

Retraction

Retracted: Removal of Methylene Blue from Aqueous Solution Using Black Tea Wastes: Used as Efficient Adsorbent

Adsorption Science and Technology

Received 28 November 2023; Accepted 28 November 2023; Published 29 November 2023

Copyright © 2023 Adsorption Science and Technology. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

This article has been retracted by Hindawi, as publisher, following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of systematic manipulation of the publication and peer-review process. We cannot, therefore, vouch for the reliability or integrity of this article.

Please note that this notice is intended solely to alert readers that the peer-review process of this article has been compromised.

Wiley and Hindawi regret that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation.

The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

References

- [1] A. Ullah, M. Zahoor, W. U. Din et al., "Removal of Methylene Blue from Aqueous Solution Using Black Tea Wastes: Used as Efficient Adsorbent," *Adsorption Science & Technology*, vol. 2022, Article ID 5713077, 9 pages, 2022.

Research Article

Removal of Methylene Blue from Aqueous Solution Using Black Tea Wastes: Used as Efficient Adsorbent

Azmat Ullah,¹ Muhammad Zahoor ,² Waqar Ud Din,³ Mian Muhammad,¹ Farhat Ali Khan,⁴ Aamir Sohail,⁵ Riaz Ullah ,⁶ Essam A. Ali,⁷ and H. C. Ananda Murthy ⁸

¹Department of Biochemistry, University of Malakand, Chakdara, Dir Lower, Malakand, Pakistan

²Department of Chemistry, University of Malakand, Chakdara, Dir Lower, Malakand, Pakistan

³Department of Chemistry, Government College Mingora Swat, Malakand, Pakistan

⁴Department of pharmacy, Shaheed Benazir Bhutto University, Sheringal, Dir Upper, Bhutto, Pakistan

⁵MSC Construction Project management University of Bolton UK, UK

⁶Department of Pharmacognosy, College of Pharmacy King Saud University Riyadh, Riyadh, Saudi Arabia

⁷Department of Pharmaceutical Chemistry, College of Pharmacy King Saud University Riyadh, Riyadh, Saudi Arabia

⁸Department of Applied Chemistry, School of Applied Natural Science, Adama Science and Technology University, P O Box, 1888 Adama, Ethiopia

Correspondence should be addressed to H. C. Ananda Murthy; anandkps350@gmail.com

Received 20 February 2022; Accepted 28 March 2022; Published 12 April 2022

Academic Editor: Rabia Rehman

Copyright © 2022 Azmat Ullah et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

The biosorbent black tea wastes (BTW) after preliminary treatments was used in this study for the removal of methylene blue (M. B) from aqueous solution. The removal of M.B from aqueous solution was studied as a function of time, initial concentration of M. B temperature, pH, and BTW dosage. The optimum time for equilibration was achieved in 3 min. The optimum dosage of adsorbent was found to be 0.4 g. Various kinetic models were applied to the sorption kinetic data in which the obtained data was best explained by the pseudo-second-order model ($R^2 = 0.99$) with a rate constant K_2 of $0.0714\text{--}0.0763 \text{ g.mg}^{-1} \text{ min}^{-1}$. Additionally, the calculated amount of adsorption was approximately equal to the experimentally determined value. The isotherm data was best fitted to the Langmuir model rather than the Freundlich model. The intraparticle diffusion model exhibited the process to be diffusion dependent. The various organic functional groups on the surface of BTW played a significant role in the sorption of the selected dye. Consequently, BTW has the prospective to act as a potential sorbent for the removal of other contaminants from aquatic media as well.

1. Introduction

Dyes are organic compounds that are widely utilized in the textile, paper, food, printing, plastic, drinks, leather, and pharmacology industries [1–3]. In general, dyes can be categorized as anionic (acidic dyes) and cationic (basic dyes). Among them, azo dyes (anionic) are that class of dye which contain nitrogen-nitrogen double bonds and are considered to be the largest group of organic dyes but at the same time are quite toxic. Some of them have been reported to have carcinogenic potentials, whereas same effect has been observed for the metabolites formed from them after degra-

ation [4]. Thus, there is extensive need to reclaim them from industrial effluents before entering into water bodies. Despite of the harmful effects, dyes are still significant and important group of chemicals used for coloring material objects. There are over 1×10^6 commercially available dyes, with an annual output of about 7×10^5 metric tons, according to the reports [5]. As mentioned before, because of their complex aromatic structure and nonbiodegradable nature, dyes in waste water pose a significant threat to living organisms, making their detection and removal a difficult job [6].

Dyes are excessively entered into water reservoirs a number of industrial sources. These sources include textile,

paper, beauty products, food, and polymers. The continuous entry of dyes into aquatic media poses severe health problems in human beings and also destroys the ecosystem. Most of these dyes are extremely poisonous, mutagenic, and carcinogenic in nature [7–10].

Methylene blue (M.B) is an aromatic and widely used synthetic cationic dye with a high adsorption capacity that is commonly used in the dyeing of silk, wool, and cotton in the industrial process. M.B is also used to treat diseases such as duck hepatitis B, psoriasis, and West Nile virus. Despite of medicinal application, some health complication is associated with its use, for example, permanent eye damage, vomiting, gastritis, breathing difficulties, nausea, mental confusion, painful micturition, tissue necrosis, cyanosis, and methemoglobinemia-like syndromes. Methylene blue is also highly carcinogenic in living organisms. Methylene blue is not regarded as extremely toxic but its ingestion through the mouth produces a burning impression [11]. Therefore, it is exceptionally important to remove it from industrial waste water before discharging it into the environment. Different physiochemical methods have been used for the removal of various contaminants, namely, solvent extraction, membrane filtration [12–15], coagulation, and chemical oxidation [16] from wastewaters. Unfortunately, these techniques are not cost-effective and also have a number of disadvantages, including a high reagent and energy consumption, as well as the production of poisonous clay or other byproducts that must be disposed of after completion of the processes. Furthermore, certain colors used in textile industry are extremely difficult to eliminate using traditional waste disposal methods since they are light and oxidizing agent resistant and unchanged by aerobic digestion [17]. As a result, new procedures for removing dyes are being extensively employed in the industries. Although activated carbon is the most effective adsorbent but due to high cost of preparation, its use becomes impractical in most parts of the world. Therefore, scientists these days are trying to use waste biomass as greener adsorbent.

