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# APPLYING BOX-BEHNKEN DESIGN WITH STATISTICAL OPTIMIZATION FOR REMOVAL VAT ORANGE DYE FROM AQUEOUS SOLUTION USING KAOLIN

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## Abstract

Kaolin as a cheap adsorbent has been studied for removal Vat Orange 11(VO) dye from aqueous solution. Many factors have been studied that affecting the adsorption process including the quantity of kaolin (0.1-1 g), pH at (3-9) and time (12-60 min). Box-Behnken Design for three variables and three levels with response surface methods have been used to get a second-order polynomial equation for dye removal percentage (DR%) as a response. The precision of the equation obtained by the Box-Behnken Design utility for modelling and optimization by response surface methodology RSM was confirmed by the analysis of variance (ANOVA). The maximum DR% has been reached 85.88% at optimum conditions based on kaolin (0.918 g), pH=3 and time (47 min.). The data of adsorption equilibrium followed the Langmuir, Freundlich and Temkin. The kinetic model of the process of adsorption of VO dye on kaolin has been fitted a pseudo-second order.

Keywords: Box-Behnken design, Kaolin, Vat orange dye, Wastewater treatment.

## 1. Introduction

Nowadays, the process of removing or minimizing the toxic substances had become one of the most important and serious challenges facing the environment and society in our modern world. The toxic substances are usually resulting from the accumulation of waste and various industrial processes. Textile industries are one of the largest consumers and pollutants of water have harmful effects on workers' health and surface water vitality such as rivers and lakes. The effluent wastewater contains dyes with a concentration of (10-50 mg/L) which are mostly carcinogenic substances, heavy metals, suspended matter and turbidity, hardness, base and dissolved substances, such wastewater cause rapid consumption of dissolved oxygen in water [1-3].

Several methods have used for the treatment and removal of organic and inorganic pollutants of industrial water: chemical method [4, 5], Photocatalytic oxidation methods [6, 7], reverse osmosis [8, 9] and adsorption methods on porous solid surfaces [10].

Adsorption is a widely used method, especially if the used surfaces are inexpensive natural materials. The adsorption method has the advantages of being good at removing a wide range of dyes, heavy metals, and other substances. It is also flexible, simple in design, easy to operate, and is unaffected by the toxic pollutants. On the other hand, some adsorbents are expensive and have a low surface area. Furthermore, some adsorbents require regeneration or need to be disposed of appropriately. Several studies in this area had been carried out in removing dyes of wastewater effluents using various adsorbents such as activated carbon [11], agricultural wastes: sawdust, pine tree [12, 13], clays [14, 15] and food waste like eggshell [16].

In the present research, kaolin has been used as available, natural sorbent in removing dye from aqueous solution. Kaolin was used as a local, available, and cheap substance, and it has been used in a number of studies as an adsorbent [14, 15]. An experimental design method (Box-Behnken design) was investigated with three factors including, the amount of kaolin (g), pH, and time (mints) with three levels for each of them. The polynomial equation is established between the DR% (as the response) and the studied factors, by using Response Surface Methodology to find the optimum factors that give maximum dye removal. The analysis of variance (ANOVA) has been used to determine the statistical significance of the factors of the developed model.

## 2. Materials and Methods

### 2.1. Materials

Kaolin was used in this study as adsorbent materials from Gerhard Buchmann KG Tuttlingen/Germany as a powder with $\leq 75$ μm in size. It dried at 110°C in an oven for 3 hours to remove moisture. An analysis of the chemical components of Kaolin conducted in the Iraqi Geological Survey (Central Laboratories department) is presented in Table 1.

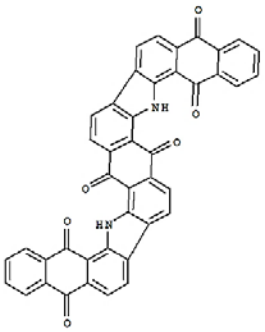
Vat Orange 11dye (VO) dye was obtained from a cotton textile factory in Baghdad without any purification. The structure and some properties of dye were shown in Table 2.

