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Isotherm Model for the Adsorption of Malachite Green on Silica Xerogel Modified with EDA/Cu^{2+}

Soni Ratia, Edi Nasra, Trisna Kumala Sari, Umar Kalmar Nizar, M. Iqbal Saputra Gemasih
Universitas Negeri Padang
edinasra@fmipa.unp.ac.id

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Abstract

Malachite green (MG) is a cationic dye that poses potential risks to aquatic organisms. This study evaluated the adsorption performance of silica xerogel derived from fly ash and modified with ethylenediamine (EDA) and copper ions (Cu^{2+}) for MG removal and identified the most suitable adsorption isotherm model. Silica xerogel was synthesized using the sol–gel method, modified with EDA and Cu^{2+}, and characterized using Fourier transform infrared (FTIR) spectroscopy. Batch adsorption experiments used 0.1 g of adsorbent and 25 mL of MG solution at initial concentrations of 100–1100 mg/L, pH 4, a stirring speed of 200 rpm, and a contact time of 90 min. Residual MG concentrations were measured using UV–visible spectrophotometry at 618 nm. FTIR spectra showed characteristic silica framework bands and spectral changes following contact with MG. Adsorption capacity increased from 22.562 mg/g at 100 mg/L to 187.416 mg/g at 900 mg/L, followed by slight decreases at 1000 and 1100 mg/L. The Langmuir model provided a better fit to the experimental data (R^2 = 0.98267) than the Freundlich model (R^2 = 0.79245), with a theoretical maximum adsorption capacity of 200.803 mg/g. These findings characterize the adsorption behavior of the modified xerogel and indicate its potential as an alternative adsorbent for removing MG from aqueous solutions under the tested conditions.

Keywords: Malachite Green; Silica Xerogel; Fly Ash; Surface Modification; Adsorption Isotherms

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INTRODUCTION

Malachite Green (MG) is a cationic dye of the triphenylmethane class; its presence in wastewater must be controlled because it has the potential to cause toxic effects on aquatic organisms and the aquatic environment. Ojediran et al. (2021) examined the potential of MG as an endocrine disruptor and evaluated its removal through adsorption. Various materials have been developed to reduce MG concentrations in solutions, including fly ash, silica, and composite materials (Ojediran et al., 2021). Romadhona et al. (2024), for example, developed a magnetized coal fly ash adsorbent to adsorb MG. Their study demonstrated that the development of adsorbents based on industrial waste is one approach that can be utilized in the treatment of water containing dyes (Romadhona et al., 2024).

Fly ash is a byproduct of coal combustion that has the potential to be utilized as a source of silica for the synthesis of adsorbent materials. Cynthia et al. (2025) reported the synthesis of silica xerogel from fly ash using the sol-gel method and identified silanol (Si–OH) and siloxane (Si–O–Si) groups through FTIR characterization (Cynthia et al., 2025). Putri et al. (2025) reported the use of silica xerogel synthesized from fly ash and obtained a maximum adsorption capacity of 21.329 mg/g at an optimal contact time of 90 minutes. This study provides a foundation for the development of fly ash-based silica xerogel as an MG adsorbent. However, the material's surface characteristics can still be modified to evaluate changes in its adsorption behavior (Putri & Nasra, 2025).

One approach to enhancing the characteristics of adsorbents is surface modification using organic compounds and metal ions. In this study, fly ash-based silica xerogel was modified using ethylenediamine (EDA) and copper ions (Cu^{2+}), hereinafter referred to as SX-EDA/Cu. EDA contains amine groups that can serve as potential interaction sites, while Cu^{2+} can coordinate with electron-donating atoms on the corresponding functional groups. Nasra et al. (2026) have reported the synthesis and characterization of fly ash-based silica xerogel functionalized with EDA and Cu^{2+} for heavy metal adsorption applications. This research provides a foundation for the development of SX-EDA/Cu materials; however, its application for the adsorption of MG dyes requires separate evaluation because the characteristics of the adsorbate and the interactions involved may differ (Nasra et al., 2026)

Adsorption evaluation is not only based on adsorption capacity but also on an analysis of the relationship between the adsorbate concentration at equilibrium and the amount of adsorbate bound to the adsorbent surface. This relationship can be explained

using the Langmuir and Freundlich isotherm models. The Langmuir model assumes adsorption at specific sites with the formation of a single layer, while the Freundlich model is an empirical model that can describe adsorption on heterogeneous surfaces. Nikmah et. al. (2023) reported that the Langmuir model is most suitable for MG adsorption using coal fly ash (Nikmah et al., 2023). Conversely, ojediran et.al. (2021) reported that the Freundlich model provides a better fit for MG adsorption using acid-functionalized corn cobs (Ojediran et al., 2021). Furthermore, nazriati et.al. (2026) evaluated the adsorption characteristics of MG using a silica aerogel/MgO composite based on sugarcane bagasse ash through isotherm, kinetic, and thermodynamic analyses (Nazriati et al., 2026). The differences in these research results indicate that the most appropriate isotherm model must be determined based on the characteristics of the adsorbent and the experimental data for each system.

