Spectral, Isotherm, Kinetic, and Thermodynamic Studies of Malachite Green Dye Adsorption from Aqueous Solutions onto Low-cost Treated Kaolin

A. Moumen[a,d,*], Y. Belhocine[b,*], F. Mechati[a], M. Karout[c], D. Charime[c], W. Boultif[b] and Z. Hattab[d]

[a]*Department of Petrochemical, Faculty of Technology, University of 20 Août 1955 Skikda, Algeria*
[b]*Laboratory of Catalysis, Bioprocess, and Environment, Department of Process Engineering, Faculty of Technology,*
University of 20 August 1955, Skikda 21000, Algeria
[c]*Department of Process Engineering, Faculty of Technology, University of 20 August 1955, Skikda 21000, Algeria*
[d]*Laboratory of Water Treatment and Valorization of Industrial Wastes, Department of Chemistry, Faculty of Sciences, Badji-Mokhtar*
University, B.P.12, Annaba 23000, Algeria

This study investigated the potential of Algerian kaolin clay exchanged with protons as an inexpensive adsorbent for malachite green (MG) dye elimination from colored effluents using batch equilibrium experiments. Before using the kaolin as an adsorbent, it was treated with HCl (0.1 M). The structural, compositional, and textural characteristics of the Tamazert kaolin (TK) and the treated Tamazert kaolin (tTK) were determined using physicochemical and mineralogical characterizations. The optimization of miscellaneous experimental parameters such as contact time, adsorbent dose, temperature, and initial pH solution was conducted through batch adsorption experiments. The findings of the experiment revealed that 30 min of contact time was adequate to achieve liquid-solid phase equilibrium. The highest adsorption capacity of MG dye was reached at 18.51 mg g^{-1}. The adsorption kinetics followed the pseudo-first-order model. The adsorption isotherms showed a correlation between the Freundlich model and the experimental results. Enthalpy changes, Gibbs free energy, and entropy were computed.

Keywords: Kaolin, Adsorption, Degradation, Malachite green, Dye removal

INTRODUCTION

Synthetic dyes find extensive use in numerous industries, including paper, textile dyeing, printing, food, plastic, pharmaceuticals, and leather [1-10]. These sectors employ a significant amount of dyes and produce large amounts of effluents, which are important sources of pollution. Their presence is regarded as the riskiest and the most damaging to the environment when compared to all other sectors because dyes are primarily consumed by the textile industries in each of their products [11-13]. Therefore, due to their toxic and non-biodegradable nature, it is essential

to remove the pollutants and colored effluents from wastewater to preserve human health and the environment [14-16]. There are miscellaneous types of dyes, namely acidic, reactive, basic, azo, diazo, *etc.* [1].

Malachite green (MG) dye constitutes a serious risk to the environment and is difficult to be removed from aqueous solutions [17]. MG is extensively used in the dyeing textile industry, as well as in the aquatic business as a fungicide and bactericide [18-20]. Although it is widely used, MG is a very controversial substance due to its alleged dangerous characteristics, which are known to have harmful effects on the respiratory system [21], and to cause hepatic toxicity, malignancy, anemia, and thyroid tumors [18-20].

Many different approaches have been used in wastewater treatment, including chemical oxidation,

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*Corresponding authors. E-mail: abdo_moumen@yahoo.fr; y.belhocine@univ-skikda.dz

coagulation and flocculation, membrane separation, and advanced oxidation [21-29]. However, these methods are either technically challenging or not economically viable [30-32]. For example, advanced oxidation, because it requires very high temperatures and pressures, leads to costly practical applications [33,34], and for coagulation, the formation of colloids during coagulation leads to the contamination of wastewater [35].

Among these approaches, the adsorption process is considered an appropriate procedure to remove tiny particles, molecules, or ions from wastewater [36]. The adsorption process is simple, quick, inexpensive, and non-toxic [37,38].

An adsorption study using kaolin clay was carried out in this investigation as an effective, convenient, eco-friendly, and economical adsorbent to remove the MG dye from polluted water [29,39-41]. In recent years, numerous types of adsorbents were prepared from several clays like bentonite, kaolin, montmorillonite, sepiolite, *etc.*, and have been employed to remove dyes from wastewater [42]. Due to its low price and wide availability, clay is an appropriate alternative to other materials. Their structure and composition affect their physicochemical characteristics such as specific surface area, ion exchange capacity, and reactivity [43,44]. In addition, the exchangeable ions can help to eliminate organic and inorganic pollutants from wastewater [45].

