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Application for fitting of Langmuir and Freundlich equations Studies on the experimental data of adsorption of Methylene Blue Dye by Cobalt Oxide Surface

Neda Ibrahim Mahdi
Department of Chemistry, Collage of Science, Al-Mustansiriyah University, Baghdad, Iraq

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Corresponding Author

E-mail: nedaibrahem7@gmail.com

Abstract

This study is based on the investigation of the adsorption property of Methylene blue onto Co_3O_4 as a surface. the optimum conditions of this study were determined and it was found that the dose of Co_3O_4 was 50 mg, the pH of adsorption dye was 10, the initial concentration of dye was 15 mg/L, and temperature found to be 45 C°. Langmuir and Freundlich isotherm models were used for investigating the equilibrium adsorption of Methylene blue dye onto Co_3O_4. The thermodynamic study shows that the adsorption of the MB dye process was non-spontaneous and endothermic under the study conditions.

Introduction:

Adsorption is a vital technique for pollution elimination from wastewater and gas separation [1]. Adsorption kinetics and thermodynamics are particularly essential in the industry. For the synthesis of diverse adsorbents, it is vital to explore both the thermodynamics and kinetics of this process [2]. Adsorption technology is frequently used in the treatment of water and wastewater procedures because it is quick, efficient, and reasonably priced approach [1].

Dyes are used to color items in numerous sectors, including food, paper, carpets, rubber, plastics, cosmetics, and textiles [3]. The dumping of coloured effluent from these businesses into natural waterways has caused numerous significant problems, including increased chemical oxygen demand (COD)and effluent toxicity, as well as reduced penetration of light, which has a negative impact on photosynthetic processes [4].

The Langmuir and Freundlich isotherms are two of the most commonly used models to study adsorption processes which help to understand the adsorption behavior of dyes onto solid surface [5]. Overall, fitting adsorption isotherm models to experimental data was done, and the model with the greatest fitting was chosen to define equilibrium adsorption [6]. The model's desorption parameters were acquired and utilized to characterize the absorption quality once the right adsorption model was chosen [7-12].

Many papers and studies have explored the use of various sorbents to remove MB (Methylene Blue) dye from water such as kaolinite [13], bamboo-based activated carbon [14], agro-industrial wastes [15], nano-zerovalent iron [16], nano-poly acrylonitrile (PAN) [17], Polyethylene Glycol Crosslinked-CD Polymers [18], activated carbon derived from waste orange and lemon peels [19], and hierarchically porous, activated starbons [20]. Most of the surfaces of this literature are expensive for preparation, consuming high energy and more time to process. The aim of this study is to use a highly efficient surface of Co_3O_4 and low-cost for the removal of MB dye from aqueous solutions.

Materials and methods

Chemicals

Methylene blue(MB), Co_3O_4 “Sigma-Aldrich”, sodium hydroxide (NaOH)“BDH”, and hydrochloric acid (HCl)“Fluka” were analytical reagents.

Apparatus

In this investigation, a water bath that shakes “BS-11 digital, JETO Korea, TECH”, pH metre“Hana”, and “UV-visible spectrophotometer” “Cary 100,VARIAN CO.”wereemployed.

Solutions

A ready solution of methylene blue (100 mg/L) was made by dissolving 0.1 gm of methylene blue in 100 ml of distilled water. A series of MB concentrations (5, 10, 15, 20, 25, 30, and 35 mg/L) were prepared by transferring (1.25, 2.50, 3.75, 5, 6.25, 7.50 and, 8.75 ml) MB standard solution, then it was diluted with distilled water to the mark in 25 ml volumetric flasks.

To examine the removal of MB by employing Co_3O_4 as a surface, the pH of MB solutions was changed using ($1 * 10^{-3}$mol/L) HCl and ($1 * 10^{-3}$mol/L) NaOH (pH 2, 4, 6.6, 8, and 10).

Adsorption studies

To evaluate the impact of adsorbent dose (30-150 mg), pH (2-10), temperature (35, 45, 55°C), and initial concentration (15-30 mg/L), a set of volumetric flasks containing 50ml of (25mg/L) MB solutions were used. The solutions were then filtered, and the concentrations of MB were determined using a UV-visible spectrophotometer set at λ= 600 nm. For 60min, investigate the isotherm for MB adsorption observed at different temperatures (35,45, and 55°C) [21].