Based on previous studies, the adsorption of MG using fly ash-based silica xerogel and various modified adsorbents has been investigated. However, no studies specifically evaluating the Langmuir and Freundlich isotherm models for MG adsorption using fly ash-based silica xerogel modified with EDA/Cu were found in the reference collection used in this study. Therefore, this study focuses on evaluating the equilibrium adsorption of MG using SX-EDA/Cu by comparing the Langmuir and Freundlich isotherm models. The objective of this study is to determine the most appropriate isotherm model based on the fit to the experimental data and to evaluate the obtained isotherm parameters to explain the equilibrium adsorption characteristics of MG on this material.

METHODS

1. Materials and Tools

The materials used in this study included fly ash, hydrochloric acid (HCl), sodium hydroxide (NaOH), ethylenediamine (EDA), copper(II) chloride hydrate ($CuCl_2 \cdot 2H_2O$), Malachite Green dye, ethanol, silver nitrate ($AgNO_3$), and distilled water. The equipment used included beakers, volumetric flasks, pipettes, a magnetic stirrer, a shaker, a pH meter, an analytical balance, an oven, a furnace, filtration equipment, a Fourier Transform Infrared (FTIR) spectrometer, and a UV-Vis spectrophotometer.

2. Synthesis and Modification of Silica Xerogel

A total of 100 g of fly ash was washed with 400 mL of 3 M HCl while stirring for 2 hours at 70 °C. The mixture was then filtered, and the residue was washed with distilled water until the filtrate reached a neutral pH, then dried at 105 °C for 8 hours. About 40 g of the residue was extracted with 250 mL of 7 M NaOH at 80 °C for 1 hour with continuous stirring. The resulting extract was filtered to obtain a sodium silicate solution and then titrated with 3 M HCl to pH 7 to form a silica hydrogel. The hydrogel was aged for 18 hours at room temperature, then filtered and washed with distilled water until Cl^- ions were no longer detected using $AgNO_3$. The solid was then dried at 70 °C for 5 hours to get silica xerogel. The silica xerogel was modified with ethylenediamine (EDA) in a silica xerogel:EDA ratio of 1:2. The mixture was stirred using a magnetic stirrer for 6 hours at 80 °C. Then it was filtered and washed with ethanol to remove the unreacted EDA. The EDA-modified silica xerogel was next impregnated using a 0.1 M $CuCl_2 \cdot 2H_2O$ solution for 2 hours at a 1:1.5 ratio. The material was then washed with ethanol, dried at 70 °C, and heated in a furnace at 400 °C for 4 hours. The final material, EDA/Cu silica xerogel, was then characterized using FTIR to identify changes in functional groups after the modification process.

3. Concentration Adsorption Experiment

The adsorption experiments were conducted using the batch method by mixing 0.1 g of SX-EDA/Cu into 25 mL of MG solution. The initial MG concentration was varied to 100 - 1100 mg/L. The pH of the solution was adjusted to 4, and the mixture was stirred using a shaker at 200 rpm for 90 minutes at room temperature. After the adsorption process was complete, the mixture was filtered, and the concentration of MG remaining in the filtrate was measured using a UV-Vis spectrophotometer at a wavelength of 618 nm. The MG concentration at equilibrium (*Ce*) was determined based on the calibration curve.

4. Adsorption Capacity and Isotherm Analysis

The equilibrium adsorption capacity (Q_e) of silica xerogel EDA/Cu toward Malachite Green was calculated from the difference between the initial concentration (C_0) and equilibrium concentration (Ce) using Equation

$$qe = \frac{(C_0 - C_e)V}{m} \quad (1)$$

Where :

Qe : Equilibrium adsorption capacity (mg/g)

C_0 : Initial MG concentration (mg/L)

C_e : MG concentration at equilibrium (mg/L)

V : Volume of the solution (L)

m : Adsorbent mass (g)

The adsorption equilibrium was evaluated using the Langmuir and Freundlich isotherm models. The linearized Langmuir equation was expressed as:

$$\frac{Ce}{qe} = \frac{1}{Q_m \ K_L} + \frac{C_e}{Q_m} \quad (2)$$

Where

Q_m : Maximum adsorption capacity (mg/g)

K_L : Langmuir constant (L/mg).

