency varies, highlighting the less predictable nature of lower-temperature BC in terms of MB adsorption. The analysis of thermodynamic parameters further supports these findings.

3.5.5. Effect of Adsorbent Dose

The impact of various BC dosages on the removal efficiency of MB dye was investigated to ascertain the most effective and economical dosage for optimal dye adsorption, as shown in Figure 12. As illustrated in Figure 12a, increasing the BC dosage from 0.01 g to 0.1 g led to a significant rise in dye removal percentage from 11.57% to 56.13% at a contact time of 1 h. This upward trend continued with the 3 h data shown in Figure 12b, where the removal increased from 11.89% to 54.57%, and peaked at 95.43% at the same dosage after 5 h, as presented in Figure 12c. Notably, a downward trend was observed in the amount of dye adsorbed per unit mass of biochar (q_e). For instance, after 1 h, the q_e dropped from 14.47 mg/g at 0.01 g to 7.02 mg/g at 0.1 g. This decline suggests that although a higher dosage of BC results in greater overall dye removal, the efficiency of adsorption per unit weight of BC decreases as the dosage increases. This behavior aligns with established adsorption principles, indicating that increased adsorbent concentrations can lead to saturation of available active sites on the adsorbent surface, thus limiting the effective adsorption [24].

In the extended contact times reflected in Figure 12b,c, the percentage of dye removal continued to increase significantly. This indicates that longer contact durations facilitate more efficient interactions between the dye and the adsorbent. However, the trend of reduced q_e values persisted, underscoring that while total dye removal may be high, the specific adsorption capacity per unit mass declines. This phenomenon can be explained by the diminishing concentration gradient between the dye in solution and on the bc surface as the adsorbent dose increases, impacting the rate of diffusion and subsequently the amount of dye that can be adsorbed per gram of BC [61,62].

The optimal dosage of 0.04 g for the removal of MB dye demonstrates a significant balance between effective dye removal and adsorption efficiency per unit mass of BC. These findings underscore the importance of optimizing BC dosage to balance effective dye removal with economic viability in various adsorption applications. A similar behavior has been documented in other studies focusing on dye adsorption [23,63].