**Table 1. Composition of kaolin.**

| Component | Weight % |
|---|---|
| $SiO_2$ | 48.1 |
| $Fe_2O$ | 1.05 |
| $Al_2O_3$ | 34.59 |
| $TiO_3$ | 0.05 |
| CaO | 0.93 |
| MgO | 0.26 |
| $SO_3$ | 0.06 |
| L.O.I* | 13.3 |
| $Na_2O$ | 0.02 |
| $K_2O$ | 1.55 |

***L.O.I: loss on ignition**

**Table 2. The chemical properties of vat orange 11 dye [17].**

| Properties | |
|---|---|
| Chemical formula | $C_{42}H_{18}N_2O_6$ |
| Molecular Weight | 646.61 |
| Product Categories: | Organics |
| Structure of dye | |
| Names:<br>Dinaphtho[2,3-i:2',3'-i']benzo[1,2-a:4,5-a']dicarbazole-5,7,12,17,19,24(6H,18H)-hexone (CAS NO.2172-33-0) |  |

## 2.2. Dye solution preparation

VO dye was dissolved accurately in distilled water to obtain (10-60 mg /L) as initial dye concentration. The pH adjusted by the addition of (0.1% HCl) or (0.1% NaOH). The VO dye concentration was determined before and after adsorption at wavelength $\lambda_{max}$ = 419 nm using a UV spectrophotometer (UV-1100 chrom Tech) [18]. The calibration curve was done between the absorbance and concentration of the dye solution.

## 2.3. Adsorption studies

The batch adsorption experiments were conducted with 50 mL of VO dye solution at various initial concentrations of the dye, containing different amounts of (kaolin) (0.1-1) g. The kaolin and VO dye solution was mixed using shaker at 200 rpm at intervals of (10-60) min. at 25ºC. The dye solution was centrifuged at 3000 rpm for 20 min after the adsorption time finish and then filtered with filter paper to remove the residual of kaolin [19].

## 2.4. Equilibrium studies

To get equilibrium data by adding 0.5 g of kaolin to 50 mL of VO dye solution with initial concentrations (10-60) mg/L. Equilibrium time was taken as 65 min and the dye solution adjusted to pH =3. The DR% was calculated from Eq. (1)

$$Re\% = 100(C_o - C_e)/C_o \quad (1)$$

The amount of dye adsorbed per gram of kaolin at equilibrium time and at time, *t* was calculated from Eqs. (2) and (3), respectively [16]:

$$q_e = (C_o - C_e) \times (\frac{V}{m}) \quad (2)$$

$$q_t = (C_o - C_t) \times (\frac{V}{m}) \quad (3)$$

where $R_e\%$ percentage of dye removal at time of equilibrium, $C_o$ and $C_e$ are the initial and the equilibrium concentrations (mg/ L) of VO in solution respectively, $C_t$: concentration of dye at any time (mg/ L), $m$: weight of kaolin (g), and $V$: the volume of solution (L) [16].

## 2.5. Box-Behnken design (BBD)

In this study, the experiments were conducted by using Box-Behnken design with three-level designs for fitting response surfaces. Three independent variables were based on sorbent dosage (kaolin), contact time, and pH. Each variable had 3 levels designated by the codes (-1, 0, 1) given in Table 3. The (BBD) includes 15 experiments [20]. The initial dye concentration was fixed on (16 mg/L) that was the maximum concentration of VO dye in wastewater from the cotton textile factory [3].

The response of each experiment (dye removal %) was calculated from Eq. (4)

$$R\% = 100(C_o - C)/C_o \quad (4)$$

where $R\%$ is the percentage of dye removal, $C$ is the concentration (mg/L) of VO in the solution after the adsorption. The second-order polynomial model was used to fit the experimental data, as in Eq. (5)

$$Y = a_0 + a_1X_1 + a_2X_2 + a_3X_3 + a_{12}X_1X_2 + a_{13}X_1X_3 + a_{23}X_2X_3 + a_{11}{X_1}^2 + a_{22}{X_2}^2 + a_{33}{X_3}^2 \quad (5)$$

where $Y$ is the predicted response (DR %). ($X_1$, $X_2$, $X_3$) are coded levels of the variables and ($a_0$, $a_1$, $a_2$, $a_3$, $a_{12}$ …) are regression coefficients.

