



Green Route Synthesis and Adsorption Studies of Copper-Benzimidazole Coordination Polymer for Removal of Methyl Orange from Water

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Abstract

An imidazole-based coordination polymer formulated as $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ (**1**) (Benz = benzimidazole) was prepared via a solvent-free method of grinding. It was compared with another, which was prepared through a solution-based method using spectroscopic and powder X-ray diffraction techniques. The results obtained showed that the two materials are similar. Adsorption of methyl orange (MO) from solution using (**1**) was investigated using batch adsorption technique. The results revealed that the adsorption process followed the pseudo-second-order kinetics, and the equilibrium data fitted best into the Langmuir isotherm. The maximum adsorption capacity of (**1**) was found to be 15.01 mg/g. The prepared materials could therefore be used as an adsorbent in the treatment of wastewater containing dyes.

Keywords Coordination polymer · Adsorption · Methyl orange · Spectroscopic · Solvent-free

1 Introduction

Research involving porous coordination polymers has been on the increase globally within different research groups. This is because of their potential applications in biomedicine, catalysis, gas purification, gas separation, and gas storage, which result from their extremely high adsorption capacity, pore size distribution, surface area, and void volume [1–3]. Another advantage is that they can be synthesized with high purity and crystallinity through synthetic routes that are relatively easy and inexpensive [2, 4–7].

Generally, two major methods are used in the synthesis of coordination compounds, namely the conventional method which uses solvents, and the unconventional method which is solvent-free [8, 9]. Solvent-free synthesis of coordination compounds has been used to produce many interesting coordination complexes. It has also received a lot of attention over the years because the method is green due to the absence of solvents in the reaction system, thus reducing environmental pollution [10].

Wastewaters from industries such as textile, leather, paper, printing, and plastic industries contain dye materials, which have a negative effect on water quality and should be removed from the wastewater before they are discharged into water bodies [11–14]. The complex chemistry of dyes makes their removal very difficult using the available or known municipal waste treatment options [13]. Therefore, the need to explore other options using materials that will be more effective for the removal of dyes from wastewater.

The bio-accumulation of dyes in environmental media through discharge from industries carrying out processes like paper and textile/fabric manufacture has been indicated as a pointer towards pollution of environmental media particularly water resources [6–8]. The majority of industries that use dyes as part of their manufacturing processes tend

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to eliminate their wastewater by methods through which the dyes find their way into the environment thereby causing detrimental effects to mankind and the ecosystem at large [7]. These dyes have been indicated as a major class of organic pollutants that have carcinogenic and mutagenic potentials. Due to their widespread usage, dyes are usually encountered as components of effluents/industrial wastewater and find their way to the environment during wastewater discharge [9–12].

A $[\text{Cu}(\text{INA})_2]$ coordination complex has been prepared in the past via a solvent-free method, and it was used as a potential adsorbent for the removal of anthracene from solution [15]. The compound proved to have excellent adsorption results with a high adsorption capacity of 22.73 mg/g for the anthracene. It was also investigated for fluorescein dye removal and the results showed high fluorescein dye removal via a physical adsorption process [16].

Adsorptive dye separation from an aqueous medium is reported to be an effective and low-cost method of removing dyes [6]. Activated carbon [17, 18] and coordination polymers [18] have been presented as efficient materials for dye adsorption among other adsorbents. Recently, there has been increased research around the application of coordination polymers as adsorbents for dyes. These materials have also found application as heterogeneous catalysts [5, 18] and photocatalysts [18] for pollutant remediation [18].

Previous research by our group presented Cu^{2+} coordination compounds as catalysts and adsorbents [1, 4, 18] for removal of various environmental pollutants. However, an extensive literature search carried out by our team revealed that the available information on the solvent-free synthesis of copper-benzimidazole complexes and their application as adsorbent for the removal of methyl orange dye from aqueous solution is inadequate. Consequently, we report in this study the solvent-free synthesis of copper-benzimidazole coordination compound and its application for the removal of methyl orange dye from an aqueous solution.

2 Materials and Methods

2.1 Materials

Analytical grade reagents, $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ (99%) and benzimidazole (99%) were purchased from Sigma Aldrich, Co., Germany and were used as received without further purification. Compound (1) was prepared with modifications made to methods previously reported in literature [15, 16]. The Melting point of (1) was measured on a Gallenkamp melting point apparatus. Fourier-transform infrared (FTIR) spectra were obtained on a SHIMADU scientific model FTIR 8400 s Spectrophotometer. UV–Visible spectra were obtained on a CARY 5000 UV–Visible NIR spectrometer.

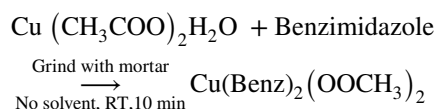
Powder X-ray diffraction (PXRD) patterns were obtained using a Bruker D8 Advance X-ray diffractometer (Bruker D8, Bruker, UK) with a $\text{CuK}\alpha$ -radiation (wavelength of monochromatic radiation, $\lambda = 1.5406 \text{ \AA}$) operating at 30 kV and 40 mA. Brunauer–Emmett–Teller surface area analysis was conducted on a NOVA 4200e BET instrument. Elemental analysis (CHN) was performed using Elemental Analyzer with PC base data system and a Sartorius Ultra Micro Balance- Model 1108 (Carlo-Erba).

2.2 Methods

2.2.1 Solvent-Free Preparation of $[\text{Cu}(\text{Benz})_2(\text{OOCCH}_3)_2]$ (1)

Benzimidazole (0.236 g, 2 mmol) and $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ (0.118 g, 1 mmol) were ground together using a mortar and pestle for 10 min. A green powdered compound was obtained. It was thereafter exposed to air for 24 h, and a change in colour from green to brick red was observed.

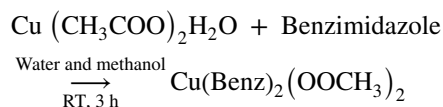
The equation for the reaction is as shown below:



2.2.2 Solvent-Based Synthesis of the $[\text{Cu}(\text{Benz})_2(\text{OOCCH}_3)_2]$ (1)

Benzimidazole (0.236 g, 2 mmol) and $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ (0.118 g, 1 mmol) were separately dissolved in methanol and distilled water respectively. They were stirred at room temperature for 3 h to obtain a brick-red powdered compound, which was filtered, washed, and dried.