The next following equations define the efficiency adsorbed amount (qe) of the adsorption of MB (equation 1), while Equation (2) defines the removal percentage(Re%)[22]:

q_e= ((C_0-Ce)*V/m…… (1)

Re% = (C_o-Ce)/C_o*100 ……. (2)

Where, qe is the mean efficiency adsorbed amount of methylene blue, which refers to the amount of MB adsorbed per unit weight of Co_3O_4 surfaces, Co is the initial concentration of MB dye (mg/L)(before adsorption), Ce is the Final concentration of the same dye(after adsorption), V is the volume of the methylene blue solution (L), and m is the weight of Co_3O_4 surfaces (gm).

The Langmuir and Freundlich isotherm models are two famous models used to describe the adsorption of molecules onto a surface. the Langmuir Isotherm assumes that the adsorption occurs on a homogeneous surface (it is meaning that only one layer of molecules can adsorb onto the surface). on the other hand, the Freundlich Isotherm supposes that the multiple layers of adsorbate molecules can be formed on the surface (Table1) [23].

Table1: isotherm equations (Langmuir and Freundlich)

Isotherm	Equation	Linearized form	Plot	Parameters
Langmuir	$q_e = q_m K_L C_e/(1+K_L C_e)$	$\frac{Ce}{qe} = \frac{1}{qmKL} + \frac{Ce}{qm}$	(Ce/qe) vs. Ce	$qm = (\text{slope})^{-1}$, K_L = (slope/intercept)
Freundlich	$q_e = K_F C_e^{1/n}$	$\text{Log}(q_e) = 1/n \log(C_e) + \log(K_F)$	$Log\, q_e$ vs. $Log\, C_e$	K_F = exp(intercept), $n = (\text{slope})^{-1}$

"q_m (mg/g) and K_L (mL/mg) are Langmuir constants; K_F (ml/mg) is Freundlich constant and n is the intensity of adsorption of Freundlich isotherm model"

Results and Discussion

Absorption spectra and a calibration curve

The MB dye's spectrophotometric spectrum showed maximum absorption at 600nm. A graph that is linear for MB dye is given in Fig.1 at the ideal circumstances explored in the technique, which depicts the concentrated variation in linearity (5-35mg/L) and molar absorptivity (Є= 0.047 L/g. cm).

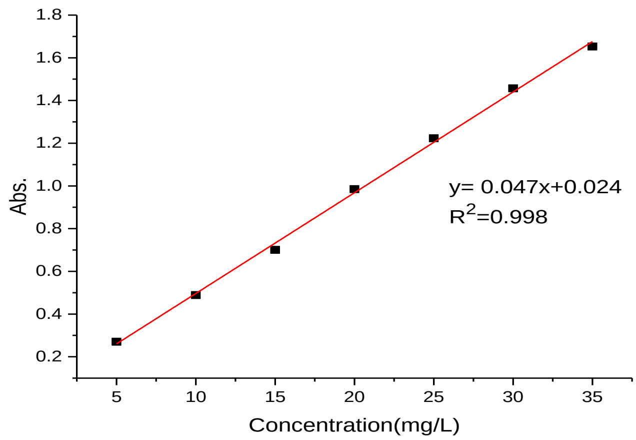

Fig.1: Calibration curveof MB dye

Adsorption studies

Adsorbent Dosage Effect

The effect of Co_3O_4 dosage on the elimination of MB dye was investigated by adding various quantities of adsorbent (30, 50, 100, and 150 mg) given a starting concentration of 20 mg/L at a temperature of 25°C for 60 min. The optimal weight was (50mg), and increasing the adsorbent dose had to make increasing in the removal percentage, which the reason for the improved removal efficiency with higher adsorbent doses might be due to increased surface area and more active sites for the adsorption of MB dye molecules(Fig.2a).

