

Comparative Adsorption of Methyl Orange Color from an Aqueous Solution Using Activated Carbon

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ABSTRACT

Background and Aim: The study was focused on the adsorption parameters of Methyl Orange dye (MO) onto Commercial Activated Carbon (CAC) in aqueous solution. The examined parameters in the adsorption process were founded on the optimization. **Materials and Methods:** Langmuir, Freundlich and Temkin models were studied at different temperatures. Time, pH, and adsorbent mass adsorption parameters were examined at 140 mg.L⁻¹ MO dye concentration at 25°C. PFO and PSO models were studied at 140 mg/L dye concentration. **Results and Discussion:** The adsorption capacity q_e of AC was 42.6 mg.g⁻¹ at 25°C. Among the isotherm models studied, the Freundlich model was found to be the optimum model with adsorption process data. Parameters in the model were found to be 0.0054 L.mg⁻¹ (K_L), 12.76 mg.g⁻¹ (q_m), 0.7827 (R^2) for the Langmuir isotherm model and 49 (1/n), 42.6 mg.g⁻¹ (K_f), 0.0680 (R^2) for Freundlich isotherm model and 33.12 mg.L⁻¹ (A), 36.37 J.mol⁻¹ (B), 0.7992 (R^2) for Temkin isotherm model. Thermodynamic studies indicated that the adsorption process was exothermic. The adsorption enthalpy (ΔH) for MO dye was found to be -63248 KJ.mol⁻¹. The negative value found for the (ΔG) indicated that adsorption of MO dye was spontaneous adsorption. The kinetic study of MO dye adsorption obeyed the Pseudo Second-Order (PSO) model. **Conclusion:** CAC is distinguished as an adsorbent that excels other adsorbents in removing the MO dye from aqueous solutions compared to the mentioned literature due to its chemical and physical properties.

Keywords: Methyl Orange (MO), Activated carbon (AC), Thermodynamics, Adsorption isotherm models, Kinetics models.

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INTRODUCTION

Synthetic dyes were monitored in wastewater from various industries, such as food processing, cosmetics, plastics, and medicines.¹⁻⁴ Usually, synthetic dyes have a hard chemical structure and are highly stable to light, oxidation, and biodegradation.^{5,6} Removing the dye from wastewater is a necessary practice with the increased use of dye in industry, which harms the environment.⁵ Dyes are insoluble and highly toxic compounds. If it is drained directly into the receiving waters, it poses an environmental hazard, and is toxic to aquatic life as a hazardous pollutant.⁷ They may be mutagenic and carcinogenic, posing serious health risks to humans such as damaged kidney, liver, brain, and Central Nervous System (CNS) function.⁸ According

to the reports, the MO dye is toxic and causes irritation when it comes into contact with the skin, as well as when it is swallowed or inhaled.⁸ Figure 1 shows the structure of MO⁸ MO is an anionic dye used in different industries, such as pharmaceutical industries, research laboratories, and food industries.⁹ There are several methods reported to remove MO dye in an aqueous solution such as physical and chemical treatments, coagulation, and chemical oxidation.¹⁰⁻¹⁴ Among the methods, the sorption method is immensely applied in manufacturing processes for the separation, removal, and purification of dyes from aqueous solutions.^{15,16} It is an economical, simple, and efficient method.¹⁵⁻¹⁹ The most popular adsorbent for adsorption processes is AC, which is popular due to its efficiency, adaptability, wide adsorption capacity, surface area, and microporous structure. Therefore, AC is a good adsorbent.²⁰⁻²² Examining MO dye adsorption characteristics on AC under various experimental settings was the primary goal of this work. Together with adsorption kinetics and desorption, the effects of concentration, time, pH solution, and other variables were also investigated.



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MATERIALS AND METHODS

Analytical grade reagents were used throughout this project, and all materials were used directly out of the package. MO was purchased from Loba Chemie in India. Nen Tech Ltd., (UK) sells Activated Carbon (AC) with a particle diameter, surface area, and pore volume of 300-500 m, 500-3000 m².g⁻¹, and 1.4 cm³.g⁻¹, respectively. Al-Degs *et al.* revealed the physiochemical characteristics of activated carbon.²³ Because of their numerous bulky functional groups and high specific surface area (820 m²/g), activated carbons can absorb significant amounts of the dye MO.^{24,25} A Spectrophotometer (Japan) Optima SP-3000 nano was used to determine the residual concentration of MO dye in solution and an Ezdo pH meter-pp-201 was used to measure pH (Taiwan). This experiment made use of a water bath shaker from Daihan Scientific in Korea.