The equation for the reaction is as shown below:



The synthesized (1) was oven-dried for 12 h at 150 °C and thereafter stored in a desiccator.

2.2.3 Calibration Curve

100 mg/L methyl orange (MO) stock solution was prepared by dissolving 0.1 g of MO in 1000 ml of deionized water. Lower concentrations (1–8 mg/L) were obtained by serial dilution of the stock. Absorbance of the prepared standard solutions was measured at the wavelength where the solutions absorbed the maximum fraction of light (λ_{max} of 464 nm) using a SHIMADZU UV-1650pc Ultraviolet–Visible (UV–Vis)

spectrophotometer and it was used to plot the calibration curve in Fig. 1.

2.2.4 Batch Adsorption Studies

Adsorption of MO dye onto (**1**) was performed using procedures that have been reported previously [16–20]. Briefly, 0.01 g each of (**1**) was added to the prepared solutions and the mixtures were shaken for 2 h. The adsorbent was filtered off after equilibration and the filtrates were immediately analyzed using a UV–Visible spectrophotometer. The effects of concentration, temperature, time, pH, and adsorbent dosage on the dye adsorption were studied to obtain the equilibrium quantity (q_e) of the dye adsorbed using Eq. 1 as reported previously [16–20].

$$q_e = \left(\frac{C_o - C_e}{m} \right) v \quad (1)$$

where C_o and C_e are initial and final concentrations of the adsorbates in mg/L respectively, v = volume of the solution used (ml) and m = mass (g) of the adsorbents.

The effect of time was investigated between 20 and 90 min, while the effect of temperature was studied between

30 and 50 °C. The effect of pH was investigated by varying the pH between 2 and 10 using 0.1 M HCl or 0.1 M NaOH, while the effect of adsorbent dosage was studied with 0.01–0.05 g of (**1**). The effect of the adsorbate concentration was studied using 1–8 mg/L of MO.

3 Results and Discussion

The physicochemical properties of the synthesized $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ (**1**) are presented in Table 1. The melting points of (**1**) prepared by solvent-free and solvent-based methods were higher when compared with the precursor, benzimidazole. CHN analysis of the two compounds gave the percentage of carbon, hydrogen, and nitrogen that are present in them. The compounds were found to be insoluble in ethanol, but soluble in DMF. The percentage yield for the solvent-free (mechanochemically synthesized) product was higher than the solvent-based. In general, the elemental (CHN) analysis results for the solvent-based and solvent-free synthesized compounds were identical indicating that the two methods produced similar compounds.

Fig. 1 Calibration curve of methyl orange (MO)

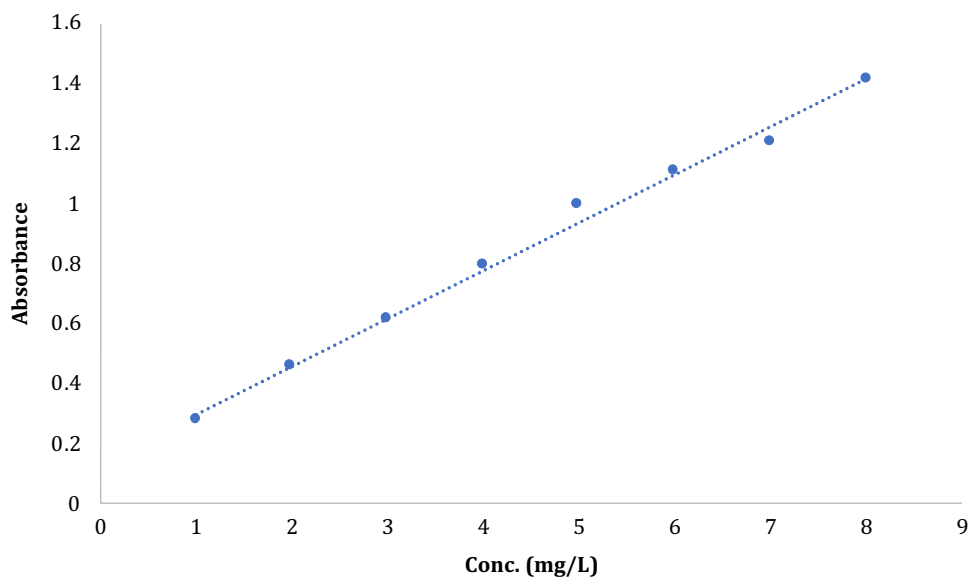


Table 1 Physicochemical properties of $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ (**1**)

Ligands/CPs	Appearance	Yield (%)	Molecular weight (g/mol)	Melting point (°C)	Elemental analysis % Found (% calculated)		
					C	H	N
Benzimidazole	White		118	171.4	–	–	–
$[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ solvent-free	Brick red powder	85	391.05	361	49.04 (54.82)	4.12 (3.34)	14.30 (18.24)
$[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ solvent-based	Brick red powder	70	391.05	360	49.04 (48.65)	4.12 (3.22)	14.30 (15.71)

3.1 FTIR Analysis

FTIR spectra of (**1**) prepared using solvent-based and solvent-free methods showed that the $\nu(\text{C}=\text{N})$ absorption band at 1620 cm^{-1} found in benzimidazole ligand shifted to 1604 cm^{-1} in (**1**). The $\nu(\text{M}-\text{N})$ was seen at 476 cm^{-1} in the solvent-based and solvent-free synthesized (**1**) as previously reported [20, 21]. Other FTIR absorption bands and frequencies of (**1**) are shown in Table 2, while the spectra for the ligand and synthesized compounds are shown in Fig. 2. The similarity in spectra for the solvent-free and solvent-based synthesized compounds is another indication that they are similar.

FTIR spectra of (**1**) obtained after the adsorption studies (Fig. 3b) showed peaks at 2912 and 2832 cm^{-1} , which could be attributed to $-\text{CH}_3$ stretching from the MO dye [22, 23]. These peaks were absent in the FTIR spectra of (**1**) before the adsorption of MO dye (Fig. 3a).

3.2 Scanning Electron Microscopy (SEM) Analysis

Figure 4 shows the SEM images of (**1**) before and after the adsorption study. Before the adsorption studies (Fig. 4a),

the rough surface of the compound with protrusions that extended into the matrix was observed. However, after the adsorption of MO (Fig. 4b), the image became blurred with no visible protrusions, which could be due to the presence of the dye molecules in the pores of the compound [3, 5, 24].