**Table 3. Rang of codes used in the Box-Behnken design for variables and their real experimental values.**

| Code | Variables | | |
|---|---|---|---|
| | Time (minute) | kaolin (g) | pH |
| | $X_1$ | $X_2$ | $X_3$ |
| -1 | 12 | 0.1 | 3 |
| 0 | 36 | 0.55 | 6 |
| 1 | 60 | 1 | 9 |

## 3. Results and Discussion

### 3.1. Response surface of dye adsorption

In this study, Minitab 16 software was used for statistical analysis of all results. The experimental design results of the DR% are shown in Table 4 includes 15 runs. As seen in Table 4, the real dye removal % was obtained from experiments that are approximately near to predicted values of the established model.

**Table 4. Results of Box-Behnken design with coded and real values of independent variables.**

| Exp. No. | Coded Variables | | | Real Variables | | | dye removal % Y | |
|---|---|---|---|---|---|---|---|---|
| | Time $X_1$ | Kaolin $X_2$ | pH $X_3$ | Time (min.) | Kaolin (g) | pH | Experimental values | Predicted values |
| 1 | -1 | -1 | 0 | 12 | 0.1 | 6 | 32.96 | 31.9329 |
| 2 | 1 | -1 | 0 | 60 | 0.1 | 6 | 46.37 | 46.7279 |
| 3 | -1 | 1 | 0 | 12 | 1 | 6 | 63.74 | 63.3821 |
| 4 | 1 | 1 | 0 | 60 | 1 | 6 | 60.89 | 61.9171 |
| 5 | -1 | 0 | -1 | 12 | 0.55 | 3 | 59.95 | 60.3476 |
| 6 | 1 | 0 | -1 | 60 | 0.55 | 3 | 78.4 | 77.4126 |
| 7 | -1 | 0 | 1 | 12 | 0.55 | 9 | 62.44 | 63.4274 |
| 8 | 1 | 0 | 1 | 60 | 0.55 | 9 | 60.09 | 59.6924 |
| 9 | 0 | -1 | -1 | 36 | 0.1 | 3 | 39.08 | 39.7095 |
| 10 | 0 | 1 | -1 | 36 | 1 | 3 | 84.709 | 84.6692 |
| 11 | 0 | -1 | 1 | 36 | 0.1 | 9 | 53.99 | 54.0298 |
| 12 | 0 | 1 | 1 | 36 | 1 | 9 | 56.338 | 55.7085 |
| 13 | 0 | 0 | 0 | 36 | 0.55 | 6 | 69.78 | 69.2300 |
| 14 | 0 | 0 | 0 | 36 | 0.55 | 6 | 68.68 | 69.2300 |
| 15 | 0 | 0 | 0 | 36 | 0.55 | 6 | 69.23 | 69.2300 |

The analysis of variance (ANOVA) was used to analyse the experimental results, as shown in Table 5, using p-value and F-value to determine the significance of the regression coefficients of the model equation. It can be observed that very small P-values (<0.05) may indicate the parameters are significantly different from zero at the 95% confidence level. In this case, all parameters ($X_1$, $X_2$, $X_3$) and their interactions were statistically significant [16].

$R^2$, $R^2$ (pred), $R^2$ (adj) values 0.9977, 0.9664, 0.9936 respectively showed convergent, that considered as a valid statistical model. *R*, *Raj*, and *Rpred* are convenient to obtain the fit and the predictive of a created model. The response function for VO dye removal % is shown in Eq. (6).

$$Y = 69.230 + 3.333X_1 + 11.660X_2 - 3.660X_3 - 5.775{X_1}^2 - 12.465{X_2}^2 + 1.765{X_3}^2 - 4.065X_1X_2 - 5.200X_1X_3 - 10.820X_2X_3 \quad (6)$$

The response surface methodology (RSM) was used to find the optimum response. (RSM) is a collection of mathematical and statistical techniques useful for experimental design (like Box-Behnken design), regression modelling and optimization methods [20].

