

Mechanism and Isotherm Modeling of Effective Adsorption of Malachite Green as Endocrine Disruptive Dye using Acid Functionalized Maize Cob (AFMC)

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Abstract

Cationic Malachite green has been identified as a candidate of endocrine disruptive compound found in the environment. In this study, the mechanism and isotherm modeling of effective adsorption of cationic malachite green dye onto acid functionalized maize cob (AFMC) was investigated by batch technique. The operational parameters such as initial concentration (100 – 600 mg/L); contact time (10 – 120 mins) and pH (3 – 10) influenced the removal efficiency and quantity adsorbed. Maximum of 99.3% removal efficiency was obtained at optimum conditions. AFMC physicochemical properties (surface area 1329 m²/g and particle size 300µm<Φ<250µm) enhanced its efficiency. Based on R²>0.97 and consistently low values of SSE, X², HYBRID and MSPD adsorption statistical error functions (ASEF), equilibrium data were best fitted to Freundlich isotherm. Kinetic data were best described by pseudo second-order model with consistent R²>0.98 and validated by ASEF. The mechanism of process was better described by intraparticle diffusion. Evidence of adsorption process was confirmed by change in morphology and surface chemistry determined by SEM and FTIR respectively. The performance of AFMC enlisted it as a sustainable and promising low-cost adsorbent from agro-residue for treatment of endocrine disruptive dye polluted water.

Keywords: Adsorption; EDC; Maize cob; Kinetics; Mechanism; Operational Parameters

Introduction

A healthy environment is a necessity for the actualization and realization of Sustainable Development Goals (SDG). However, the global release of Endocrine Disruptive Chemicals (EDC) through unregulated anthropogenic activities is troublesome. This Endocrine Disruptive Chemicals (EDC) have been identified as emerging contaminants that can hamper hormonal system at little doses leading to the following hazardous effects: cancerous tumors, birth defects, and developmental disorders^{1,2}. EDC can cause hormonal dysfunction, deficit brain disorder, body deformation, breast, prostate, and thyroid cancers, many cases of sexual development problems. The hazardous effects of EDC are not alien to some of the negative effects of some dyes and antibiotics released into the environment. malachite green dyes³⁻⁷.

Cationic dye of interest in this study is Malachite Green (MG). This has been reported to be toxic, calcinogenic and mutagenic. Malachite green could cause damage to humans and animals through direct inhalation and ingestion contact leading to various negative effects such as carcinogenesis, mutagenesis, tetragenesis, respiratory toxicity and reduced fertility^{8,9}. Systems and sensory organs of the body have been reported to be adversely affected by malachite green dyes^{10,11}. MG does not biodegrade easily; it is known to be resilient to fading on exposure to light and water. More so, its removal from contaminated water via common conventional techniques (biological and chemical precipitation) is tough. However, its affinity for dissociation in solution makes it prone to liquid-solid adsorption. Cleaner environment and sustainable cities as part of the sustainable development goals would be difficult to achieve if the problem arising from endocrine disruptive chemicals are not totally combatted. Some of the explored methods of waste water treatment are advanced oxidation, adsorption, photocatalytic degradation and biodegradation^{12,13}. Exceptional among this treatment technique is adsorption owing to its ease of operation, low cost, adaptation to broad range of dyes and design flexibility^{14,15}. A number of sorbents have been reported effective in uptake of this EDC dye (MG Dye): *Ocinum gratissimum*⁴, magnetic biochar¹⁶, *Opuntia ficus-indica* activated carbon¹⁷, almond gum¹⁸, *Carica papaya* wood¹⁹. Strength of selectivity and increase in the capacity of the sorbent with high removal efficiency could be enhance by biomass functionalization. This has necessitated our interest in modification of our sustainable low cost agro-residue, maize cob. As a result, in this study, orthophosphoric acid has been used to functionalized and modified maize cob as low cost agro-waste with the focus of achieving better sequestration. In this study, Acid Functionalized Maize Cob (AFMC) was developed purposely to effectively biosorb malachite green cationic dye as a candidate of endocrine disruptive chemical. Mechanistic and isotherm modeling of biosorption were explored. Statistical validity of the models using different error models was also investigated. Pre-and-post-adsorption characterization by surface morphology using Scanning Electron Microscopy (SEM) and surface chemistry by FTIR.

Materials and Methods

Acid Functionalized Maize Cob (AFMC) as Low Cost Adsorbent. Maize cobs obtained from Landmark University (Agro-based University) were screened and cleaned thereafter dried at 105 °C for 5 hours in Gen lab oven, crushed, grounded and screened to 106 µm. Acid activation was carried out following the procedure in our previous study²⁰ and elsewhere in other literature²¹ using 0.5 M ortho-phosphoric acid (H₃PO₄). Detailed typical procedure for preparation of Acid Functionalized Maize Cob (AFMC) was explicitly presented in the supplementary material (SI).