Effect of pH

Various pH levels (2, 4, 6.6, 8, and 10) were studied to determine the effect of pH on methylene blue adsorption on Co_3O_4 surfaces. All experiments were carried out for 60 min at a concentration of dye of 20 mg/L, an adsorbent dosage of 50 mg, and at a temperature of 25°C (Fig.2b and Fig. 3). The greatest removal percentage was discovered at pH = 10, and the removal percentage gradually increased as the pH increased. The reason for the increased removal percentage at higher pH values could be attributed to the surface charge factor, at higher pH levels (pH > 7), MB dye is a cationic dye that attracts and binds positively charged with the negatively charged surface (the surface of the Co_3O_4 adsorbent might become more negatively charged due to the dissociation of hydroxyl groups) [24].

Effect of Initial MB concentration

The adsorption assay was completed for MB at different concentrations of 15, 20, 25, and 30 mg/L was completed using a 0.1 g adsorbent dosage, pH 6.6, and 25 °C for 60 minutes, as demonstrated in Fig.2c. The adsorption sites get saturated on the Co_3O_4 surface when the initial concentration of MB dye is increased due to the number of dye molecules in the solution becoming higher than from the available adsorption sites on the Co_3O_4 surface. So, the removal percentage decreases as the adsorption capacity reaches its limit [25].

Effect of Temperature

Figure 1d showed that a higher percentage of MB dye removal is achieved at a temperature of 45°C. At 45°C, the Co_3O_4 adsorbent exhibits higher efficiency in removing MB dye from the solution compared to other temperatures. Thermal energy increase Leads to Increased surface activity of Co_3O_4, making more adsorption sites available for MB dye molecules to bind at 45°C (Fig.2d) [26-28].

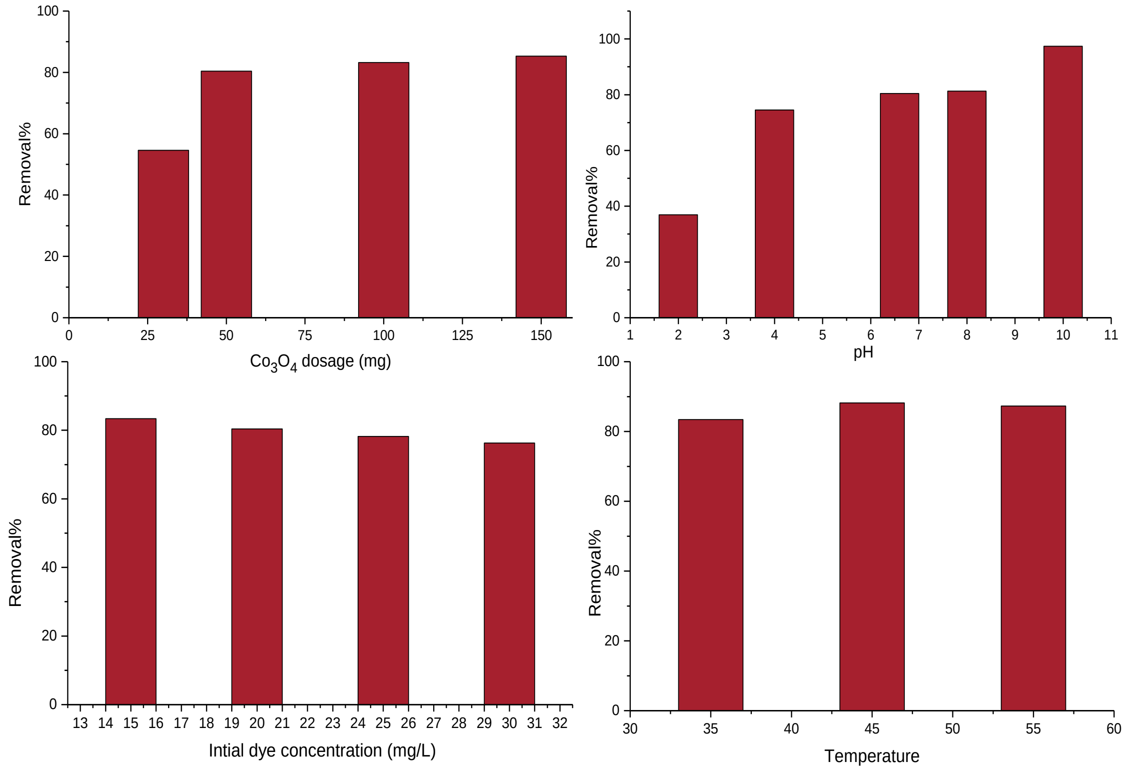

Fig.2: The ideal conditions for an adsorption reaction of MB dye (a) adsorbentdosage, (b) pH, (c) concentration, and (d) temperature.

