Mass Transfer Modeling of Acid Violet 17 Adsorption onto Activated Carbon

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Abstract: This study was conducted to determine the suitability of the observed equilibrium and kinetic model, as well as the removal of acid violet 17. The batch experimental variables include the dyes initial concentration and the ratio of the adsorbent mass to the volume of the solution. The adsorption equilibrium data were examined using three equilibrium models i.e., Linear, Freundlich, and Langmuir. The observed model is limited by two forms of the proposed kinetic model. The first model just considers the convection mass transfer of the dyes to the external surface of the adsorbent, while the second model also takes into account the solute diffusion in particles. The experiment result showed that the adsorption process follows the Langmuir correlation with the adsorption capacity of 185.19 mg/g. From this study, it was found that the adsorption is controlled by the diffusion stage inside the particle. The dyes removal reach 58%, which was achieved when 0.0015-gram adsorbent was used per ml solution. The results of this study can contribute to the design of the commercial scale adsorption process.

1 INTRODUCTION

In recent years, the production of dyes continues to grow in line with the growth of the industries using dyes as coloring agents such as paper, textile, cosmetic, and leather tanning (Jain & Gogate, 2017). At present, approximately 10 thousand dyestuff products on the market (Goswami & Phukan, 2017). The chemical structure of dyes is a complex aromatic molecule. Besides, they contain various functional groups. This causes toxic and non-biodegradable properties (Yu et al., 2019). They are very stable, so they tend to accumulate in nature. Globally, the annual consumption of dyes in the chemical industry reaches 7x105 tons, and it was estimated that 10-15% of which was disposed into the water stream, causing serious environmental pollution (Gamoudi & Srasra, 2019), (Belbel et al., 2018).

Acid violet 17 is the anionic azo dyes and is usually used as a coloring agent for textile and leather (Saleem, Pirzada & Qadeer, 2007). The biggest source of dye contamination in the water stream is the textile industry (Li, Mu & Yang, 2019). Disposal of the dyes from industries to the water stream will cause many environmental problems. The presence of dyes in the water, even at low quantity, is undesirable. It will decrease the beauty of the surroundings and threatens human health and the environment (Boudechiche et al., 2019). Besides, its presence in the water body will reduce the penetration of light needed for photosynthesis of aquatic biota (Daoud et al., 2019).

It was reported that the intake of organic dyes into the body could cause damage to several important organs such as the liver, digestive system, and nervous system of human beings (Gao et al., 2019). Other harmful effects of dyes are carcinogenic, mutagenic, and cause kidneys dysfunction (Abd-Elhamid et al., 2019). Therefore, the removal of dyes from the flow of industrial wastewater before being discharged into a water body is a must.

There are several processing methods available to eliminate dyes from wastewater, such as adsorption, photocatalytic degradation, biodegradation, coagulation-flocculation, ion exchange, membrane technology, and electrochemical oxidation (Liu et al., 2019). Among the processing methods for dyes removal from liquid waste, adsorption is still considered to be a more efficient and inexpensive technology because the process is simple, operated at ambient temperature and pressure, and cost-efficient (Thinakaran et al., 2008), (Khorasani & Shojaosadati, 2019).

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Various adsorbents are available to remove dyes from aqueous solutions such as clay, activated carbon, clay, zeolite, carbon nanotubes, graphene oxide, polymeric materials, and agricultural waste (Rajabi, Mahanpoor & Moradi, 2019), (Vahdati-Khajeh et al., 2019). In practice, the most widely used adsorbent for removing dyes from aqueous solutions is activated carbon (Ferreira et al., 2017). Besides having high efficiency, activated carbon has an economic advantage because it can be prepared from agricultural waste materials that are rich in cellulose and lignin (Herrera-González, Caldera-Villalobos & Peláez-Cid, 2019).

Adsorption is one of the most important separation technologies, where the dissolved compound is selectively attracted to the surface of the adsorbent through the mass transfer process resulting in the accumulation of the compound at the surface or the interphase of the adsorbent. Adsorption can occur physically due to physical force such as hydrogen bonding, polarity, Vander Waals forces, and dipole-dipole interactions, etc., or chemically due to the exchange of electrons, where adsorbate is chemically bonded to the surface of the adsorbent (Kausar et al., 2018). In the phenomenon of adsorption, the interaction between solute and adsorbent is very specific, and this depends on the mutual interactions which are influenced by their physical and chemical properties (Benabi et al., 2019).