**Table 5. Analysis of variance for %dye removal.**

| Source | DF | Seq SS | Adj SS | Adj MS | F | P |
|---|---|---|---|---|---|---|
| Regression | 9 | 2621.16 | 2621.16 | 291.24 | 241.38 | 0.000 |
| time(min), $X_1$ | 1 | 88.84 | 88.84 | 88.84 | 73.63 | 0.000 |
| ads.(g), $X_2$ | 1 | 1087.57 | 1087.57 | 1087.57 | 901.38 | 0.000 |
| pH, $X_3$ | 1 | 107.17 | 107.17 | 107.17 | 88.82 | 0.000 |
| $X_1^2$ | 1 | 93.72 | 123.12 | 123.12 | 102.05 | 0.000 |
| $X_2^2$ | 1 | 589.78 | 573.73 | 573.73 | 475.51 | 0.000 |
| $X_3^2$ | 1 | 11.50 | 11.50 | 11.50 | 9.53 | 0.027 |
| $X_1X_2$ | 1 | 66.10 | 66.10 | 66.10 | 54.78 | 0.001 |
| $X_1X_3$ | 1 | 108.16 | 108.16 | 108.16 | 89.64 | 0.000 |
| $X_2X_3$ | 1 | 468.31 | 468.31 | 468.31 | 388.13 | 0.000 |
| Pure Error | 2 | 0.60 | 0.60 | 0.30 | | |
| Total | 14 | 2627.19 | | | | |
| $S$ = 1.09844 PRESS = 88.2069 | | | | | | |
| $R^2$ = 99.77% , $R^2$ (pred) = 96.64% , $R^2$ (adj) = 99.36% | | | | | | |

Equation 6 was used to determine the optimum condition of the variable that gives the higher DR%. Then, two experiments were done at the optimal conditions to approve the validity of the predicted optimal response as shown in Table 6. The average removal efficiency of dye from the two experiments was 85.05 %, whereas the predicted value was 85.88%. Both result values (the predicted and experimental) are convergent, which means RSM is a very useful method for optimizing [16].

**Table 6. The experimental and predictive value of VO dye removal% at optimal conditions of variables.**

| Variables | Optimum value | VO dye removal % | |
|---|---|---|---|
| | | Predictive | Experimental |
| Time (min.), $X_1$ | 46.9 | | |
| Kaolin (g), $X_2$ | 0.918 | 85.88 | 85.05 |
| pH, $X_3$ | 3 | | |

The 3D response surface and contour graphs are shown in Fig. 1. It shows the relationships between VO dye removal %(response) and the three variables (Time, Kaolin dose, and pH). All these graphs were drawn at 0 code level. The effect of the Kaolin dosage is shown in Fig. 1(a), it can be seen that with increasing Kaolin Dosage, dye removal increasing and reaching a maximum at range (0.9-1) g of kaolin. On the other hand, the increase of the time led to a slight increase in dye removal. The effect of contact time is shown in Fig. 1(b). It is clear that the increase of time led to an increase in the DR% until reaching maximum at the time range of about (50-60 mints) at pH = 3. Furthermore, an increase of the DR% was obtained with the acidic medium, the high pH, the low dye removal, Figs. 1(b) and (c). Moreover, Fig. 1 has pointed out the interaction intensity between variables. It is noted from the contour graph that kaolin dose and pH are more effective than that of time. Fig. 1(a) shows that at pH = 6, the DR% increases with the increase in the Kaolin dosage, not with increasing time. When the Kaolin Dosage is 0.55 g, the DR% remains in the range of 60-70%, even with increasing time. This indicates that time has less significant effect on the rate of dye removal. In Fig. 1(b), when Kaolin Dosage is 0.55 g and pH = 9, the DR% is low (from 64% to 68%), even when the time increases to more than 50 mins, but at pH = 3, the DR% increases

with time and the maximum removal reaches more than 76%. Therefore, at low pH, DR% is high. Fig. 1(c) shows that when fixing the time at 36 mints, the DR% increases in the acidic medium, and also with the increase in the Kaolin Dosage. The maximum removal is more than 80%. At this point the highest removal is obtained at the acidic medium and when the kaolin dose is increased.