Physicochemical and Spectroscopic Characterization of AFMC

Determination of pH of AFMC. pH determination of AFMC was done by boiling 1 g AFMC in 100 mL distilled water for a period of 5 min. This was allowed to cool and its pH value was measured using ATPH-6 pH meter.

Determination of AFMC bulk density. Weight difference divided by the volume as depicted in Archimedes' principle was used for bulk density determination as depicted in Eq. 1²²

$$\text{Bulk density} = \frac{w_2 - w_1}{v} \quad (1)$$

W₁ = Weight of empty measuring cylinder,

W₂ = combination of AFMC mass and the crucible

V = volume

Determination of AFMC moisture Content. Moisture content was determined typically by introducing 5 g AFMC into initially weighed crucible and heated for 1 hr at 105 °C. Evaluation of the moisture content was done using Eq. 2⁴³

$$\% \text{ Moisture content} = \frac{w_2 - w_3}{w_2 - w_1} \times 100 \quad (2)$$

W₁ = Weight of crucible

W₂ = Initial weight of crucible with sample

W₃ = Final weight of crucible with sample

Determination of AFMC surface area by Saer's method. The AFMC surface area was determined using the Sear's method. This involves acidifying 0.5 g of each adsorbent with 0.1 M HCl to a pH 3 – 3.5. The volume was made up to 50 mL with distilled water after addition of 1 g of NaCl. The titration was carried out with standard 0.1 M NaOH at 298K to pH 4, and then to pH 9.0 following the procedure reported in the literature^{23,24}. The volume needed to raise the pH from 4 to 9 was noted and surface area evaluated using Eq. 3:

$$S(\text{m}^2/\text{g}) = 32V - 25 \quad (3)$$

Batch Biosorption Studies

Preparation of Malachite Green Adsorbate. Analytical grade reagents were used all through the study. Stock solution of 1000 mg/L MG dye (Figure 1) solution was prepared by dissolving 1 g MG salt in 1000 mL distilled water. Lower working concentration was prepared (100 – 600 mg/L) by serial dilution.

Biosorption Operational Parameters

Various Operational parameters relevant to this study was carried out following reported method^{3,4,25}. Effect of pH was determined by varying the pH values between 3 and 10 via dropwise addition of 1 M HCl or NaOH where applicable. Effect of initial MG concentration was investigated by introduction of 1 g AFMC into different

concentrations of MG dye (100 – 600 mg/L). Variation of time as done to investigate the effect of contact time from 10 to 120 minutes. All through the study, the adsorbate- adsorbent system was agitated on the Orbital shaker in order to increase effective collision in the system. Measure of residual concentration at maximum wavelength of 617 nm on done using double beam Libra Biochrom 5505 v1.0.4 PCB 1500 coupled with water peltier system UV-Vis spectrophotometer.

Theory

Biosorption Isotherm and Kinetic Modeling and Statistical Error Validity

Equilibrium biosorption data obtained from the study of were analyzed using six of two-parameter models (Freundlich²⁶, Langmuir¹⁴, Temkin²⁷, Dubinni-Raduskevich²⁸, Halsey²⁹ and Jovanovic³⁰). Similarly, both kinetics and mechanism models were fitted to Pseudo first-order³¹, Pseudo-second-order³², Elovich³³, Fractional power³⁴, Intraparticle³⁵ and liquid film³⁶ diffusion models. Estimation of the quantity adsorbed and percentage removal efficiency was done using Eq. 4 and 5³⁷⁻³⁹:

$$Q_e = \frac{(C_0 - C_e)V}{W} \quad (4)$$

$$\% RE = \frac{C_0 - C_e}{C_0} \times 100 \quad (5)$$

Presented in Table 1 and 2 are the descriptions of both isotherm, kinetics and mechanism models used in this study

Adsorption Statistical Error Function (ASRF) models. In most cases, determination of best fitting relationship and finalizing the best isotherm and kinetics model have always been through the use of linear correlation coefficient (R^2) values. However. Owing to inherent bias from this transformation, the following four rigorous statistical error function models were used: Sum of square error (SSE)⁴⁰; Hybrid fractional error function (HYBRID)²⁹; Nonlinear chi-square test (χ^2)⁴¹; Marquardt's Percent Standard Deviation (MPSD)⁴², Presented in Table 2 are the equation of the Adsorption Statistical Error Function (ASRF) Models from Eq.24 - 27.