A lot of studies have been reported on various low-cost adsorbent for dye removal from water [8–19]. Out of which waste biomasses and commercial byproducts have been intensively investigated for their potential to eliminate dye from aqueous media, as they are readily available and cheap green sources, e.g., corncob, modified leaves of plants, and rice husk [18].

Due to the massive consumption of tea around the world, numerous researchers have employed tea waste as an excellent adsorbent. After water, tea is the world's second most consumed nonalcoholic liquid. In 2013, global tea output was 5.1 million tons, with China accounting for 36% of total production, followed by India (21.2%), Kenya (7.8%), Sri Lanka (7.0%), Turkey (4.8%), Vietnam (4.6%), and Iran (4.6%). The remaining 15.3% comes from other countries. Turkey is the world's fifth largest tea grower, with an annual output of 212,400 tons and the highest per capita tea consumption. Turkey produces around 150,000 tons of tea annually (dry basis) and 30,000 tons of tea waste per year [20]. The tea wastes are discarded as such which finally finds its way to river and canals.

The aim of present study was to use the tea wastes in a useful way as a biosorbent for methylene blue. Different isothermal and kinetic models were employed to estimate the adsorption parameters of selected dye on tea waste-based biosorbent.

2. Materials and Methods

2.1. Materials. Black tea wastes (BTW) was collected from local market. The BTW sample was treated with hot double distilled water to remove dust and other materials. The treatment was applied to remove certain pigments remained in BTW. After treatment, the BTW was dried in an electric oven at 75–80°C for 12–14 h. The dried BTW sample was then grinded into a fine powder form using a grinder. The dried and grinded BTW was then stored in a glass reagent bottle till further studies. Methylene blue (M.B) was provided by the Institute of Chemical Sciences University of Malakand. The chemicals NaOH and HCl were purchased from Merck (Germany). The schematic diagram of the whole process is shown in Figure 1.

2.2. Batch Adsorption Experiments. In batch sorption experiments, 30 mL of M.B solution in a 100 mL glass reagent bottle were contacted with specified amount of adsorbent. Different concentration solutions of M.B were obtained from 800 mg.L⁻¹ stock solution using dilution method. For adsorption kinetics about 0.40 g of BTW was added to 30 mL M.B solution during kinetic study and stirred for different interval of time. During isotherm studies, different initial concentrations of M.B, i.e., from 100 mg.L⁻¹ to 400 mg.L⁻¹ in 30 mL volume were contacted with 0.4 g of BTW powder. The adsorption isotherm data was then interpreted for different isotherm models. For optimum dose of BTW, different masses of BTW were added in the range of 0.10–0.50 g to a series of flasks containing 30 mL solution each. Out of the tested doses, 0.40 g amount was selected as optimum dose of BTW for the subsequent experiments. The pH on M.B adsorption was studied in range 1–12 where pH of solutions were adjusted through solution of NaOH and HCl. The M.B concentration after adsorption in each experiment was determined at 665 nm using a double beam UV/visible spectrophotometer (Shimadzu Japan).

2.3. Calibration Curve of M.B. As mentioned above, the concentrations of the remaining dye in solution were determined at 665 nm. To avoid tedious calculations, a calibration curve as shown in Figure 2 was constructed.

2.4. Data Analysis. The experimental data was fed (equations (1) and (2)) to calculate different adsorption parameters:

$$Q = \frac{(X_o - X_t)}{m} \times V, \quad (1)$$

$$\%R = \frac{(X_o - X_t)}{X_o} \times 100. \quad (2)$$

In equation (1), Q is the amount of M.B adsorbed at different time interval, X_o and X_t are the initial concentration