### 3.2. Isotherm adsorption models

It is important to study the adsorption isotherms, to explain the relationship at equilibrium between adsorbate molecules and adsorbent surface at a given temperature. Therefore, various isotherm models were studied, and their constant parameters were calculated. That expresses the surface characteristic and affinity of the adsorbent. Three models were chosen to explicate interaction between (VO dye) and kaolin, by Eq. (7) to (11) [21, 22].

Langmuir $$\frac{C_e}{q_e} = \frac{C_e}{q_{max}} + 1/k_L q_{max} \quad (7)$$

Freundlich $$q_e = k_F \times {C_e}^{1/n} \quad (8)$$

$$lnq_e = lnk_F + \frac{1}{n} lnC_e \quad (9)$$

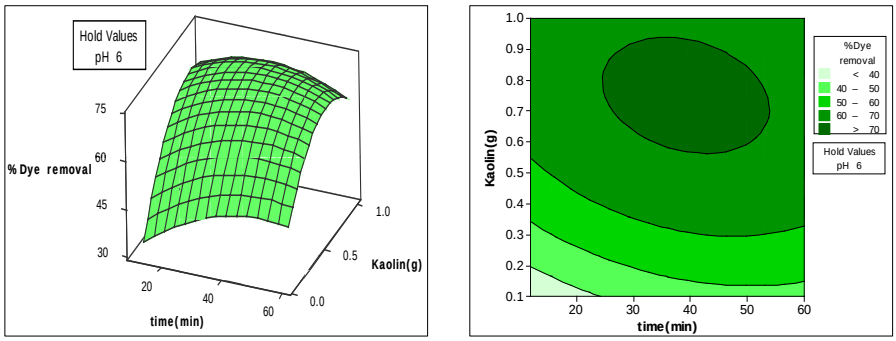
Temkin $$q_e = B_1 lnk_T + B_1 lnC_e \quad (10)$$

$$B_1 = RT/b_1 \quad (11)$$

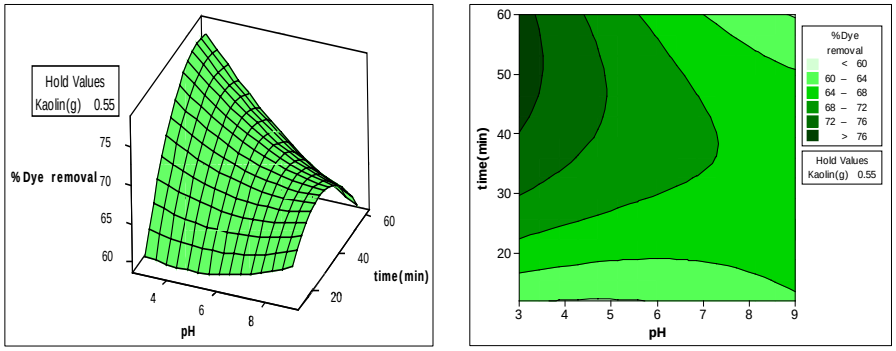
where $C_e$ (mg/ L) is the concentration of VO solution at equilibrium, $q_e$ (mg/g) is the adsorption capacity of equilibrium, $q_{max}$ is the maximum adsorption capacity and $k_L$ (L/mg) is a constant related to the adsorption energy. Both $k_F$ (mg/g) and $n$ Freundlich constants are associated with adsorption capacity and adsorption intensity. $B_1$ is a constant for the heat of adsorption (J/mol). $T$ is the absolute temperature (298 K°) and $R$ is the general gas constant 8.314 J/(mol.K°). $k_T$ is Temkin isotherm equilibrium binding constant (L/g). $b_1$ is Temkin isotherm constant.

