Chemically Modified *Allium cepa* Peel as a Low-Cost Adsorbent for Methylene Blue Removal: Kinetics, Isotherms, and Thermodynamic Analysis

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***Abstract-** Synthetic dyes such as methylene blue (MB) are persistent water pollutants due to their structural stability, toxicity, and resistance to biodegradation. This study evaluates the adsorption efficiency of $KMnO_4/H_2SO_4$-modified* **Allium cepa** *peels for MB removal from aqueous solutions. The adsorbent was characterized using SEM, FTIR, and XRD to confirm morphological modification, functional-group participation, and its predominantly amorphous structure. Batch experiments revealed that adsorption was highly pH-dependent, with optimal removal (91.3%) at pH 3, and equilibrium was attained at 100 min. The Temkin isotherm provided the best fit to the equilibrium data (R^2 = 0.99), indicating heterogeneous adsorption with an adsorption energy constant BT of 0.22 J mol^{-1}. Kinetic analysis showed strong conformity to the pseudo-second-order model ($R^2 \approx 0.99$), with an adsorption capacity (Qe) closely matching experimental values, confirming chemisorption as the rate-determining step. Thermodynamic evaluation yielded negative $\Delta G°$ values (-4.42 to −26.59 kJ mol^{-1}), demonstrating spontaneous adsorption, while the negative enthalpy change ($\Delta H°$ = −44.53 kJ mol^{-1}) confirmed exothermic interactions. Overall, the chemically modified onion peel exhibited strong potential as a low-cost, sustainable biosorbent for dye-laden wastewater treatment and offers a promising alternative to commercial activated carbon.*

***Keywords:** Allium cepa peel, Adsorption, Methylene blue, Biosorption, Kinetics, Isotherms, Thermodynamics.*

I. INTRODUCTION

Water contamination from industrial effluents is a serious environmental problem, especially in poorer nations where regulations are frequently not enforced properly [1]. The United Nations estimates that almost 80% of wastewater worldwide is released back into the environment untreated, which exacerbates the problem of water pollution [7]. Among the various contaminants discharged into waterways, synthetic dyes from the food, paper, leather, and textile sectors are particularly problematic because of their toxicity, persistence, and lack of biodegradation. Common cationic dyes like methylene blue (MB) are widely used in a variety of sectors, such as leather, paper, and textiles. Its longevity and possible toxicity have prompted environmental concerns in light of its widespread use. Research has indicated that MB has the potential to seriously pollute the environment, disrupt photosynthesis, and pose health risks to both humans and animals [7]. Furthermore, MB is well-known for its deep colour and slow rate of degradation, both of which can seriously harm aquatic ecosystems if they are dumped into bodies of water [8]. Due to these features, effective removal methods must be developed in order to lessen the influence on the environment.

Coagulation, flocculation, membrane filtration, advanced oxidation processes, and adsorption are some of the techniques that have been investigated for the removal of dyes from wastewater [5]. Adsorption is among the most efficient methods because of its high removal rate and low cost, especially when utilising bio-based adsorbents, claim Krishnappa et al. [5]. Because of its ease of use, low cost, and efficiency, adsorption has become a popular method among them. Although activated carbon is frequently used as an adsorbent for the removal of dyes, its high cost of production and regeneration issues have prompted research into more affordable and environmentally friendly adsorbent alternatives. Due to their structural makeup, which offers functional groups for dye binding, agricultural waste materials—which are easily accessible and biodegradable—have become well-known as efficient adsorbents. One viable substitute for the adsorptive removal of colours from aqueous solutions is onion peel, a common agro-waste.

As a by-product of processing onions (Allium cepa), onion peel is made up of proteins, cellulose, hemicellulose, lignin, and flavonoids. It contains functional groups like hydroxyl (-OH), carboxyl (-COOH), and amine ($-NH_2$) that can interact with dye molecules. On the other hand, low active site availability and poor surface characteristics frequently limit the adsorption capability of raw onion peel. Its potential for wastewater treatment applications was highlighted by Agarry et al. [8], who showed that chemically modified onion skin (CMOS) could successfully remove heavy metals like cadmium (II) from aqueous solutions. Chemical changes are therefore used to improve its adsorptive capabilities. Various chemical treatments have been employed to enhance the surface area, porosity, and availability of functional groups in biomass-based adsorbents, such as acid, alkali, and functionalization with metal oxides. For example, Kwarteng et al. [1] showed that KOH-activated onion peel biochar had a high surface area of 1725.6 m^2/g, which resulted in a maximum MB adsorption capacity of 454.54 mgg^{-1}, with adsorption kinetics according to the pseudo-second-order model. Characterised by Raman, SEM, XRD, and BET analyses, the biochar exhibited graphene-like structures, which contributed to its high adsorption efficiency; isotherm studies showed strong conformity with the Langmuir model ($R^2 = 0.9958$); and the biochar showed significant regeneration capabilities over three consecutive cycles, maintaining significant adsorption efficiency. Similarly, studies by Saka et al. [2] examined the use of cold plasma-treated and formaldehyde-treated onion skins as biosorbents to remove MB from aqueous solutions. With sorption capacities of 250 mgg^{-1} for onion skins treated with cold plasma and 166.67 mg/g for onion skins treated with formaldehyde, the adsorption data aligned well with the Langmuir isotherm model. Sorption rates complied with pseudo-first-order kinetics. The pseudo-first-order and pseudo-second-order equations are two models that have been used extensively to study the adsorption kinetics of MB onto modified bio-waste adsorbents. Lagergren devised the pseudo-first-order model, which postulates that the number of available adsorption sites is exactly proportional to the rate of adsorption. Ho and McKay's pseudo-second-order model [6] makes the assumption that chemisorption is the rate-limiting phase. Furthermore, the Temkin, Freundlich, and Langmuir isotherm models are commonly used to analyse adsorption equilibrium. The Freundlich isotherm takes into consideration heterogeneous adsorption, the Temkin model takes into consideration the effects of indirect adsorbate-adsorbate interactions, and the Langmuir isotherm represents monolayer adsorption on a homogeneous surface. Optimising dye removal procedures and creating efficient treatment systems requires an understanding of these adsorption models.

Using chemically modified *Allium cepa* peel, this study examines the adsorptive removal of methylene blue dye from model effluent. Assessing the effectiveness of onion peel as a cheap biosorbent and determining how chemical alteration affects its adsorption capacity are the main goals. Adsorption kinetics and isotherm models are also used to forecast the interaction between the dye molecules and the adsorbent and to comprehend the adsorption mechanism. By providing an alternative to traditional and costly adsorbents, the study's findings add to the expanding corpus of research on bio-waste valorisation for wastewater treatment. This research advances the use of agricultural waste materials in environmental remediation, addressing the demand for sustainable and affordable water treatment techniques.