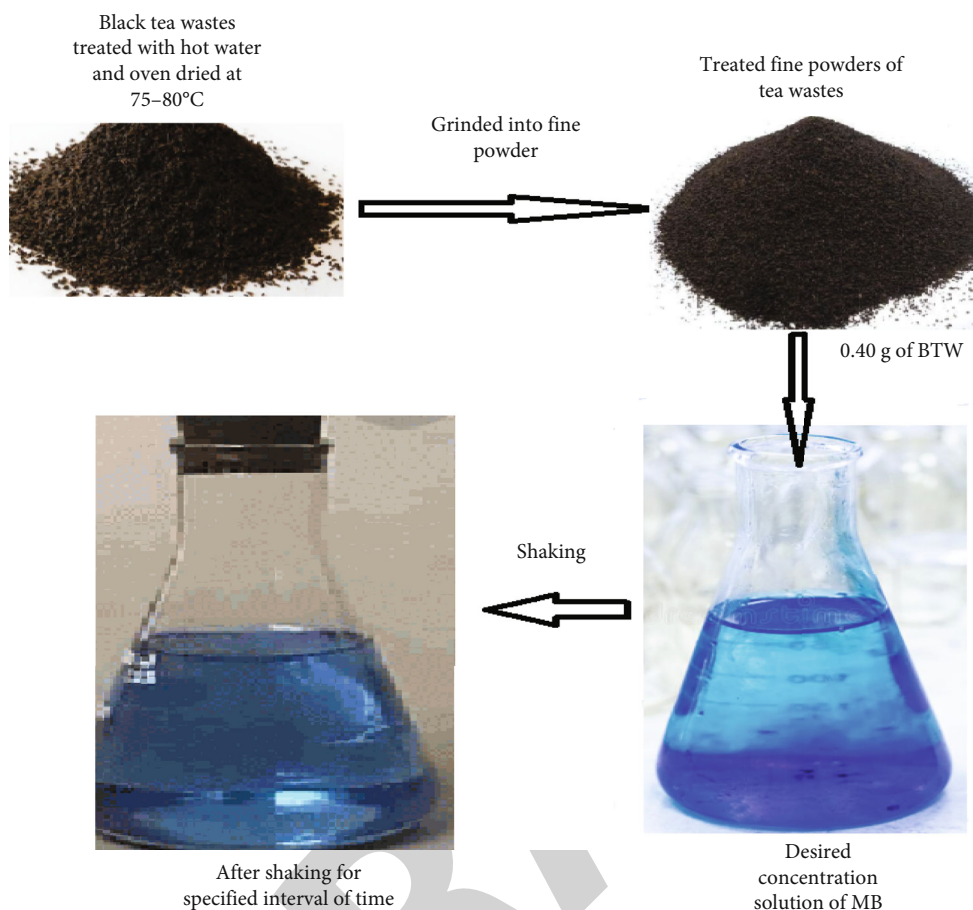


FIGURE 1: Schematic representation of decolorization of M.B by BTW.

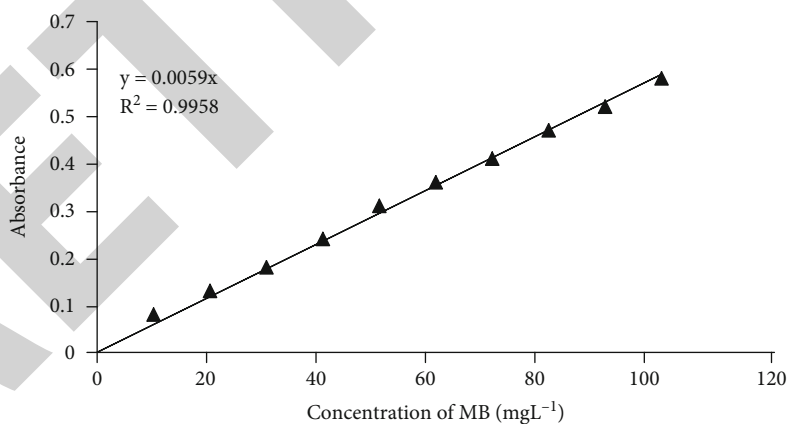


FIGURE 2: Calibration curve for methylene blue.

and concentration at time t of M.B dye, respectively, whereas m is the mass of BTW in grams and V is the volume in liter (L) of M.B dye solution. %R represents the percent removal of M.B dye.

3. Results and Discussion

3.1. *Effect of pH on Percent % Removal of M.B.* The effect of pH on the adsorption of M.B on BTW is graphically shown in Figure 3 as studied in the pH range of 1-12.

The maximum adsorption occurs at the pH 8 and then the adsorption rate decreased. The high degree of adsorption at pH 8 may be due some positive interaction of certain functional groups such as amino, hydroxyl, and ethereal present on the surface of BTW with that of the M.B [21]. These oppositely charged ions attract each other strongly at pH 8.

3.2. *Effect of BTW Dosage.* The effect of BTW on the adsorption of M.B is shown in Figure 4. With the increase in BTW

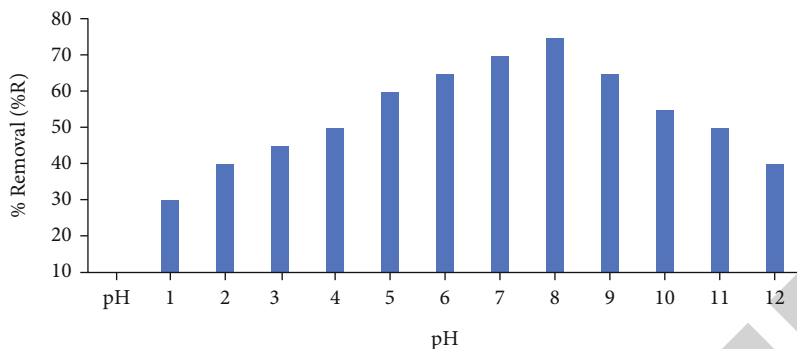


FIGURE 3: Effect of pH on methylene blue removal.

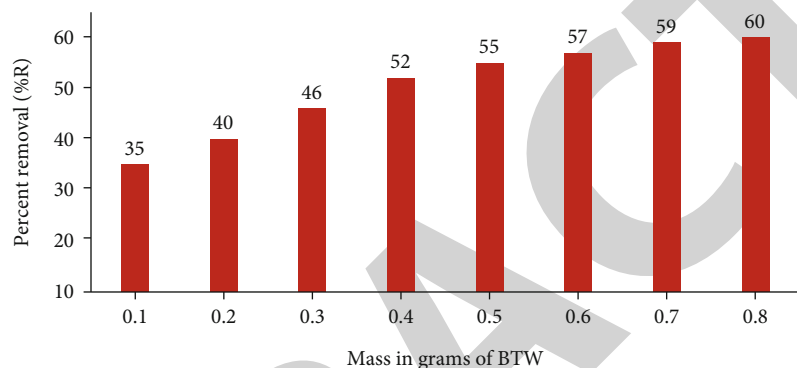


FIGURE 4: Effect of dosage on M.B removal.