Langmuir model is based on the assumption that: the adsorption is a monolayer of solute molecules (dye) on the surface of the adsorbent. The adsorption is assumed to take place at specific homogenous sites with the adsorbent. All adsorption sites are identical and are energetically equivalent [21]. Freundlich isotherm model is used to describe multilayer adsorption on a heterogeneous surface. The Temkin isotherm model has assumed a uniform distribution of energies over surface adsorption sites [22].

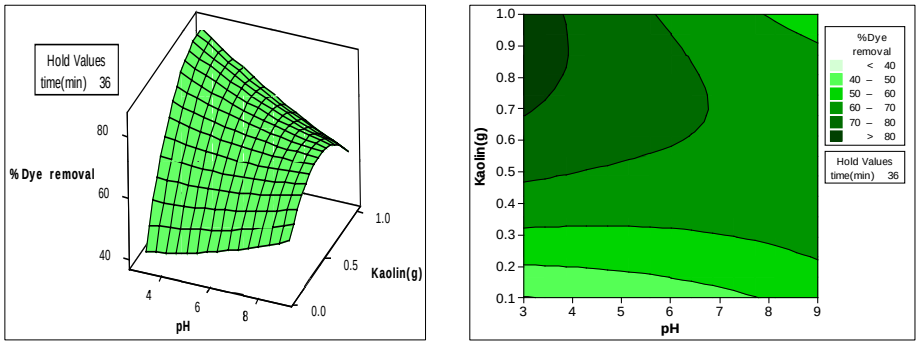
The equilibrium data analysis was shown in Table 7 for the three models; as a comparison, each of, the Langmuir, Freundlich, and Temkin models are indicated a good fit for the experimental data, with $R^2$ = 0.9844, 0.9678, 0.9827, respectively. Table 7 shows that the value of $n$ >1, $n$ is a constant that measures the degree of non-linearity between the solution concentration and adsorption. If the $n$ value is equal to 1, this indicates that the adsorption is a liner, and n value of less than one, indicates that the adsorption process is chemical, whereas $n$ value of more than 1, indicates that the adsorption is a favourable as a physical process. Based on the $n$ value given in Table 7, the adsorption process is physical [22]. Table 8 shows the maximum adsorption capacities of kaolin for different adsorbents.




**(a) Kaolin dose and time.**




**(b) time and pH.**




**(c) Kaolin dose and pH.**

**Fig. 1. 3D response surface and contour graphs of dye removal % and its relations.**

**Table 7. Result of the adsorption Isotherm at 25C°, pH =3, sorbent (kaolin) = 0.5 g.**

| Langmuir | | | Freundlich | | | Temkin | | |
|---|---|---|---|---|---|---|---|---|
| $q_{max}$ mg/g | $k_L$ L/mg | $R^2$ | $K_F$ mg/g | $n$ | $R^2$ | $B_1$ J/mol | $k_T$ L/g | $R^2$ |
| 3.369 | 0.0621 | 0.9844 | 0.325 | 1.7615 | 0.9678 | 0.7698 | 0.566 | 0.9827 |

**Table 8. Maximum adsorption capacities of kaolin for different adsorbents.**

| Adsorbate | Adsorption capacity, $q_{max}$ (mg/g) | References |
|---|---|---|
| Cadmium (II) | 41.84 | [23] |
| Chromium (III)/ (VI) | 11.60 | [23] |
| Zinc (II) | 250.00 | [23] |
| Methylene Blue dye | 52.76 (25°C), (pH 6.0) | [24] |
| Malachite Green dye | 128 (25°C), (pH 6.3) | [25] |
| Procion red H-EGXL dye | 4.51 (pH 3.1) | [19] |
| Basic yellow dye | 1.818 | [26] |
| Anionic dye Congo red | 5.94 (pH 10) | [27] |

### 3.3. Kinetic studies

The equilibrium time between adsorbate and adsorbent was studied in the range of 10-120 min for two different initial concentrations 20 and 40 mg/L of VO dye. The amount of kaolin (0.5 g) was added to 50 mL of the VO dye solution. Figure 2 shows the results of dye removal was increased at the beginning until reached equilibrium after about (65 min) for both concentrations of dye.