Kaolin is a mineral clay made up of kaolinite $Al_2Si_2O_5(OH)_4$, a substance that has been utilized in a range of technical applications [46-55]. Kaolin clay is a promising adsorbent that is gaining popularity due to its low cost, environmental friendliness, and abundance [56]. Furthermore, due to its electrostatic nature, which is promoted by the charge heterogeneity at its surface, kaolin is designated as a good natural adsorbent [57,58]. On the other hand, some studies showed that the edges possess both alumina (Al_2O_3) and silica (SiO_2) sites, which act as ion exchange sites whose charges could change with the pH of the medium [58]. Many studies have investigated the ability of natural and modified kaolin clays originating from different countries to remove various dyes such as MG from aqueous solutions [56].

The potential use of Algerian-treated kaolin as an adsorbent for the elimination of MG dye from an aqueous solution was explored by X-ray diffraction (XRD), X-ray Fluorescence (XRF) and Fourier-transform infrared spectroscopy (FTIR). Batch studies were performed and optimized using several parameters such as adsorption temperature, pH solution, mixing time, and adsorbent load. Adsorption isotherms were investigated using Freundlich and Langmuir models, as well as adsorption kinetics using pseudo-first and second-order kinetic models and thermodynamic characteristics.

MATERIALS AND METHODS

Materials

The Tamazert Kaolin (originating from North-East-Algeria) was provided by the company of sanitary ceramics (El-Milia). All other chemicals employed in this work were of analytical reagent grade and used without any further purification. Hydrogen chloride (HCl) and malachite green (MG) were purchased from Merck Chemical Company. The 3D molecular structure of the MG (dye) was retrieved from the PubChem database server [59] and represented in Fig. 1 using Jmol [60].

Methods

Preparation of clay adsorbent and dye solution. The treated kaolin was prepared by the following method: Raw kaolin was washed with distilled water and H_2O_2 solution at 70 °C, and then treated with HCl (0.1 M) for two days at room
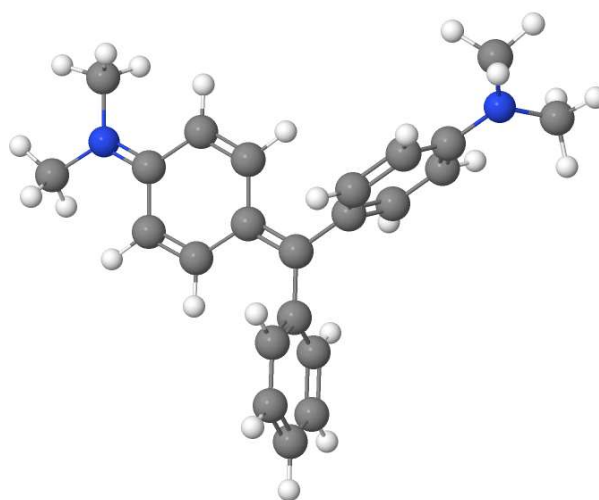

Fig. 1. Molecular structure of MG (cationic dye).

temperature until saturation [61]. After that, filtration was used to separate the sample (treated kaolin), which was then dried at 105 °C. This procedure was repeated until the chloride ion was no longer present, as confirmed by adding a drop of 0.1 M $AgNO_3$ to the filtrate. The desired amount of dye was dissolved in distilled water to make a stock dye solution of 1000 mg l^{-1}.

Batch adsorption experiments. The performance of the kaolin clay as an adsorbent in the degradation of MG was evaluated. All the samples were prepared and placed in 250 ml glass flasks containing 50 ml of the MG solution with C = 20 mg l^{-1}, then they were stirred at a speed of 300 rotations per minute. By varying one parameter while leaving the others constant, the effects of various parameters such as contact time, pH, adsorbent dose, and temperature were explored. The adsorption isotherms and kinetics were also examined to understand how these factors affect the adsorption capacity. The ranges of experimental variables that were explored are as follows: pH of the prepared solutions (2, 4, 6, 8, and 10), the adsorbent dosage values ranging from 0.1 to 1 g, contact time (5, 10, 15, 20, 25, 30, 35, and 40 min), and temperature (20, 30, and 40 °C). Single measurements were obtained every 5 min at regular intervals and examined by UV spectroscopy. The plot of absorbance versus concentration is shown in Fig. 2.