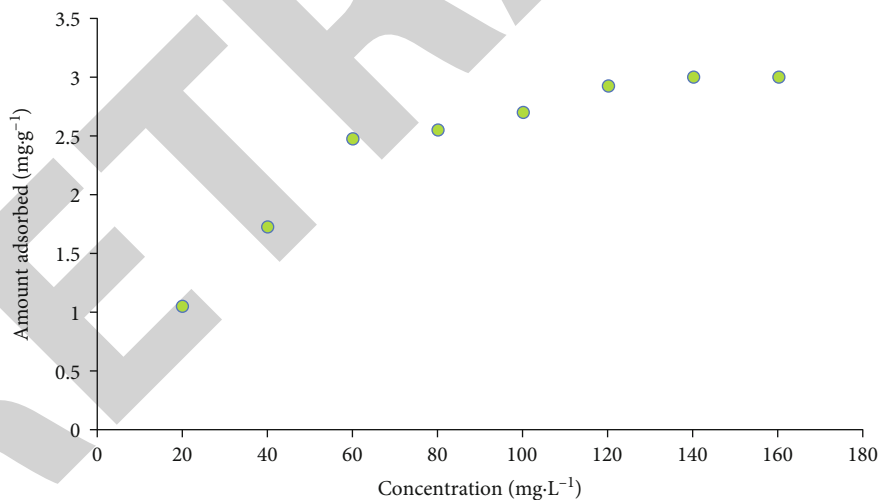


FIGURE 5: Adsorption isotherm plot of M.B on BTW.

TABLE 1: Adsorption isotherm parameters of M.B on BTW.

Isotherm model	Parameter	Values	R^2
Langmuir	K_L	0.0740	0.997
	X_M	3.367	
	K_f	2.32	
Freundlich	$\frac{1}{n}$	0.367	0.927

amount, the uptake of M.B molecules also increased and then becomes steady after 0.40 g. Thus, 0.4 g of BTW was selected as maximum sorbent dosage that was then used in subsequent experiments. The rise in the percent removal of dye with increasing dose is due to the fact that the larger amount of mass provided a larger number of active sites for the biosorption, which causes the rise in the percent of removal as already reported in the literatures by Alencar et al. [22] and Tural et al. [23].

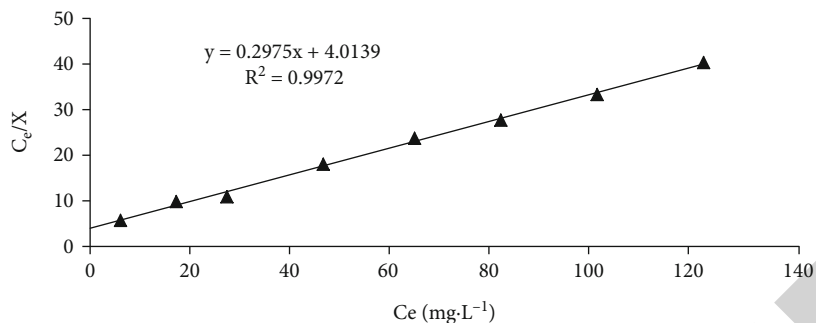


FIGURE 6: Langmuir isotherm plot for adsorption of M.B on BTW.

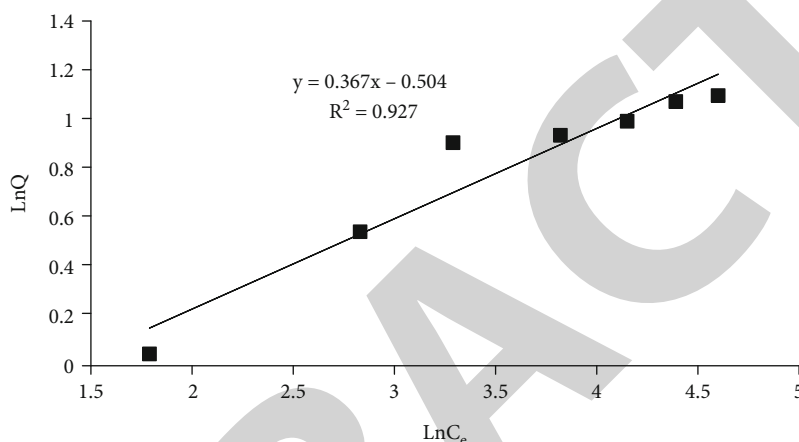


FIGURE 7: Freundlich isotherm plot for adsorption of M.B on BTW.

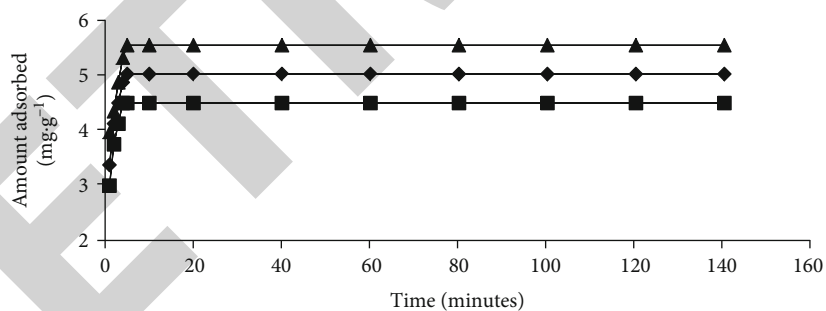


FIGURE 8: Adsorption kinetic plot of M.B on the surface of BTW at different temperatures.

3.3. *Adsorption Isotherm Studies.* The effect of dye concentration on adsorption studied in range 20 mg.L^{-1} – 140 mg.L^{-1} is shown in Figure 5. It is clearer from the Figure 4 that as concentration of M.B increases from 20 mg.L^{-1} to 100 mg.L^{-1} , the rate of adsorption increases. The plot becomes almost linear at 100 mg.L^{-1} , further increase in concentration of M.B has no effect on the removal of M.B from aquatic media. At lower initial concentrations of M.B, there were fairly insufficient dye molecules for the large number of available active sites, hence resulting in rapid uptake by sorbent. With the increase in the initial concentrations of M.B, a gradual decrease in the percentage removal of dye was observed which was due to the saturation of the available active sites [24].

3.4. *Langmuir Isotherm Model.* Linear equation [25] for the Langmuir isotherm model is given:

$$\frac{C_e}{X_e} = \frac{1}{K_L} X_m + \frac{C_e}{X_m}, \tag{3}$$

where X_e is the equilibrium concentration, X_m is the maximum sorption capacity (mg.L^{-1}), and K_L is the Langmuir isotherm constant. The value of K_L is related to free energy of sorption process, which corresponds to the affinity of between the surface of sorbate molecules and sorbent. It also defines the mode of adsorption [26, 27].