II. MATERIALS AND METHODS

2.1. Chemicals and Plant Materials Collection

The *Allium cepa* peels were collected from a popular market situated in Ikotun, latitude 6.549° N, and longitude 3.268° E, Lagos State, Nigeria. Analytical-grade reagents were used without purification. Methylene blue (MB) dye, sodium hydroxide (NaOH), Sulphuric Acid (H_2SO_4), and Potassium permanganate (VII) ($KMnO_4$) with a purity of ≥ 99.5% were obtained from Sigma-Aldrich, India. The solutions were prepared using deionized water. The MB dye concentrations were determined using a spectrometer at 635 nm [9]. A pH meter (HANNA HI.9828) was used to measure the pH values.

2.2. Chemical modification of oxidized *Allium cepa* peels

The collected *Allium cepa* peels were air-dried, manually cleaned, and rinsed with distilled water before being pulverized using an electric blender and subsequently a factory grinder to obtain a uniform texture, followed by sieving through a laboratory test sieve. A 12 g portion of the sieved material was weighed into a conical flask and chemically modified by adding 100 mL of 0.1 M $KMnO_4$ and 100 mL of H_2SO_4, after which the mixture was stirred on a magnetic stirrer at 60 °C for 24 h to enhance oxidative activation. The reaction mixture was cooled, thoroughly washed with distilled water to remove residual reagents, and oven-dried at 80 °C for 48 h. The dried material was then removed, sun-dried to eliminate remaining moisture, and crushed to obtain a fine adsorbent powder suitable for subsequent applications. This procedure aligns with established methods for chemical activation of agricultural biomass [17].

2.3. Characterization of chemically oxidized *Allium cepa* peels

The chemically modified *Allium cepa* peels were characterized using instrumental analyses. X-ray diffraction (XRD) images were obtained using Cu Kα radiation (k=0.152) with a Siemens D5005 diffractometer. Fourier Transform Infrared Spectroscopy (FT-IR) was conducted using FTIR with a Bruker Vertex-80 spectrometer to identify the chemical species coating the *Allium cepa* peels. The surface morphology was examined using Scanning Electron Microscope (SEM; JEOL/JEM-1230).

2.3. Batch Adsorption Studies Experiment

The adsorption behavior of MB was investigated using batch experiments employing a one-factor-at-a-time approach. Table 1 presents the experimental conditions evaluated across pH, contact time, adsorbent dosage, dye concentration, and temperature. In each experiment, 1 gL^{-1} of adsorbent was mixed with 1000 mL of anionic or cationic dye solutions (100–500 mgL^{-1}) in Erlenmeyer flasks and agitated at 150 rpm. Following equilibration, the mixtures were filtered through Whatman No. 1 filter paper, and the residual dye concentrations were determined using a UV–Vis spectrophotometer at λ = 635 nm for MB. Measurements were further validated using an Apel 101 spectrophotometer equipped with a 410 nm filter. The selected wavelengths align with those reported in previous studies [10]. The removal efficiency and sorbed dyes were calculated using Equations (1) and (2), with experiments performed in triplicate.

$$\% R = \frac{Co-Ce}{Co} \times 100 \qquad (1)$$

where Co is the initial concentration (mgL^{-1}) of the dye and Ce is the equilibrium concentration in solution (mgL^{-1}).

$$Q_e = \frac{(Co-Ce)}{m} \times V \qquad (2)$$

Where Q_e is the equilibrium adsorption capacity (mgg^{-1}), V is the volume of aqueous solution (L), and m is the dry mass of the *Allium cepa* peels (g). Through the adsorption experiments, the amount of the dye filtered and removed at multiple intervals was evaluated using the equation

$$Q_t = \frac{(Co-Ct)}{m} \times V \qquad (3)$$

where C_t represents the concentration in liquid (mgL^{-1}) at different periods "t."

Table 1. Batch experiments at different operating parameters.

Effect of parameters	Adsorbent mass (g)	Contact time (min)	pH	Concentration (mgL^{-1})	Temperature (°C)
Effect of pH	0.6	100	3-11	400	27
Effect of adsorbent dose	0.2-1.0	100	3	400	27
Effect of contact time	0.6	20-120	3	400	27
Effect of concentration	0.6	100	3	100-500	27
Effect of temperature	0.6	100	3	400	27-67

2.5. Kinetic, Isotherm Adsorption, and Thermodynamics Model

2.5.1. Kinetic Models

Various kinetic models, such as pseudo-first-order [11] and pseudo-second-order [12], were used in the present study. Equations (4) and (5) give the expressions of the linear forms of the pseudo-first-order model and the pseudo-second-order model, respectively:

$$\text{Log}\ (Q_e\text{-}Q_t) = \text{Log}\ (Q_e) - (\frac{K_1}{2.303})t \qquad (4)$$

$$\frac{t}{Qt} = \frac{1}{K_2 {Q_e}^2} + \frac{1}{Q_e} t \qquad (5)$$

Where k_1 (min^{-1}) represents the rate constant of the pseudo-first-order, k_2 ($g\ mg^{-1}.min^{-1}$) represents the rate constant of the pseudo-second-order model, Qt (mg/g) and Qe (mg/g) are the amounts of methylene blue and indigo carmine dyes adsorbed at time t (min) and equilibrium, respectively.

2.5.2. Adsorption Isotherm Models

The adsorption isotherm models, such as Langmuir [13, 14] and the Freundlich [15] models, were applied in the present study. Equations (6) and (7) give the expressions of the linear forms of the Langmuir and the Freundlich adsorption models, respectively:

$$\frac{Ce}{Qe} = \frac{1}{Qm\ Kl} Ce \ + \frac{1}{Qm} \qquad (6)$$

$$\text{Log}\ Q_e = \frac{1}{n} \log Ce + \text{Log}\ Kf \qquad (7)$$

Where C_e (mgL^{-1}) is the equilibrium methylene blue and indigo carmine concentration; Q_e (mg/g) is the amount of indigo carmine adsorbed at equilibrium, K_l (Lmg^{-1}) is the Langmuir constant, K_f (Lg^{-1}) is the Freundlich constant, Q_m (mg/g) is the Langmuir maximum amount adsorbed, and n is the Freundlich model exponent.

The essential features of the isotherm were expressed in terms of the dimensional constant, separation factor (R_L), defined by the relationship:

$$R_L = \frac{1}{1+KL\ Co} \qquad (8)$$

C_o is the initial concentration (mg/L), and K_L is the Langmuir equilibrium constant (L/mg).