3.3 Thermogravimetric (TGA), and Derivative Thermogravimetric (DTG) Analysis

The TGA analysis of (**1**) shown in Fig. 5 revealed one stage decomposition which started at about $285\text{ }^{\circ}\text{C}$ and ended at $390\text{ }^{\circ}\text{C}$ accounting for the complete decomposition of the compound (i.e., weight loss of 100%). The DTG curve correlates with the one-step decomposition pattern obtained in the TGA curve with decomposition commencing at about $125\text{ }^{\circ}\text{C}$ and ending at around $295\text{ }^{\circ}\text{C}$ in the DTG curve.

Table 2 FTIR spectroscopy analysis result of $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ (**1**)

Ligands/Mofs	$\nu(\text{C}=\text{N})\text{ cm}^{-1}$	$\nu(\text{C}=\text{C})\text{ cm}^{-1}$	$\nu(\text{C}-\text{N})\text{ cm}^{-1}$	$\nu(\text{M}-\text{N})\text{ cm}^{-1}$	$\nu(\text{N}-\text{H})\text{ cm}^{-1}$
Benzimidazole	1620.26	1410.01	1246.06		3450.77
$[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ solvent-based	1604.83	1467.86	1240.27	476.43	
$[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ solvent-free	1604.83	1467.85	1240.27	476.43	

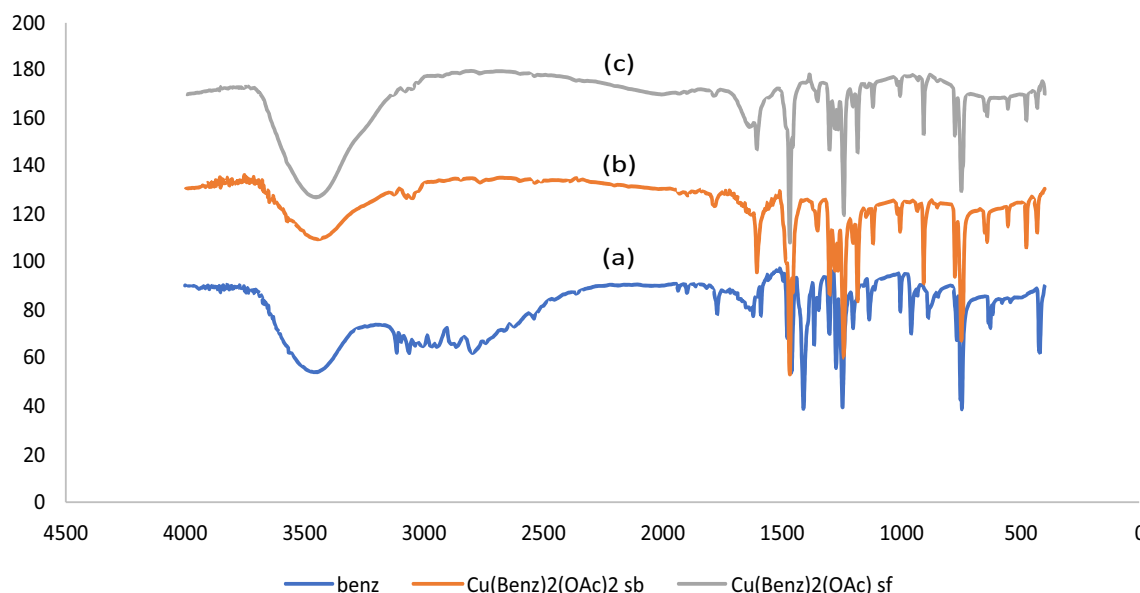


Fig. 2 FTIR spectroscopy result **a** Benzimidazole; **b** $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ **1** *sb* solvent-based, **(c)** $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ **1** *sf* solvent-free

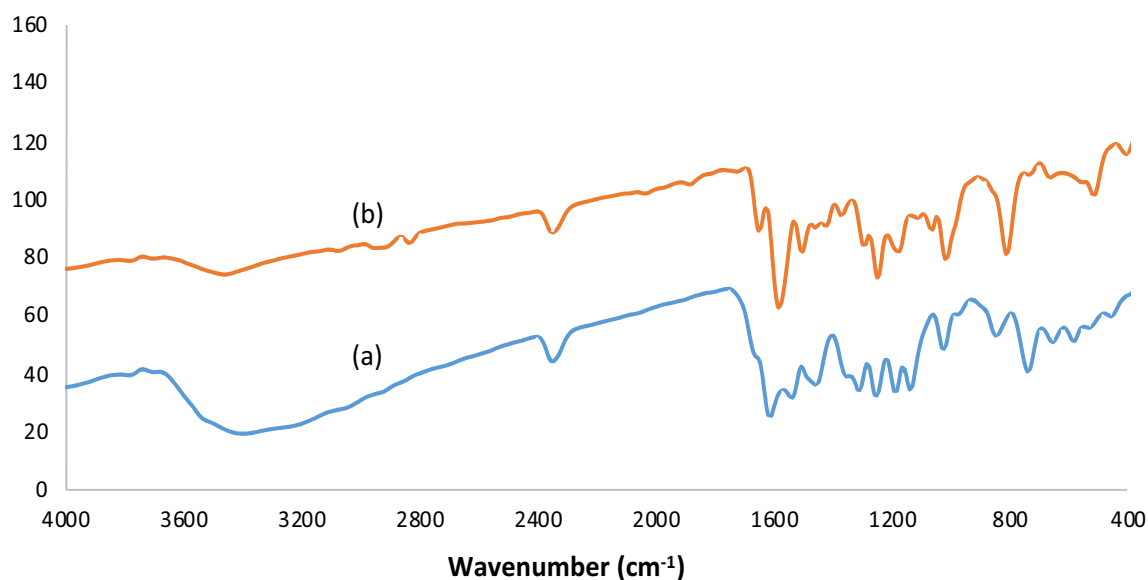


Fig. 3 FTIR spectra of $[\text{Cu}(\text{Benz})_2(\text{OOCH}_3)_2]$ **1** (a) before adsorption; (b) after adsorption