The experimental values of constants in equation are given in Table 1. After plotting C_e/X_e versus C_e , linear plot

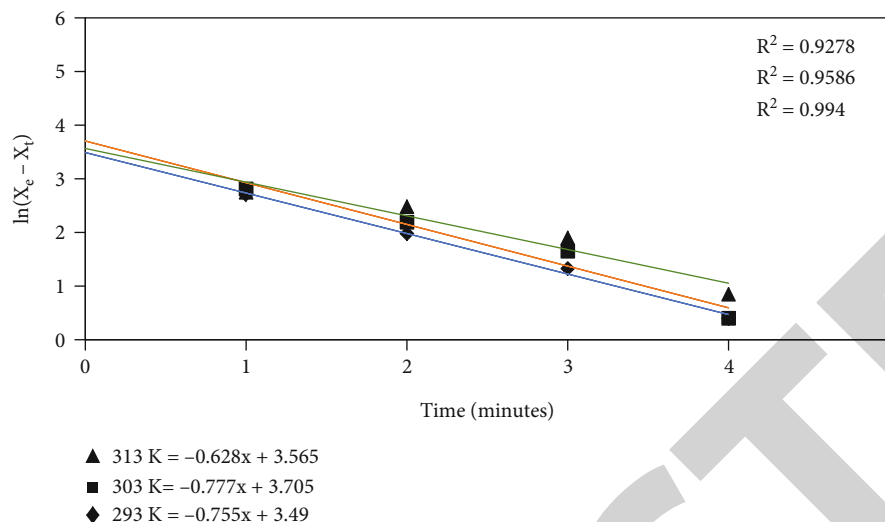


FIGURE 9: Pseudo-first-order kinetic plot of M.B onto BTW.

TABLE 2: Constant of pseudo-first-order kinetics.

Temperature	293 K	303 K	313 K
q_{\max}	0.175	0.167	0.235
K_1	3.49	3.705	3.565
R^2	0.994	0.960	0.9278

TABLE 3: Pseudo-second-order kinetic constants.

Temperature	$q_t \times 10^{-4} \text{mg.g}^{-1}$	K_2	R^2
293 K	33.33	0.0762	0.9985
303 K	0.0714	0.9980
313 K	0.9902

was obtained as shown in Figure 6. It indicates that the experimental data fitted well to the Langmuir isotherm model, verifying monolayer M.B adsorption on a homogeneous surface of BTW [26]. The value of X_m was calculated from the slope of plot while K_L (affinity constant or energy constant) was calculated from the intercept and its values are given in Table 1.

3.5. Freundlich Isotherm Model. The Freundlich isotherm model [28] is given.

$$\log q_e = \log K_f + \frac{1}{n} \log C_e, \quad (4)$$

where $1/n$ and K_f are Freundlich isotherm constants and can be obtained by plotting $\log X_e$ versus Q as shown in Figure 7. The constant K_f indicates the quantity of the M. B adsorbed on the surface of the BTW, while the constant $1/n$ shows the heterogeneity of the system or adsorption intensity. The value of $1/n < 1$ shows normal adsorption [29]. The R^2 value (0.927) was far away from unity as com-

pared to same parameter value of the Langmuir model. This reveals that the Freundlich model could not be obeyed by the experimental data.

3.6. Adsorption Kinetics. Adsorption kinetics is one of the most features of such processes that dictate the solute uptake level/the adsorption ability of biosorbent that help in deciding the possible applicability of the adsorbent to be used in industry [30]. The effect of contact time on adsorption is shown in Figure 8. It is clear from the figure that the rate of adsorption is higher in the first three minutes which then slows down. The faster rate is due to availability of active sites on the surface of BTW at start of experiments. As the sites are saturated with M.B molecules, the rate of adsorption slows down. It is due to the increasing competition between the residual dye molecules for the remaining active sites on the surface of biosorbent. Similar results have been reported earlier by Tang et al. [31], Kumar and Jena [32], Pang et al. [33], and Heidarinejad et al. [34].

3.7. Pseudo-First-Order Model. To calculate the kinetic constants of M.B adsorption on BTW, the Lagergren pseudo-first-order rate (equation (5)) was applied to the adsorption kinetic data.

$$\text{Log}(X_e - X_t) = \log X_e - \left(\frac{K_1}{2.303} \right) t, \quad (5)$$

where X_e is the amount of M.B adsorbed at equilibrium time and X_t is the amount of M.B adsorbed at time t . K_1 is the pseudo-first-order rate constant. The constants in the equation were obtained by plotting time (t) vs. $\ln X_e - X_t$ (Figure 9). The values of constants are given in Table 2. The results obtained for this model were not in accordance with the experimental data. The lower value of R^2 is less than unity at temperature 303 and 313 K showing that this model does not fit well the kinetic data.

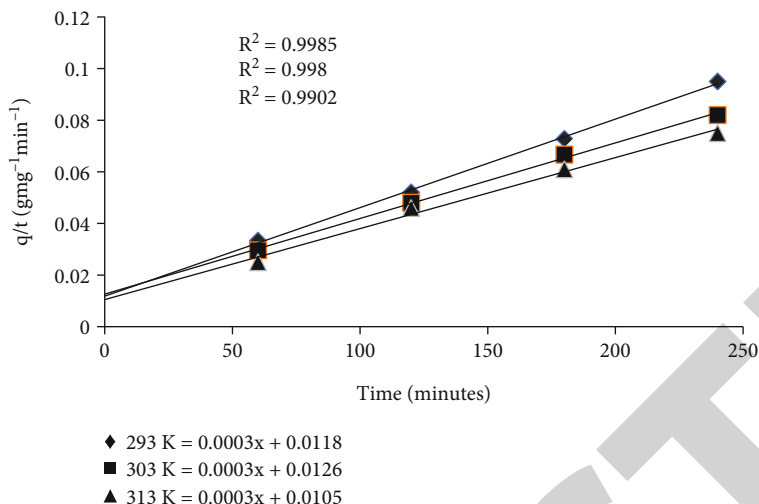


FIGURE 10: Pseudo-second-order kinetic plot of M.B onto BTW.