The value of the separation factor (R_L) provides important information about the nature of adsorption. The value of (R_L) indicates the type of Langmuir isotherm to be irreversible $R_L = 0$), favorable ($0<R_L<1$), linear ($R_L =1$), or unfavorable ($R_L >1$). When $K_L>1$, sorption is favorable [16]

2.5.3. Thermodynamic Studies

The thermodynamic parameters depend on the reaction process, and they can help in the detailed study of the adsorption process mechanism. This experimental study considered different temperature values. The enthalpy (ΔH°) in $kJ\ mol^{-1}$, entropy (ΔS°) in $kJ\ mol^{-1}\ K^{-1,}$ and standard free energy (ΔG°) in $kJ\ mol^{-1}$, for sorption of the dye onto adsorbents (*Allium cepa* peels) were determined with Eqs. (10-12).

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \qquad (9)$$

$$\ln Kc = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \qquad (10)$$

$$\Delta G^\circ = -RT \ln K_c \qquad (11)$$

where the universal gas constant (R) is in J mol-1 K^{-1}, Temperature (T) in kelvin (K), and K_c is the adsorption equilibrium constant.

III. RESULTS AND DISCUSSION

3.1 Characterisation of Adsorbent

3.1.1 Scanning Electron Microscope (SEM) Analysis

The SEM micrographs (Fig. 1) reveal clear differences in the morphology of the modified *Allium cepa* peels before and after adsorption. Before adsorption (Fig. 1a–b), the surface exhibited a heterogeneous, rough texture with visible pores and irregular cavities. Such structural roughness is characteristic of chemically activated lignocellulosic biomass and provides abundant active binding sites for pollutant uptake [18]. After adsorption (Fig. 1c–d), the pores appeared noticeably reduced or entirely obscured, suggesting that the dye molecules had occupied the available surface sites. The smoother surface seen post-adsorption further confirms effective attachment of methylene blue molecules onto the oxidized onion peel matrix. Similar morphological transformations have been reported for $KMnO_4$ and acid-modified agro-waste adsorbents, supporting the occurrence of surface saturation during adsorption [19].

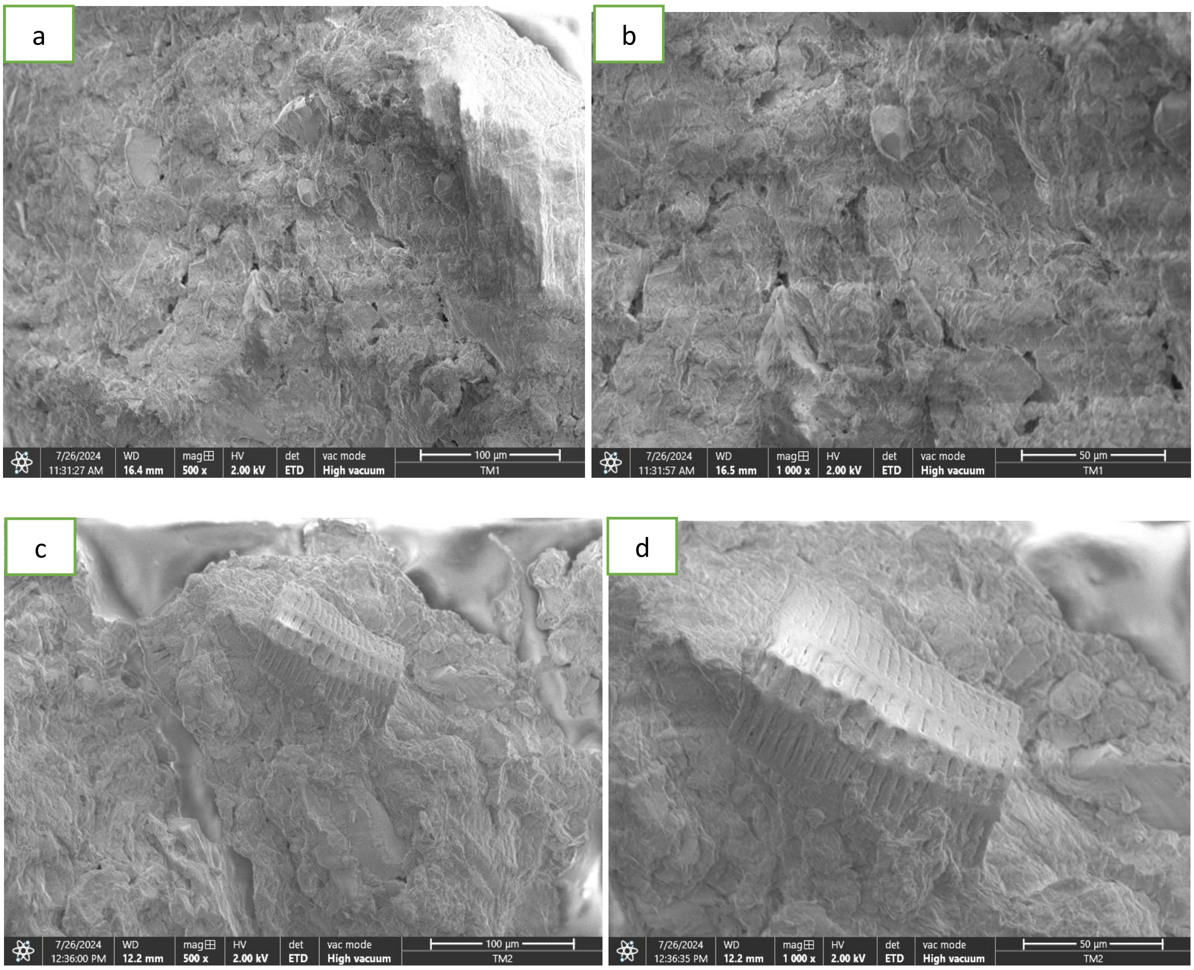

Figure 1. SEM images of modified Allium cepa peels before (a–b) and after adsorption (c–d)

3.1.2 Fourier Transform Spectroscopy (FTIR)

The FTIR spectra (Fig. 2; Table 2) revealed characteristic functional groups responsible for interaction with methylene blue. Broad absorption around 3280–3288 cm^{-1} corresponds to –OH stretching vibrations of alcohols, indicating the presence of hydroxyl groups capable of hydrogen bonding. Peaks observed at 2922–2926 cm^{-1} are associated with C–H stretching of alkanes, while the band near 1058 cm^{-1} confirms C–O stretching of aliphatic ethers. A distinct peak at 1595 cm^{-1} represents C=C stretching in alkenes. After adsorption, slight shifts and reductions in peak intensity were observed, signifying the involvement of these functional groups in dye–adsorbent interactions. This indicates that adsorption was driven mainly by hydrogen bonding, π–π interactions, and possibly electrostatic attraction, consistent with previous studies on modified onion peel and similar biomass adsorbents [20].