Adsorption of the MO dye

The batch approach was used to research the dye's adsorption characteristics. 100 mL of the MO dye were combined with an adsorbent mass of AC, and the mixture was stirred for a predetermined amount of time. The amount remaining of MO dye adsorbed by the activated carbon could be determined by the difference between the initial and final concentrations after the final dye Concentration (Ce) in the aqueous solution was measured. Following the investigation described below, the effects of carbon mass, MO concentration, pH, ionic strength, and temperatures on MO dye adsorption were examined. At 480 nm, the MO dye concentrations that were still in solution were measured spectrophotometrically. Using a series of standard samples with concentrations between 20-200 mg.L⁻¹, a linear calibration curve was produced. With a correlation coefficient squared (R²) of 0.9924, a linear calibration graph was produced. Every experiment was done in triplicate, and the standard deviation was used to calculate the measurement uncertainty, which was always judged to be fewer than 2%.

Effects of the MO dye on the contact time, pH, mass and temperatures of the adsorbent of the solution

The following approach was used to examine how agitation time affected the adsorption of the MO dye: 100 mL solutions containing 140 mg.L⁻¹ of MO dye were first mixed with 50 mg of AC and stirred for periods ranging from 0 to 225 min and pH 7 at 25°C. The final dye concentration was calculated. The agitation duration employed in the following studies was determined to be 125 min, which was adequate to yield a reasonable value of the Kd. The following adsorption investigations were conducted: In a 100 mL volumetric flask, 140 mg.L⁻¹ of MO dye concentration was prepared. The final MO dye concentration was calculated after stirring the flask for 120 min. The acidity of the solution's effect on MO dye adsorption, the effects of pH (1.0, 3.0, 4.8, 5.7, 7.0, 9.0, 10.0, and 12.0), and activated carbon mass (25, 50, 75, 100, 125,

150, 175 and 200 mg) were investigated. The pH was checked at the start and end of the experiment, and it was then changed with either acid or base to reach the desired pH. Eight solutions with a pH of 7 and varying MO dye concentrations (20-200 mg.L⁻¹) each received a mass of 100 mg of AC. The isotherm was studied at different temperatures (25, 30, 40, and 50°C). The difference between the initial (C_i, mg.L⁻¹) and final (C_e, mg.L⁻¹) MO dye concentrations can be used to compute the concentration of adsorbed dye (q_e, mg.g⁻¹). Distribution coefficient K_d (L.g⁻¹) was used to express the MO dye's absorption.

$$q_e = \frac{(C_i - C_e)}{m} X V \dots \dots \dots (1)$$

$$K_d = \frac{q_e}{C_e} \dots \dots \dots (2)$$

Effect of MO dye concentration

To study the impact of dye concentration, eight solutions were made, ranging in concentration from 20 to 200 mg. L⁻¹, 100 mg of AC in 100 mL, and stirred for 125 min at 25°C. The final MO dye concentration was calculated.

RESULTS AND DISCUSSION

Effects of the MO dye on the contact time, pH, mass and temperatures of the adsorbent of the solution

Figure 2a shows the affected of adsorption process of MO dye onto AC at time passing. The K_d value was increased (0-100 min) and reached equilibrium with the passage of time. Maybe the increased adsorption at initial contact time is due to the available active site in AC. A short contact time is desirable for economic reasons, and a K_d smaller than 1 L.g⁻¹ is generally considered acceptable (Ramesh *et al.* 2017). For the following experiments, a contact time of 125 min was chosen, yielding a K_d of 0.426 L.g⁻¹. In Figure 2b, the impact of pH is depicted in relation to the adsorption of dye by AC. The K_d values increased from 0.2 L.g⁻¹ at pH 1 to 0.75 L.g⁻¹ at pH 7, then decreased with increasing pH from 8-12. At lower and higher pH, the K_d values decreased, which may be due to the increase in H⁺ and OH⁻ concentration, which may attack the available active adsorption site AC adsorbent.^{7,26,27} The K_d values of the AC adsorbent mass are determined at MO dye concentration, which is a crucial parameter in adsorption experiments. Figure 2c illustrates the impact of adsorbent dosage

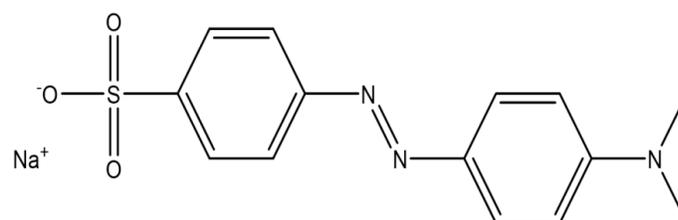


Figure 1: Chemical structure of Methyl Orange.