The Freundlich isotherm was evaluated using the following linear equation:

$$\log Q_e = \log K_F + \frac{1}{n} \log C_e \quad (3)$$

Where K_F is the Freundlich adsorption constant and (n) represents the adsorption intensity. The adsorption data were fitted using linear regression, and the coefficient of determination (R^2) was used to evaluate the fit of each isotherm model

RESULTS

1. FTIR

The FTIR spectrum in Figure 1 shows differences in the absorption band characteristics of SX-EDA/Cu before and after contact with Malachite Green (MG). In the SX-EDA/Cu sample, the main absorption bands were identified at 3446.85 cm^{-1}, corresponding to O–H stretching, and 1632.77 cm^{-1}, associated with H–O–H bending vibrations. In addition, the bands at 1018.43 and 805.29 cm^{-1} were attributed to asymmetric

and symmetric Si–O–Si stretching vibrations, respectively, which are characteristic of the silica framework..

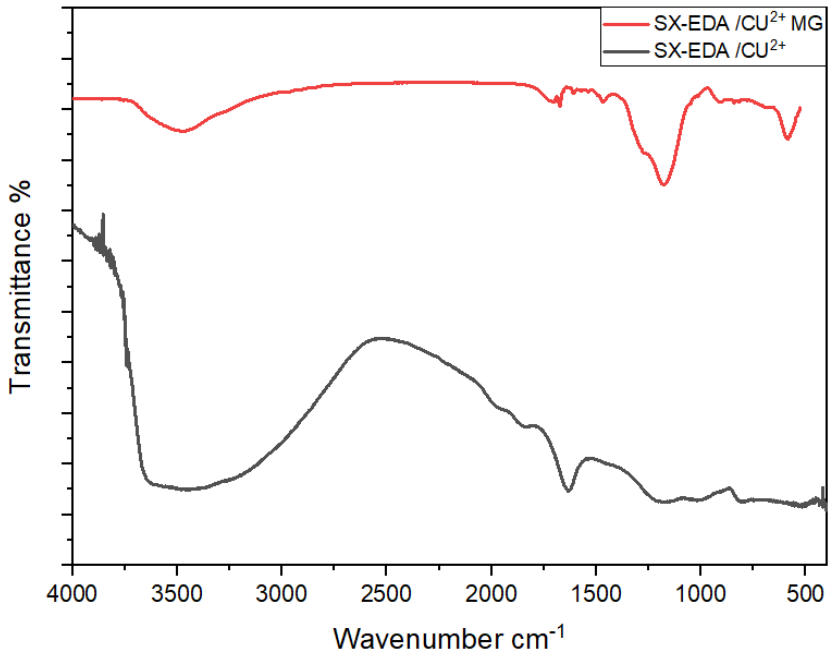


Figure 1. FTIR Spectrum Silika Xerogel EDA/CU before and after contact with malachite green

After contact with MG, the SX-EDA/Cu-MG sample exhibited shifts in the absorption band positions to 3467.51 cm^{-1}, 1617.45 cm^{-1}, and 1074.94 cm^{-1}, which were associated with the O–H stretching region, H–O–H bending region, and Si–O–Si stretching vibrations, respectively. Additional absorption bands appeared at 1587.35, 1519.48, and 1374.05 cm^{-1}, together with bands at 726.13 and 458.99 cm^{-1}. Overall, the comparison of both spectra revealed changes in absorption band positions and the appearance of additional bands after contact with MG.

2. The Effect of Initial Concentration on Adsorption

The effect of initial Malachite Green concentration on the adsorption capacity of silica xerogel EDA/Cu was investigated. The equilibrium concentration (Ce) and adsorption capacity (Qe) obtained at each initial concentration (C_0) are presented in Table 1

Table 1. Experimental Adsorption Capacity of Malachite Green at Different Initial Concentrations.

C_0	C_e	Q_e
100	6.9020	22.562
200	4.8604	47.552
300	7.7047	72.298
400	16.3839	94.19242
500	57.4137	110.0882
600	73.9759	130.3353
700	115.0444	145.3664
800	120.8604	169.0996
900	148.1409	187.4159
1000	258.6294	184.6209
1100	360.0508	182.6142

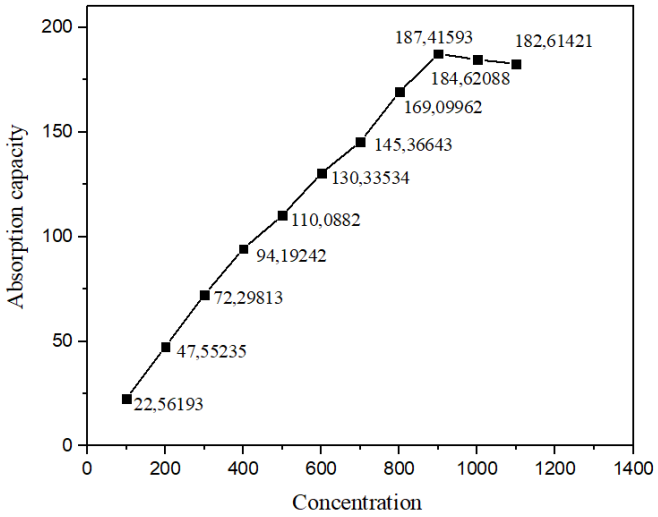


Figure 2. Experimental Adsorption Capacity of Malachite Green at Different Initial Concentrations