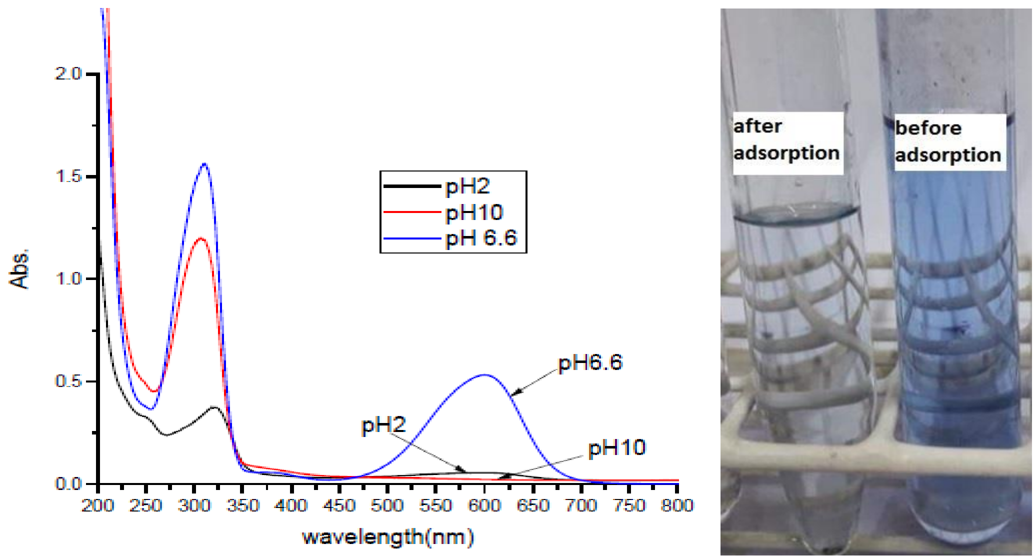

Fig.3: UV-visible absorption spectrum of (20 mg/L) MB mixture prior to adsorption method, after adding 50 mg Co_3O_4 solution for 60 min and 45 °C at three different pH (2, 6.6, and 10)

Adsorption isotherm

Langmuir and Freundlich adsorption Isotherm.

Linear regression was employed in this investigation to obtain the best-fitted isotherm. The linear Langmuir isotherm models had the greatest R^2values (0.986), the best fit of the Langmuir parameters indicated that the Langmuir is other favored and that the adsorption process is largely monolayer and homogenous(Fig.4). The Freundlich linear isotherm at three distinct temperatures (Fig.4 and Table 2, 3). The high R^2 value of 0.967 at 45°C suggested good adsorption behavior at that temperature. Furthermore, Values less than one represent low adsorption intensity, values 1-2 indicate the adsorption intensity of mediums, while values 2-10 imply a high level of adsorption. 1/n values range from 0 to 2, with lower value suggest in gamore variable adsorption surface [29- 31].

Table2: The values of adsorption isotherms used for the Langmuir and Freundlich plotting at 45°C.

Ci (mg/L)	Ce(mg/L)	qe*(mg/g)	Ci-Ce	log Ce	Log qe	Ce/qe
10	3.446	6.553	6.553	0.537	0.816	0.525
15	6.851	8.148	8.148	0.835	0.911	0.840
20	11.382	7.553	8.617	1.056	0.935	1.320
25	15.595	8.340	9.404	1.193	0.973	1.658
30	19.510	10.489	10.489	1.290	1.020	1.860

***volume of dye solution 50 mL(0.05L) and weight of the Co_3O_4 surface 50 mg(0.05 g).**

Table3: linear correlated coefficient obtained from the application of Langmuir and Freundlich isotherm of the experimental results of the adsorption of MB dye on the Co_3O_4 surface at pH 10 using a certain range of concentration and at 45°C