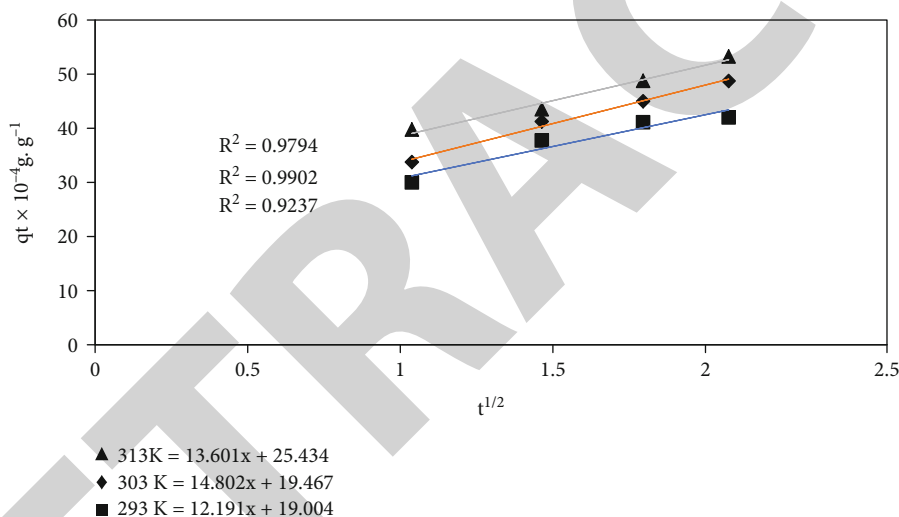


FIGURE 11: Intraparticle diffusion plot.

TABLE 4: Constant of intraparticle diffusion.

Temperature	293 K	303 K	313 K
K_{diff}	12.191	14.802	13.601
C	19.004	19.467	25.434
R^2	0.9237	0.9902	0.9794

3.8. Pseudo-Second-Order Model. This model can be given as follows [30, 35]:

$$\frac{t}{X_t} = \frac{1}{K_2 X_e} + \frac{t}{X_t} \quad (6)$$

This is the linear form of the pseudo-second-order kinetic model where X_e is the amount of M.B adsorbed at equilibrium, X_t is the amount of M.B adsorbed at time t , and K_2 is the pseudo-second-order rate constant. The values of constants (Table 3) were obtained by plotting t/X_t vs. time

(Figure 10). The value of R^2 is almost equal unity at all temperatures showing that the kinetic data could be better accommodated by this model and adsorption process is chemical in nature where the sharing or exchange of electrons between sorbate and biosorbent has been taken place. Similar results have been reported by Li et al. [36], X. Li and Y. Li [37], Weng et al. [38], Ravi and Pandey [39], Pathania et al. [40], and Qian et al. [41].

3.9. Intraparticle Diffusion Model. Intraparticle diffusion mechanism was used to investigate the diffusion mechanism of adsorption process. For intraparticle diffusion, equation (7) [42] is used:

$$q_{t=K_{diff}} t^{1/2} + C. \quad (7)$$

In equation (7), K_{diff} ($\text{mgg}^{-1} \text{min}^{-1}$) is intraparticle diffusion rate constant, q_t is the amount of M.B adsorbed on BTW at time (t), $t^{1/2}$ is the square root of time, and C is

the thickness of boundary layer. A linear plot (Figure 11) was obtained by plotting q_t vs. $t^{1/2}$. The thickness of boundary layer is obtained from the intercept of the linear plot, while K_{diff} ($\text{mgg}^{-1} \text{min}^{-1}$) was obtained from the slope of linear plot. The values of constants obtained from linear plot are tabulated in Table 4. It was observed from the linear plot of intraparticle diffusion plot that the process of M.B adsorption on BTW is a diffusion-controlled process [43].

4. Conclusions

In this study, black tea wastes were used as biosorbent for the removal of M.B from aqueous media. The methylene blue was efficiently recovered from solution through the biosorbent. The sorption of methylene blue was studied as a function of time, initial concentration of methylene blue, temperature, pH of methylene blue, and sorbent dosage. The kinetic studies were recorded for three different temperatures, i.e., 293 K, 303 K, and 313 K. The equilibrium time for the removal of methylene blue on black tea wastes was achieved within 3 min. The fast rate of kinetics shows that methylene blue dye adhered to the surface of adsorbent very quickly. The adsorption isotherm data was well explained by the Langmuir adsorption isotherm while the adsorption kinetic data by pseudo-second-order kinetic model rather than pseudo-first-order kinetic model. Maximum removal of methylene blue occurs at pH 8. The optimum dosage of black tea wastes was selected 0.4 g in all experiments. The adsorbent was effective in the removal of selected dye, and its use could be extended to other dyes as well.

Data Availability

All the available data are incorporated in the MS.

Conflicts of Interest

Authors declared that they have no conflict of interest.

Acknowledgments

Authors wish to thank the researchers supporting project number (RSP-2021-45) at the King Saud University, Riyadh, Saudi Arabia, for their financial support.