Figure 2 shows that the adsorption capacity of silica xerogel EDA/Cu increased with increasing initial Malachite Green concentration from 100 to 900 mg/L. The adsorption capacity increased from 22.562 mg/g at 100 mg/L to 187.416 mg/g at 900 mg/L. However, at higher initial concentrations, the adsorption capacity slightly decreased to 184.621 mg/g at 1000 mg/L and 182.614 mg/g at 1100 mg/L. The highest experimental adsorption capacity was therefore obtained at an initial concentration of 900 mg/L, reaching 187.416 mg/g

3. Langmuir Adsorption Isotherm

The Langmuir isotherm analysis was performed using the C_e/Q_e ratio as a function of Ce. The equilibrium data are presented in Table 2.

Table 2. Equilibrium adsorption parameter data for malachite green

C_e	Q_e	C_e / Q_e
6.9020	22.562	0.3059
4.8604	47.552	0.1022
7.7047	72.298	0.1066
16.3839	94.19242	0.1739
57.4137	110.0882	0.5215
73.9759	130.3353	0.5676
115.0444	145.3664	0.7914
120.8604	169.0996	0.7147
148.1409	187.4159	0.7904
258.6294	184.6209	1.4009
360.0508	182.6142	1.9716

The data in Table 3 were then analyzed using the Langmuir isotherm model for Malachite Green (MG) on EDA/Cu-modified silica xerogel based on the linear relationship model based on the C_e/Q_e ratio to obtain the Langmuir isotherm curve shown in Figure 3.

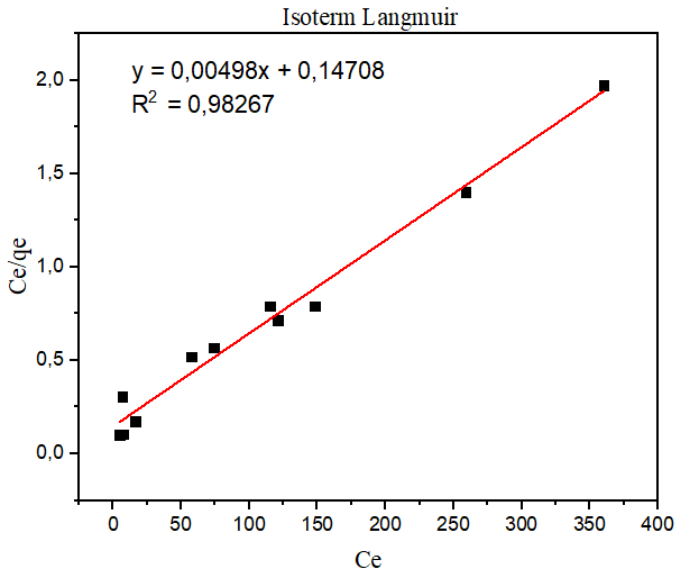


Figure 3. Langmuir Adsorption Isotherm of Malachite Green on EDA/Cu-Modified Silica Xerogel

The Langmuir isotherm shows a linear relationship between (Ce) and (Ce/Qe). The resulting linear regression equation is as follows:

$$\frac{C_e}{q_e} = 0{,}00498x + 0{,}14708 \quad (4)$$

$$R^2 = 0{,}98267$$

With :

C_e = Equilibrium concentration (mg/L)

Q_e = Adsorption capacity at equilibrium (mg/g)

The Langmuir isotherm parameters, including the maximum adsorption capacity (Qm), Langmuir constant (KL), and coefficient of determination (R^2), are presented in Table 3

Table 3. Langmuir Isotherm Parameters for Malachite Green Adsorption onto EDA/Cu-Modified Silica Xerogel

Parameter	Value
Maximum Adsorption Capacity Q_m (mg/g)	200.803 mg/g
Langmuir Constant K_L (L/mg)	0.03388 L/mG
Coefficient of determination R^2	0.98267

The Langmuir isotherm analysis yielded a maximum adsorption capacity (Qm) of 200.803 mg/g and a Langmuir constant (KL) of 0.03388 L/mg, with a coefficient of determination (R^2) of 0.98267.

4. Freundlich Adsorption Isotherm

The Freundlich isotherm model was analyzed using the relationship between log C_e and log Q_e. The adsorption equilibrium data are presented in Table 4. Using the equation:

Table 4. Equilibrium adsorption parameter data for malachite green

Log C_e	Log Q_e
0.83897767	1.353376166
0.68667256	1.677171963
0.88675548	1.859127053
1.21441684	1.974015944
1.75901558	2.041740763

Log C_e	Log Q_e
1.86909019	2.115062198
2.06086554	2.162464137
2.08228405	2.22814263
2.17067487	2.272806494
2.41267796	2.266280806
2.55636373	2.261534576

The data in Table 4 were then analyzed using the Freundlich isotherm model for Malachite Green (MG) on EDA/Cu-modified silica xerogel based on the linear relationship between log Ce and log qe to obtain the Freundlich isotherm curve shown in Figure 4.