Isotherm models	constant	value
Langmuir linear	Regression equation(y)	0.085x+0.272
	R^2	0.986
	q_m	11.764
	K_L	0.312
Freundlich linear	Regression equation(y)	0.249x+0.686
	R^2	0.967
	1/n	0.249
	K_f	1.985

"K_L: Langmuir constant, qm: (mg/g) is the maximum theoretical adsorption capacity, n is related to adsorption capacity, and K_f: Freundlich constant"

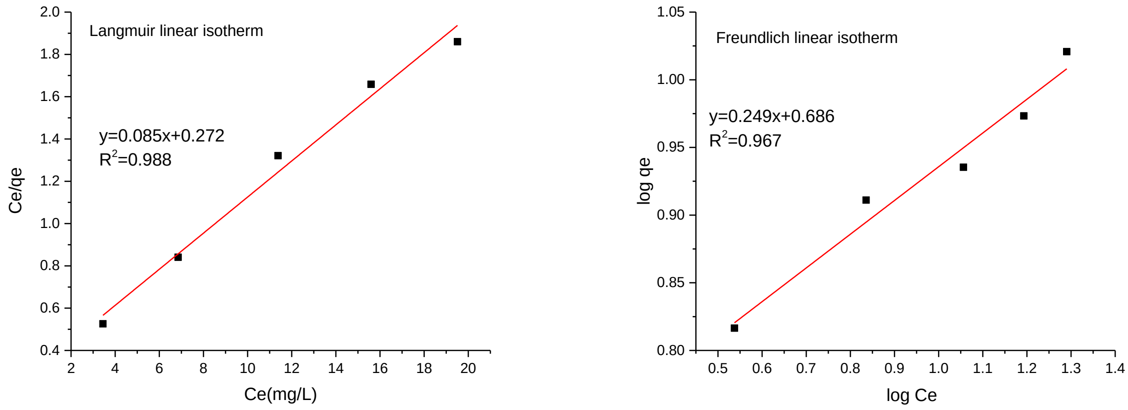

Fig.4: Langmuir and Freundlich linear isotherm at MB dye, 45 °C, 50 mg of Co_3O_4 for 60 min., and pH 10.

In the plot of Ce/qe against Ce in Fig.4, straight lines were seen at all concentrations, showing that both adsorbents' adsorption was well matched to the Langmuir isotherm. The high Coefficient of determination value (R^2 =0.988). The discovery suggests a strong affinity between the surface of the adsorbent and the MB, that's critical in the absorption process. The isotherm of Freundlich (linear forms) for both of the adsorbents utilized in this work is depicted in Figure 4. Based on the high Coefficient of determination (R^2=0.967), it is possible to conclude that the MB adsorption isotherm employing R^2 provides a superior match to the Langmuir model.

Thermodynamic study

To find the nature of adsorption, Thermodynamic study as applied on the removal MB dye on the Co_3O_4 surface by using Vant's Hoff equation:

$$\mathbf{lnKd} = \frac{\mathbf{\Delta S^\circ}}{\mathbf{R}} - \frac{\mathbf{\Delta H^\circ}}{\mathbf{RT}}$$

Kd is the adsorption equilibrium constant can be calculated by:

$$Kd=\frac{Cad(\frac{mg}{L})}{Ce(\frac{mg}{L})}$$

where C_{ad} is Ci-Ce, Ci is initial concentration(mg/L), and Ce is concetration after adsorption(mg/L) (Table 4).

The values of ΔH° can be calculated from the slope of the linear Vant's Hoff plot of lnKd versus 1/T (Slope= - ΔH°/R) (Figure 5). Gibbs free energy (ΔG°) and entropy (ΔS°) are calculated from the following equations:

$$\Delta G^\circ=-R\ T\ LnKd$$

$$\Delta S^\circ=(\Delta H^\circ-\Delta G^\circ)/\ T$$

where R is the gas constant (8.314 J/mol.k), and T is the temperature(k).