References

- [1] M. Anantha, S. Olivera, C. Hu et al., "Comparison of the photocatalytic, adsorption and electrochemical methods for the removal of cationic dyes from aqueous solutions," *Environmental Technology & Innovation*, vol. 17, article 100612, 2020.
- [2] F. K. G. Sardi, M. Behpour, Z. Ramezani, and S. Masoum, "Simultaneous removal of Basic Blue41 and Basic Red46 dyes in binary aqueous systems via activated carbon from palm bio-waste: optimization by central composite design, equilibrium, kinetic, and thermodynamic studies," *Environmental Technology and Innovation*, vol. 24, article 102039, 2021.
- [3] H. G. Choi and S. W. Yu, "Biosorption of methylene blue from aqueous solution by agricultural bioadsorbent corncob," *Environmental Engineering Research*, vol. 24, no. 1, pp. 99–106, 2019.
- [4] H. Gao, S. Zhao, X. Cheng, X. Wang, and L. Zheng, "Removal of anionic azo dyes from aqueous solution using magnetic polymer multi-wall carbon nanotube nanocomposite as adsorbent," *Chemical Engineering Journal*, vol. 223, pp. 84–90, 2013.
- [5] B. Lellis, C. Z. F. Polonio, J. A. Pamphile, and J. C. Polonio, "Effects of textile dyes on health and the environment and bioremediation potential of living organisms," *Biotechnology Research and Innovation*, vol. 3, no. 2, pp. 275–290, 2019.
- [6] H. Zollinger, *Azo Dyes and Pigments. Color Chemistry: Synthesis, Properties and Applications of Organic Dyes and Pigments*, VCH, New York, NY, 1987.
- [7] Kamaljit Singh and Sucharita Arora, "Removal of Synthetic Textile Dyes From Wastewaters: A Critical Review on Present Treatment Technologies," *Critical Reviews in Environmental Science and Technology*, vol. 41, no. 9, pp. 807–878, 2011.
- [8] D. A. Yaseen and M. Scholz, "Textile dye wastewater characteristics and constituents of synthetic effluents: a critical review," *International Journal of Environmental Science and Technology*, vol. 16, no. 2, pp. 1193–1226, 2019.
- [9] W. Gan, X. Shang, X. H. Li, J. Zhang, and X. Fu, "Achieving high adsorption capacity and ultrafast removal of methylene blue and Pb^{2+} by graphene-like TiO_2/C ," *Colloids and surface A: Physicochemical and engineering Aspects*, vol. 561, pp. 218–225, 2019.
- [10] M. Ehrampoosh, G. H. Moussavi, M. Ghaneian, S. Rahimi, and M. Ahmadian, "Removal of methylene blue dye from textile simulated sample using tubular reactor and $\text{TiO}_2/\text{UV-C}$ photocatalytic process," *Iranian Journal of Environmental Health Sciences and Engineering*, vol. 8, pp. 35–40, 2011.
- [11] S. Rangabhashiyam and S. Lata, "Biosorption characteristics of methylene blue and malachite green from simulated wastewater onto *Carica papaya* wood biosorbent," *Surfaces and Interfaces*, vol. 10, pp. 197–215, 2018.
- [12] M. Zahoor, M. Wahab, S. M. Salman, A. Sohail, E. A. Ali, and R. Ullah, "Removal of doxycycline from water using *Dalbergia sissoo* waste biomass based activated carbon and magnetic oxide/activated bioinorganic nanocomposite in batch adsorption and adsorption/membrane hybrid processes," *Bioinorganic Chemistry and Applications*, vol. 2022, Article ID 2694487, 17 pages, 2022.
- [13] M. Muneeb Ur Rahman Khattak, M. Zahoor, B. Muhammad, F. A. Khan, R. Ullah, and N. M. AbdEl-Salam, "Removal of heavy metals from drinking water by magnetic carbon nanostructures prepared from biomass," *Journal of Nanomaterials*, vol. 2017, Article ID 5670371, 10 pages, 2017.
- [14] M. Zahoor, A. Ullah, and S. Alam, "Removal of enrofloxacin from water through magnetic nanocomposites prepared from pineapple waste biomass," *Surface Engineering and Applied Electrochemistry*, vol. 55, no. 5, pp. 536–547, 2019.
- [15] A. Ullah, M. Zahoor, S. Alam, R. Ullah, A. S. Alqahtani, and H. M. Mahmood, "Separation of levofloxacin from industry effluents using novel magnetic nanocomposite and membranes hybrid processes," *BioMed Research International*, vol. 2019, Article ID 5276841, 13 pages, 2019.
- [16] I. Muhammad, A. Kalsoom, M. I. Khan et al., "Pollution, toxicity and carcinogenicity of organic dyes and their catalytic bio-remediation," *Current Pharmaceutical Design*, vol. 25, no. 34, pp. 3645–3663, 2019.