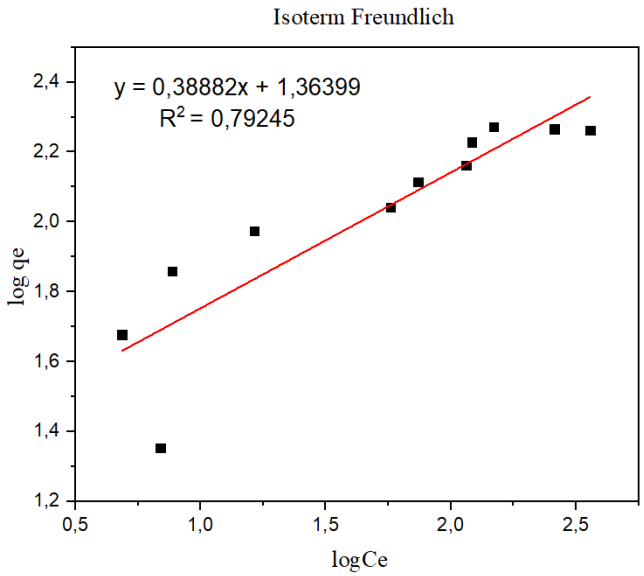


Figure 4. Freundlich Adsorption Isotherm of Malachite Green on EDA/Cu-Modified Silica Xerogel

The Freundlich isotherm showed a linear relationship between the logarithm of equilibrium concentration (log Ce) and the logarithm of adsorption capacity (log Qe). The linear regression equation was obtained as follows:

$$\log Qe = \log K_F + \frac{1}{n} logCe \quad (5)$$

With :

Ce = equilibrium concentration (mg/L)

Qe = adsorption capacity at equilibrium (mg/g)

K_F = freundlich constant

n = adsorption intensity parameter

The results of the linear regression using the Freundlich model yield the following equation:

$$\log Q_e = 0{,}38882 \log C_e + 1{,}36399 \quad (6)$$

$$R^2 = 0{,}79245$$

The Freundlich isotherm parameters, including the adsorption capacity constant (KF), adsorption intensity parameter (n), and coefficient of determination (R^2), are presented in Table 5.

Table 5. Freundlich Isotherm Parameters for Malachite Green Adsorption onto EDA/Cu-Modified Silica Xerogel

Parameter	Value
K_F	23.1201
n	2.5719
R^2	0.79245

The Freundlich isotherm analysis yielded a Freundlich constant (KF) of 23.1201 and an adsorption intensity parameter (n) of 2.5719, with a coefficient of determination (R^2) of 0.79245.

5. Comparison of Langmuir and Freundlich Isotherm

The adsorption data were evaluated using the Langmuir and Freundlich isotherm models. The Langmuir model exhibited a higher coefficient of determination (R^2 = 0.98267) than the Freundlich model (R^2 = 0.79245). This indicates that the Langmuir model provided a better fit to the experimental adsorption data than the Freundlich model. A comparison of the Langmuir and Freundlich isotherm models based on their determination coefficients and characteristic parameters is presented in Table 6.

Table 6. Comparison of Langmuir and Freundlich Isotherm Parameters for Malachite Green Adsorption

Isotherm model	R^2	Main parameter
Langmuir	0.98267	Q_m = 200.803 (mg/g)
Freundlich	0.79245	K_F = 23.1201; n = 2.5719

DISCUSSION

1. Analysis and Interpretation of FTIR Results

Based on the FTIR characterization results, the EDA/Cu-modified silica xerogel (SX-EDA/Cu) exhibited absorption bands at 3446.85, 1632.77, 1018.43, and 805.29 cm^{-1}. The band at 3446.85 cm^{-1} corresponds to O–H stretching vibrations originating from silanol groups (Si–OH) and adsorbed water molecules. The band at 1632.77 cm^{-1} is associated with H–O–H bending vibrations of adsorbed water. Furthermore, the bands at 1018.43 and 805.29 cm^{-1} correspond to asymmetric and symmetric Si–O–Si stretching vibrations, respectively. Asymmetric Si–O–Si stretching generally occurs at 1000–1200 cm^{-1}, while symmetric stretching is observed around 800 cm^{-1} (Ellerbrock et al., 2024). Similar absorption bands have been reported in fly ash-derived silica xerogel, with Si–OH and Si–O–Si bands appearing at 3388 and 985 cm^{-1}, respectively (Cynthia et al., 2025)

The presence of EDA functional groups in modified silica can be identified through C–N and CH_2 vibrations. The CH_2 stretching vibration of EDA occurs at 2840–2980 cm^{-1}, while C–N vibrations appear at 1000–1250 cm^{-1}. However, the C–N absorption region overlaps with the Si–O–Si absorption region, making its identification difficult (Nasra et al., 2026). In this study, the clearly identified absorption bands correspond to O–H, H–O–H, and Si–O–Si vibrations. Separate C–N and CH_2 bands are not observed in the reported spectrum. Therefore, the FTIR results primarily confirm the characteristic functional groups of the silica framework, particularly O–H and Si–O–Si vibrations. However, the incorporation of EDA cannot be conclusively confirmed from the FTIR spectrum alone because the C–N and CH_2 absorption bands were not clearly identified. Additional characterization is therefore required to provide further evidence of EDA incorporation into the silica xerogel.