The positive values of ΔG^o indicated the non-spontaneity of the adsorption process of this study. The positive ΔHº and the positive ΔSº values indicate the adsorption process is endothermic with increased randomness at the interface of adsorbate and adsorbent (Table 5) [32].

Table 4: The values of equilibrium constants (**K**d) for MB dye adsorption.

Temp. (T)	1/Temp. (1/T) (k^{-1})	C_i (mg/L)	C_e (mg/L)	C_{ad}(C_i-C_e) (mg/L)	K_d(C_{ad}/C_e)	Ln K_d
308	0.003246753	20	14.65	5.35	0.365188	-1.00734
318	0.003144654		13.845	6.155	0.444565	-0.81066
328	0.00304878		12.02	7.98	0.663894	-0.40963

Table 5. Thermodynamic study of the adsorption of (20 mg/L) MB dye on the Co_3O_4 surface.

Temperature K	ΔHº kJ/mol. K	ΔGº kJ /mol	ΔSº J/mol
308	25000.2	2579.517292	72.79441788
318		2143.263468	71.87715262
328		1117.067326	72.81442279

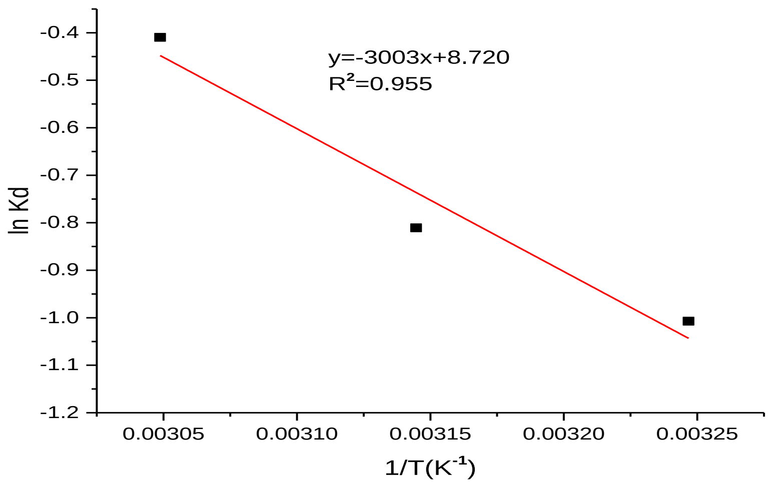

Fig.5: Van't Hoff plots of the MB dye removal at (35, 45, and 55°C) temperatures

Conclusions

This study demonstrates how cobalt oxide maybe utilized as an effective adsorbent to remove MB dye from aqueous solutions. The effects of, Co_3O_4 dose, pH, initial concentration, and temperature on adsorption capabilities were studied. Two isotherm models were studied, and the results reveal that the Langmuir models suit the data the best.

The use of cobalt oxide to remove MB dye was examined. Adsorption was studied at three different temperatures (35°C, 45°C, and 55°C). R^2 is the determination coefficient. At 45°C, the Langmuir linear isotherm most closely matches the data for MB dye removal using cobalt oxide. The entire method is based on chemical adsorption, which is endothermic, randomness, and non-spontaneous, according to the thermodynamic characteristics.

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تطبيق معادلات Langmuir و Freundlich لدراسة بيانات الامتزاز التجريبية لصبغة المثيلين الازرق على سطح أكسيد الكوبالت Co3O4

نداء ابراهيم مهدي
قسم الكيمياء، كيلة العلوم، الجامعة المستنصرية

معلومات البحث:

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الكلمات المفتاحية:

المثيلين الأزرق، Langmuir، Freundlich، اوكسيد الكوبالت، الايزوثيرم

معلومات المؤلف

الخلاصة:

الغرض من هذا البحث هو دراسة امتزاز صبغة المثيلين الزرقاء على سطح اوكسيد الكوبالت. تم دراسة دوال توازن ايزوثيرم الامتزاز Langmuir و Freundlich، وكذلك درست الظروف المثلى، إذ أظهرت أفضل ظروف مثلى تعطي أفضل إزالة هي pH 10, وزن السطح 0.05 غم، تركيز صبغة المثيلين الزرقاء 15ppm، الزمن 60 دقيقة، والحرارة 45 مئوي.