$$\text{Sum of Square Error } SSE = \sum_{i=1}^n (q_{e,cal} - q_{e,exp})^2 \quad (24)$$

$$\text{Non-linear-chi-square test } \chi^2 = \sum_{i=1}^n \frac{(q_{e,cal} - q_{e,exp})^2}{q_{e,cal}} \quad (25)$$

$$\text{Hybrid fractional error functions } HYBRID = \sum_{i=1}^n \left[\frac{(q_{e,exp} - q_{e,cal})^2}{q_{e,exp}} \right]_i \quad (26)$$

$$\text{Marquardt's Percent Standard Deviation (MPSD) } MPSD = \sum_{i=1}^n \left[\frac{(q_{e,exp} - q_{e,cal})}{q_{e,exp}} \right]^2 \quad (27)$$

Both isotherm and kinetics data were tested with the statistical error validity models.

Results and Discussion

Physicochemical Characterization. Fig. 1 shows the structure of Malachite green as cationic dye. Presented in Table S1 of the supplementary document is the physicochemical characteristics of Malachite Green (MG) indicating that it is a cationic dye with vast application. The unique physicochemical properties of AFMC were determined and summarized in Table S2. The pH determined was 6.75, surface area (1329 m²/g), 12% moisture content, 0.386 g/cm³ bulk density and approximated particle size 300 μm < Φ < 250 μm. It has been

reported that for applicability, activated carbon in the range of pH 6 to 8 is acceptable. The pH of AFMC determined as 6.75 is suitable for activated carbon (AC). Amount of water bound to activated carbon is determined via the moisture content. Lower moisture content is desirable for active activated carbon because of the competition of the water vapour with the pores of AC. Moisture content of AFMC lower than commercial activated carbon (CAC)⁴³ is suitable. Filterability of activated carbon is determined from the bulk density.

Effect of pH. Shown in Fig. S1 is the effect of pH on biosorption of MG cationic dye onto AFMC. Ionic mobility and degree of ionization as well as the surface chemistry was influence by this operational parameter. Protonation as well as ionic competition between H^+ and MG^+ zwitterion in aqueous solution for available sites was observed between pH 2 – 5 at acidic region. Higher quantity adsorbed and removal efficiency observed between pH 6 and 8 was due to deprotonation, low competition and higher aggregate of MG^+ . 74.25% removal efficiency was observed at pH 6 and beyond this, no further increase was observed therefore pH 6 was chosen as an optimum pH. Findings here is similar to what was observed in the literature^{33,41}

Effect of Initial Concentration. Fig. S2 of the supplementary document shows the result of effect on initial concentration on effective removal of EDC cationic MG dye using AFMC. Concentration gradient developed was due to bombardment of the MG^+ surrounding the active sites. Therefore, the values of the percentage removal efficiency (%RE) increased with increase in initial MG concentrations until a saturated point was reached when all the active sites were filled up. At this point the rate of binding decreases and equilibrium was reached. There was no significant increase in the percentage removal efficiency of MG even with increase in concentration when equilibrium was reach because the saturation of the active sites. It is obvious from Fig. S2 that a rapid adsorption was observed at low concentration as a result of increase in the active sites as compared to MG molecules in the bulk. Thereafter, diffusion, convection and migration of MG molecules as a result of mass transport from the bulk lead to increase in removal efficiency until a saturated point was reached^{46,47}.

Biosorption Isotherm modeling and Statistical Validity

Understanding of the binding interaction between AFMC and MG dye solution is enhanced by the study of the isotherm models. Equilibrium data were fitted to six isotherm models namely; Freundlich (Fig. 2A), Langmuir (Fig. 2B), Temkin (Fig. 2D), Dubinin-Raduskevich (Fig. 2E), Halsey (Fig. 2F) and Jovanovic (Fig. 2G). Portrayed in Figs 2 (A-G) are the isotherm models' linear plots. Better fit with $R^2 > 0.97$ were observed in Table 3a for Freundlich, Temkin, Dubinin-Raduskevich (D-R), Halsey. Equilibrium data did not fit well to Langmuir and Jovanovic considering their R^2 value less that 0.92 (Table 3a). Both Freundlich and Halsey isotherm models describe the adsorption characteristic for heterogeneous surface. The characteristics parameters of Freundlich isotherm models are K_F (adsorption capacity) and $1/n_F$ and n_F (adsorption intensity) obtained from the linear plot of $\log Q_e$ against $\log C_e$. The function of the strength of adsorption of MG onto AFMC is determined from the parameter $1/n_F$. The value of $1/n_F$ (2.1372) being above unity is an indication of a cooperative adsorption⁴⁴ The favourability of the adsorption process of MG onto AFMC could be affirmed from the Langmuir dimensionless and separation factor (R_L). The R_L value indicates the adsorption nature to either unfavourable or unfavourable. It is unfavourable if $R_L > 1$, linear if $R_L = 1$, favourable if $0 < R_L < 1$ and irreversible if $R_L = 0$. The value of R_L which ranges between 0.00377 and 0.0744 and being less than one indicated a favourable adsorption. The Dubinin-Kaganer-Raduskevich is generally applied to determine the mechanism of the MG-dye and AFMC system with a Gaussian energy distribution onto a heterogeneous surface. The $R^2 > 0.98$ is an indication of better description of equilibrium data by The DKR mean energy (E) value being less than 8 kJ indicated that the mechanism is physisorption. Studies from Bello *et al.* (2015) on scavenging of MG onto *Citrus grandis* peels further supported this findings²⁸