In general, the mechanism of solute adsorption from an aqueous solution to the surface of a solid adsorbent consists of the following three successive stages, i.e., 1) external mass transfer, where the solute moves from the bulk solution to the outer surface of the adsorbent particle; 2) intraparticle diffusion (pore volume diffusion, surface diffusion, or a combination of both mechanisms); 3) adsorption of the solute molecules on an active site of the adsorbent (Ocampo-Perez et al., 2010).

The adsorption reaction model, which is one of the most widely used to represent dyes adsorption kinetic onto activated carbon and includes pseudo-first-order and pseudo-second-order, ignores both external mass transfer and intraparticle diffusion resistance. (Dotto, Buriol & Pinto, 2014). Thus this model assumes that the adsorption kinetic is fully controlled by the rate of adsorption of solutes on the surface of the adsorbent. Therefore, this model expressed the rate of adsorption as identical to the rate of a chemical reaction (Ocampo-Pérez et al., 2012). The consequence of using this model is that neither the mass transfer coefficient nor the rate-controlling-stage can be obtained. However, to obtain a design for a reliable commercial-scale adsorption process

system, detailed and accurate information regarding mass transfer resistance and the stage which controls the adsorption rate are required. Therefore, in this study, adsorption kinetic models will be developed to determine the external as well as the intra-particle diffusion mass transfer coefficient of acid violet 17 adsorptions onto activated carbon. To solve such an adsorption kinetic model, the adsorption equilibrium correlation at the interface between the solid phase and the liquid phase is required. Hence the objective of this study is 1) to determine the correlation of equilibrium adsorption of acid violet 17 onto activated carbon; 2) to determine the external and the intra-particle diffusion mass transfer coefficients; 3) to investigate the percent dyes removal.

2 THEORY AND EXPERIMENT

2.1 Equilibrium Adsorption Correlation

Equilibrium adsorption correlation is the relationship between solute concentration in the liquid phase and solid phase at equilibrium at a certain temperature. Three adsorption equilibrium models will be investigated for their suitability with the adsorption system studied in this study, namely: the Langmuir model, the Freundlich model, and the Linear model.

The Langmuir equilibrium model is expressed in Equation (1). The Langmuir model assumes: (1) the adsorbent has a homogeneous active side and can only adsorb one layer of the adsorbate molecule, (2) there is no interaction between the absorbed molecules, (3) the same mechanism occurs in each adsorption process. The Freundlich model is stated in equation (2). The Freundlich model assumes that the adsorbent has a heterogeneous surface, and each molecule has different absorption potential. Meanwhile, related to Henry's Law, equilibrium in solid-liquid systems can be described by linear equilibrium. This linear equilibrium is limited to low equilibrium concentrations, i.e., for solute concentrations below 50 ppm. The formulation of linear equilibrium is expressed in Equation (3).

$$\frac{1}{C_{\mu,eq}} = \frac{1}{C_{\mu,max}} + \frac{1}{C_{\mu,max}\,k}\frac{1}{C_{eq}} \qquad (1)$$

$$C_{\mu,eq} = K C_{eq}^{1/n} \qquad (2)$$

$$C_{eq} = H\,C_{\mu,eq} \qquad (3)$$

where, C_{μ}, eq: the mass of solute adsorbed per mass of the adsorbent at equilibrium, C_{μ}, max: the

theoretical monolayer saturation capacity, Ceq: solute concentration in solution at equilibrium, k, K, n: constant, H: a distribution constant.