After contact with Malachite Green (MG) solution, the FTIR spectrum of SX-EDA/Cu-MG exhibited shifts in several absorption bands. The band at 3446.85 cm^{-1} shifted to 3467.51 cm^{-1}, while the band at 1632.77 cm^{-1} shifted to 1617.45 cm^{-1}. In addition, the band at 1018.43 cm^{-1} shifted to 1074.94 cm^{-1}. The band at 1074.94 cm^{-1} remains within the asymmetric Si–O–Si stretching region of 1000–1200 cm^{-1} (Ellerbrock et al., 2024). This shift of 56.51 cm^{-1} indicates a change in the vibrational environment of the silica framework after

contact with MG. Changes in the O–H and H–O–H absorption regions also demonstrate alterations in the spectral characteristics of the adsorbent following MG contact.

In addition to these shifts, the SX-EDA/Cu-MG spectrum exhibited additional absorption bands at 1587.35, 1519.48, 1374.05, 726.13, and 458.99 cm^{-1}. The band at 1587.35 cm^{-1} is associated with aromatic ring vibrations of MG, corresponding to the absorption band reported at 1588 cm^{-1} for MG-containing adsorbent materials (Abdelrahman et al., 2025). The bands at 1519.48 and 1374.05 cm^{-1} are also observed in the spectrum after MG adsorption; however, their specific functional group assignments cannot be established from the available reference data. The band at 458.99 cm^{-1} is associated with bending vibrations of the silica framework in the low-wavenumber region (Ellerbrock et al., 2024), whereas the band at 726.13 cm^{-1} is retained as an additional absorption band without a specific assignment.

Overall, the FTIR results show changes in the absorption characteristics of SX-EDA/Cu after contact with MG, particularly in the O–H, H–O–H, and Si–O–Si regions. The appearance of the band at 1587.35 cm^{-1}, which corresponds to the aromatic ring vibration reported for MG, provides additional evidence of MG presence on the adsorbent surface. These changes demonstrate alterations in the surface spectral characteristics following the adsorption process.

2. The Effect of Initial Concentration on Adsorption

The adsorption capacity of silica xerogel EDA/Cu increased progressively with increasing initial Malachite Green (MG) concentration from 100 to 900 mg/L. The adsorption capacity increased from 22.562 mg/g at 100 mg/L to 47.552, 72.298, 94.192, 110.088, 130.335, 145.366, and 169.100 mg/g at initial concentrations of 200, 300, 400, 500, 600, 700, and 800 mg/L, respectively. The highest experimental adsorption capacity was obtained at 900 mg/L, reaching 187.416 mg/g. The increase in adsorption capacity can be attributed to the increasing concentration gradient between MG molecules in the solution and the adsorbent surface, which provides a greater driving force for mass transfer and increases the probability of interaction between MG molecules and available adsorption sites (Ojediran et al., 2021). Similar increases in adsorption capacity with increasing initial MG concentration have been reported for various adsorbents, including mesoporous natural inorganic clays and other silica- or mineral-based adsorbents (Abewaa et al., 2023). Similar

concentration-dependent adsorption behavior for MG has been examined using fly-ash-derived adsorbents and other low-cost materials (Romadhona et al., 2024)

At higher initial concentrations, the adsorption capacity slightly decreased to 184.621 mg/g at 1000 mg/L and 182.614 mg/g at 1100 mg/L. The decrease in adsorption capacity at 1000 and 1100 mg/L indicates that increasing the initial MG concentration beyond 900 mg/L did not result in a further increase in adsorption uptake. This behavior suggests that the adsorption system was approaching its maximum experimental uptake under the investigated conditions. At this stage, the available adsorption sites may have become increasingly occupied, limiting further adsorption of MG molecules (Barakat et al., 2025).

Overall, the results demonstrate that an initial MG concentration of 900 mg/L produced the highest experimental adsorption capacity of 187.416 mg/g. Further increases in the initial concentration to 1000 and 1100 mg/L resulted in lower adsorption capacities of 184.621 and 182.614 mg/g, respectively. Therefore, 900 mg/L represents the initial concentration that yielded the highest adsorption capacity within the investigated concentration range.

