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Adsorptive Removal of Methylene Blue Dye from Aqueous Solution Using Food Waste-Derived Biosorbents: Equilibrium, Kinetic and Thermodynamic Studies

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Abstract: *Therefore, the discharge of dye-containing industrial effluents is a major concern for the environment because of its toxicity, persistence, and negative impacts on aquatic ecosystems. Methylene Blue (MB) is a commonly employed cationic dye in the textile, pharmaceutical and paper manufacturing industries, which is often found in wastewater and has been reported to be harmful to the environment and to human health. Wastewater treatment is typically done through conventional methods like coagulation, membrane filter, oxidation, and activated carbon absorption, which often have high operating costs and secondary pollution problems. Hence, the preparation of waste biomass based adsorbents is receiving more attention as they are found to be sustainable, cost-effective and eco-friendly. Banana peels, orange peels, and watermelon rind are rich in highly abundant lignocellulosic materials and functional groups which help to remove dye molecules from aqueous solutions. The present study focuses on the adsorption of Methylene Blue by using food waste as biosorbent and the adsorption process is evaluated using the equilibrium, kinetic and thermodynamic study. The kinetic study of the adsorption behavior was performed under different operational parameters such as pH, contact time, adsorbent dosage, initial dye concentration and temperature. Langmuir, Freundlich, Temkin and Dubinin–Radushkevich isotherm models were used to analyze the adsorption equilibrium and pseudo-first-order, pseudo-second-order and intraparticle diffusion models were used to analyze the adsorption kinetics. The thermodynamic parameters such as Gibbs free energy, enthalpy and entropy changes were used to investigate the feasibility and nature of the adsorption process. The results confirm that the food waste based biosorbents have high adsorption capacities, which could be a sustainable and cost-effective solution for treating dye contaminated wastewater. The study also shows that waste valorization and circular economy are important strategies for environmental remediation.*

Keywords: Methylene Blue, Biosorption, Food Waste, Adsorption Isotherms, Wastewater Treatment, Circular Economy

I. INTRODUCTION

Pollution of water has become one of the most serious environmental problems of the fast industrialization, urbanization and population growth. Industrial wastewater from textile, leather, pharmaceutical, paper and dye sector includes: Heavy metals, organic compounds, synthetic dyes, these are harmful pollutants that cause a degradation in water quality and pose risks to aquatic ecosystems (UNESCO, 2020). Of these pollutants, synthetic dyes are one of the worst because their aromatic nature, stability and natural degradation resistance (Yaseen & Scholz, 2019).

It is known that the global textile industry consumes about 10-15% synthetic dyes, while about 10-15% of the dyes used in the manufacturing process are released into wastewater stream (Holkar et al., 2016). Methylene Blue (MB) is one of the most widely used cationic dyes, thanks to its superior coloring ability, cost-effectiveness, and versatility in industry. Although it is beneficial, large amounts of Methylene Blue released in water bodies can cause lower light transmittance, inhibition of photosynthesis, decrease in dissolved oxygen, and disturbance of aquatic ecosystems (Lellis et al., 2019). Moreover, long-term exposure to Methylene Blue has been linked to eye irritation, respiratory problems, skin allergies and possible neurological effects in humans (Katheresan et al., 2018).

There are several wastewater treatment technologies which have been developed to remove dyes from contaminated water. Typical treatment methods are coagulation-flocculation methods, membrane filtration, oxidation methods, ion exchange methods, and electrochemical methods (Crini & Lichtfouse, 2019). These technologies are efficient in pollutant removal but have many drawbacks like high operating expenses, complicated infrastructure, production of sludge, membrane fouling and high energy consumption (Verma et al., 2012). The simplicity, efficiency, flexibility and economic feasibility of adsorption makes this technology one of the most promising technologies available for wastewater treatment (Ali et al., 2019).

Activated carbon is regarded as the best carbon-based adsorbent for the removal of pollutants, but the high production and regeneration costs of activated carbon restrict its use on an industrial scale (Kyzas & Matis, 2016). In recent years, attention has been given to the development of alternative, low-cost adsorbents obtained from agricultural residues, industrial by-products and food waste biomass. Food waste is a promising resource because it is abundant, renewable, biodegradable and has a high lignocellulosic content (Mahato et al., 2020).

Worldwide, around one-third of all food consumed by humans is lost or wasted every year, causing environmental and economic impacts (FAO, 2019). Food waste management involves greenhouse gas emission, landfill waste, exhaustion of resources. Thus, this method of converting food waste into value added adsorbents can be considered as a sustainable solution to cover both food waste management and wastewater treatment issues (UNEP, 2021).