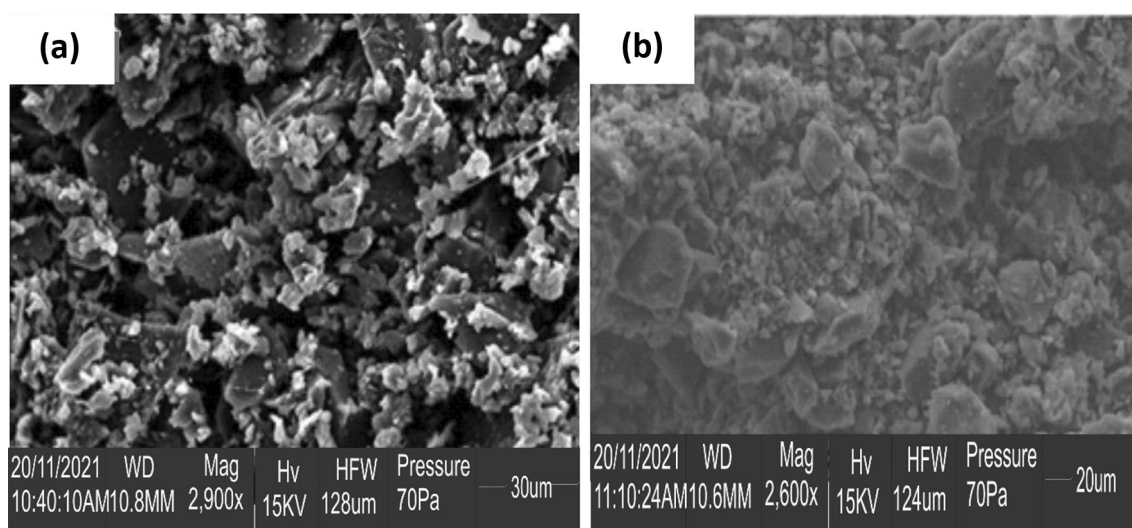


Fig. 4 SEM images of (**1**), **a** before adsorption, **b** after adsorption

3.4 BET Analysis

The results of the BET measurements obtained from the N_2 adsorption experiments are shown in Fig. 6. BET-specific surface area value of $260.61 \text{ m}^2 \text{ g}^{-1}$ was obtained before the adsorption studies, while $240.65 \text{ m}^2 \text{ g}^{-1}$ was obtained after adsorption. The lower value of BET surface area obtained after adsorption could be ascribed to the presence of the MO molecules on (**1**), which led to the blockage of the pores [3].

3.5 Powder x-Ray Diffraction Analysis

PXRD patterns of (**1**) were obtained before and after the adsorption study, and are presented in Fig. 7. The patterns obtained showed a prominent peak at 2θ value of about 48.2568° in (**1**) before adsorption, which also appeared in the PXRD pattern of (**1**) after adsorption of MO. Peaks observed at 2θ values of 51.1145° , 57.5416° and 62.1098° in (**1**) before adsorption were observed at 2θ values of about 50.9018° , 52.5819° and 60.1076° after adsorption, which

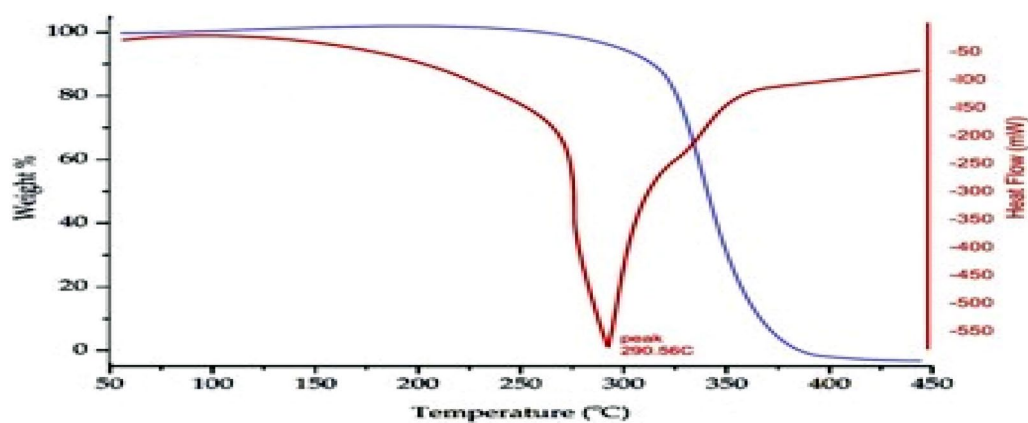


Fig. 5 TGA and DTG analysis of (1)

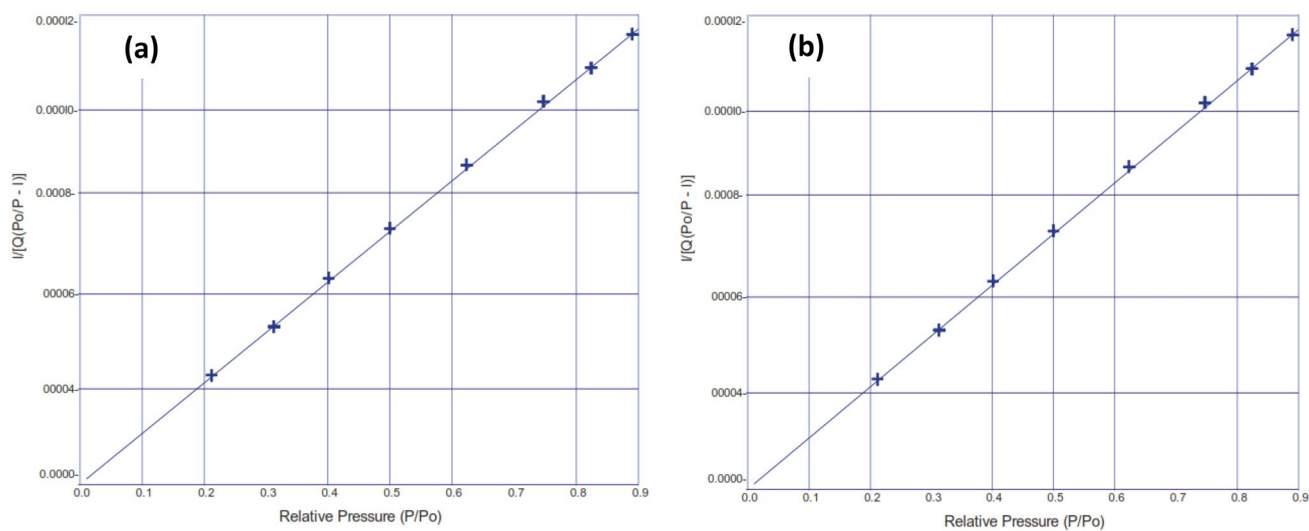
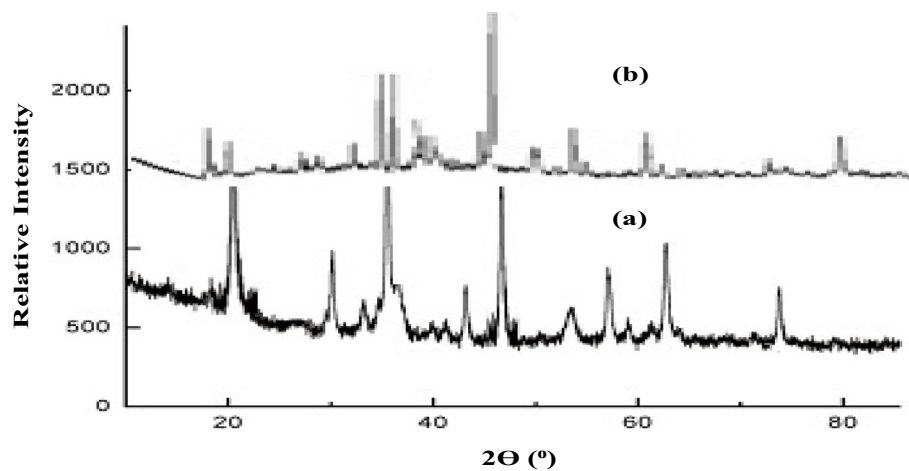