- [17] Q. Zhou, W. Q. Gong, Y. B. Li et al., "Biosorption of methylene blue onto spent corn cob substrate kinetics, equilibrium and thermodynamic studies," *Water Sci, Technol*, vol. 63, no. 12, pp. 2775–2780, 2011.
- [18] S. T. Ong, P. S. Keng, A. W. Chong, S. L. Lee, and Y. T. Hung, "Tartaric acid modified rice hull as a sorbent for methylene blue removal," *American Journal of Environmental Sciences*, vol. 6, no. 3, pp. 244–248, 2010.
- [19] K. Malmström, J. Savolainen, and E. O. Terho, "Allergic alveolitis from pine sawdust," *Allergy*, vol. 54, no. 5, pp. 532–533, 1999.
- [20] E. Pelvan and M. Özilgen, "Assessment of energy and exergy efficiencies and renewability of black tea, instant tea and ice tea production and waste valorization processes," *Sustainable Production and Consumption*, vol. 12, pp. 59–77, 2017.
- [21] R. Singh, T. S. Singh, J. O. Odiyo, J. A. Smith, and J. N. Edokpayi, "Evaluation of methylene blue sorption onto low-cost biosorbents: equilibrium, kinetics, and thermodynamics," *Journal of Chemistry*, vol. 2020, Article ID 8318049, 11 pages, 2020.
- [22] W. S. Alencar, E. Acayanka, E. C. Lima et al., "Application of *Mangifera indica* (mango) seeds as a biosorbent for removal of Victazol Orange 3R dye from aqueous solution and study of the biosorption mechanism," *Chemical Engineering Journal*, vol. 209, pp. 577–588, 2012.
- [23] B. Tural, E. Ertaş, B. Enez, S. A. Fincan, and S. Tural, "Preparation and characterization of a novel magnetic biosorbent functionalized with biomass of *Bacillus Subtilis*: Kinetic and isotherm studies of biosorption processes in the removal of Methylene Blue," *Journal of Environmental Chemical Engineering*, vol. 5, no. 5, pp. 4795–4802, 2017.
- [24] W. Hassan, U. Farooq, M. Ahmad, M. Athar, and M. A. Khan, "Potential biosorbent, *Haloxylon recurvum* plant stems, for the removal of methylene blue dye," *Arabian Journal of Chemistry*, vol. 10, pp. 1512–1522, 2017.
- [25] I. Langmuir, "The constitution and fundamental properties of solids and liquids: part I—solids," *Journal of the American Chemical Society*, vol. 38, no. 11, pp. 2221–2295, 1916.
- [26] F. Silva, L. Nascimento, M. Brito, K. da Silva, W. Paschoal Jr., and R. Fujiyama, "Biosorption of methylene blue dye using natural biosorbents made from weeds," *Materials*, vol. 12, no. 15, p. 2486, 2019.
- [27] S. Pal, S. Ghorai, C. Das, S. Samrat, A. Ghosh, and A. B. Panda, "Carboxymethyl tamarind-g-poly (acrylamide)/silica: a high performance hybrid nanocomposite for adsorption of methylene blue dye," *Industrial and Engineering Chemistry Research*, vol. 51, no. 48, pp. 15546–15556, 2012.
- [28] H. M. F. Freundlich, "Over the adsorption in solution," *Journal of Physical Chemistry*, vol. 57, pp. 4574–4578, 1906.
- [29] M. Zbair, Z. Anfar, H. Khallok, H. A. Ahsaine, M. Ezahri, and N. Elalem, "Adsorption kinetics and surface modeling of aqueous methylene blue onto activated carbonaceous wood sawdust," *Fullerenes, Nanotubes, and Carbon Nanostructures*, vol. 26, no. 7, pp. 433–442, 2018.
- [30] M. Zahoor, "Removal of crystal violet from water by adsorbent prepared from Turkish coffee residue," *Tenside Surfactants Detergents*, vol. 49, no. 2, pp. 107–113, 2012.
- [31] Y. Tang, Y. Zeng, T. Hu, Q. Zhou, and Y. Peng, "Preparation of lignin sulfonate-based mesoporous materials for adsorbing malachite green from aqueous solution," *Journal of Environmental Chemical Engineering*, vol. 4, no. 3, pp. 2900–2910, 2016.
- [32] A. Kumar and H. M. Jena, "Removal of methylene blue and phenol onto prepared activated carbon from Fox nutshell by chemical activation in batch and fixed-bed column," *Journal of Cleaner Production*, vol. 137, pp. 1246–1259, 2016.
- [33] J. Pang, F. Fu, Z. L. J. Ding, N. Li, and B. Tang, "Adsorption behaviors of methylene blue from aqueous solution on mesoporous birnessite," *Journal of the Taiwan Institute of Chemical Engineers*, vol. 77, pp. 168–176, 2017.
- [34] Z. Heidarinejad, O. Rahmanian, M. Fazlzadeh, and M. Heidari, "Enhancement of methylene blue adsorption onto activated carbon prepared from Date Press Cake by low frequency ultrasound," *Journal of Molecular Liquids*, vol. 264, pp. 591–599, 2018.
- [35] H. Ren, Z. Gao, D. Wu, J. Jiang, Y. Sun, and C. Luo, "Efficient Pb(II) removal using sodium alginate-carboxymethyl cellulose gel beads: preparation, characterization, and adsorption mechanism," *Carbohydrate Polymers*, vol. 137, pp. 402–409, 2016.
- [36] Y. Li, X. Zhang, R. Yang, G. Li, and C. Hu, "Removal of dyes from aqueous solutions using activated carbon prepared from rice husk residue," *Water Science and Technology*, vol. 73, no. 5, pp. 1122–1128, 2016.
- [37] X. Li and Y. Li, "Adsorptive removal of dyes from aqueous solution by KMnO_4 -modified rice husk and rice straw," *Journal of Chemistry*, vol. 2019, Article ID 8359491, 9 pages, 2019.
- [38] C. H. Weng, Y. T. Lin, and T. W. Tzeng, "Removal of methylene blue from aqueous solution by adsorption onto pineapple leaf powder," *Journal of Hazardous Materials*, vol. 170, no. 1, pp. 417–424, 2009.
- [39] L. M. Ravi and L. M. Pandey, "Enhanced adsorption capacity of designed bentonite and alginate beads for the effective removal of methylene blue," *Applied Clay Science*, vol. 169, pp. 102–111, 2019.
- [40] D. Pathania, S. Sharma, and P. Singh, "Removal of methylene blue by adsorption onto activated carbon developed from *Ficus carica* bast," *Arabian Journal of Chemistry*, vol. 10, pp. S1445–S1451, 2017.
- [41] W. C. Qian, X. P. Luo, X. Wang, M. Guo, and B. Li, "Removal of methylene blue from aqueous solution by modified bamboo hydrochar," *Ecotoxicology and Environmental Safety*, vol. 157, pp. 300–306, 2018.
- [42] V. O. S. Neto, D. Q. Melo, and T. C. de Oliveira, "Evaluation of new chemically modified coconut shell adsorbents with tannic acid for Cu (II) removal from wastewater," *Journal of Applied Polymer Science*, vol. 131, pp. 40744–40755, 2014.
- [43] T. K. Sen, S. Afroze, and H. M. Ang, "Equilibrium, kinetics and mechanism of removal of methylene blue from aqueous solution by adsorption onto pine cone biomass of *Pinus radiata*," *Water, Air, & Soil Pollution*, vol. 218, no. 1–4, pp. 499–515, 2011.