on the K_d . After the 100 mg dose, the K_d practically approached a constant value. Initially, when AC absorbed mass increased, the K_d values increased. With the increase in adsorbent dosage from 25 mg to 200 mg, the K_d increased from 0.099 to 0.0454. Because there were more accessible sorption surface and adsorption sites, the K_d increased. For subsequent MO experiments 100 mg of adsorbent was chosen. By increasing the temperatures, Figure 2d showed a decrease in K_d , which might be caused by the dissociation of the adsorption bonds between the active sites in the AC and MO dye.^{28,29}

Effect of MO dye concentration

The effect of MO dye concentration on adsorption was investigated at pH7, 25°C, 125 min shaking, and an AC amount of approximately 100 mg. The K_d values increased as concentration increased, as shown in Table 1. The K_d values increased from 0.069 L.g⁻¹ at 20 mg.L⁻¹ to 0.83 L.g⁻¹ at 200 mg.L⁻¹, maybe due to an increase in the available sites for MO dye adsorption.²⁷ The K_d changed only slightly above a concentration of 140 mg.L⁻¹, maybe due to the limited available sites of the AC mass for the

adsorption of MO dye at higher concentrations. As a result, 140 mg.L⁻¹ was chosen as the initial dye concentration for studying the effects of other conditions.

Isotherms adsorption assessment

The adsorption isotherm displays the distribution of MO dye and AC adsorbent at equilibrium within the sorption process. The examination of isotherm models is important until the optimum model is determined to represent the adsorption process at various temperatures.³⁰ The Langmuir model indicates that the sorption process occurs at a homogenous active site on the adsorbent.³¹ The Langmuir isotherm equation is shown as follows:

$$\frac{C_e}{q_e} = \frac{1}{kl q_m} + \frac{C_e}{q_m} \dots \dots \dots (3)$$

Where C_e , q_e , q_m , V , and kl the final concentrations (mg.L⁻¹), adsorption capacity (mg.g⁻¹), theoretical adsorption capacity, and constant isotherm Langmuir, respectively. The K_d and q_m values can be calculated by linearly plotting $\frac{C_e}{q_e}$ vs. C_e . The Freundlich isotherm indicates that the adsorption process occurs between adsorption capacity and dye concentration on a heterogeneous