Fig. 6 BET surface area plot of (1), **a** before adsorption, **b** after adsorption

Fig. 7 PXRD pattern of (1), **(a)** before adsorption, **(b)** after adsorption



indicates that the structural integrity of the compound was not compromised after the adsorption process [3, 5, 25].

3.6 Effect of Initial Dye Concentration

The effect of initial dye concentration was studied at 25 ± 2 °C for 60 min using 0.01 g of adsorbent and the result is presented in Fig. 8. The amount of MO adsorbed on (1) increased with an increase in concentration up to 6 mg/L and then started to decrease. The maximum q_e value obtained was 14.79 mg/g. This indicates that at concentrations below 6 mg/L, there were still unoccupied adsorption sites that got occupied as concentration increased [17, 19–21]. At 6 mg/L concentration, all the adsorption sites were saturated with dye molecules. Hence, desorption began thereafter.

3.7 Effect of Contact Time

The effect of time was investigated at 25 ± 2 °C with an MO concentration of 6 mg/L for 90 min. A plot of the quantity adsorbed at different contact time is shown in Fig. 9. The

adsorption process was rapid at the beginning of the experiment, reaching equilibrium after 60 min. This is a result of the availability of adsorption sites on (1) at the initial stage which became occupied after 60 min leading to decreased adsorption [17–21].

3.8 Effect of Temperature

The effect of temperature on adsorption of MO dye on (1) was studied for 60 min at a pH of 6, using 0.01 g of adsorbent and 6 mg/L of MO. The temperature was varied from 30 to 50 °C and the result is presented in Fig. 10. The results showed that the amounts of adsorbed MO dye adsorbed reached equilibrium at about 45 °C with an uptake of 12.35 mg/g, which is consistent with previous study [20].

3.9 Effect of pH

The effect of pH on adsorption of MO on (1) was studied for 60 min at pH values of 2–10 using 0.01 g of adsorbent, 6 mg/L of MO at 25 ± 2 °C. The result presented in Fig. 11

Fig. 8 Effect of initial concentration on the adsorption of MO onto (1)

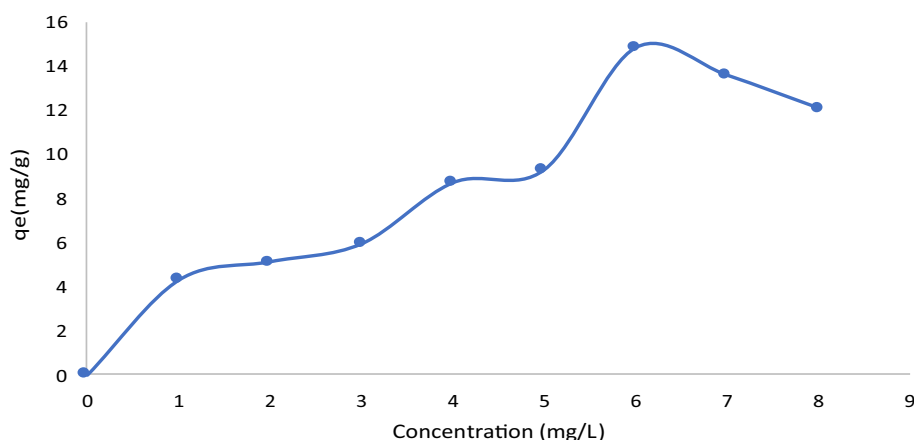


Fig. 9 Effect of contact time on the adsorption of MO onto (1)

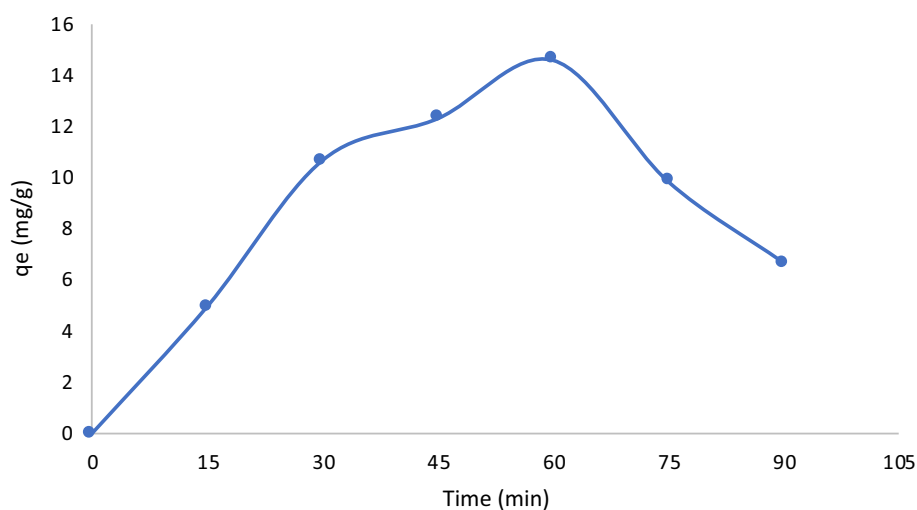


Fig. 10 Effect of temperature on the adsorption of MO onto (1)