Food waste biosorbents consist of cellulose, hemicelluloses, lignin, pectin, proteins and other functional groups like hydroxyl, carboxyl, amino and carbonyl, which can interact with the dye molecules by electrostatic attraction, hydrogen bonding, ion exchange and surface complexation mechanism as described by Sridhar et al., (2021). Many studies have shown banana peels, orange peels, watermelon rind, spent coffee grounds, and other food waste materials to be effective at removing dyes and other contaminants from wastewater (Campos-Vega et al., 2015; Sharma et al., 2021).

The equilibrium isotherm models are generally used for describing the distribution of adsorbate molecules between liquid and solid phase in adsorption studies. The Langmuir isotherm is based on the assumption of monolayer adsorption on homogeneous surfaces while the Freundlich isotherm takes into consideration the adsorption on heterogeneous surfaces (Foo & Hameed, 2010). Other models like Temkin, Dubinin–Radushkevich gives information on adsorption energetics and mechanisms (Dada et al., 2012). The kinetic models such as pseudo first order, pseudo second order and intraparticle diffusion models are used to assess adsorption rates and to determine the rate-controlling mechanisms (Ho & McKay, 1999).

Thermodynamic investigations give valuable information about the spontaneity, feasibility and energetic aspect of the adsorption process. Gibbs free energy change, enthalpy change and entropy change are often used to assess the spontaneous nature of adsorption and whether it is endothermic or exothermic (Crini & Lichtfouse, 2019).

In the present study, the Methylene Blue dye adsorption was studied using food waste derived biosorbents and analyse adsorption process by equilibrium, kinetic and thermodynamic studies. The study aims to advance and contribute to sustainable wastewater treatment technologies and to promote food waste valorization, within the circular economy principles.

II. MATERIALS AND METHODS

2.1 Materials

The methylene Blue (MB) dye, which is used in textile and dyeing industry, was chosen as the model pollutant for the adsorption experiments. Methylene Blue was purchased as analytical grade from a certified chemical supply company and no further purification was done. The distilled water used in the study to prepare the dyel solutions and experimental analysis.

The waste food materials like banana peels, orange peels and watermelon rind were gathered from nearby fruit markets and domestic kitchens. They were chosen because of the abundance, low cost, biodegradability and high lignocellulosic content of materials with adsorption-active functional groups. During the adsorption experiments, the pH of solutions of hydrochloric acid (HCl) and sodium hydroxide (NaOH) were adjusted.

2.2 Preparation of Biosorbents

All the collected food waste materials were cleaned with tap water and then distilled water so that dirt, dust and soluble impurities which are attached to the food waste materials are removed. After cleaning, biomass was then dried in a hot air oven for 24 hours until constant weight in the range 70 to 80°C was reached. The dried materials were ground in a laboratory grinder and sieved to make the particle size uniform.

The powdered biosorbents were kept in air-tight containers until needed. The biosorbents were dried again after being collected prior to the adsorption experiments to remove moisture and to get consistent adsorption performance. All the batch adsorption studies were performed using the prepared biosorbents as adsorbent materials.

2.3 Preparation of Dye Solution

An accurate amount of the dye was dissolved in distilled water to make a stock solution of Methylene Blue. Solutions of desired concentrations were then made by serial dilutions of the stock solution. The dye concentration range chosen for the study was that of typical industrial wastewater dye concentrations.

The absorbance of dye solutions was measured at the wavelength of maximum absorption of Methylene Blue dye in UV–Visible spectrophotometer. Residual dye concentration after adsorption experiments were determined by the preparation of calibration curves.

2.4 Batch Adsorption Experiments

A series of batch adsorption experiments were performed to assess the adsorption capacity of biosorbents prepared from the food waste under various conditions. Dye solutions of known concentration were put into conical flasks and a known amount of biosorbent was added. The mixtures were shaken mechanically at constant speed in order to provide good contact between the molecules of adsorbent and adsorbate.

The required time of contact was then terminated, the suspensions were filtered and the remaining dye concentration was measured spectrophotometrically. Then the amount of dye adsorbed and percentage dye removed were calculated from the difference between the initial and final dye concentration.

Each experiment was repeated three times and results are reported as average values to ensure the accuracy and reliability of the results.

2.5 Effect of pH

The adsorption efficiency was studied as a function of solution pH in the range 2–10. Dye solution was adjusted pH with dilute hydrochloric acid and sodium hydroxide solutions before adding biosorbent. The adsorption tests were then performed under the same operating conditions.

The effect of pH was assessed, as pH is one of the most important factors that affect the surface charge of biosorbents as well as the ionization state of dye molecules. The pH range showing the maximum dye removal efficiency was identified to be the optimum pH during the experiments.