Statistical Error Validity on Isotherm model. Studies have shown that determination of best isotherm model does not only depend on R^2 value. Statistical validity model has been introduced to further justify the suitability of the best isotherm model to describe the adsorption process^{50,60}. Table 3b has shown the values of the experimental and calculated quantity adsorbed, q_e, exp and q_e, cal , respectively. Four mostly used statistical validity models in adsorption studies explored are: SSE, HYBRID, X^2 and MPSD. Adsorption Statistical Error Function (ASRF) has always been the most reliably validity parameter in justifying the best isotherm model for the equilibrium studies. In order to determine the isotherm best model, coupled with the higher R^2 value, there must be closeness between the data of the q_e, cal and q_e, exp alongside a low value of the ASRF^{61,62}. Considering Table 3b, Freundlich, Temkin and Halsey isotherm models fits well into these conditions for fitness. From Table 4, the R^2 values (0.9726 for Freundlich, 0.9726 for Halsey and 0.9846 for Temkin) are closer to unity with consistent agreement between q_e, exp and q_e, cal (254.13 mg/g and 293.43 mg/g for Freundlich; 254.13 mg/g and 243.75 mg/g for Temkin; 254.13 mg/g and 293.51 mg/g for Hasley).

Effect of Contact time at various initial concentrations. Importance relevant parameter that controls the transfer and build-up of charges from the bulk to the pore active site in all transfer media is the contact time. Effect of contact time was studied from 10 – 120 minutes at six different initial concentration from 100 – 600 mg/L as depicted in Fig S3 of the supplementary document. Based on the results, rapid adsorption was observed in the first 30 minutes due increase attractive forces between the active sites and MG molecules as a result of van der Waals forces and electrostatic attractions. Between 60 and 90 mins not significant increase in adsorption capacity and removal efficiency was observed due to attainment of saturation and equilibrium. A fast diffusion onto the external surface of AFMC was followed by fast pore diffusion into the intraparticle matrix as a result of the participation of the functional groups until equilibrium was attained where 93% removal efficiency was achieved. The reaction was allowed to proceed till 90 mins beyond which to increase was observed as depicted in Fig 5. This finding is supported by the report of Hamdaoui et al., 2008⁴⁵ as well as Figen and Bayrak, 2017⁴⁶

Kinetics and Mechanism model of MG Sequestration

The rate of binding of MG onto AFMC was determine by the adsorption kinetics which also helps in gaining insight into the mechanism of the sorption process. Across various concentrations from 100 – 600 mg/L, the kinetic data were fitted to the following kinetics and mechanism models: Pseudo first-order (PFO)(Fig 3A), Pseudo second order (PSO) (Fig 3B), Elovich (Fig 3C), Fractional power (power function) (Fig 3D); Intraparticle Diffusion (Fig 3E) and Liquid film diffusion (Fig 3F). Based on the evaluated data presented in Table 4b, correlation coefficient R^2 of pseudo-second-order (>0.99) is highest among all the kinetics model explored. The R^2 value is consistently higher and increases as the concentration increases. The k_2 initial pseudo second-order adsorption rate constant increases from 23.92 to 105.26 mg/g/min suggesting a rapid kinetic process. The error bars on the kinetic plots from Figs 3A-D, showed that the kinetic models were validated using statistical error functions. The consistency of the calculated adsorption capacity (q_e, cal) with the experimental adsorption capacity (q_e, exp) coupled with lower values of the statistical error function validity data as observed in SSE, HYBRID, X^2 and MSPD further supported the PSO as the best kinetic model in this study. Presented in Fig. 3D is the Fractional power plot for adsorption of MG onto AFMC. The parameter v and k being positive, greater than unity and increase with increase in concentration suggested rapid kinetic process. The close agreement between q_e, exp and q_e, cal are indications of the best fitting of the kinetic data to fractional power model. At low concentration, the R^2 values were far away from unity, however, better regression coefficients were obtained with higher concentration indicating the applicability of the adsorbent, AFMC, to removal of pollutant at higher concentrations of MG dye

The choice of the best fit kinetic model were adjudged not only with correlation coefficient but also with the statistical error validity functions. It has been established that, the model with higher R^2 value, nearness between q_e, exp and q_e, cal and lower data of statistical error function, would be chosen as the best descriptive model⁴⁷⁻⁴⁹.