The solution concentration allowed us to calculate the quantities q_t and q_e

$$q_e = \frac{C_0 - C_e}{m} V \qquad (1)$$

$$q_t = \frac{C_0 - C_t}{m} V \qquad (2)$$

Where C_0 and C_e refer to the initial and equilibrium dye concentrations in solution (mg l^{-1}), V is the volume of the solution in liter and is the mass of adsorbent in g, C_0 and C_t are the dye concentration at t = 0, and at any time in mg l^{-1} respectively.
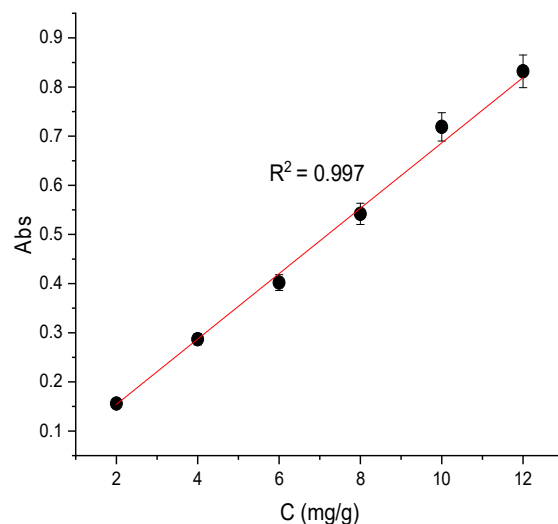

Fig. 2. Calibration curve for the standard solution of MG Dye

Characterization of the samples. The elemental composition of the kaolin sample was determined by XRF analysis with a Siemens-type apparatus (SRS 3000). The powder XRD patterns of the crystalline structure were recorded on a PANalytical X'Pert PRO diffractometer. The infrared spectra (FT-IR) of TK and tTK were obtained using a Thermo Scientific Nicolet iS10 spectrometer with wavelengths ranging from 4000 to 500 cm^{-1}.

RESULTS AND DISCUSSION

Experimental Characterization of TK and tTK

The chemical composition of TK. The chemical composition analysis of TK is reported in Table 1. The results show that TK is rich in oxides of silica SiO_2 (43.82%) and aluminum Al_2O_3 (36.66%), the remaining oxides are present in small amounts. The loss on ignition (LOI) of pure kaolin was 16.36%.

Table 1. Chemical Composition (wt.%) of the TK

Oxides	SiO_2	Fe_2O_3	Al_2O_3	$CaCO_3$	CaO	MgO	MnO	LOI
(wt.%)	43.82	0.55	36.66	1.35	1.05	0.05	0.1	16.36

FT-IR and XRD Analysis of TK and tTK

The FT-IR spectra of TK and tTK in the 500-4000 cm^{-1} region are presented in Fig. 3. The peaks around 3600 cm^{-1} as well as those at 937 and 912 are ascribed to hydroxyl groups. The bending modes of the water molecules are responsible for the band appearing around 1670 cm^{-1}. The Si-O and Si-O-Si stretching vibrations are assigned to the broad bands at 1119 and 1032 cm^{-1} [62]. After chemical treatment, no remarkable changes were found as shown in Fig. 3.

The XRD diffractograms of TK and tTK obtained after chemical treatment are compared in Fig. 4. The primary phase of the TK sample is kaolinite, with a small number of crystalline quartz phases (2θ = 12, 20, 22, 25, and 56°) and muscovite. Protons replaced the interlayer cations during acid treatment.

In addition, acid treatment with HCl (0.1 M) dissolves major impurities such as calcite and dolomite, as well as traces of smectite and illite/smectite interlayered in tTK, thus resulting in its higher crystallinity [63]. The mineral structure of the clay is not influenced by the modest acid concentration (0.1 M) used in this work. The surface was substantially cleaned for better adsorption without damaging the mineral structure of the clay.

The Effect of Different Experimental Parameters

The potential of our prepared adsorbent in the degradation of MG dye was evaluated by considering the following parameters: contact time, adsorbent dose, temperature, and pH values.

MG removal rate as a function of the contact time. The impact of contact time on the removal rate of MG dye is examined in Fig. 5.

Figure 5 shows that the removal rate of the pollutant by the adsorbent was fast until the optimum time, and then it slowed down and finally reached saturation after reaching equilibrium. At the flow rate for t = 20 min, the removal efficiency rate was higher due to the large adsorbent surface area available for MG adsorption, then, only a small increase in MG removal was observed over time. The contact time studied revealed that the adsorption capacity was at its highest after 30 min.