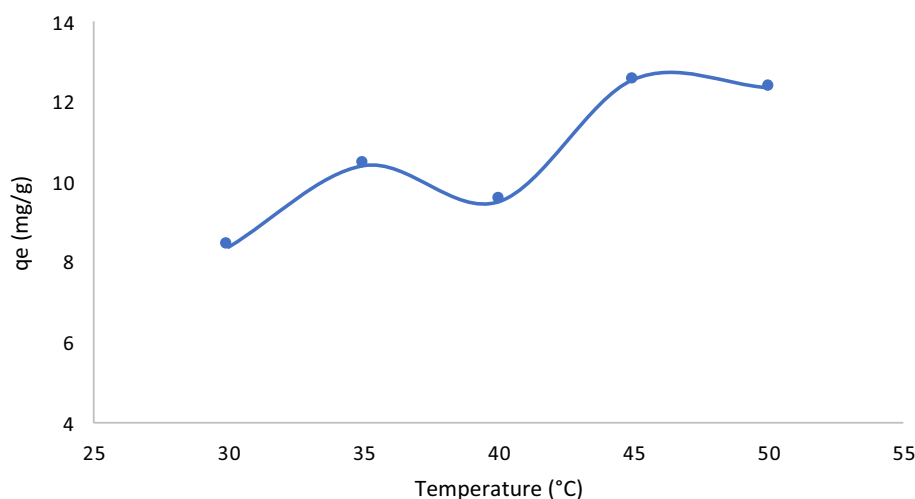
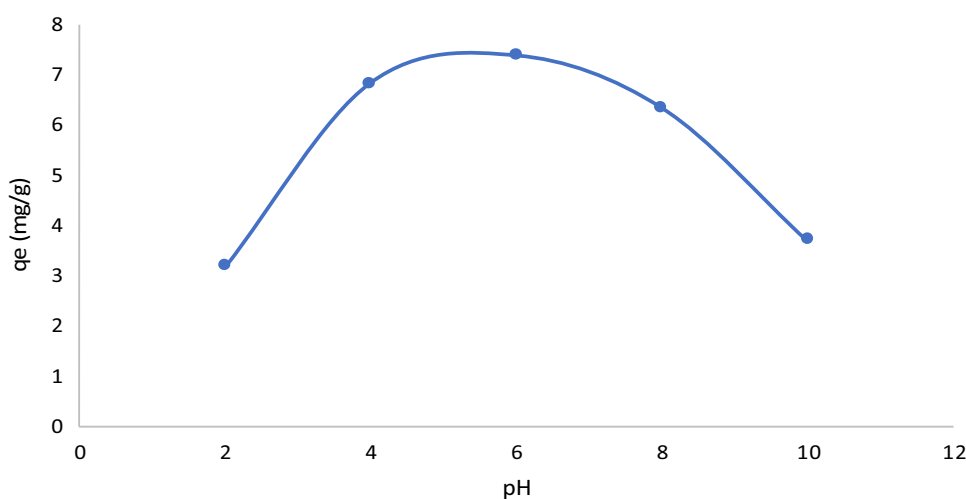


Fig. 11 Effect of pH on the adsorption of MO onto (1)



revealed that the amounts of dye adsorbed increased up to the pH of 6 and decreased thereafter. The anionic nature of methyl orange enabled it to adsorb more with increasing pH [3]. The maximum adsorption of 7.36 mg/g uptake was obtained at a pH of 6 and decreased with a further increase in pH.

3.10 Effect of Adsorbent Dosage

The effect of adsorbent dosage on the adsorption of MO on (1) was studied for 60 min at pH 6 using 0.01–0.05 g of adsorbent, and 6 mg/L of MO at 25 ± 2 °C. The result presented in Fig. 12 revealed that the amount of MO adsorbed decreased rapidly with increasing adsorbent dosage. A high uptake of 14.60 mg/g was obtained with 0.01 g of the adsorbent. The decrease in the uptake of the adsorbate as the amount of the adsorbent is increased could be a result of the aggregation of adsorbent particles at higher dosages leading to a decrease in the surface area, and an increase in

diffusion path length, which reduced the availability of the MO molecules and thus making it insufficient to cover all the exchangeable sites on the adsorbent [26].

3.11 Adsorption Kinetic Models

The adsorption of MO onto (1) was further studied using the pseudo-first-order and pseudo-second-order kinetic models. A comparison of the adsorption kinetics data was also made.

The pseudo-first-order kinetic model for the adsorption of MO onto (1) was investigated using Eq. 2.

$$\log(q_e - q_t) = \log q_e - \left(\frac{k_1}{2.303} \right) t \quad (2)$$

k_1 and q_e were respectively determined from the slope and the intercept when a graph of $\log(q_e - q_t)$ against t was plotted as shown in Fig. 13. The small R^2 value obtained indicates that the pseudo-first-order kinetic model did not fit

Fig. 12 Effect of adsorbent dosage on the adsorption of MO onto (1)