Pseudo second order fit perfectly well into this condition and thus the best kinetic model to describe the sequestration of MG dye onto AFMC. Supporting this claim is the finding of Dehbi et al., 2020⁵⁰

Figs 3E and 3F show the linear plots of Intraparticle Diffusion (IPD) and Liquid film diffusion (LFD) models. Both the rate controlling step and the diffusion mechanism were explored using IPD because its R^2 values were consistently higher than that of the LFD. IPD would be the only rate determining step if its plot begins from the origin. Contrary to this, the plot of q_t against $t^{1/2}$ did not begin from the origin hence IPD it is not the only rate determining step. However, the value of the thickness C of the adsorbent calculated from the IPD model being greater than zero across all concentrations indicated that the thickness of boundary layer participated in adsorption process. It is suggested that since $C > 0$ from the evaluated parameters in Table 4b, other diffusion model maybe involved in determining the rate controlling step⁵⁰.

Surface Morphology and Surface Chemistry Post Adsorption Characterization

Evidence of the adsorption process was justified by morphological characterization of AFMC before and after adsorption onto MG using scanning electron microscopy (SEM). More so, surface chemistry were investigated by functional groups determination using Fourier Transform Infrared (FTIR) spectroscopy. Before adsorption, dry and particle-like-crake nature with presence of pores are evident all through the micrographs at different magnifications as portrayed in Figs 4(A-B). However after adsorption as depicted in Figs 4(C-D), there was disappearance of crakes, impregnated of pores with MG dye solution and robustness of AFMC adsorbent are morphology evidence of the adsorption process.

Depicted in Figs 5 (A-B) are the FTIR spectra of AFMC before and after adsorption. Surface chemistry of AFMC before and after adsorption was investigated using FTIR. Broad band at 3390.70 cm^{-1} is attributed to O-H stretching which disappeared after adsorption as an evidence of its participation in the adsorption process. Aliphatic C-H stretching band at 2920.98 cm^{-1} was also found to decrease after adsorption. Carbonyl group, -C=O stretching vibration attributed to the lignin aromatic groups was assigned to 1714.28 cm^{-1} and 1667.29 cm^{-1} . Ascribed to -C=C- bending of the Aromatic ring are the signals observed between 1515.10 cm^{-1} – 1427.71 cm^{-1} while -CH_3 bands as a result of deformation is observed at 1372.37 cm^{-1} . Several bands between 1200 and 800 cm^{-1} are ascribed to characteristic carbohydrate bands while at 1049.93 cm^{-1} , C–O vibrational band assigned to cellulose is observed at 1049.93 cm^{-1} . Shift in bands and disappearance of functional groups confirmed their participation in the adsorption process⁵¹

Conclusion

Adsorption however has been identified as sustainable technique because it is cost effective, easy, unique, efficient, low-cost technique and it has high performance and wide pH range for removing contaminant. From this study, maize cob as agrowaste residue was successfully acid functionalized resulting in AFMC. AFMC was used to treat malachite green cationic dye synthetic waste water. Prepared AFMC was characterized by $1329\text{ m}^2/\text{g}$ surface area, 12% moisture content, 0.386 bulk density which enhanced the adsorptive capacity. Effective sequestration of cationic malachite green dye was carried out by batch technique and was found to depend on operational parameters. 84.25% removal efficiency was attained at optimum concentration between 500 – 600 mg/L at pH 6, with rapid and fast kinetics at contact time 90 minutes. Based on higher $R^2 > 0.97$ and lower statistical validity models, equilibrium data indicating heterogeneous multilayer adsorption was best described by Freundlich isotherm models as supported by Halsey and Temkin isotherm models. Physisorption adsorption process was determined from DKR free energy value less than 8 kJ. Kinetic data were best fitted to Pseudo second order with consistent higher R^2 value > 0.98 , closeness between $q_{e, \text{exp}}$ and $q_{e, \text{cal}}$ and lower values of statistical validity models (SSE, HYBRID, X^2 and MSPD). Mechanism of the adsorption process was diffusion governed. Finally, this

study is relevant to sustainable development goals because of the sustainable low cost agro-residues (maize cob) used for environmental solution in order to obtain sustainable cities and clear water environment.