2.2 Kinetic Model

In designing a system of adsorption processes, the analysis of adsorption kinetic is a very important stage. From the kinetic analysis, information about the time needed to achieve the adsorption equilibrium will be obtained. Furthermore, from the kinetic analysis, the adsorption mechanism will be known (Magdy & Altaher, 2018). Adsorption of solute in porous particles will involve the following stages i.e., external mass transport, intra-particle diffusion, and adsorption on an active site inside the pores. (Ocampo-Perez et al., 2011). In this work, kinetic models were developed on the basis of the following general assumptions: (i) intraparticle diffusion occurs by pore volume diffusion mechanism that follows Fick's law of diffusion, (ii) adsorption reaction at the surface of the adsorbent occurs instantaneously, (iii) as an adsorbent, the activated carbon particles have a spherical shape, (iv) within the adsorbent particles the diffusion occurs only in the radial direction, (v) adsorption takes place under isothermal conditions.

2.2.1 The First Kinetic Model

In the first kinetic model, solute adsorption is considered to only occur from the bulk of the liquid to the outer surface of the adsorbent. Thus there is no gradient of solute concentration within the adsorbent particles. At such condition, the rate of decreasing mass of solute in the liquid phase is equal to the rate of mass transfer of the solute from the bulk solution to the outer surface of the adsorbent, and is mathematically formulated in the following equation:

$$\frac{dC}{dt} = -\frac{k_c A}{V_l}\left(C - C^*\right) \qquad (4)$$

where: t: time, kc: the convection mass transfer coefficient, A: the outer surface area of the adsorbent, C: the solute concentration at the bulk of solution, C*: the solute concentration at the liquid phase interphase, Vl: total solution volume.

Meanwhile, the solute transfer rate from the solution to the adsorbent is equal to the rate of solute increase in the adsorbent, as stated in the following equation.

$$\frac{dC_\mu}{dt} = \frac{k_c A}{m_p}\left(C - C^*\right) \qquad (5)$$

Where: Cμ: solute concentration in the adsorbent,

mp: the mass of adsorbent. The initial condition are: at t=0, C = Co and Cμ = 0. The correlation between the concentration in the solid phase and in the liquid phase at the interface follows the Langmuir model. This correlation acts as a boundary condition.

2.2.2 The Second Kinetic Model

The second kinetic model assumes that the adsorbent is a porous medium so that the mass solute transfer occurs in three stages, namely: (1) convection of the solute from liquid bulk to the outer surface of the adsorbent through the film layer on the outer surface of the adsorbent, (2) intraparticle liquid phase diffusion of solute from the outer surface of the adsorbent to the active site of the adsorbent through the pore, (3) attachment of the solute to the active site of the adsorbent in the pore.

$$\left(-D_e\, 4\pi\, r^2 \left.\frac{\partial C}{\partial r}\right|_r\right) -$$

$$\left(-D_e\, 4\pi (r + \Delta r)^2 \left.\frac{\partial C}{\partial r}\right|_{r+\Delta r}\right) =$$

$$4\pi\, r^2 \Delta r\, \frac{\partial}{\partial t}\left(\varepsilon C + C_\mu \rho_{ads}\right) \qquad (6)$$

$$D_e\, 4\pi \left(\frac{\Delta r}{2}\right)^2 \left(\frac{\partial C}{\partial r}\right) =$$

$$\frac{4}{3}\pi \left(\frac{\Delta r}{2}\right)^3 \left(\varepsilon + \frac{\rho_{ads}}{H}\right)\left(\frac{\partial C}{\partial t}\right) \qquad (7)$$

$$k_c\, 4\pi\, R^2 \left(C_L - C_L^*\right) = D_e\, 4\pi\, R^2 \left(\frac{\partial C}{\partial r}\right)_{surface} \qquad (8)$$

In the second model, it is assumed that diffusivity and porosity are uniforms in all parts of the adsorbent. While, as in the first model, the equilibrium between the solute concentration in the liquid and solid phases at the interface follows the Langmuir model. The second model was developed by applying the principles of solute mass balance in the particle and manifested in three differential equations i.e. equation (6) which applicable at the middle of the solid adsorbent, equation (7) which applicable at the center of the solid adsorbent and equation (8) which applicable on the surface of the adsorbent, where, C: concentration of solute in the liquid which fills the adsorbent pore, ρads :density of the adsorbent, ε: porosity of the adsorbent, R: adsorbent radius, r: position in adsorbent in radial system, De: the effective diffusivity of the solute, CL: solute

concentration in the bulk of the solution, CL*: solute concentration in the solution at the interface.