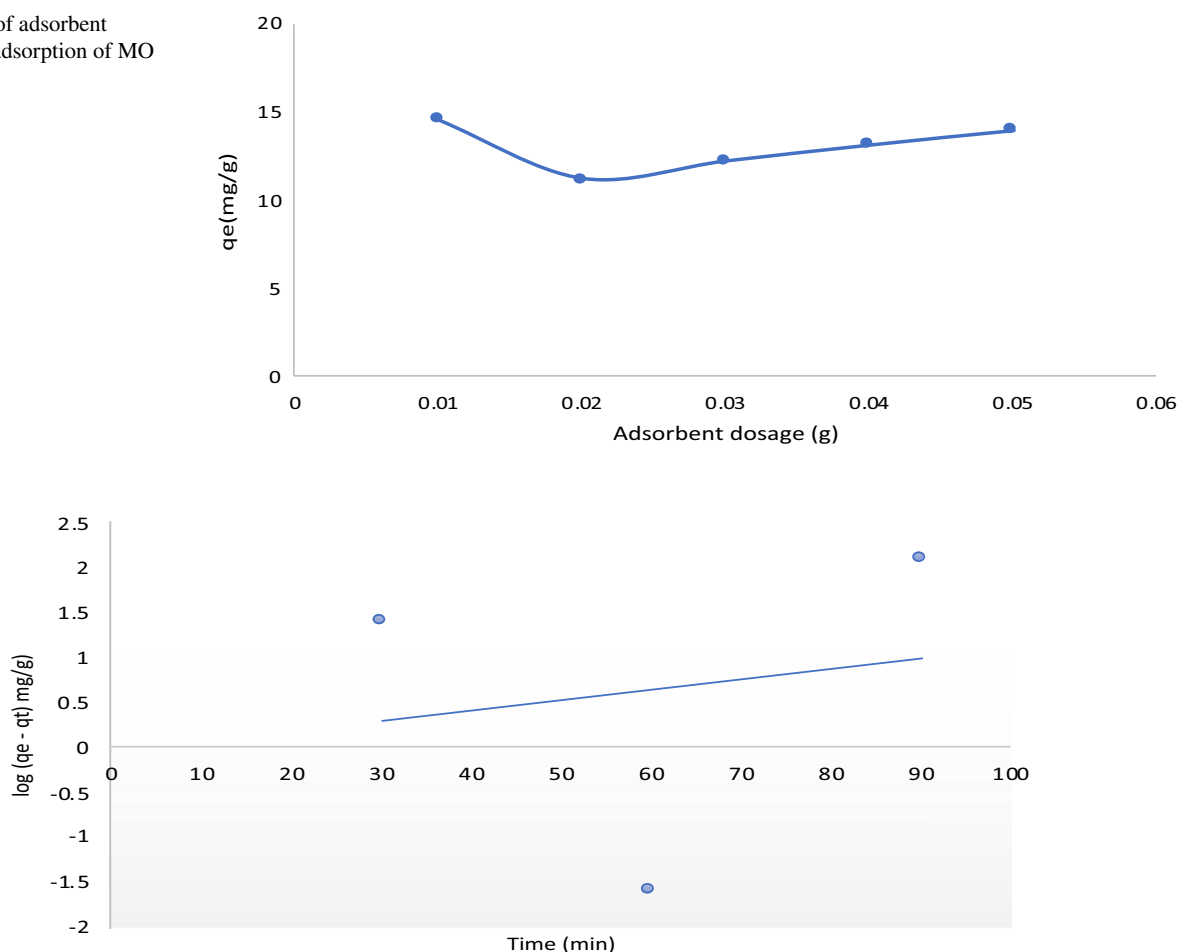


Fig. 13 Pseudo-first-order kinetics plot of MO onto (1)

the experimental data and thus not suitable for the adsorption process [3, 22].

The pseudo-second-order kinetic model for the adsorption of MO onto (1) was investigated using Eq. 3:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (3)$$

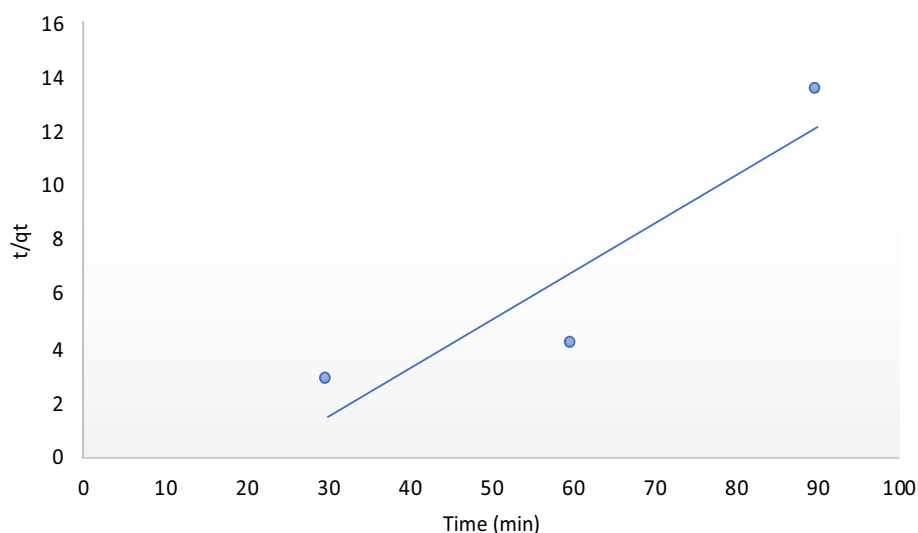
where q_e = amount adsorbed at equilibrium (mg/g), q_t = amount adsorbed (mg/g) at time, t , k_2 is the pseudo-second-order rate constant. A plot of t/q_t against t for the pseudo-second-order kinetic model shown in Fig. 14 gave a straight-line graph with a correlation coefficient (R^2) value of 0.99. The value of k_2 and q_e were obtained from the intercept and slope of the graph respectively.

Comparing the amount of MO adsorbed at equilibrium, the correlation coefficient (R^2), and the linearity of the plot of the kinetic models shows that the adsorption process favors the pseudo-second-order kinetics model more. The co-existence of physisorption and chemisorption is thus established for the adsorption of methyl orange on (1) [3,

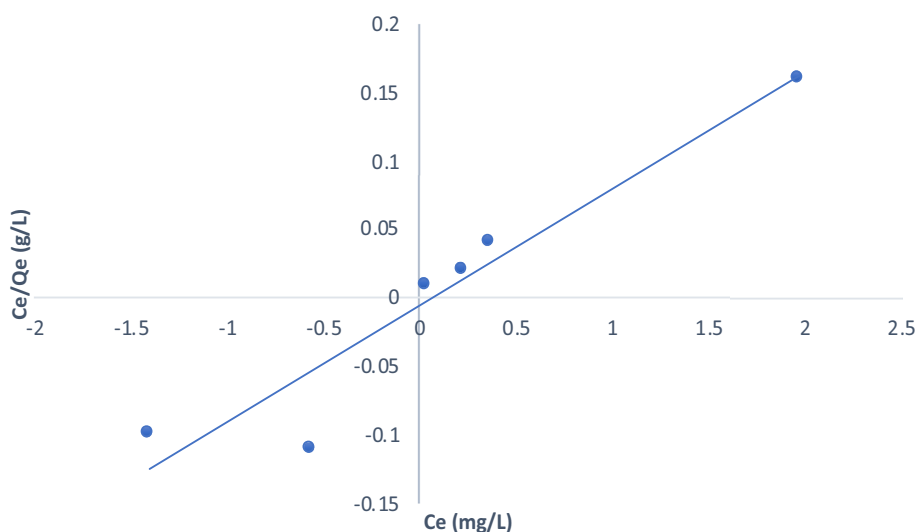
24, 27, 28]. Table 3 gives the summary of the values for the kinetic parameters from both kinetic models.