It can be concluded that the equilibrium time is independent of concentration. Adsorption of the dye at equilibrium was increased with VO concentration from 20 to 40 mg/L because the diffusion of dye molecules will accelerate from the solution to the adsorbent surface that will take 65 min as equilibrium time [16].

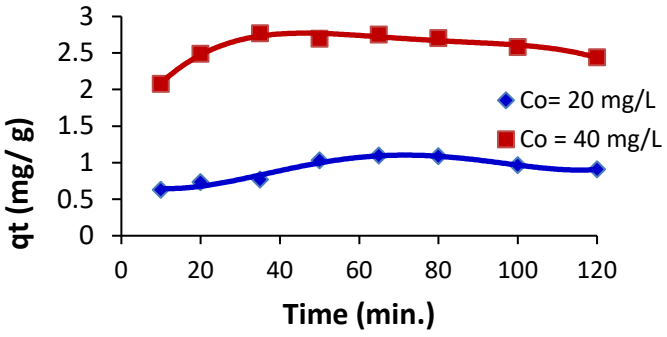



**Fig. 2. The effect of contact time to VO dye adsorption onto kaolin at pH =3 and 0.5 g kaolin, 50 ml of dye solution.**

The study of kinetic adsorption gives information about the mechanism of sorption [23, 24]. In this study, three kinetic models were used to explain the experimental data of dye adsorptions from aqueous solution. These models are given in equations (12-15) [23-25].

Pseudo-first-order model, $\ln(q_e - q_t) = lnq_e - k_1 t$ (12)

Pseudo-second-order model, $\frac{t}{q_t} = \frac{1}{k_2 {q_e}^2} + \frac{t}{qe}$ (13)

where $q_t$ and $q_e$ (mg/g) are the adsorption capacity at equilibrium and any time, $t$ (min), respectively, $k_1$ is the rate constant of pseudo-first-order adsorption (1/min), $k_2$ is the rate constant of pseudo- second- order adsorption (g/mg.min).

$$h = k_2 {q_e}^2 \quad (14)$$

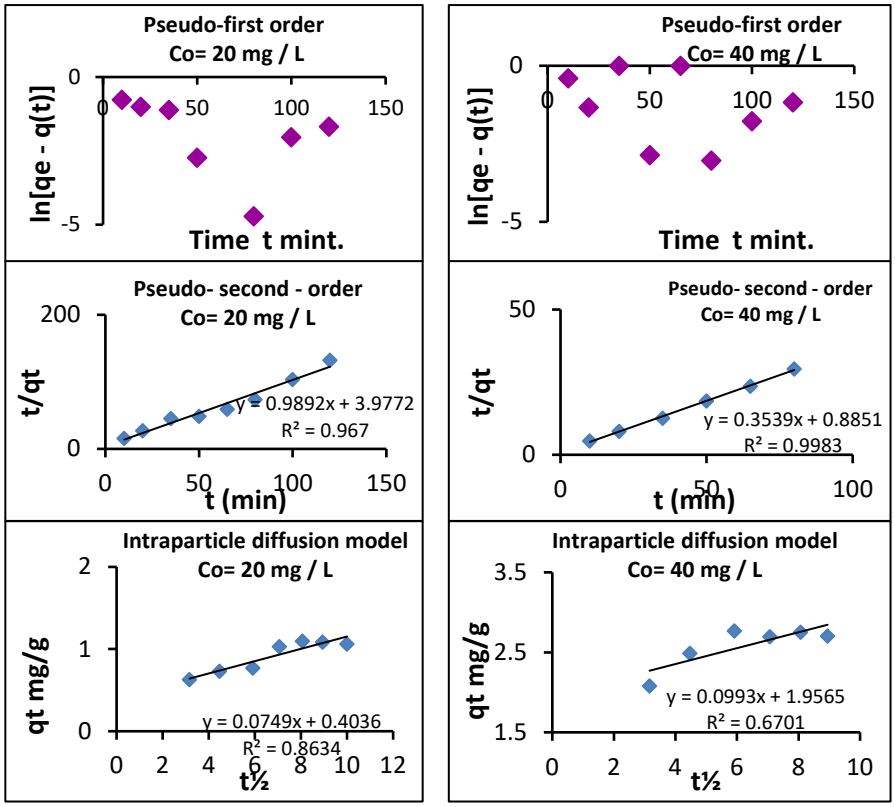
where $h$: the initial sorption rate (mg/g.min) [24]