The effect of pH. According to Fig. 6, the adsorption of the MG as a function of pH indicates that the amount of adsorbed MG increased from 9.01 mg g^{-1} at pH = 2 to
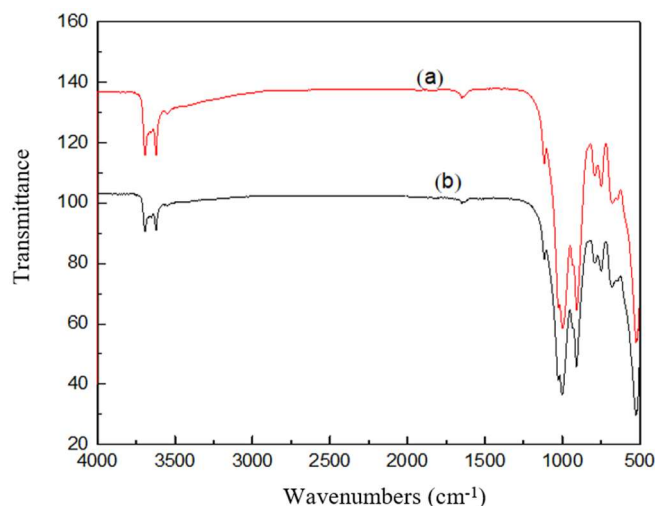

Fig. 3. IR spectra of (a) TK and (b) tTK.

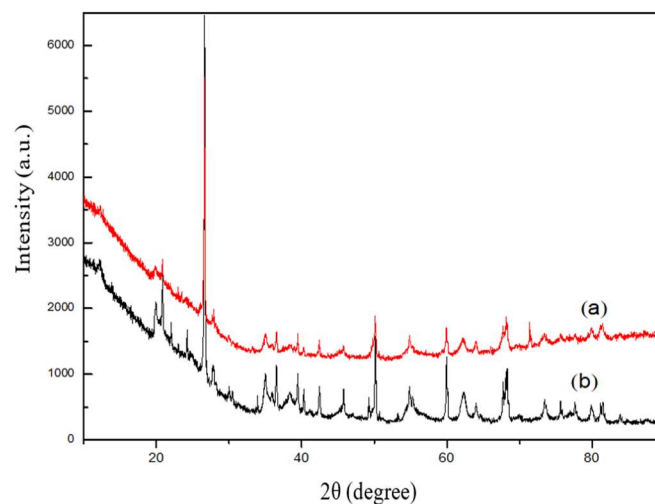**Fig. 4.** XRD Diffractograms of (a) TK and (b) tTK.

9.49 mg g^{-1} at pH = 10. The electrostatic repulsion between the positively charged MG ion and the positively charged sites on the adsorbent surface may be the reason for the drop in adsorption at low values of pH. Higher pH values resulted in a negatively charged adsorbent surface, therefore, favoring the adsorption of positively charged dye cations *via* electrostatic attraction. As a result, higher pH values favor the adsorption of MG dye. A similar trend was observed by Ullah *et al.* [64], Bello *et al.* [65], Oyelude *et al.* [66], Shirmardi *et al.* [67], and Mounin *et al.* [68]. In the rest of this study, the pH value was fixed at 10.
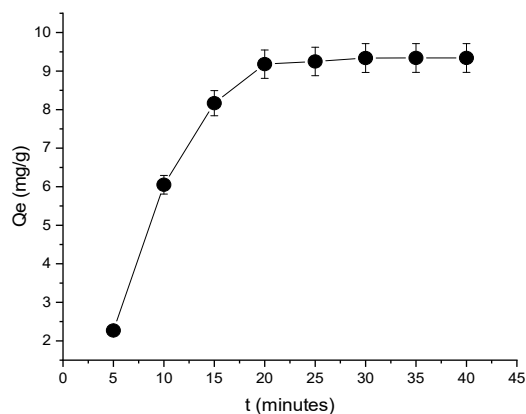
Fig. 5. Evaluation of the contact time effect on the adsorbed quantity. (Reaction conditions: adsorbent dose = 0.1 g, temperature = 30 °C and stirring speed = 300 rpm).
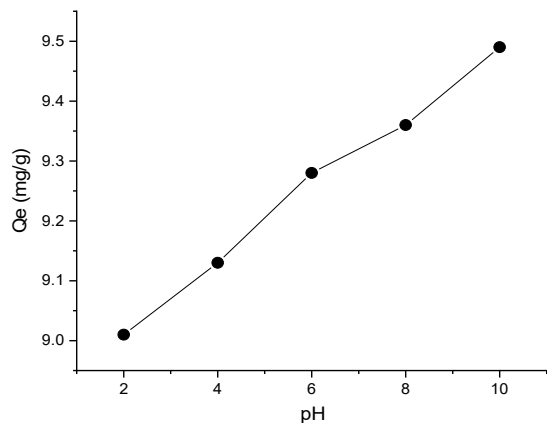