2.3 Experiment

The acid violet 17 was obtained from a local chemical supplier. The activated carbon, having particle size <100 μm and bulk density of 150 - 440 kg/m3, was purchased from Merck Millipore. In all batch adsorption experiments, activated carbon and acid violet 17 were mixed in 250 ml beaker while stirring continuously at a constant speed, in which the volume of acid violet 17 solution was 100 ml. Experimental variables include: initial concentration of acid violet 17 (75 ppm, 125 ppm, 175 ppm, 225 ppm, 275 ppm) and mass of activated carbon used (0.1 g; 0.1125 g; 0.125 g; 0.1375 g; 0.15 g). The concentration of acid violet 17 in solution at certain times and equilibrium was measured using a UV-Vis spectrophotometer - Agilent 8453.

3 RESULTS AND DISCUSSION

3.1 Equilibrium Analysis

In this study, the equilibrium condition was assumed to be achieved by allowing the batch adsorption process to take place overnight. In this study, an investigation of three equilibrium models was carried out, i.e., Langmuir, Freundlich, and linear equilibrium. The plot of equilibrium data obtained from the experiment and plot of equilibrium models is presented in Figure 1.
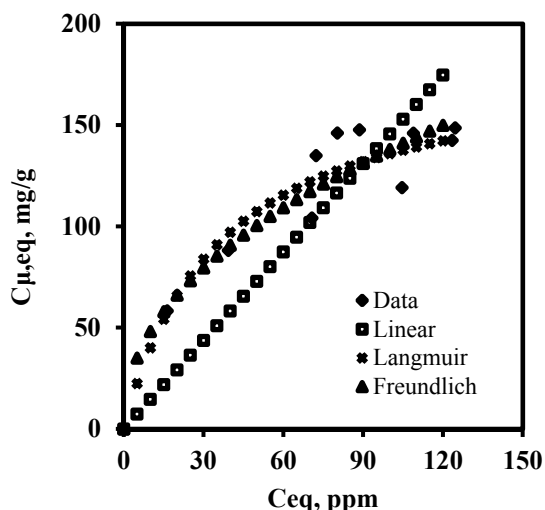
Figure 1: Plot the experimental equilibrium data and the equilibrium model.

The equilibrium parameters and the coefficient of determination ($R2$) for each equilibrium model observed for this system are presented in Table 1.

Table 1: The equilibrium parameters obtained for each observed equilibrium model.

The observed equilibrium model	The equilibrium parameters		R^2
Linear	H	0.69 g/l	0.78
Freundlich	K	16.76	0.88
	$^1/_n$	0.48	
Langmuir	k	0,28 l/mg	0.93
	$C_{\mu,max}$	185.19 mg/g	

From the coefficient of determination, it can be concluded that the Langmuir equilibrium model is the most appropriate for this system. This result has shown that the adsorption process takes place on a homogeneous solid surface, and the adsorbed dyes form a monolayer on the surface of activated carbon. Furthermore, based on Langmuir's equilibrium parameters obtained from this experiment, it can be seen that the maximum adsorption capacity of the activated carbon is 185.19 mg/g.

3.2 Kinetic Analysis

Kinetic models that are often used to observe adsorption kinetic include the pseudo-first-order and pseudo-second-order equations. The advantage of the model is that the equation is simple, but both models ignore the mass transfer or diffusion step that occurs in adsorption and only taking into account the rate of attachment of the solute to the surface of the adsorbent (Yao & Chen, 2018).

In contrast, in this study the rate of attachment of solutes to the surface of the adsorbent was considered to be very fast so that the rate of the adsorption was controlled by the rate of solute transfer across the liquid film to the outer surface of the adsorbent and/or the rate of solute diffusion within adsorbent particles.

The governing equations for the mass transfer in the liquid film and the intraparticle diffusion are partial differential equations, as expressed in the first and second kinetic models. To obtain the mass transfer coefficient of the solute in the liquid film and the effective diffusivity of the solute within the particles, the two models, are solved numerically using the Matlab program.

3.2.1 The Solution of the First Kinetic Model

The kinetic solution, which is expressed as changes in solute concentration in liquid bulk with respect to time, for the first model, is presented in Figure 2. The solution applied the Langmuir model for the calculation of concentration in the interface.