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Data Availability

All data generated or analyzed during this study are included in this manuscript (and its Supplementary Information files).

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List of Tables

Types of Adsorption Models	Non-linear expression	Linear Expression	Parameters Nomenclature and description
Langmuir	$Q_e = \frac{Q_{max}K_L C_e}{1+K_L C_e}$	$\frac{1}{Q_e} = \frac{1}{Q_{max}} + \frac{1}{Q_{max}K_L C_e}$ (6)	K _L is the Langmuir isotherm constant (L.mg ⁻¹) related to the binding energy of adsorption. Q _{max} is the maximum monolayer coverage capacity (mg.g ⁻¹), R _L dimensionless separation factor indicating the nature and favourability of adsorption process. From slope and intercept of linear plot of C _e /Q _e vs 1/C _e , K _L and Q _{max} were determined.
		$R_L = \frac{1}{1+K_L C_o}$ (7)	
Freundlich	$Q_e = K_F C_e$	$\log Q_e = \log K_F + \frac{1}{n_F} \log C_e$ (8)	C _e equilibrium concentration of the MG dye adsorbate (mgL ⁻¹); Q _e amount of MG dye adsorbed at equilibrium per unit weight of AFMC (mg g ⁻¹); K _F Freundlich indicator of adsorption capacity, 1/n _F Intensity of the adsorption indicating the surface heterogeneity and favourability of the adsorption process. 1/n _F and K _F were determined from slope and intercept of linear plot of log Q _e vs log C _e
Temkin	$Q_e = \frac{RT}{b_T} \ln(A_T C_e)$	$Q_e = \frac{RT}{b_T} \ln A_T + \frac{RT}{b_T} \ln C_e$ (9)	b _T is the Temkin isotherm constant related to the heat of adsorption and A _T is the Temkin isotherm equilibrium binding constant (Lg ⁻¹). R= universal gas constant (8.314 J/mol/K) T= absolute Temperature in Kelvin. B = RT/b _T = Constant related to heat of sorption (J/mol) obtained either from intercept or slope
DKR	$Q_e = Q_{DKR} \exp^{-A_{D-R} \epsilon^2}$	$\ln q_e = \ln Q_{DKR} - A_{DKR} \epsilon^2$ (10)	Q _{DKR} is the theoretical adsorption isotherm saturation capacity (mg/g) obtained from intercept. A _{DKR} is the D-R isotherm constant (mol ² /kJ ²) related to free sorption energy obtained from the slope. ε is Polanyi potential determined by the expression = RT ln(1+1/C _e). E is the mean adsorption free energy helpful in determining the adsorption nature (physisorption or chemisorption of the adsorption process). Q _{D-R} and A _{D-R} were determined from intercept and slope of linear plot of ln q _e vs ε ²
		$\epsilon = RT \ln \left[1 + \frac{1}{C_e} \right]$ (11)	
		$E = - \left[\frac{1}{\sqrt{2A_{D-R}}} \right]$ (12)	
Halsey	$Q_e = \exp \left[\frac{\ln K_H - \ln C_e}{n_H} \right]$	$\log Q_e = \left[\left(\frac{1}{n_H} \right) \ln K_H \right] - \left(\frac{1}{n_H} \right) \ln C_e$ (13)	K _H is Halsey isotherm constant; n _H is the Halsey isotherm exponent. Both were determined from linear plot of log Q _e vs ln C _e
Jovanovic	$Q_e = Q_J [1 - \exp^{(K_J C_e)}]$	$\ln Q_e = \ln Q_{max} - K_J C_e$ (14)	K _J is Jovanovic isotherm constant (L g ⁻¹) determined from the slope of plot of ln q _e against C _e

Table 1: Adsorption Isotherm Models (Non-linear and linear models with the description of parameters)⁴⁷⁻⁵¹

Kinetic & Mechanism Models	Linear Expression	Parameters Nomenclature and description
Pseudo First order (PFO)	$\log(q_e - q_t) = \log q_e - \frac{K_1 t}{2.303}$ (15)	q _e is the quantity of adsorbate at equilibrium per unit weight of the