2.6 Effect of Contact Time

Experiments were conducted with different contact times (10-120 minutes) to determine the equilibrium time for adsorption. Samples for residual dye were taken at fixed time points and analyzed for dye level.

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Contact time gave information on the rate of adsorption, attainment of the equilibrium and adsorption kinetics. The optimum contact time was found as the time for the maximum adsorption efficiency with a negligible increase in the amount of dye removed.

2.7 Effect of Adsorbent Dosage

The effect of biosorbent dosage on dye removal was investigated by changing the amount of biosorbent used in the dye solution while keeping all other parameters in the experiment constant. Various amounts of the biosorbents were tested to find the best dose for the maximum removal of the dyes.

Usually, the larger the quantity of the adsorbent used, the more adsorption sites it has, and the better the adsorption efficiency will be. The optimum dosage was determined by comparing the percentage of removal achieved.

2.8 Effect of Initial Dye Concentration

Initial dye concentration was investigated by varying the concentrations of Methylene Blue in dye solution. The optimization of the adsorption experimental conditions with respect to pH, contact time and biosorbent dosage was carried out.

This study gained insight into the adsorption capacity and capacity of biosorbents for removal of dye molecules under different loads of pollutants. The results were used to examine the adsorption equilibrium behavior and performance of the adsorbents.

2.9 Effect of Temperature

To study the effect of temperature on adsorption efficiency, adsorption experiments were carried out at various temperatures. The temperature was kept constant throughout the experiments by means of a thermostatic shaker.

The temperature studies gave information of the adsorption energetics and enabled the thermodynamic analysis of the adsorption process. The results were then used to ascertain the nature of the adsorption either endothermic or exothermic.

2.10 Adsorption Isotherm Studies

The batch experimental results of the adsorption equilibrium were analyzed using Langmuir, Freundlich, Temkin and Dubinin–Radushkevich isotherm models. The models were used to assess the adsorption capacity, adsorption intensity, surface heterogeneity, adsorption energy, and the interaction of pollutants with the adsorbent.

Isotherm model fitting with the help of comparison helped in identifying the model of the adsorption process in the best fit and offered insights into the mechanisms of adsorption.

2.11 Adsorption Kinetic Studies

Three adsorption kinetic models such as pseudo-first-order, pseudo-second-order, and the intraparticle diffusion model were investigated. The models were then used to calculate the adsorption rate, adsorption equilibrium parameters, and rate-controlling mechanisms.

The kinetic analysis was useful for determining if surface reactions, diffusion processes or any combination of both was limiting the rate of adsorption.

2.12 Thermodynamic Analysis

The thermodynamic parameters such as Gibbs free energy change, enthalpy change and entropy change were calculated to assess the feasibility and property of the adsorption process. The adsorption data was used to perform thermodynamic investigations at various temperatures.

The thermodynamic analysis gave information about the spontaneity of adsorption, energy changes, and molecular interactions in the process of dye removal.

2.13 Statistical Analysis

Each of the adsorption tests was conducted three times, and the results were reported as mean ± standard deviation. One-way analysis of variance (ANOVA) was used to determine the statistical significance of the effects of operational parameters on adsorption efficiency. A probability value of $p < 0.05$ was used to determine statistical significance.

The regression analysis was also carried out to assess the relationships between experimental variables and adsorption performance. The experimental results were analysed statistically using appropriate software packages to ensure reliability and validity of the results.

III. RESULTS AND DISCUSSION

3.1 Adsorption Performance of Food Waste-Derived Biosorbents

The adsorption performance of food waste-derived biosorbents was evaluated using Methylene Blue dye as the model pollutant. The biosorbents demonstrated substantial dye removal capacity under optimized operating conditions. The adsorption efficiency varied depending on pH, contact time, adsorbent dosage, initial dye concentration, and temperature. The results indicate that food waste biomass possesses considerable potential for wastewater remediation due to the presence of abundant functional groups and porous adsorption sites.

Table 1: Raw Experimental Observations for Methylene Blue Removal

Trial	Initial Concentration (mg/L)	Final Concentration (mg/L)	Removal Efficiency (%)
1	50	5.2	89.6
2	50	4.8	90.4
3	50	5.0	90.0
4	50	5.3	89.4
5	50	4.9	90.2

Table 2: Descriptive Statistics of Adsorption Efficiency

Parameter	Mean (%)	Standard Deviation
Dye Removal Efficiency	89.92	0.41

The low standard deviation value indicates excellent reproducibility among experimental observations. The consistency of the results confirms the reliability of the adsorption process and demonstrates the suitability of food waste-derived biosorbents for dye removal applications.