Intraparticle diffusion model: $q_t = k_t t^{0.5} + C$ (15)

where $k_t$: the intraparticle diffusion rate constant (mg / g. $\text{min}^{0.5}$), $C$: constant, reference to the thickness of the boundary layer [25].

Figure 3 shows the pseudo-first-order model was given the lowest fit with experimental data for both concentrations of dye (20 and 40 mg/L). The pseudo-first-order kinetic model was described sorption based on the sorption capacity of solids [23].

Pseudo-second-order model had the best fitting as shown in Fig. 3 and Table 8 with high $R^2$ (coefficient of determination) value of (0.967 for 20 mg/L) and (0.9958 for 40 mg /L). This model was assumed that one dye molecule is sorbed onto two sorption sites on the sorbent surface; the rate of sorption follows second order chemisorptions [24]. The experimental value of ($q_{e\ exp}$) and calculated value ($q_{e\ cat}$) are convergent for both initial dye concentrations. Each ($q_e$) and ($h$) increase as initial dye concentrations increase, while $k_2$ decreases [16, 25].




**(a) 20 mg/L dye solutions** **(b) 40 mg/L dye solutions**

**Fig. 3. Different kinetic models of VO dye adsorption by kaolin.**

From Table 9 and Fig. 3, it can be seen that the intraparticle model with less $R^2$ value than pseudo-second order (0.8634, 0.6701) for both initial dye concentrations, plotting qt versus $t^{0.5}$, passing through the origin (intercept value $C = 0$) leads to the intraparticle diffusion is the only rate-limiting step. Intercept $C$ has value when the film diffusion also occurs, regarding as a measure of the boundary layer thickness. This means that adsorption processes not only followed the intraparticle diffusion but also the film diffusion. The parameters $k_t$ and $C$ increase as concentrations of dye increase from 20 mg/L to 40 mg/L. The high value of $k_t$ confirmed the rapid transfer [30, 31].

**Table 9. Kinetics parameters for adsorption of VO dye onto kaolin.**

| Models | Parameters | 20 mg/l | 40 mg/l |
|---|---|---|---|
| Pseudo-second order Eqs. (13) and (14) | $k_2$ (g/mg.min) | 0.246 | 0.1415 |
| | $q_{e\ exp}$ (mg/g) | 1.094 | 2.750 |
| | $q_{e\ cal}$ (mg/g) | 1.011 | 2.8257 |
| | $h$ (mg/g.min) | 0.2514 | 1.1298 |
| | $R^2$ | 0.967 | 0.9983 |
| Intraparticle diffusion model Eq. (15) | $k_t$ (mg/g.min$^{0.5}$) | 0.0749 | 0.0993 |
| | C (mg/g) | 0.4036 | 1.9565 |
| | $R^2$ | 0.8634 | 0.6701 |

## 4. Conclusions

The study has investigated the use of cheap natural clay minerals such as kaolin in removing VO dye from its aqueous solution using the adsorption process. The results showed that kaolin could be used to adsorb industrial dyes and to decolorizing industrial wastewaters from the textile factory. The maximum efficiency of dye removal reached to 85.88% at the optimum conditions, of kaolin dosage (0.918 g), pH (3), time (47 min.). At an initial dye concentration of 16 mg/L, the interaction intensity between variables showed that kaolin dose and pH were more effective on dye removal. The equilibrium data of adsorption of the dye followed the Langmuir, Freundlich and Temkin models with $R^2$ = 0.9844, 0.9678, 0.9827, respectively. The kinetic data with the pseudo-second-order model were given a better fit than the pseudo-first-order kinetic model and Intraparticle diffusion model.

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