	$h_1 = k_1 q_e \quad (16)$	adsorbent (mgg^{-1}), q_t is the amount of adsorbed at any time (mgg^{-1}) and k_1 is the pseudo first-order rate constant (min^{-1}) and h_1 initial pseudo first-order rate constant ($\text{mgg}^{-1}\text{min}^{-1}$). q_e and k_1 were determined respectively from intercept and slope of the linear plot of $\log_e q_t$ vs t
Pseudo Second-order (PSO)	$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (17)$	k_2 is the pseudo second-order rate constant (min^{-1}) h_2 is initial pseudo second-order adsorption rate constant ($\text{mgg}^{-1}\text{min}^{-1}$). q_e and k_2 were determined respectively from slope and intercept of the linear plot of t/q_t vs t .
	$h_2 = k_2 q_e^2 \quad (18)$	
Elovich	$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln(t) \quad (19)$	q_t is the amount of adsorbate per unit mass of adsorbent at time (t), and α and β are the constants slope and intercept of the determined from the linear plot of q_t versus $\ln(t)$. α is the initial adsorption rate (mg/g-min); β is the desorption constant (g/mg) during any one experiment. The slope is $1/\beta$ while the intercept is $1/\beta \ln(\alpha\beta)$.
Fractional Power (Power Function)	$\log(q_t) = \log(k) + v \log(t) \quad (20)$	q_t is the amount of adsorbate per unit mass of adsorbent, k is a constant, t is time, and v is a positive constant (<1). The parameters v and k are obtainable from slope and intercept of a linear plot of $\log(q_t)$ versus $\log(t)$.
Intraparticle Diffusion (IPD)	$q_t = k_{id} t^{0.5} + C \quad (21)$	k_{id} is the intraparticle diffusion rate constant ($\text{mg.g}^{-1}\text{min}^{0.5}$) and C is the thickness of the adsorbent determined from slope and intercept of linear plot of q_t vs $t^{0.5}$
Liquid Film Diffusion (LFD)	$\ln(1-F) = -K_{LFD} t + C \quad (22)$	F is fractional attainment to equilibrium and K_{LFD} is the rate coefficient for particle-diffusion controlled process corresponding to the particle size of the adsorbent. $-K_{LFD}$ was determined from the linear plot of $\ln(1-F)$ vs t .
	$F = \frac{[q_t^n]}{[q_e^n]} \quad (23)$	

Table 2: Kinetics and Mechanism modeling of adsorption⁴¹⁻⁵²

<i>Type of Isotherm</i>	<i>Model parameters</i>	<i>Evaluated value</i>
Freundlich	k_f	2.1373
	$1/n_f$	2.1373
	n_f	0.4679
	R^2	0.9726

Langmuir	Parameters	Values
	$Q_{max} (mgg^{-1})$	64.5161
	$K_L (Lmg^{-1})$	0.01023
	R_L	0.140-0.494
	R^2	0.9149
Temkin	Parameters	Values
	$b_T (J mol^{-1})$	10.555
	$\beta (Lg^{-1})$	234.72
	$A_T (Lg^{-1})$	0.03078
	R^2	0.9846
D-R	Parameters	Values
	Q_d	330.135
	A_{DKR}	5×10^{-4}
	$E (J/mol)$	3.163×10^1
	R^2	0.989
Halsey	Parameters	values
	$1/n_H$	-2.1373
	n_H	-0.4679
	K_H	6.4286
	R^2	0.9726
Jovanovic	Parameters	values
	Q_{max}	13.1615
	K_j	0.0348
	R^2	0.9082

Table 3a: Isotherm models' parameters and for adsorption of malachite green onto AFMC

ASRF Models	Freundlich	Langmuir	Temkin	D-R	Halsey	Jovanovic
$q_{e, exp} (mg/g)$	254.13	254.13	254.13	254.13	254.13	254.13
$q_{e, cal} (mg/g)$	293.43	36.3703	243.75	29.81	293.51	320.7
R^2	0.9726	0.9149	0.9846	0.989	0.9726	0.9082
SSE	1544.49	47419.29	107.7444	50319.46	1550.784	4431.565
$HYBRID$	6.077559	186.5946	0.423974	198.0068	6.102327	17.43818
X^2	5.263572	1303.791	0.442028	1688.006	5.283583	13.81841
$MPSD$	0.023915	0.734249	0.001668	0.779155	0.024013	0.068619

Table 3b: Adsorption Statistical Error Function (ASRF) Data on Adsorption Isotherm Models