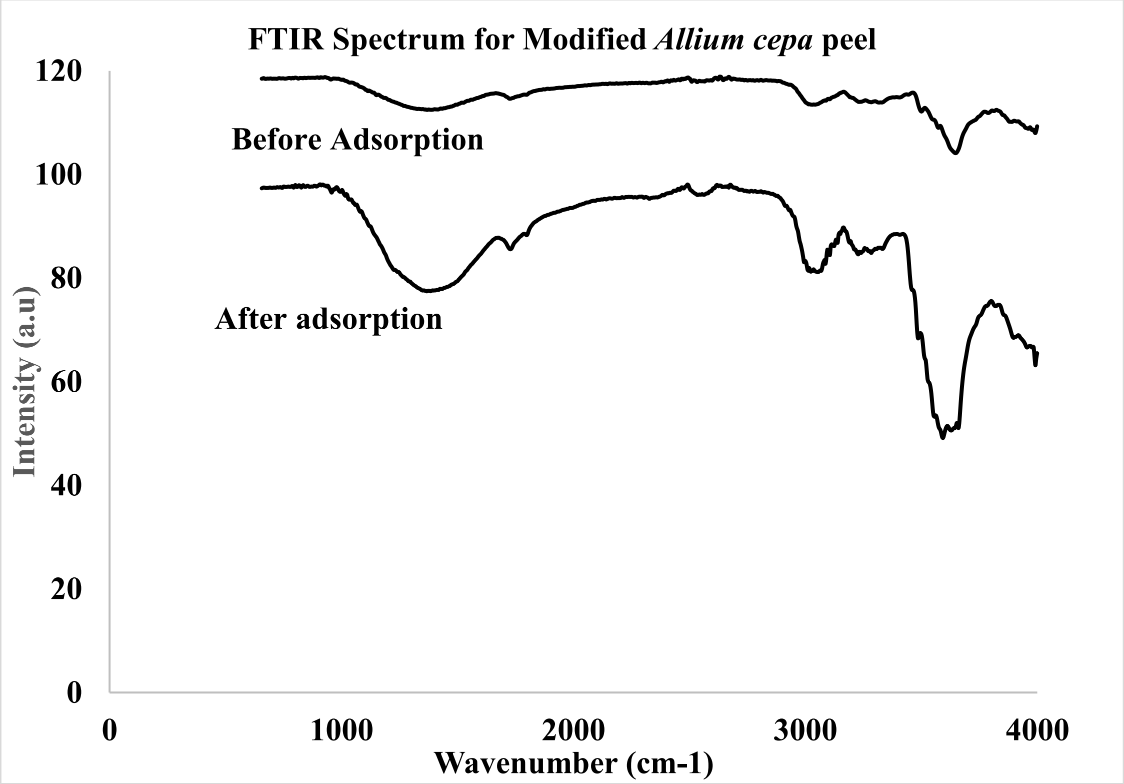

Figure 2. FTIR spectra of *Allium cepa* peel adsorbent before and after MB adsorption.

Table 2: Adsorption band of modified *Allium cepa* peels before and after adsorption

ABSORPTION BAND cm^{-1}	SELECTED GROUPS	FUNCTIONAL GROUP
3288.8	-0-H stretch	0-H for Alcohol
2922.2	-C-H stretch	C-H for Alkane
1058.6	-C-0 stretch	C-0 for Aliphatic Ether
1595.3	-C=C stretch	C=C Alkene
3280.1	-O-H stretch	O-H for Alcohol
2926,0	-C-H stretch	C-H for Alkane

3.3.1. X-ray diffraction (XRD) Analysis

The XRD pattern (Fig. 3) shows a predominantly amorphous structure, typical of lignocellulosic materials. The broad diffraction peaks indicate limited crystallinity, which is advantageous for adsorption because amorphous regions provide more flexible and accessible active sites. The absence of strong crystalline peaks suggests that chemical oxidation did not induce long-range structural ordering. Similar amorphous characteristics have been reported for modified onion peel–based adsorbents and are associated with improved dye uptake due to enhanced porosity and surface functionalisation [21].

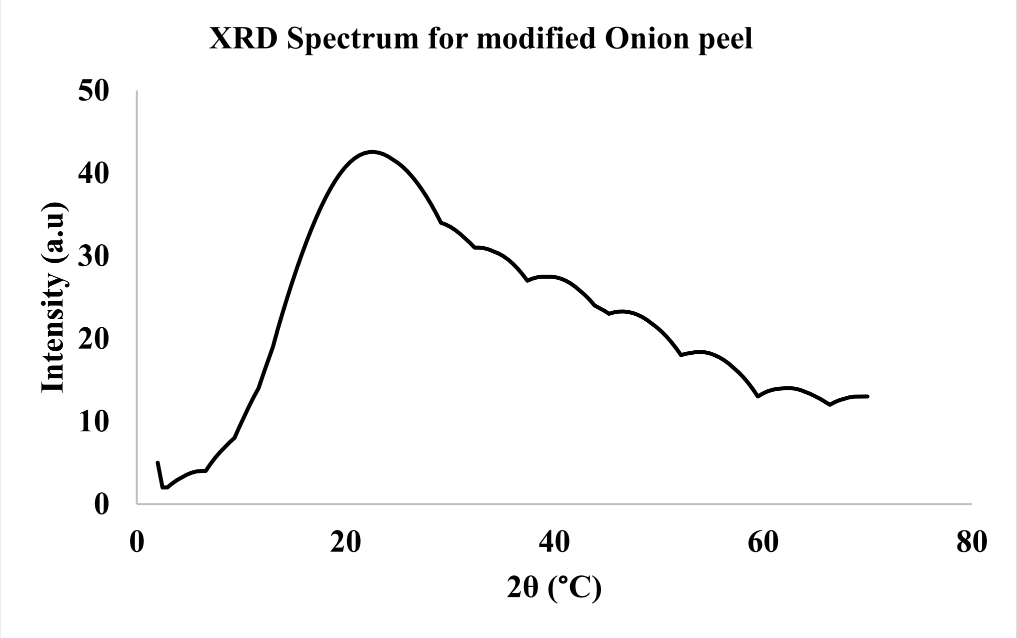

Figure 3. XRD diffraction pattern of chemically oxidized *Allium cepa* peel showing its amorphous structure.

3.2 Effect of Operating Parameters

3.2.1 Effect of Contact Time

The influence of contact time on methylene blue removal is presented in Fig. 4. A rapid increase in adsorption was observed within the first 20–60 minutes, attributable to the abundance of available active sites on the fresh adsorbent surface. Equilibrium was achieved at 100 minutes, where maximum removal reached 87.5%. Beyond this point, the rate of adsorption slowed due to saturation of the active sites. This behaviour aligns with the typical two-phase adsorption process widely reported for biosorbents: an initial fast external surface binding stage followed by slower intraparticle diffusion [22].