Fig. 6. Effect of pH on the adsorbed quantity. (Reaction conditions: adsorbent dose = 0.1 g, temperature = 30 °C, stirring speed = 300 rpm, initial concentration = 20 mg l^{-1}, and contact time = 30 min).

The effect of temperature. The effect of temperature on the adsorbed quantity of MG as a function of time is illustrated in Fig. 7. All the tests were conducted at 20, 30, and 40 °C with all other variables held constant.

The results show that the adsorption capacity of tTK for MG increases with the rising of temperature from 20 to 30 °C. However, rising the temperature from 30 to 40 °C, resulted in a modest reduction in the adsorbed amount of MG.
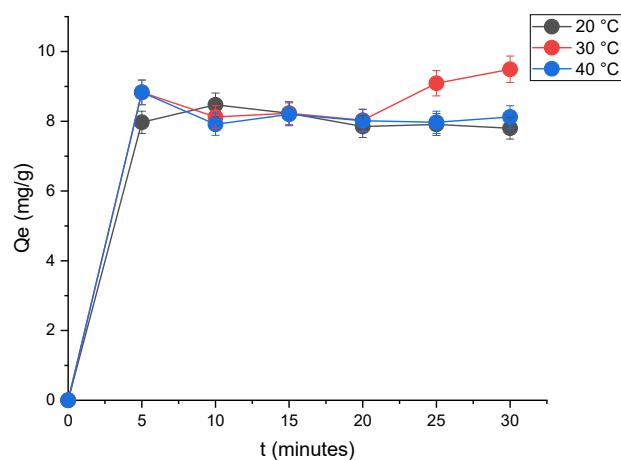

Fig. 7. Amount of MG adsorbed on tTK at different temperatures. (Reaction conditions: adsorbent dose = 0.1 g, pH = 10, stirring speed = 300 rpm, initial concentration = 20 mg l^{-1}, and contact time = 30 min).

Similar findings were reported by Emmanuel *et al.* [69] and Yusop *et al.* [70]. At lower temperatures, the removal of MG from the aqueous solution was more efficient. As a result, the optimum temperature for MG adsorption was found to be 30 °C, indicating an exothermic adsorption process [71].

Effect of the adsorbent dose. Figure 8 illustrates the adsorption capacity of MG as a function of the adsorbent dose.

Increasing the dose of tTK from 0.1 g to 0.3 g resulted in a rapid decrease in the amount of adsorbed MG. However, the amount of MG adsorbed on the tTK decreased slightly when the dose of adsorbent was increased beyond 0.3 g (Fig. 8), in agreement with similar works [72,73]. In summary, increasing the dose of adsorbent above 0.1 g increased the number of active sites; as the amount adsorbed increases, particle agglomerations may occur, reducing the total adsorption surface area, and therefore, the amount of MG adsorbed.

Adsorption Isotherms and Kinetic Studies

The correlation at equilibrium between the concentrations of the solid phase of the adsorbent and the solution phase was carried out by adsorption isotherm experiments. The suitability of the adsorption of MG dye was tested by several adsorption isotherm models. Freundlich and
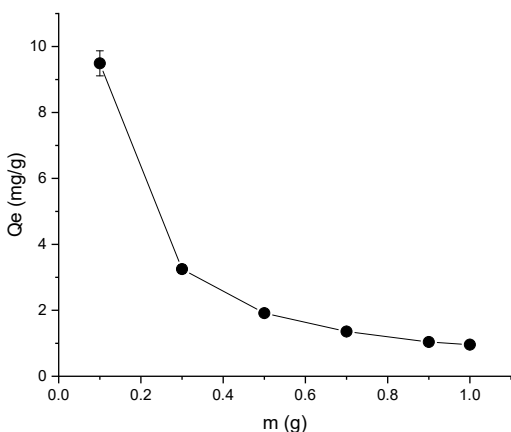
Fig. 8. Amount of MG adsorbed on tTK for different masses of adsorbent (Reaction conditions: pH = 10, temperature = 30 °C, stirring speed = 300 rpm, initial concentration = 20 mg l^{-1}, and contact time = 30 min).