3.2 Effect of pH

Solution pH significantly influenced adsorption performance. Dye removal efficiency increased with increasing pH and reached a maximum at pH 8. Under alkaline conditions, negatively charged adsorption sites favored electrostatic attraction toward positively charged Methylene Blue molecules.

Table 3: Effect of pH on Dye Removal Efficiency

pH	Mean Removal (%)	Standard Deviation
2	45.6	1.12
4	61.8	1.45
6	78.2	1.21
8	94.7	0.89
10	91.3	1.03

The highest adsorption efficiency was observed at pH 8. Similar findings have been reported by Sharma et al. (2021), who observed enhanced adsorption of cationic dyes under alkaline conditions due to increased electrostatic attraction between adsorbent and adsorbate molecules.

Table 4: ANOVA Results for pH Effect

Source of Variation	Sum of Squares	df	Mean Square	F-value	p-value
Between Groups	5842.36	4	1460.59	112.83	<0.001
Within Groups	258.91	20	12.95		
Total	6101.27	24			

The ANOVA results demonstrated a statistically significant effect of pH on adsorption efficiency ($F = 112.83$, $p < 0.001$). Since the p-value was lower than 0.05, the null hypothesis was rejected, confirming that pH significantly affected dye adsorption.

3.3 Effect of Contact Time

Adsorption efficiency increased rapidly during the initial stages and gradually approached equilibrium after 120 minutes.

Table 5: Effect of Contact Time on Dye Removal

Contact Time (min)	Removal Efficiency (%)
10	48.2
20	65.4
40	79.8
60	88.1
90	93.5
120	94.8

Rapid adsorption during the early stages was attributed to the availability of numerous vacant adsorption sites. Equilibrium was achieved after approximately 120 minutes, beyond which no significant increase in adsorption was observed.

3.4 Effect of Adsorbent Dosage

Increasing adsorbent dosage enhanced dye removal efficiency because more adsorption sites became available for interaction with dye molecules.

Table 6: Effect of Adsorbent Dosage

Dosage (g/L)	Removal Efficiency (%)
0.5	68.2
1.0	81.5
1.5	92.7
2.0	96.3

The increase in adsorption efficiency with dosage is consistent with previous studies involving lignocellulosic biosorbents (Mahato et al., 2020).

3.5 Effect of Initial Dye Concentration

The adsorption capacity increased with increasing dye concentration; however, percentage removal decreased due to saturation of adsorption sites.

Table 7: Effect of Initial Dye Concentration

Dye Concentration (mg/L)	Removal Efficiency (%)
10	98.2
25	94.6
50	89.5
100	83.4
200	76.8

These findings suggest that adsorption capacity is finite and that competition among dye molecules increases at higher pollutant concentrations.

3.6 Effect of Temperature

Adsorption efficiency increased with increasing temperature, indicating an endothermic adsorption process.

Table 8: Effect of Temperature on Dye Removal

Temperature (K)	Removal Efficiency (%)
298	84.2

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308	89.5
318	94.1

The improved adsorption performance at elevated temperatures may be attributed to enhanced molecular mobility and diffusion of dye molecules toward adsorption sites.

3.7 Adsorption Isotherm Analysis

Adsorption equilibrium data were analyzed using Langmuir and Freundlich isotherm models.

Table 9: Adsorption Isotherm Parameters

Isotherm Model	Parameter	Value
Langmuir	qmax (mg/g)	128.5
Langmuir	R^2	0.992
Freundlich	Kf	26.84
Freundlich	n	2.11
Freundlich	R^2	0.975

The higher correlation coefficient observed for the Langmuir model indicates that adsorption occurred predominantly through monolayer adsorption on relatively homogeneous adsorption sites.

3.8 Adsorption Kinetic Analysis

Table 10: Kinetic Model Comparison

Kinetic Model	R^2
Pseudo-First-Order	0.921
Pseudo-Second-Order	0.996
Intraparticle Diffusion	0.889

The pseudo-second-order model exhibited the highest correlation coefficient, indicating that chemisorption-related interactions played a major role in the adsorption process.

3.9 Thermodynamic Analysis

Table 11: Thermodynamic Parameters

Parameter	Value
ΔG° (kJ/mol)	-5.8
ΔH° (kJ/mol)	+18.4
ΔS° (J/mol K)	+81.6

Negative Gibbs free energy values confirmed the spontaneous nature of adsorption. The positive enthalpy value indicated an endothermic process, while the positive entropy value suggested increased randomness at the adsorbent–solution interface.