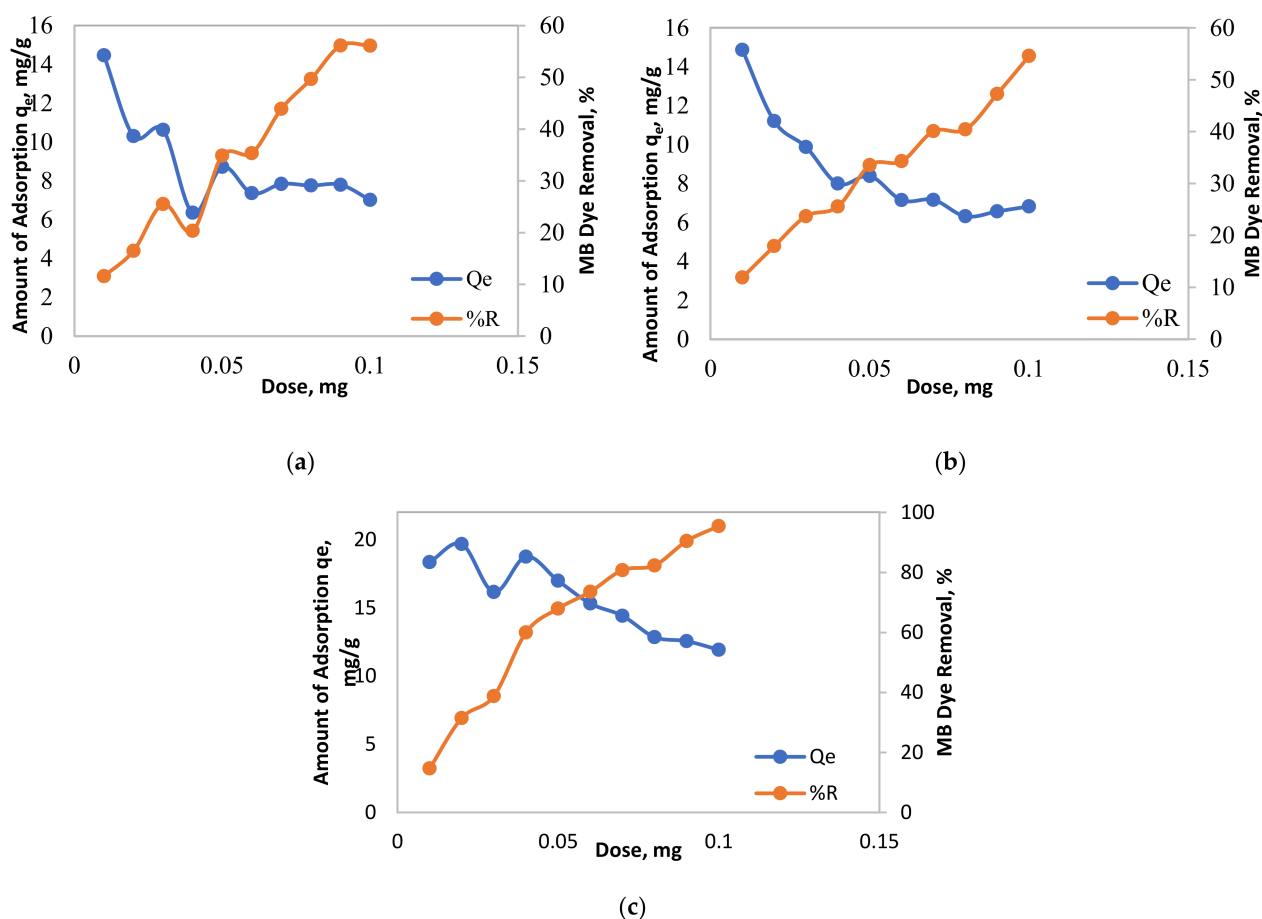


Figure 12. Effect of adsorbent dosage of MB dye onto BC produced at 600 °C: volume of MB solution = 50 mL, and initial dye concentration = 50 ppm. (a) 600 °C at 1 h. (b) 600 °C at 3 h. (c) 600 °C at 5 h.

The results presented in Figure 12a–c highlight the dual relationship between BC dosage and its efficacy in dye removal. Higher dosages effectively improve the total MB removal percentage but also lead to decreased efficiency in terms of dye adsorbed per unit mass of adsorbent.

4. Comparison of Biochars for Methylene Blue Removal

Biochar adsorption performance toward MB varies widely depending on the precursor type, activation strategy, and pyrolysis conditions. Agricultural residues, lignocellulosic wastes, and algal biomass have been extensively investigated as sustainable feedstocks for producing low-cost adsorbents. Table 5 summarizes representative materials derived from different biomass precursors, highlighting their preparation conditions and maximum adsorption capacities toward MB.

As shown in Table 5, the results show that maximum adsorption capacities reported for chemically modified BCs, such as Fe₃O₄-composite banana peel BC (862 mg/g) and luffa vine activated BC (559.11 mg/g), are significantly higher than those obtained in the present work. This enhancement is mainly attributed to intensive surface modification, increased surface area, and the introduction of highly active functional groups that promote strong electrostatic and electron-transfer interactions with MB molecules [64,65].

Similarly, chemically activated rice husk BC exhibited a relatively high adsorption capacity (359.7 mg/g), confirming the strong influence of alkali activation and micropore development on MB uptake [66]. Other modified agricultural BCs, such as N-doped or acid-activated rice straw BCs, showed moderate adsorption capacities in the range

of 136–145 mg/g, further supporting the role of surface functionalization in enhancing adsorption performance [67].

In contrast, unmodified or thermally produced BCs, such as bamboo BC (86.6 mg/g) and neem shell BC (134.13 mg/g), exhibit adsorption capacities closer to or slightly higher than those observed in this study [68,69]. This indicates that the OMSW-derived BC produced here falls within the expected performance range for non-chemically activated biochars, despite the absence of additional modification steps.

Table 5. Comparison of OMSW-derived BC with previously reported adsorbents for MB removal.