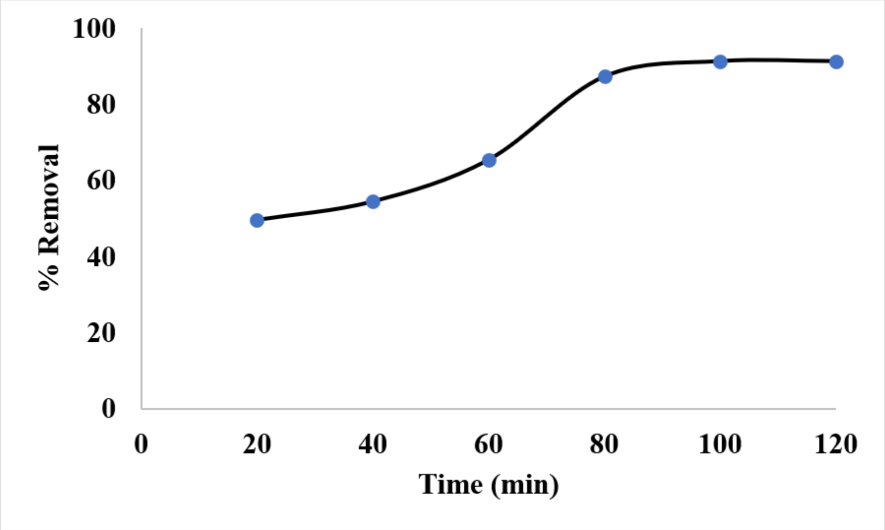

Figure 4. Effect of contact time on MB adsorption

3.2.2 Effect of Adsorbent Dosage

As shown in Fig. 5, increasing the adsorbent mass from 0.2 to 1.0 g led to a progressive rise in the percentage removal, with optimal performance at 0.6 g (89.0%). This trend is expected, as higher dosages provide a greater number of binding sites and a larger surface area for adsorption. However, beyond a certain dosage, the removal efficiency may plateau due to particle aggregation and site overlap, reducing effective surface exposure [23]. A similar pattern has been reported for onion-based biosorbents used in dye removal [24].

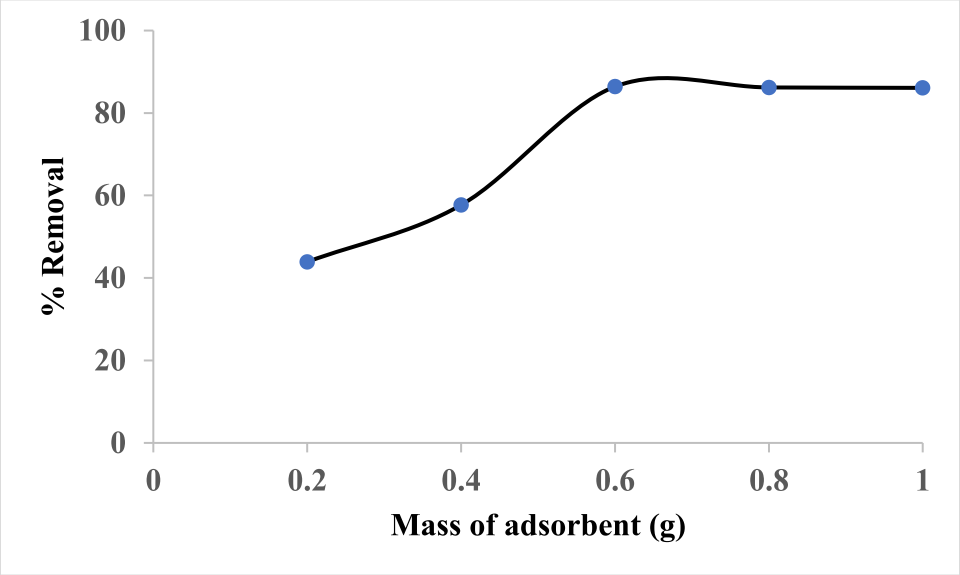

Figure 5. Effect of adsorbent dosage on MB removal efficiency

3.2.3 Effect of Initial Concentration

The effect of initial dye concentration (100–500 mg/L) is illustrated in Fig. 6. Maximum adsorption occurred at 400 mg/L, after which removal efficiency declined slightly. At low concentrations, the ratio of available binding sites to dye molecules is high, allowing efficient uptake. As concentration increases, binding sites become saturated, and diffusion resistance within the adsorbent pores begins to limit further adsorption. This behaviour is consistent with monolayer adsorption mechanisms and has been documented in several dye–biosorbent systems [25].

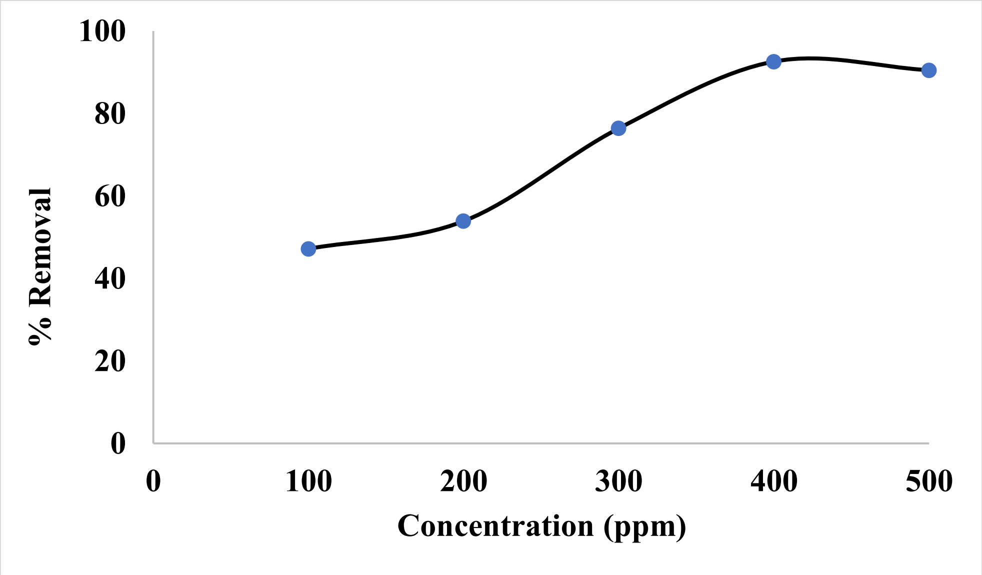

Figure 6. Effect of initial dye concentration on MB adsorption.

3.2.4 Effect of Temperature

Figure 7 shows a decrease in methylene blue uptake as temperature increased from 27 °C to 67 °C, with maximum removal (79.5%) at 27 °C. The decline at higher temperatures suggests that adsorption is exothermic, meaning elevated thermal energy weakens the interactions between dye molecules and active sites. Additionally, increased temperature may destabilise functional groups on the adsorbent surface, reducing available binding sites. Exothermic adsorption behaviour has similarly been reported for modified agro-waste materials [26].