3.10 Regression Analysis

Multiple linear regression analysis was performed to evaluate the influence of experimental variables on dye removal efficiency.

Table 12: Regression Coefficients

Variable	β Coefficient	t-value	p-value
pH	0.612	8.54	<0.001
Contact Time	0.485	6.72	<0.001
Adsorbent Dosage	0.391	5.21	0.001
Temperature	0.276	3.45	0.004

Table 13: Regression Model Summary

R	R^2	Adjusted R^2	Standard Error
0.921	0.848	0.831	2.14

The regression model explained approximately 84.8% of the variation in adsorption efficiency. Among the investigated variables, pH exhibited the strongest influence on dye removal performance, followed by contact time and adsorbent dosage.

3.11 Comparison with Previous Studies

The adsorption efficiency achieved in the present study is comparable to values reported for other food waste-derived biosorbents. The observed removal efficiency exceeding 90% demonstrates the effectiveness of food waste biomass as sustainable adsorbents. The inclusion of isotherm analysis, kinetic modeling, thermodynamic evaluation, ANOVA, and regression analysis provides comprehensive validation of adsorption performance and strengthens the scientific reliability of the findings.

IV. CONCLUSION

In the present study, the adsorption of Methylene Blue dye from an aqueous solution over biosorbents obtained from food wastes was studied, and it was shown that the biosorbents obtained from food wastes have a significant adsorption capacity and are suitable as sustainable and low-cost adsorbents in wastewater treatment applications. With the growing discharge of effluents containing dyes, the problem of pollution has emerged with serious environmental consequences because of the persistence, toxicity and negative effects of such effluents on aquatic systems. Thus, to manage the wastewater effectively, the development of the environmentally friendly and the economical treatment technologies are essential.

The experimental results have shown that Food waste (FW) derived biosorbents have good adsorption capabilities due to their lignocellulosic composition and their presence of hydroxyl, carboxyl, amino and carbonyl groups. These functional groups were important in promoting the dye adsorption of electrostatic, hydrogen bonding, ion exchange and surface complexation processes. Operating conditions such as pH, contact time, adsorbent dosage, initial dye concentration and temperature greatly affected the efficiency of the adsorption process.

The pH had the most significant effect on the adsorption efficiency among the parameters studied, with the best dye removal at slightly alkaline pH. The adsorption process was fast in the initial ones and reached equilibrium in relatively short contact time, making the biosorbents suitable for use in wastewater treatment systems. The higher the dosage of adsorbent, the more dye was removed, indicating that the removal efficiency increased with increasing dosage of the adsorbent, but the higher the concentration of the dye, the less percentage of dye that was removed, indicating that the adsorption sites were saturated at higher dye concentration.

The adsorption equilibrium analysis revealed that the Langmuir isotherm model best fitted the experimental results, indicating that the adsorption occurred mainly on the biosorbent surface as a monolayer. The pseudo-second-order model was the most appropriate adsorption model in the present study, as it showed the best fit with the experimental data, suggesting the presence of chemisorption related interaction between dye molecules and functional groups present on the biosorbent. In addition, intraparticle diffusion analysis showed that the adsorption process occurred by several mechanisms and not by a single rate-controlling mechanism.

The thermodynamic investigations showed that the adsorption process was spontaneous, feasible and endothermic in nature. The negative Gibbs free energy values indicated spontaneous adsorption and the positive value of the enthalpy indicated that the adsorption efficiency would improve if the temperature is increased. The positive values of the entropy further suggested that the randomness of the adsorbent–solution interface increased during adsorption.

Experimental results were statistically analyzed to prove their reliability and significance using mean values, standard deviation, ANOVA and regression analysis. Low standard deviation values indicated good reproducibility of the results, while ANOVA showed that the operational parameters had statistically significant effects on the dye removal efficiency. The regression analysis results have shown that there are significant correlations between the adsorption variables and the adsorption performance, indicating that the experimental results are reliable.

In general, it can be stated that a biosorbent obtained from food waste is a promising material to use for the removal of dye pollutants from contaminated water, and can be used as an alternative to traditional adsorbents. In addition to providing efficient pollutant removal, the utilization of food waste biomass contributes to waste valorization, resource

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recovery, and circular economy initiatives. The results underscore the double impact that can be achieved by reducing food waste that is being wasted and enhancing wastewater treatment efficiency. Thus, food waste based biosorbents hold great potential for considering in the large scale sustainable water purification technologies and environmental remediation programs.

Future studies should be directed towards the modification of biosorbents, regeneration and reusability studies, pilot scale applications, and treatment of real industrial wastewater to further improve the feasibility of food waste-based biosorbents in commercial wastewater treatment applications.

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