3. Langmuir Adsorption Isotherm

The equilibrium adsorption data of Malachite Green (MG) onto silica xerogel EDA/Cu were analyzed using the Langmuir isotherm model. The linear plot of Ce/Qe against Ce produced the equation $Ce/Qe = 0.00498Ce + 0.14708$, with a coefficient of determination (R^2) of 0.98267. The R^2 value indicates a strong fit between the Langmuir model and the experimental equilibrium data.

The Langmuir model assumes that adsorption occurs on a relatively homogeneous surface with a finite number of adsorption sites, resulting in monolayer coverage of adsorbate molecules on the adsorbent surface (Nandoost et al., 2022). Based on the Langmuir equation, the maximum theoretical adsorption capacity (Qm) of silica xerogel EDA/Cu was 200.803 mg/g, while the Langmuir constant (KL) was 0.03388 L/mg. The Qm value represents the theoretical maximum adsorption capacity when the available adsorption sites are occupied by MG molecules. This value was higher than the highest experimental adsorption capacity of 187.416 mg/g obtained at an initial MG concentration of 900 mg/L.

The applicability of the Langmuir model has also been demonstrated in other studies involving MG adsorption onto modified biomass-based adsorbents. Yildiz et al. (2024) reported that MG adsorption onto activated carbon derived from okra stalk waste followed

the Langmuir isotherm, with a theoretical maximum adsorption capacity of 119.05 mg/g. Compared with that value, the theoretical adsorption capacity of SX-EDA/Cu in the present study was higher, reaching 200.803 mg/g. However, this difference should be interpreted cautiously because adsorption capacity is influenced by adsorbent characteristics, surface properties, and experimental conditions. This comparison further highlights the potential of EDA/Cu-modified silica xerogel as an adsorbent for MG removal (Yildiz et al., 2024)

Similar applications of the Langmuir model have been reported for MG adsorption using other adsorbents. The Langmuir model provided the best fit for MG adsorption onto sodium dodecyl sulfate-modified bentonite (Bishwas et al., 2023). The Langmuir model has also been applied to describe MG adsorption equilibrium using modified bagasse fly ash (Gebre Meskel et al., 2024). These findings support the applicability of the Langmuir model for describing the equilibrium adsorption data of MG onto silica xerogel EDA/Cu.

4. Freundlich Adsorption Isotherm

The equilibrium adsorption data of Malachite Green (MG) onto silica xerogel EDA/Cu were also analyzed using the Freundlich isotherm model. The linear plot of log Qe against log C_e produced the equation log Qe = 0.38882 log C_e + 1.36399, with a coefficient of determination (R^2) of 0.79245. This value indicates that the Freundlich model can describe the relationship between the equilibrium concentration and adsorption capacity, although its correlation is lower than that of the Langmuir model (R^2 = 0.98267). The Freundlich model is based on the assumption of adsorption on heterogeneous surfaces, where adsorption sites possess different energies and adsorption may occur in multiple layers. Consequently, the Freundlich model is commonly used to describe adsorption systems with nonuniform surface characteristics (Kalam et al., 2021).

The Freundlich parameters were calculated from the slope and intercept of the linear regression equation. The adsorption capacity constant (KF) was obtained as 23.1201, while the adsorption intensity parameter (n) was 2.5719. The corresponding value of 1/n was 0.38882, which falls within the range of 0 to 1. This range generally indicates favorable adsorption, suggesting that MG adsorption onto silica xerogel EDA/Cu is favorable according to the Freundlich model. The KF parameter represents the relative adsorption capacity, whereas n describes the adsorption intensity of the adsorbent toward the adsorbate (Nandoost et al., 2022).

The Freundlich model has also been reported to describe MG adsorption using different types of adsorbents. For example, adsorption of MG onto coal-associated soil was better represented by the Freundlich model than by the other isotherm models evaluated. The reported coefficient of determination was 0.9568, indicating a strong correlation between the experimental data and the Freundlich equation. This result was associated with the heterogeneous adsorption characteristics of the coal-associated soil surface. Such findings demonstrate that the Freundlich model can be suitable for describing MG adsorption on adsorbents with heterogeneous surface properties (Sundararaman et al., 2021).

A similar result was reported for MG removal using date seeds as a natural adsorbent. The Freundlich model provided the best fit to the experimental equilibrium data compared with the other isotherm models evaluated in that study. This finding indicates that the Freundlich equation can describe adsorption systems in which the adsorbent surface contains sites with different adsorption affinities. However, the suitability of an isotherm model may differ depending on the characteristics of the adsorbent and the adsorption system. Therefore, the results from other MG adsorption studies serve as comparisons rather than direct predictions of the behavior of silica xerogel EDA/Cu (Sorour et al., 2024)