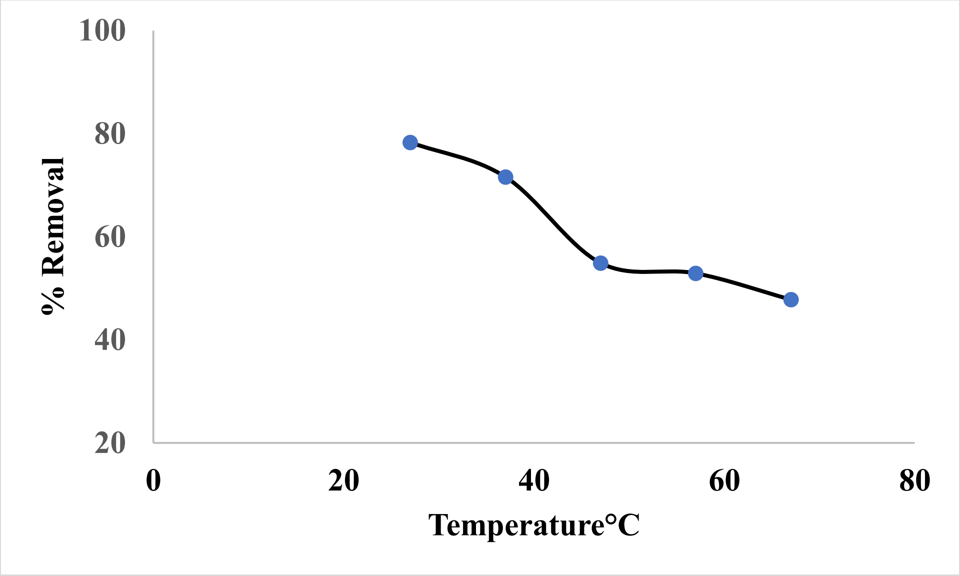

Figure 7. Effect of temperature on MB removal efficiency.

3.2.5 Effect of pH

As depicted in Fig. 8, the highest removal efficiency (91.3%) was obtained at pH 3. Under acidic conditions, protonation of the adsorbent's surface functional groups creates a positively charged surface, facilitating strong electrostatic attraction with the negatively charged components of the dye molecule. At higher pH values, surface deprotonation reduces electrostatic interactions, leading to decreased adsorption. The observed trend agrees with earlier reports showing enhanced adsorption of cationic and anionic dyes under acidic conditions, depending on the adsorbent's protonation behaviour [27]

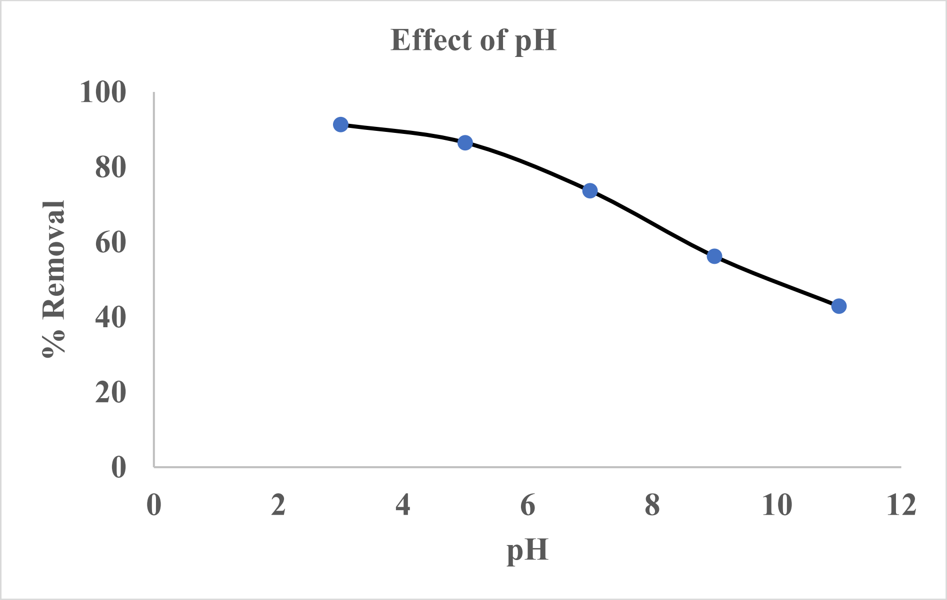

Figure 8. Effect of pH on MB removal efficiency.

3.3.1 Adsorption Isotherm Model

The equilibrium data were fitted to Langmuir, Freundlich, and Temkin models (Figs. 9–11; Table 3). The Temkin model produced the highest correlation coefficient ($R^2 = 0.99$), indicating the best description of the adsorption process. This implies that the heat of adsorption decreases linearly with surface coverage, suggesting a heterogeneous surface with variable binding energies. The Freundlich model ($R^2 = 0.96$) also showed good agreement, further supporting the heterogeneous nature of the adsorbent surface. The Langmuir model demonstrated poor fit ($R^2 = 0.02$), indicating that monolayer adsorption on a uniform surface is not the dominant mechanism.

The Temkin dominance corresponds with literature on chemically modified onion peels, which often exhibit heterogeneous adsorption due to diverse oxygen-containing functional groups [28].

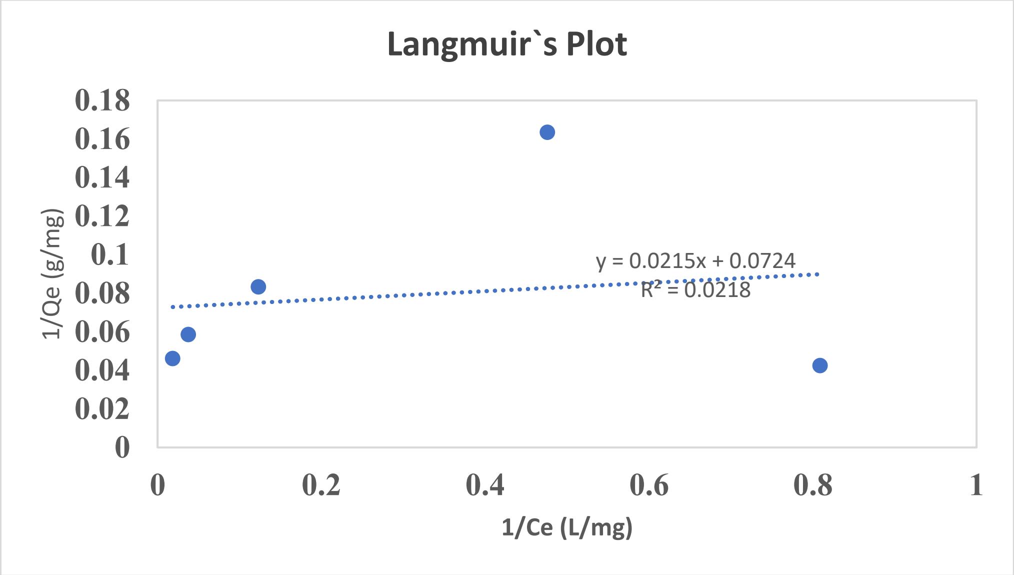

Figure 9. Langmuir isotherm plot for MB adsorption.