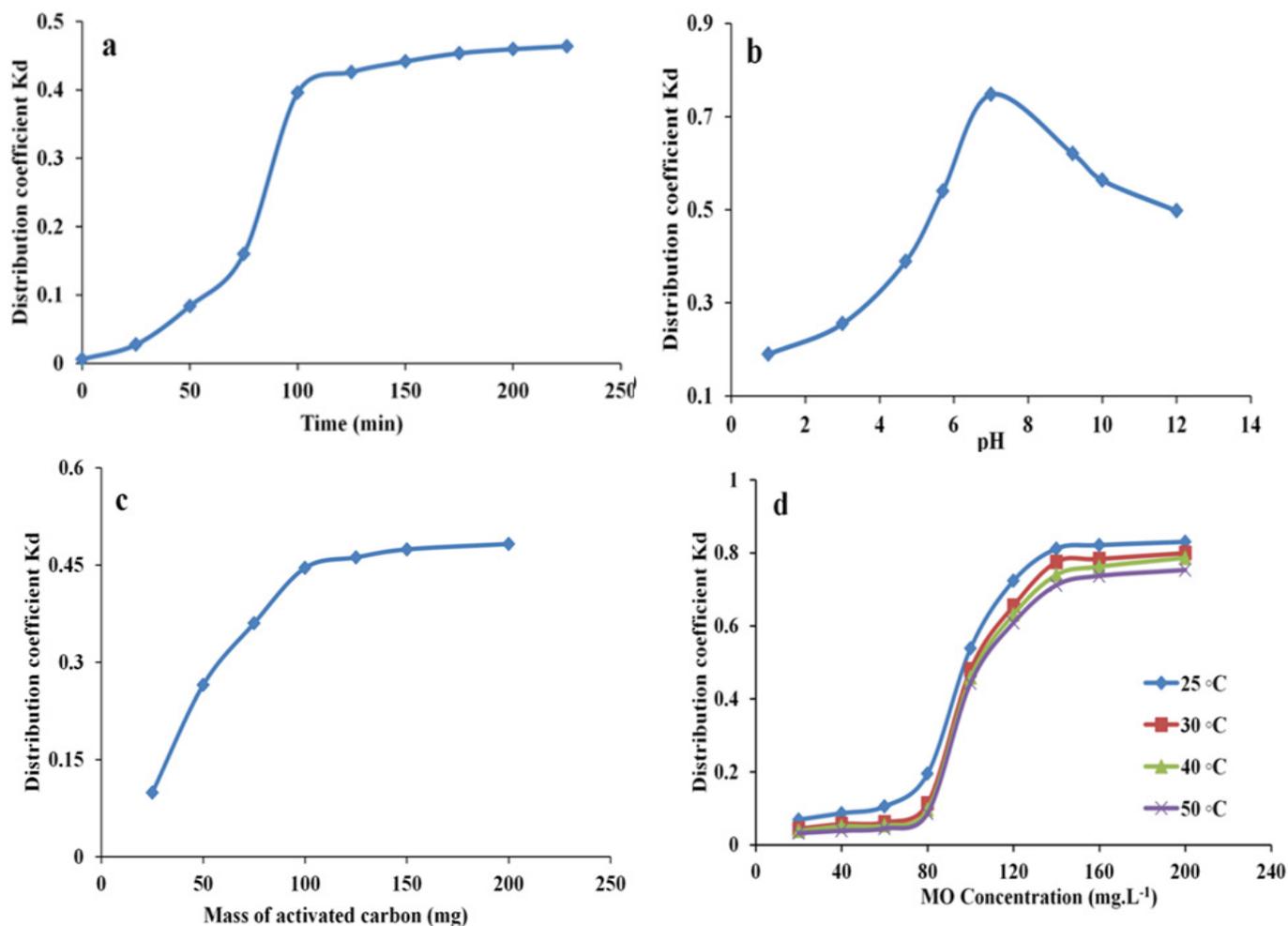


Figure 2: (a) Effects of contact time, (b) pH, (c) Mass and (d) Temperatures on the adsorption of MO dye.

Table 1: Effect of MO dye concentration.

MO dye concentration (mg.L ⁻¹)	Distribution coefficient K _d (L.g ⁻¹)
20	0.069
40	0.087
60	0.106
80	0.195
100	0.539
120	0.724
140	0.812
160	0.821
200	0.830

Table 2: Isotherm models parameters for the adsorption of MO dye onto AC were found.

Model	25°C	30°C	40°C	50°C
Langmuir				
q _m	12.76	12.76	6.73	5.49
K _L	0.0054	0.0054	0.0058	0.0059
R ²	0.7827	0.7827	0.7870	0.7841
Freundlich				
K _f	42.6	92.6	116.9	164.9
1/n	0.49	0.47	0.46	0.45
R ²	0.9680	0.9316	0.9212	0.9077
Temkin				
A	33.12	34.47	34.81	35.16
B	36.37	35.84	35.42	34.83
R ²	0.7992	0.7707	0.7635	0.7607

adsorbent surface at equilibrium³¹. As follows is the Freundlich equation:

$$qe = KfCe^{1/n} \dots \dots \dots (4)$$

The plot of ln q_e against ln C_e was used to calculate the values of K_f and 1/n from the intercept and slope, respectively.

The Temkin model predicts a decrease in the heat of adsorption for molecules in the layer as a result of the adsorption process.³²

The Temkin isotherm's linear form is represented as:

$$qe = B \ln A + B \ln Ce \dots \dots \dots (5)$$

Where B, and A the heat of adsorption-related Temkin constant, and the constant of equilibrium binding (L.mg⁻¹), respectively. A Plot of q_e vs lnC_e will show the constants A and B.

In Table 2, all the R² values and parameter constants acquired from the three models are listed. The values of R² in the Freundlich model were higher than the R² values in the Langmuir and Temkin models at different temperatures. Which demonstrated that

the Freundlich model was the most suitable for describing the adsorption of dyes on AC.³³ Indicating exothermic adsorption, the K_f shown increased with the temperatures increased. The values of 1/n were less than 1, which indicates the adsorption condition was acceptable.³³

Adsorption Thermodynamics

For the adsorption of MO dye into AC, the values of entropy ΔS (J⁻¹.mol.K) and enthalpy ΔH (KJ.mol⁻¹) were determined by using the equation (6):

$$\log \left(\frac{qe}{Ce} \right) = \frac{\Delta S_{ads}}{2.303R} - \frac{\Delta H_{ads}}{2.303RT} \dots \dots \dots (6)$$

The symbols R and T represent the value of the gas constant (8.314 J⁻¹.mol.K) and the value of the absolute temperature (K), respectively. The slope and intercept of the plot log (q_e/C_e) vs 1/T can be used to calculate ΔH and ΔS, respectively. According to the negative value of ΔH (-632.48 KJ.mol⁻¹), the MO dye adsorption onto AC is an exothermic reaction. The adsorption process positive value (2.82J.mol⁻¹.K⁻¹) of ΔS demonstrated the reduced unpredictability at the solid-liquid interface. According to Table 3, the values of (ΔG) decreased from -207.88 KJ.mol⁻¹ at 298K to -278.38KJ.mol⁻¹ at 323K, indicating that MO was adsorbing on AC exothermically on its own.