From Figure 2, it can be seen that for lower concentrations, the first model kinetic is better suited to this adsorption system. It is shown that for lower solute concentrations, the results obtained from the model are closer to the data obtained from the experiment.

Meanwhile, the mass transfer coefficient of liquid films (kc), obtained from the application of the first kinetic model at an experiment with an adsorbent mass 0.1 gram, was presented in Table 2.

The AAD parameter is the absolute average deviation used to determine the level of suitability of the model to experimental data. The AAD parameter is defined in Equation 9, where: C_e = dyes concentration obtained from the experimental result, C_m = dyes concentration obtained from the observed kinetic model, N = the number of the experimental data.

$$AAD = \frac{\sum_i^N \left| \frac{C_e - C_m}{C_e} \right|}{N} \quad (9)$$
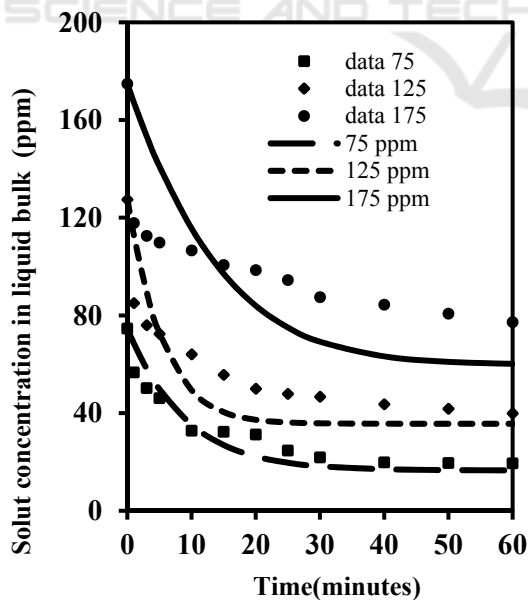
Figure 2: Plot solute concentration in the liquid bulk versus time at various initial solute concentrations. The results of the first kinetic model were expressed as line, while the experimental data were expressed as dot.

Table 2: Mass transfer coefficient at various solute initial concentration applying the first kinetic model at an experiment with an adsorbent mass 0.1 gram.

Initial concentration (ppm)	kc (cm/minute)	AAD (%)
75	1.24 x 10-4	25.1
125	1.97 x 10-4	19.7
175	1.37 x 10-4	17.9
225	8.66 x 10-5	16.5
275	8.55 x 10-5	19.2

Meanwhile, the kc values at a various ratio of the adsorbent mass to the solution volume are presented in Table 3.

Table 3: Mass transfer coefficient at various ratio adsorbent mass to solution volume applying the first kinetics model when the initial solute concentration 275 ppm.

Ratio the adsorbent mass to the solution volume (g/ml)	kc (cm/minute)	AAD (%)
0.001	8.56 x 10-5	15.7
0.001125	8.38 x 10-5	22.0
0.00125	1.05 x 10-4	23.2
0.001375	9.46 x 10-5	22.5
0.0015	8.78 x 10-5	22.6

From the results obtained, it can be seen that the change in solute concentration and the ratio of the mass of adsorbent to the volume of solution does not significantly influence the mass transfer coefficient. The value of the mass transfer coefficient is in the range of 8.38 x 10-5 - 1.97 x 10-4 cm/minute, with the absolute average deviation in the range of 15.7% - 25.1%.

3.2.2 The Solution of the Second Kinetic Model

The value of the mass transfer coefficient and effective diffusivity obtained by applying the second kinetic model are presented in Table 4 for various initial solute concentrations and Table 5 for the various ratio of the mass of adsorbent to the volume of solution.

Table 4: The value of the mass transfer coefficient and effective diffusivity obtained by applying the second kinetic model for various initial solute concentrations.

Initial concentration (ppm)	kc cm/minute)	De (cm2/ minute)	AAD (%)
75	0.28	2.13 x 10-5	7.6
125	0.25	2.13 x 10-5	13.5
175	0.19	1.06 x 10-5	14.4
225	0.15	7.38 x 10-6	7.9
275	0.10	5.52 x 10-6	9.8

By applying the second kinetic model, the mass transfer coefficient value tends to decrease with increasing solute concentration. It also found that the ratio of the adsorbent mass to the solution volume does not have much effect on the mass transfer coefficient.