In the present study, the Freundlich model produced an R^2 value of 0.79245, which was lower than the Langmuir R^2 value of 0.98267. This difference indicates that the Langmuir equation provides a better fit to the experimental equilibrium data of MG adsorption onto silica xerogel EDA/Cu. A comparable result was reported for MG adsorption using magnetized coal fly ash, where the Langmuir model provided a better fit than the Freundlich model. The similarity in the preferred isotherm model provides a relevant comparison because both studies involved adsorbents derived from fly ash. Nevertheless, the higher Langmuir R^2 in the present study only indicates a better mathematical fit and does not independently prove that adsorption occurs exclusively through monolayer formation (Romadhona et al., 2024)

5. Comparison of Langmuir and Freundlich Isotherm

The comparison of the Langmuir and Freundlich isotherm models revealed a clear difference in their ability to describe the equilibrium adsorption of Malachite Green (MG) onto silica xerogel EDA/Cu. The Langmuir model exhibited a higher coefficient of determination ($R^2 = 0.98267$) than the Freundlich model ($R^2 = 0.79245$), indicating a better

fit to the experimental adsorption data. Furthermore, the maximum theoretical adsorption capacity (Qm) calculated using the Langmuir model was 200.803 mg/g, which was relatively close to the highest experimental adsorption capacity of 187.416 mg/g. The difference between these values was approximately 6.62%, indicating that the experimental adsorption capacity approached the theoretical value predicted by the Langmuir model. A comparable result was reported for MG adsorption onto *Rumex abyssinicus*-derived activated carbon, where Langmuir provided a better fit than Freundlich, although the Koble–Corrigan model showed the highest overall correlation (Abewaa et al., 2023)

The difference in the fitting performance of the two models can be understood from their respective theoretical assumptions. The Langmuir model describes adsorption as monolayer coverage on a homogeneous surface with a finite number of adsorption sites, whereas the Freundlich model describes adsorption on heterogeneous surfaces with different adsorption energies. These differences provide a theoretical basis for evaluating the equilibrium behavior of adsorbate molecules on different adsorbent surfaces. The selection of an appropriate isotherm model should therefore consider the characteristics of the adsorbent and its interaction with the adsorbate (Musah et al., 2022)

The predominance of the Langmuir model has also been observed in other MG adsorption systems. Adsorption of MG using manganese oxide nanoparticles was better described by the Langmuir model, which produced an R^2 value of 0.973 under the reported experimental conditions. This finding indicates that the Langmuir model can provide a strong representation of MG adsorption equilibrium in systems involving metal oxide-based adsorbents. Nevertheless, differences in adsorbent composition and surface properties may result in different adsorption behaviors. Therefore, the results from these studies provide comparative evidence rather than direct confirmation of the adsorption mechanism in the present system (Hasan et al., 2023)

A similar comparison was reported for MG adsorption using porous organic polymers. The Langmuir model produced an R^2 value of 0.97412, which was higher than the Freundlich value of 0.8137. This difference demonstrates that the Langmuir equation provided a better fit to the experimental equilibrium data for that adsorbent. The similarity with the present study is reflected in the higher correlation obtained from the Langmuir model compared with the Freundlich model. These findings further support the use of the

Langmuir equation to describe the equilibrium adsorption data of MG onto silica xerogel EDA/Cu under the investigated conditions (Melhi et al., 2024)

The results have important implications for the potential application of silica xerogel EDA/Cu as an adsorbent for MG removal from aqueous solutions. The theoretical maximum adsorption capacity of 200.803 mg/g provides an estimate of its adsorption performance based on the Langmuir model and can serve as a reference for further adsorbent development. Nevertheless, this study has limitations because the selection of the most suitable isotherm model was primarily based on the coefficient of determination obtained from linear regression. A higher R^2 value does not independently establish the actual adsorption mechanism or prove that adsorption occurs exclusively through monolayer formation. Further studies using nonlinear regression, additional statistical error parameters, and real wastewater samples are recommended to evaluate the adsorption behavior and practical applicability of silica xerogel EDA/Cu more comprehensively.

CONCLUSION

Silica xerogel modified with ethylenediamine (EDA) and copper ions (Cu^{2+}), synthesized from fly ash, demonstrated potential as an adsorbent for Malachite Green (MG) removal from aqueous solutions. FTIR analysis identified hydroxyl (O–H), water bending (H–O–H), and siloxane (Si–O–Si) groups in the SX-EDA/Cu sample. After contact with MG, shifts in these absorption bands and the appearance of an additional band at 1587.35 cm^{-1}, associated with aromatic ring vibrations of MG, indicated changes in the adsorbent's surface characteristics following adsorption. The highest experimental adsorption capacity reached 187.416 mg/g at an initial MG concentration of 900 mg/L. The Langmuir model provided a better fit to the adsorption equilibrium data (R^2 = 0.98267) than the Freundlich model (R^2 = 0.79245), with a theoretical maximum adsorption capacity of 200.803 mg/g. These findings highlight the potential of fly-ash-derived silica xerogel modified with EDA/Cu as an alternative adsorbent for MG removal, while further studies on adsorbent regeneration and its performance in real wastewater are needed to assess its practical application.

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