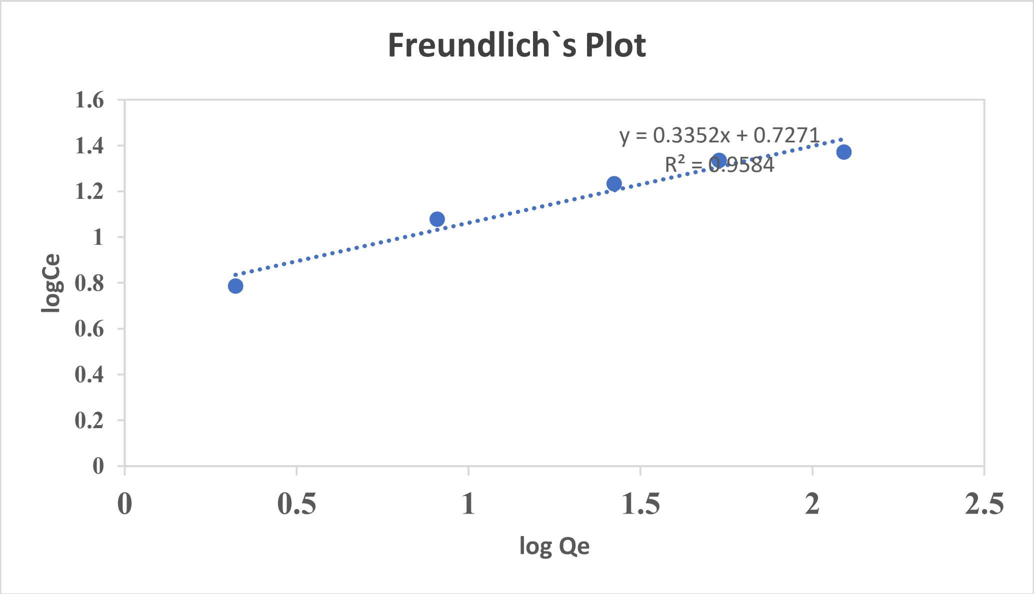

Figure 10. Freundlich isotherm plot for MB adsorption.

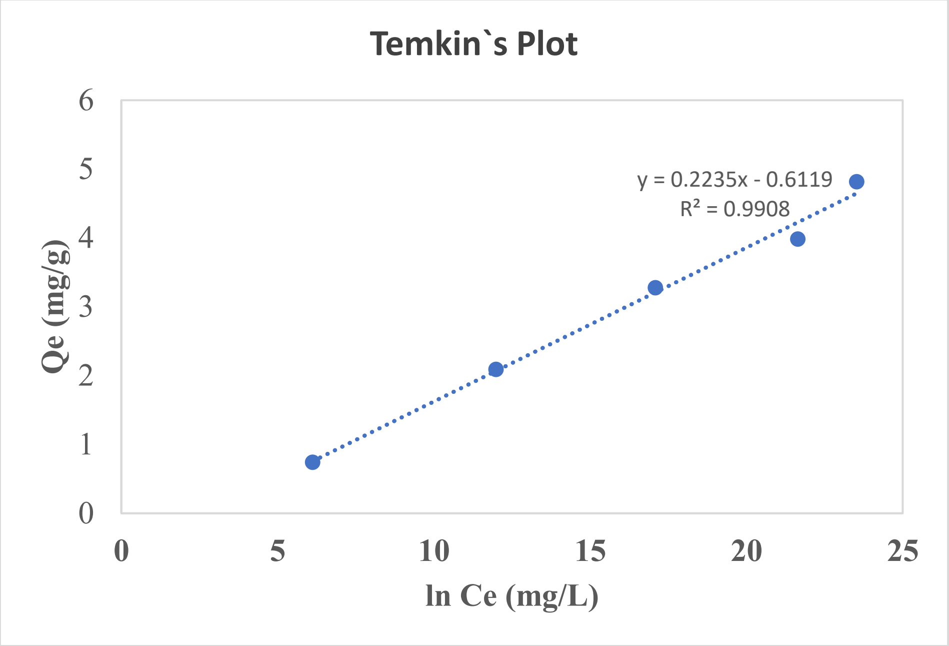

Figure 11. Temkin isotherm plot for MB adsorption.

Table 3: Adsorption Isotherms Variable/Constant of MB

ISOTHERMS	MBD Values
LANGMUIR	
Q_{max} (mgg^{-1})	13.81
K_L (Lg^{-1})	0.07
R_L	0.03
R^2	0.02
FREUNDLICH	
n	2.98
K_F (Lg^{-1})	5.33
R^2	0.96
TEMKIN	
B_T $Jmol^{-1}$	0.22
K_T	0.06
R^2	0.99

3.4 Adsorption Kinetics Studies

Kinetic evaluation using pseudo-first-order and pseudo-second-order models (Figs. 12–13) revealed that the pseudo-second-order model provided a superior fit, showing an R^2 value close to unity. This indicates that chemisorption—likely involving electron sharing, electrostatic attraction, and surface functional group interaction—governs the adsorption process. The dominance of the pseudo-second-order model is consistent with earlier findings on modified lignocellulosic adsorbents used for dye removal [29].

3.4.1 Pseudo-First Order Model

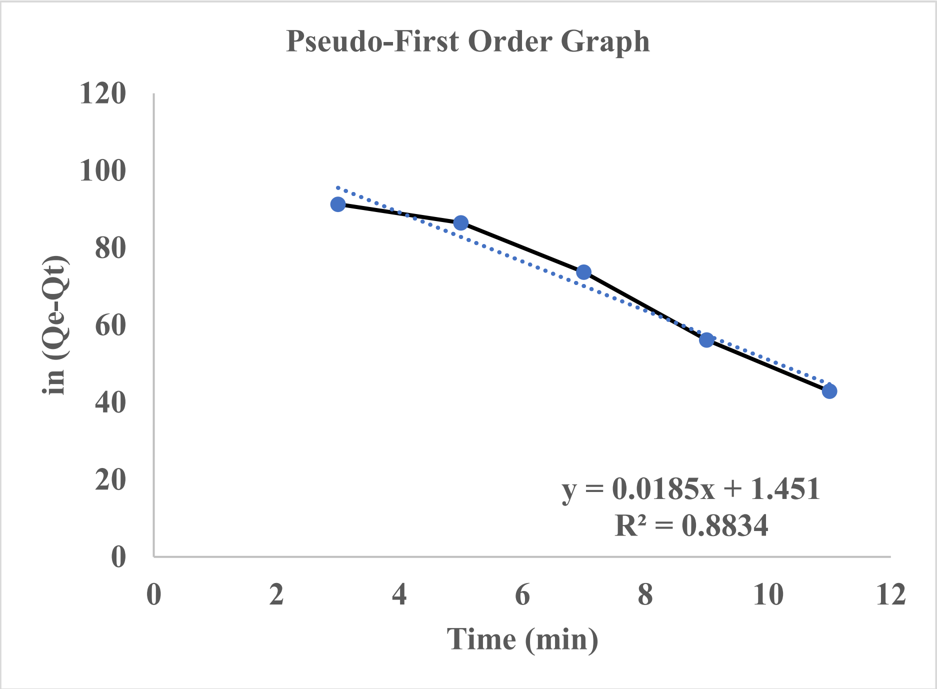

Figure 12. Pseudo-first-order kinetic plot.