Kinetics assessment

To examine and determine the effectiveness and rate of adsorbate at equilibrium time, we studied the kinetics of adsorption. The PFO and PSO models were studied and examined to find the most suitable model for the experimental data from the adsorption process of MO dye onto AC. The following formulas (7) and (8) were used to compute the values of k₁ and q_e (mg.g⁻¹) by plotting a graph of log (q_e-q_t) vs't' (min), and values of k₂ and q_e (mg.g⁻¹) were founded by plotting t/q_e vs't' (min).

PFO model formula:

$$\log(qe - qt) = \log qe - \left[\frac{k_1}{2.303} \right] t \dots \dots \dots (7)$$

PSO model formula:

$$\frac{t}{qt} = \frac{1}{k_2 qe^2} + t/qe \dots \dots \dots (8)$$

Where q_e and q_t equal the quantity of MO adsorbed (mg.g⁻¹) at equilibrium and at any other time during the adsorption process, respectively. Table 4 shown the R² values for the PFO and PSO models in the range 0.7103-0.9188 and 0.9902-0.9924 for MO. The q_e (exp) values at PFO model did not match with the q_e (cal) values, therefore, the PFO model does not fit with the adsorption of MO dye into AC. The value of q_e (exp) was in agreement with the q_e (cal). Table 4 showed that indicating that PSO model is the suitable kinetic model for MO into AC. The adsorption process was controlled via chemisorption.

Table 3: Thermodynamic parameters for the adsorption of MO dye onto activated carbon.

ΔH (KJ ⁻¹ .mol)	ΔS (J ⁻¹ .mol.K)	ΔG (KJ ⁻¹ .mol)			
		298 K	303 K	313 K	323K
- 632.48	2.82	-207.88	-221.98	-250.18	-278.38

Table 4: Kinetic parameters for the adsorption of MO dye onto AC at different initial concentrations.

C_i (mg.L ⁻¹)	q_{exp} (mg.g ⁻¹)	PFO			PSO		
		k_1	q_{cal}	R^2	k^2	q_{cal}	R^2
60	5.5	6.67X10 ⁻³	0.177	0.9188	1.61X10 ⁻⁴	17.4	0.9924
100	32.19	1.38X10 ⁻³	0.491	0.728	2.08X10 ⁻³	35.71	0.9902
140	56.41	2.3X10 ⁻³	0.494	0.7103	6.64X10 ⁻⁴	46.95	0.9912
200	81.49	4.84X10 ⁻³	0.877	0.7142	2.11X10 ⁻⁴	106.38	0.9914

Table 5: Recent reported adsorption capacities of MO dye.

Adsorbent	Adsorption capacity q_e (mg.g ⁻¹)	Reference
Rayon-based AC fibers.	7000	34
AC from coffee waste.	1017	35
Pomelo peel-derived porous carbon.	680.3	36
AC from <i>Aloe vera</i> .	196	37
AC from coconut shell	3	38
Mesoporous carbon material	294.1	39
Ag NPs-coated AC	27.48	8
Activated Carbon	42.6	Present work

Comparison of the Adsorption of MO dye with Different Adsorbents

The adsorptive capacities of the adsorbents used in this study were compared to those of others reported in the literature, and the results are shown in Table 5. As a comparison parameter, the adsorption capacity (q_e) was used. The q_e value obtained in this study was consistent with and higher than most previous studies, indicating that the AC used in this study could easily remove MO dye from solutions.

CONCLUSION

This study focuses on the environmental impact of utilizing AC as an adsorbent to remove MO dye from solution. The MO dye's ability to bind to AC increased with increasing temperatures, AC amount, and pH. The maximum adsorption capacity of the MO dye on AC was 42.6 mg.g⁻¹ at 25°C, and the equilibrium adsorption data were better fitted to the Freundlich model than to the Langmuir and Temkin models. At the investigated temperatures range, the adsorption of MO dye onto AC was found to be spontaneous (negative ΔG values) and exothermic

(negative ΔH value) in nature. The increased randomness at the solid-solution interface during MO dye adsorption onto the AC is clearly shown by the positive entropy (ΔS). The PSO kinetic model was followed in the dye onto AC.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

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