Kinetics Model parameters	Various Concentrations					
	100 mgL ⁻¹	200 mgL ⁻¹	300 mgL ⁻¹	400 mgL ⁻¹	500 mgL ⁻¹	600 mgL ⁻¹
<i>Pseudo first-order</i>						
q_e, exp (mg/g)	34.243	84	132.3	181.02	230.3	279.3
q_e, cal (mg/g)	13.9284	38.256	34.546	101.555	83.483	124.48
k_1 (min ⁻¹)	0.0621	0.0506	0.0414	0.05	0.0407	0.5269
h_1 (mg/g/min)	0.864954	1.935754	1.430204	5.07775	3.397758	65.58851
R^2	0.5443	0.5827	0.7156	0.6609	0.6989	0.689
SSE	412.683	2092.514	9555.845	6314.687	21555.23	23969.23
HYBRID	12.0516	24.91088	72.22861	34.88392	93.59631	85.81895
X^2	29.62889	54.69767	276.6122	62.17997	258.199	192.5549
MPSD	0.351943	0.296558	0.545946	0.192708	0.40641	0.307264
<i>Pseudo Second-order</i>						
q_e, exp (mg/g)	34.243	84	132.3	181.02	230.3	279.3
q_e, cal (mg/g)	34.1296	84.746	133.333	181.8182	232.5581	285.7143
k_2 (g/mg/min)	0.02054	0.00276	0.003583	0.00318	0.00174	0.00123
h_2 (mg/g/min)	23.9234	19.8412	63.694	105.263	94.339	101.0101
R^2	0.9986	0.9833	0.9992	0.9996	0.9989	0.9983
SSE	0.01286	0.556516	1.067089	0.637123	5.099016	41.14324
HYBRID	0.000376	0.006625	0.008066	0.00352	0.022141	0.147308
X^2	0.000377	0.006567	0.008003	0.003504	0.021926	0.144001
MPSD	1.1x10 ⁻⁵	7.89x10 ⁻⁵	6.1 x10 ⁻⁵	1.94 x10 ⁻⁵	9.61 x10 ⁻⁵	5.27x10 ⁻⁴
<i>Elovich</i>						
q_e, exp (mg/g)	34.243	84	132.3	181.02	230.3	279.3
q_e, cal (mg/g)	33.468	79.753	130.269	179.519	226.119	273.279
α (g.min ² /mg)	8.95x10 ²⁷	10.3x10 ¹⁷	9.26x10 ¹²	2.42x10 ¹²	1.93x10 ¹²	0.494x10 ¹²
β (g.min/mg)	2.0881	0.5732	0.2555	0.1758	0.1375	0.1079
R^2	0.4294	0.0791	0.8596	0.9643	0.7837	0.699
SSE	0.600625	18.03701	4.124961	2.253001	17.48076	36.25244
HYBRID	0.01754	0.214726	0.031179	0.012446	0.075904	0.129797
X^2	0.017946	0.226161	0.031665	0.01255	0.077308	0.132657
MPSD	0.000512	0.002556	0.000236	6.88E-05	0.00033	0.000465
<i>Fractional Power</i>						
q_e, exp (mg/g)	34.243	84	132.3	181.02	230.3	279.3
q_e, cal (mg/g)	33.461	79.463	130.303	179.558	226.059	273.189
ν (min ⁻¹)	0.0144	0.021	0.031	0.0329	0.0331	0.0349
k_3 (mg/g)	31.232	71.8621	112.331	153.391	192.93	231.153
$k_{3\nu}$ (mg/g/min)	0.449741	1.509104	3.482261	5.046564	6.385983	8.06724
R^2	0.4325	0.0651	0.8676	0.9686	0.7911	0.7043
SSE	1118.675	6311.031	16970.79	32229.26	51087.71	74613.16
HYBRID	33.43221	79.42101	130.241	179.4922	225.9928	273.1192

X^2	77685.77	300525.3	547445	979612.8	1543435	2137913
$MPSD$	0.999139	0.999472	0.999524	0.999634	0.999707	0.999745

Table 4a: Parameters of kinetics models with associated Statistical Validity Data.

Mechanism model parameters of adsorption of MG onto FMC						
Mechanism models Parameters	Various Concentrations					
	100 mgL ⁻¹	200 mgL ⁻¹	300 mgL ⁻¹	400 mgL ⁻¹	500 mgL ⁻¹	600 mgL ⁻¹
<i>Intraparticle Diffusion</i>						
$k_p(\text{mg/g/min}^{0.5})$	0.1671	0.7151	1.3105	1.8328	2.481	3.209
C	31.772	72.793	116.82	160.5	200.79	240.6
R^2	0.513	0.1304	0.945	0.9823	0.8944	0.822
<i>Liquid Film Diffusion</i>						
	100 mgL ⁻¹	200 mgL ⁻¹	300 mgL ⁻¹	400 mgL ⁻¹	500 mgL ⁻¹	600 mgL ⁻¹
K	0.0628	0.0506	0.0421	0.0679	0.0487	0.0521
C	0.899	0.814	1.343	0.578	1.015	0.808
R^2	0.544	0.583	0.716	0.661	0.699	0.689

Table 4b: Mechanism model parameters of adsorption of MG onto AFMC