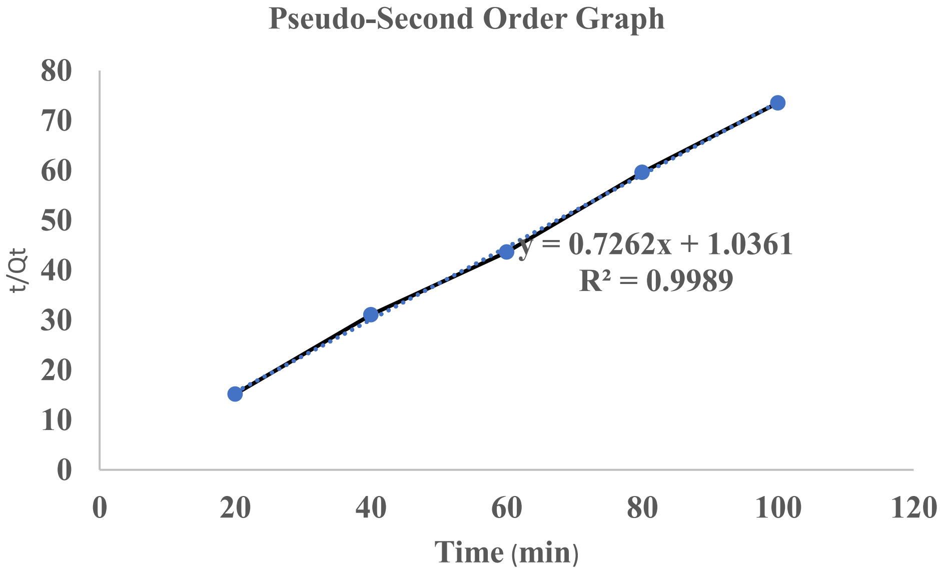

Figure 12. Pseudo-second-order kinetic plot.

3.5. Thermodynamics Studies

The thermodynamic parameters (Fig. 14; Table 4) provide insight into the feasibility and nature of the adsorption process. The negative values of ΔG° at all temperatures examined confirm that the adsorption of methylene blue onto modified onion peel is spontaneous. The magnitude of ΔG° became more negative with increasing temperature, indicating enhanced spontaneity and stronger adsorbent–adsorbate interaction at lower temperatures. The negative ΔH° value confirms that the process is exothermic, consistent with the temperature effect results. The negative ΔS° suggests decreased randomness at the solid–solution interface during dye binding, a trend common in exothermic biosorption systems [30].

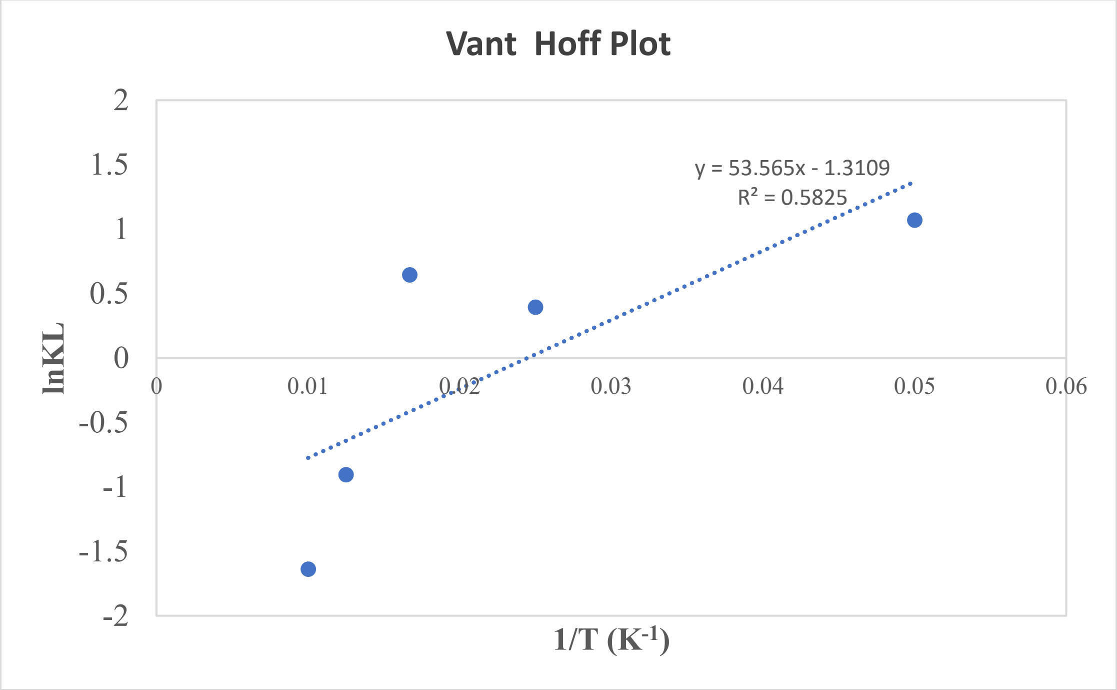

Figure 14. Van't Hoff plot for thermodynamic evaluation of MB adsorption.

Table 4: Thermodynamic Parameters Values

TEMP (K)	$\ln K_L$	ΔG° (kJ/mol)	ΔH° (kJ/mol0	ΔS°(J/molK)
300	1.0694	-442.042	- 445.3394	-10.9888
310	0.9325			
320	0.6450	1785.7225		
330	-0.9060	-2658.9669		
340				

IV. CONCLUSION

Chemically modified *Allium cepa* peels demonstrated strong potential as an efficient, low-cost adsorbent for methylene blue removal. The $KMnO_4/H_2SO_4$ treatment significantly enhanced surface activity, enabling high adsorption capacities under optimized conditions. Adsorption followed pseudo-second-order kinetics and conformed best to the Temkin isotherm, indicating heterogeneous chemisorption. Thermodynamic analysis confirmed that the process was spontaneous and exothermic. Owing to its abundance, biodegradability, and performance, modified onion peel is a promising sustainable material for wastewater treatment applications.

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