Biomass Precursor	Modification/Activation	Key Conditions	Adsorption Capacity, mg/g	Ref.
Banana peel	FO ₃ O ₄ green modification	pH 6.1, 313 K	862	[64]
Luffa vine waste	One-step carbonization	700 °C	559.11	[65]
Neem shell	Pyrolysis	pH 8.5–9	134.13	[68]
Rice husk	KOH/KOH-NaOH Activation (Chemical activation + pyrolysis)	High-temperature activation	359.7	[66]
Lantana camara	Pyrolysis BC	600 °C, pH 3–12	-	[70]
Rice straw	Modified N-doped/H ₃ PO ₄	Variable pH	136.9–145.8	[67]
Bamboo	Pyrolysis BC	500 °C	86.6	[69]
Bamboo	KOH Activation	900 °C, pH 11	67.46	[66]
Agricultural BC (date pits, jute sticks, etc.)	Natural/ZnCl ₂ Activation	-	Variable	[71]
OMSW	Pyrolysis (No Chemical Activation)	400–600 °C, 1–5 h	Up to 17	Current Study

The OMSW-derived BC in this study showed adsorption capacities (up to 17 mg/g) in a comparatively lower range than highly modified adsorbents; however, this material was produced without any chemical activation or post-treatment. Despite this, it still demonstrated effective MB adsorption performance, confirming that pyrolysis conditions alone (temperature and residence time) can significantly influence adsorption behavior.

Importantly, although chemically modified adsorbents exhibit higher capacities, they often involve additional cost, multi-step synthesis, and environmental concerns due to chemical usage. Therefore, the OMSW BC offers a sustainable and low-cost alternative, making it suitable for large-scale wastewater treatment applications.

5. Conclusions

This study demonstrated that pyrolysis temperature and residence time significantly control the physicochemical properties and adsorption performance of BC derived from OMSW for MB removal. The results showed that increasing pyrolysis temperature enhanced BC properties. At 400 °C, the BC exhibited very low surface area (1.34–1.9 m²/g) and limited porosity, resulting in moderate MB removal performance. Increasing the temperature to 500 °C improved the surface area to 6.99–44.19 m²/g with a corresponding increase in pore volume (up to 0.026 cm³/g), leading to higher adsorption efficiency. The most effective performance was obtained at 600 °C, where the BC achieved a maximum surface area of 135.23 m²/g and pore volume of 0.075 cm³/g, which resulted in the highest MB removal efficiency of 99.57% under optimal conditions (5 h contact time, 50 mg/L MB).

Adsorption performance was strongly dependent on operational parameters. Increasing initial MB concentration from 10 to 100 mg/L led to a reduction in removal efficiency due to saturation of active sites, while increasing BC dosage from 0.01 to 0.1 g improved dye removal up to 95.43% after 5 h. pH also has a significant impact, as the adsorption behavior

was influenced by surface charge interactions between BC and MB molecules. Kinetic analysis revealed that MB adsorption onto all BC samples followed the pseudo-second-order model ($R^2 \approx 0.99$), confirming that the process is mainly controlled by chemisorption and surface interactions.

Equilibrium studies were best fitted by the Langmuir isotherm under most conditions (R^2 up to 0.9963), indicating monolayer adsorption on relatively homogeneous active sites. However, at higher temperature and extended contact times, the Freundlich isotherm also became relevant, suggesting increased surface heterogeneity and multilayer adsorption behavior. Thermodynamic results confirmed that MB adsorption was spontaneous, with negative ΔG° (−38.6 to −47.6 kJ/mol), and generally endothermic in nature, indicating that higher temperatures enhance adsorption efficiency. An increase in entropy ($\Delta S^\circ > 0$) suggests greater disorder at the BC–MB interface, further enhancing adsorption efficacy.

Overall, the findings confirm that OMSW can be effectively converted into high-performance BC for dye removal applications, with optimal performance achieved at 600 °C due to its superior surface area, porosity, and adsorption capacity. However, the study was conducted using synthetic MB solutions; therefore, future work should investigate real wastewater systems containing salts and another pollutant, which may affect adsorption behavior. Additionally, regeneration studies and multi-cycle reuse experiments are necessary to evaluate the long-term applicability and economic feasibility of the BC.

Future research should focus on assessing the economic feasibility of BC production under different pyrolysis conditions. Integrating BC production with bioenergy or bio-fuel recovery systems could enhance process sustainability, reduce net energy costs, and improve the overall viability of large-scale applications.

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