Langmuir isotherms are among the most frequently utilized models [74].

Isotherm of Langmuir. According to the Langmuir model, monolayer adsorption happens on a homogeneous surface, at certain sites [75]. The Langmuir isotherm linear form can be represented by the following equation:

$$\frac{C_e}{Q_e} = \frac{1}{K_L Q_m} + \frac{C_e}{Q_m} \tag{3}$$

Where C_e is the equilibrium dye concentration (mg l^{-1}), Q_e (mg g^{-1}) is the amount of MG adsorbed at equilibrium, Q_m (mg g^{-1}) is the amount of MG adsorbed at saturation and K_L is Langmuir constant (l mg^{-1}). The Langmuir isotherm may be described in terms of a dimensionless separation factor R_L, as follows:

$$R_L = \frac{1}{1 + K_L C_0} \tag{4}$$

Where C_0 is the maximum initial concentration (mg l^{-1}). The value of R_L points to the shape of Langmuir isotherm to be either unfavorable ($R_L > 1$), linear ($R_L = 1$), irreversible ($R_L = 0$), or favorable ($0 < R_L < 1$).

Isotherm of Freundlich. The adsorption on heterogeneous surfaces can be described by the Freundlich

isotherm model [76]. The Freundlich isotherm linear form is expressed by the following equation:

$$ln\ Q_e = ln\ K_F + \frac{1}{n}\ ln\ C_e \tag{5}$$

Where K_F is the Freundlich constant which measures the adsorption capacity and $\frac{1}{n}$ is a constant that indicates the adsorption intensity of the system.

Both kinetic models (pseudo-first-order and pseudo-second-order) were used to investigate the adsorption process of MG on tTK.

The Langmuir isotherm for the adsorption of MG is shown in Fig. 9. The Langmuir isotherm is a uniform distribution on the surface of the adsorbent. Additionally, the Langmuir model has also been evaluated through equation y = 0.054x + 0.429. The plot of C_e/q_e as a function of C_e adequately satisfies the Langmuir isotherm due to the large value of the coefficient of determination R^2 (0.973). Langmuir isotherm constants (Q_m, K_L) were determined from the straight line slope and intercept, as reported in Table 2. The Freundlich linear isotherm plot of lnQ_e *vs.* lnC_e for MG adsorption on tTK is illustrated in Fig. 10.

According to the plot of lnQ_e *vs.* lnC_e (Fig. 10), the coefficient of determination R^2 for the adsorption of MG dye on tTK is close to 1 (0.998), showing a better correlation with the Freundlich model. The isotherm Freundlich constants, K_F, and n, are obtained from the plot's interception and slope, respectively (Table 2).
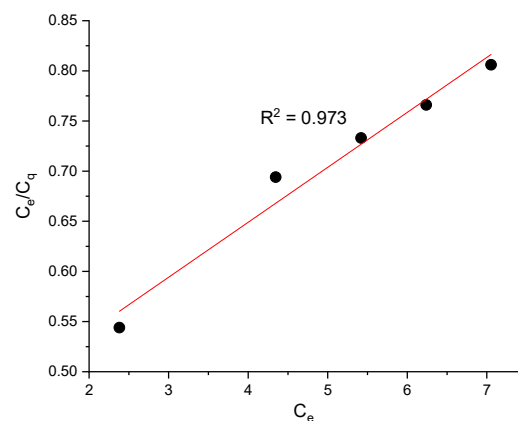

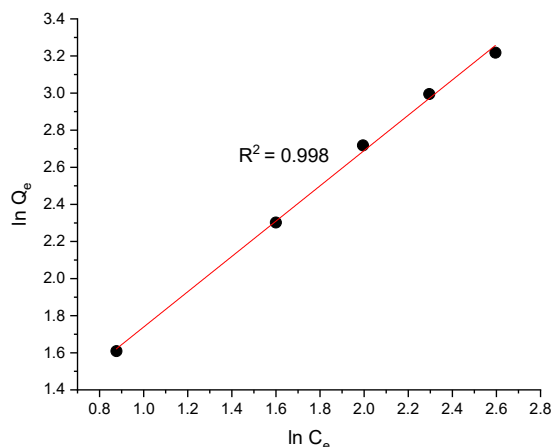**Fig. 9.** Langmuir linear isotherm for MG adsorption on tTK.