3.12 Adsorption Isotherms

The data obtained from the adsorption study were subjected to the Langmuir and Freundlich adsorption isotherms. The mathematical expression of the Langmuir adsorption isotherm is given by Eq. 4. A linear plot with an R^2 value of 0.91, and Q_e value of 11.76 mg/g was obtained when C_e/Q_e was plotted against C_e (Fig. 15). The separation factor (R_L value) given by Eq. 5 shows the nature of the adsorption process. It is either favorable ($0 < R_L < 1$), linear ($R_L = 1$), irreversible ($R_L = 0$), or unfavorable ($R_L > 1$). The R_L values obtained at 25 °C ranged from 0.0659 to 0.0087 indicating that the experimental conditions were favorable for the adsorption process [5, 25, 29]. It also confirmed that the adsorption data in this study fitted the Langmuir isotherm at the optimum conditions [5].

Fig. 14 Pseudo-second order kinetics plot of MO onto (1)**Table 3** Summary of the kinetic parameters for the adsorption of MO onto (1)

Parameters/Adsorbent	Pseudo-first-order kinetics			Pseudo-second-order kinetics		
	R ²	q _{e(calc)}	K ₁	R ²	q _{e(calc)}	K ₂
(1)	0.59	2.88	0.03	0.99	11.81	0.02

Fig. 15 Langmuir isotherm for the adsorption of MO onto (1)

$$\frac{C_e}{Q_e} = \frac{1}{K_a Q_m} + \frac{C_e}{Q_m} \quad (4)$$

$$R_L = \frac{1}{(1 + K_a C_o)} \quad (5)$$

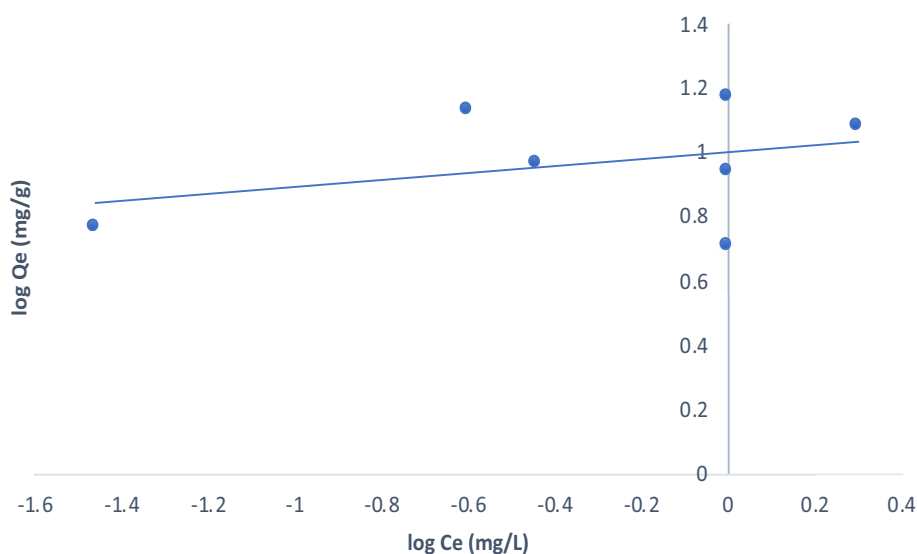
where Q_m is the maximum adsorption capacity (mg/g), Q_e is the amount of adsorbate per unit mass of the adsorbent at equilibrium (mg/g), C_e is the equilibrium solution

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

The logarithmic form of the Freundlich model of adsorption isotherm is given by Eq. 6:

$$\log Q_e = \log K_f + \left(\frac{1}{n}\right) \log C_e \quad (6)$$

Q_e is the amount adsorbed at equilibrium per gram of the adsorbent (mg/g), C_e is the equilibrium concentration of the adsorbate in the solution (mg/L). K_f and n are the Freundlich

Fig. 16 Freundlich isotherm for the adsorption of MO onto (**1**)**Table 4** Adsorption isotherms parameters for the adsorption of MO onto (**1**)

Parameter	Langmuir model			Freundlich model		
	R^2	Q_m	K_α	R^2	K_F	$1/n$
	0.91	15.01	6.59	0.11	7.79	0.38

model constants related to the adsorption capacity and adsorption intensity of the adsorbent. The plot of $\log Q_e$ against $\log C_e$ gave a straight-line graph with a slope of $\frac{1}{n}$ and $\log K_f$ as the intercept. The low R^2 value of 0.11 obtained for the Freundlich isotherm indicates that it is the less fit of two models applied to the experimental data (Fig. 16).

The parameters of the adsorption isotherms in Table 4 showed that the adsorption process followed the Langmuir model with the highest R^2 value of 0.91. The monolayer adsorption capacity, Q_m was 15.01 mg/g, and $1/n$ was 0.38. These results further revealed that the adsorbate formed a homogenous layer on the surface of the adsorbent with uniform distribution, and the process was favorable based on the $1/n$ value [3].

4 Conclusion

The results obtained from this study show that the adsorption of methyl orange dye onto (**1**) was dependent on various factors such as adsorbent dose, contact time, initial adsorbate concentration, and temperature. The equilibrium data was subjected to two isotherm models and the best fit was found to be the Langmuir adsorption isotherm with an R^2 value of 0.91. The adsorption data followed the pseudo-second-order kinetic model indicating a process of chemisorption for the adsorption of MO dye on **1**. Therefore, the compound (**1**) could be used as a potential adsorbent in the removal of methyl orange from industrial effluents.

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Data availability Data available on request due to privacy restrictions.

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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