Table 5: The value of the mass transfer coefficient and effective diffusivity obtained by applying the second kinetic model for the various ratio of the mass of adsorbent to the volume of the solution when the initial solute concentration 275 ppm.

Ratio the adsorbent mass to the solution volume (g/ml)	kc (cm/ minute)	De (cm2/ minute)	AAD (%)
0.001	0.13	6.54 x 10-6	4.1
0.001125	0.12	6.06 x 10-6	9.7
0.00125	0.11	8.22 x 10-6	12.6
0.001375	0.10	1.05 x 10-5	12.2
0.0015	0.10	1.36 x 10-5	11.8

The mass transfer coefficient obtained by applying the second kinetic model is in the range of 0.10 – 0.28 cm/minute. This value is far greater than the mass transfer coefficient obtained from the application of the first kinetic model, which is in the range of 8.38 x 10-5 - 1.97 x 10-4 cm/minute.

Meanwhile, the value of the effective diffusivity tends to decrease from 2.13 x 10-5 cm2/minute to 5.52 x 10-6 cm2/minute when the solute concentration increases from 75 ppm to 275 ppm and it tends to increase from 6.54 x 10-6 cm2/minute to 1.36 x 10-5 cm2/minute when the ratio of the adsorbent mass to the solution volume is increased from 0.001 g/ml to 0.0015 g/ml.

When the second kinetic model was applied, the absolute average deviation value is in the range of 4.1% -14.4%. It was much lower than its value when the first kinetic model was applied, which was in the range of 15.7% - 25.1%. This shows that, compared to the first model, the second model is more suitable for this case. Furthermore, these results also show that in this case, the transfer of solutes into the particle is controlled by the solute diffusion step within the adsorbent particle. The determining factor for the controlling stage is the pore structure of the particles. The pore structure will determine the resistance of solute diffusion within the particle.

3.3 Dyes Removal

The ability of adsorbent to absorb dyes is shown by its removal value. Basically, the removal of the dye is the mass of the dyes separated from the solution to the initial mass of the dye.

The acid violet removal profile overtime at various mass fractions of the adsorbent in the solution when the initial solute concentration 275 ppm was presented in Figure 3. The profile shows that up to 60 minutes, the percentage of dyes removal is still increasing. This means that up to 60 minutes, the adsorption process has not finished.

It can also be seen that the greater the ratio of the mass of adsorbent to the volume of solution, the greater the removal of the dyes. The highest removal achieved was 58% when used 0.0015 g adsorbent per ml solution.
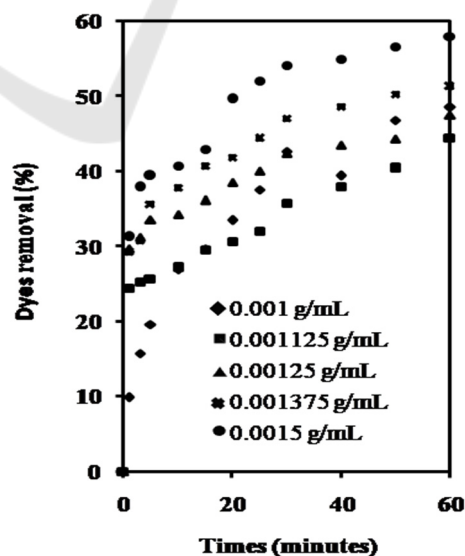
Figure 3: Acid violet 17 removals at various ratio adsorbent mass to solution volume, when the initial solute concentration 275 ppm.

4 CONCLUSIONS

Adsorption of acid violet 17 to activated carbon takes place on a homogeneous solid surface form a single layer at the surface. The maximum dyes removal capacity is 185.19 mg/g. From the kinetic study, it was known that the mechanism of acid violet 17 adsorptions on activated carbon is determined by the diffusion stage in the particle. The percent dyes removal was 58% achieved when 0.0015 g adsorbent was used per ml solution.

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