Fig. 10. Freundlich linear isotherm for MG adsorption on tTK.

Table 2. The Parameters of Langmuir and Freundlich Equations

Models	Parameters	Values
Langmuir	Q_m (mg g^{-1})	18.518
	K_L (l g^{-1})	0.125
	R^2	0.973
	R_L	0.284
Freundlich	$1/n$	0.951
	K_F (mg g^{-1} (l g^{-1})$^{1/n}$)	2.139
	R^2	0.998

Table 2 shows the parameters of the Langmuir and Freundlich equations. The Freundlich model appears to better suit the experimental data. The size of the exponent $1/n$ denotes the favorability of the solid adsorbent for the adsorbate being examined, if $1/n < 1$ then the adsorption is favorable. According to the obtained results, the Freundlich model was the best-suited isotherm for the adsorption of MG on the tTK.

The pseudo-first order. The pseudo-first-order equation [77] can be written as:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \tag{6}$$

Where: t is the time (min), q_e and q_t are the amount of MG

adsorbed (mg g^{-1}) at equilibrium and at any time respectively, and k_1 is the rate constant of adsorption (min^{-1}).

The pseudo-second-order. The pseudo-second-order model [78] is represented by the following equation:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \tag{7}$$

Where: k_2 is pseudo-second order constant (g mg^{-1} min^{-1}) The results of pseudo-first order and pseudo-second order kinetic models are presented in Figs. 11 and 12.
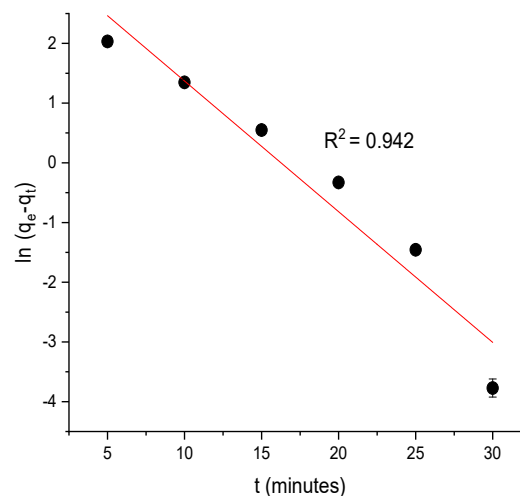

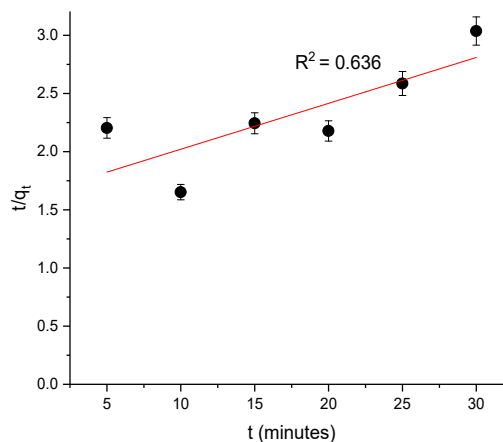**Fig. 11.** Pseudo-first-order model of adsorption kinetics.

Fig. 12. Pseudo-second-order model of adsorption kinetics.

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Table 3. Kinetics Parameters for MG Adsorption on tTK

Pseudo-first-order			Pseudo-second-order		
q_e	k_1	R^2	q_e	k_2	R^2
(mg g^{-1})	(min^{-1})		(mg g^{-1})	(g mg^{-1} min^{-1})	
9.674	0.218	0.942	20	0.001	0.636

According to the results obtained using the two kinetic models, the pseudo-first-order model provided a high correlation coefficient (R^2) value of 0.942 for the tTK, while the pseudo-second-order model produces an R^2 of 0.636. The q_e and k values of both models were determined from the slope and the ordinate at the origin of the plot of t/q_t against t and $\ln(q_e - q_t)$ *versus* t respectively, as reported in Table 3.

The correlation coefficient (R^2) for the pseudo-first-order 0.942 is greater than that of the second-order model 0.636 (Table 3). The results showed that the adsorption process is better described by the pseudo-first-order model, thus indicating that the rate-determining step may be a physical process.

THERMODYNAMIC PARAMETERS

The experimental results at different temperatures (20, 30, and 40) were used to compute the thermodynamic parameters of the adsorption of MG on tTK. The following equations were used to calculate the standard change in Gibbs free energy ($\Delta G°$), enthalpy ($\Delta H°$), and entropy ($\Delta S°$).
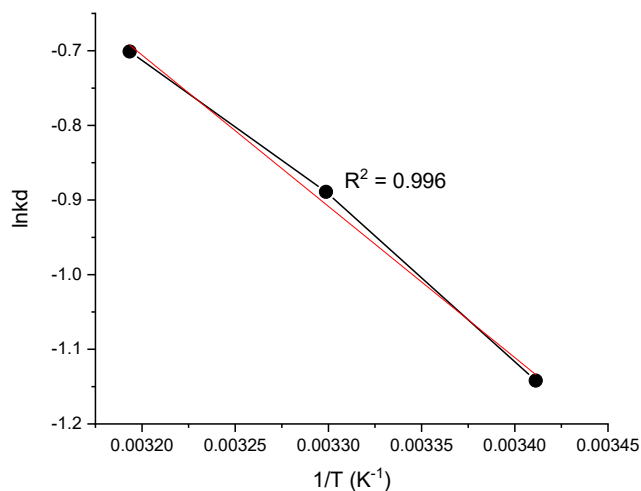
$$\Delta G° = -RT \ln K_d \qquad (8)$$

$$\Delta G° = \Delta H° - \Delta S° \qquad (9)$$

$$\ln k_d = \frac{\Delta S°}{R} - \frac{\Delta H°}{RT} \qquad (10)$$

Where K_d is the equilibrium constant. The values of ($\Delta S°$) and ($\Delta H°$) are measured from the slope and intercept of the linear plot of $\ln K_d$ as a function of (1/T) as illustrated in Fig. 13.

The thermodynamic parameters for the adsorption of MG on tTK are reported in Table 4.

Fig. 13. Linear plot of thermodynamic parameters.

Table 4. Thermodynamic Parameters for the Adsorption of MG on tTK

T	$\Delta H°$	$\Delta S°$	$\Delta G°$
(K)	(kJ mol^{-1})	(J mol^{-1} K^{-1})	(kJ mol^{-1})
293			2.78
303	16.85	48.05	2.24
313			1.82

According to Table 4, the positive value of the change in $\Delta G°$, $\Delta H°$, and $\Delta S°$ suggests an endothermic non-spontaneous adsorption process [79,80]. The value of the enthalpy ($\Delta H°$) was found to be less than 40 kJ mol^{-1}, which is indicative of the physisorption process of MG dye on the tTK surface [81].

To highlight the efficiency of the studied treated kaolin for MG dye removal from aqueous solutions, a comparison with other adsorbents reported in the literature was made in Table 5, where it can be seen that tTK has one of the highest q_m values with a short contact time, and small sample volume, implying its potential use as an efficient adsorbent for MG removal.

CONCLUSIONS

The capacity of treated Algerian kaolin to act as a good adsorbent for MG dye elimination was established in this

Table 5. Comparison of Maximum Adsorption Capacity of tTK Adsorbent with other Adsorbent Materials for the Removal of MG

Adsorbents	Maximum adsorption capacity (mg g^{-1})	Contact time (min)	Sample volume (ml)	Ref.
Cellulose powder	2.42	30	100	[82]
Tamarind fruit shell	1.95	60	100	[71]
Commercial activated carbon	8.27	240	50	[83]
Potato stem powder	27.0	18	50	[84]
Casuarina equisetifolia needle	77.6	180	20	[85]
Persian kaolin	52	60	250	[86]
Bentonite clay	8	10	100	[87]
Catha edulis stem	5.62	60	50	[88]
Brazilian kaolin	128	240	100	[56]
Neem sawdust (Azadirachtaindica)	4.3	14	50	[89]
Coconut shell-based activated carbon	214.63	120	100	[65]
tTK	18.51	30	50	This work

study. Batch experiments were used to investigate the effects of pH value, contact time, adsorbent mass, and temperature. The ideal parameters for obtaining the largest quantity of adsorption were found to be pH 10, mass 0.1 g, and temperature 30 °C; additionally, the equilibrium of the adsorption can be reached in 30 min. The adsorption process follows the Freundlich isotherm model. The maximal adsorption capacity observed for the treated kaolin was 18.51 mg g^{-1} at ambient temperature. According to the kinetic study, the pseudo-first-order equation was the appropriate model for describing the kinetics adsorption of MG. Furthermore, thermodynamic parameters show that the entire adsorption process is endothermic. These results demonstrated the effectiveness of acid-treated kaolin for MG removal from water.

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