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Adsorption Kinetics and Isotherm Models: A Review

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Adsorption Kinetics describes the rate at which solute is adsorbed and the resident time of the adsorbates on the solid-liquid interface. Adsorption isotherms play important role in determining the interaction between adsorbate and adsorbent and the optimum adsorption capacity of adsorbent. This article considered selected adsorption kinetics and isotherms models. Pseudo first order, Pseudo second order, Elovich, Bhattacharya and Venkobachar, and Natarajan and Khalaf were adsorption kinetics reviewed on the assumption that the process behaves as heterogeneous reaction at solid-liquid interface. Adsorption kinetics equation presented takes the form of straight line, the slopes and intercepts of the plots are used to determine adsorption capacity of adsorbent, rate constant, rate of adsorption and intraparticle diffusion. Value of correlation coefficient obtained is used in determining the adsorption kinetics model that best describe the adsorption process. Langmuir, Freundlich, Radlich-Peterson, Temkin and Dubinin-Radushkevich adsorption isotherms were presented. Their slopes and intercepts provide insight on adsorption affinity, mean free energy, whether the adsorption is physisorption or chemisorptions, single or multilayer. Adsorption kinetics and isotherms reviewed provide essential information required for understanding adsorption process.

Keywords: Adsorption, kinetics, isotherms, pseudo first order, Langmuir

1. Introduction

Adsorption is a process where attractive forces associate a solute (adsorbate) to the surface of an adsorbent until equilibrium in the concentration of the adsorbate is attained and no further net adsorption occurs (Yousef, Qiblawey, and El-Naas, 2020; Mhemeed, 2018). It could also be defined as the formation of layer of adsorbate on the surface of an adsorbent by the attraction of Van der Waals' forces. The process involves the accumulation of adsorbate at an interface relative to its concentration in the bulk solution (Hu and Xu, 2020). The adsorbent used for adsorption consists of porous medium with a high surface area (Yousef *et al.*, 2020). According to Da ąrowsk (2001), adsorption of solute onto adsorbent takes the following stages: diffusion of molecules from the bulk phase towards the interface space (external diffusion); diffusion of molecules inside the pores (internal diffusion); diffusion of molecules in the surface phase (surface diffusion); and adsorption-desorption elementary processes.

Adsorption could be physical (physisorption) or chemical (chemisorption). Physical adsorption is a result of weak, short range electrostatic

attractive forces arising from dipole moment (Van der Waals); and chemisorption involves bond formation between the adsorbate and the adsorbent. It is important not to confuse adsorption with absorption. While adsorption is the attraction and/or bonding on the surface of adsorbent, absorption is the process by which solute is taken across a membrane or taken up into a structure. Adsorption is one of the most effective means of removing contaminants from solutions (Ademiluyi and Nze, 2016).

Adsorption Kinetics is an important factor that defines the effectiveness of sorption. Kinetics of adsorption basically describes the rate at which solute is adsorbed and the resident time of the adsorbates on the solid-liquid interface. Adsorption velocity depends upon the number of particles adsorbing on the adsorbent surface per second and the number of particles colliding in unit area per second (Meera and Ganesan, 2015).

Adsorption isotherms are useful in describing the interaction between adsorbate and adsorbent. They play important roles in determining the

optimum adsorption capacity of adsorbent and indicate how efficient an adsorbent can adsorb, and also allows an estimation of the validity of adsorbent's application (Prasat, Muthirulan and Kannan, 2014). Isotherms also represent the amount of adsorbate that binds on the surface of adsorbent depending on the material available in the solution and are important requirements in designing a system for adsorption.

2. Adsorption Kinetics

Adsorption kinetics describes the rate of retention or release of a sorbate from aqueous solution to solid-phase interface. In adsorption, linear or non-linear analysis of the kinetics is applied. The goodness of fit index is applied to determine the model that best describe the process (Musah, Yisa, Suleiman, Mann, Shaba and Aliyu, 2018). Kinetics of adsorption onto the surface of activated carbon can be ascertained by the following models:

i. Pseudo first order (Largergren model)

The model considers the rate of change that occurs in the uptake of adsorbate at a particular reaction time to be directly proportional to the difference in the concentration and rate at which the adsorbate is removed with time. The model also known as Largergren model is represented by the equation below (Edet and Ifelebuegu, 2020; Santuraki and Muazu, 2015):

$$\frac{dq_t}{dt} = k_1 (q_e - q_t) \quad (1)$$

Where:

q_e is the adsorption capacity of the adsorbent at equilibrium (mg.g^{-1}).

q_t is the adsorption capacity of the adsorbent at time t (mg.g^{-1}).

k_1 is the rate constant for pseudo first order adsorption (min^{-1}).

After equation 1 is integrated and boundary conditions are applied,

$t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_e$, the integrated form of the equation (1) becomes:

$$\log (q_e - q_t) = \log (q_e) - \frac{k_1}{2.303} t \quad (2)$$

When values of $\log (q_e - q_t)$ were correlated linearly with t . The plot of $\log (q_e - q_t)$ versus t gives a relationship that is linear from which k_1 and q_e can be determined from the slope and intercept of the plot.

ii. Pseudo second order

According to Ademiluyi and Nze (2016), the adsorption kinetics rate equation for pseudo second order is expressed as:

$$\frac{dq_t}{dt} = k_2 (q_e - q_t)^2 \quad (3)$$

Where:

k_2 is the rate constant of the pseudo second order adsorption ($\text{g.mg}^{-1}.\text{min}^{-1}$).

The integrated form of the equation for the boundary conditions $t = 0$ to $t = t$ and $q_t = 0$ to $q_t = q_e$ becomes:

$$\frac{1}{q_e - q_t} = \frac{1}{q_e} + kt \quad (4)$$

Equation 4 is the integrated form of the rate law of pseudo second order reaction whose linear form is rearranged and expressed in equation 5 (Ebelegi, Ayawei and Wankasi, 2020; Ademiluyi and Nze, 2016):

$$\left[\frac{t}{q_t} \right] = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} (t) \quad (5)$$

If the initial rate of adsorption, h ($\text{mg.g}^{-1}.\text{min}^{-1}$) is:

$$H = k_2(q_e) \quad (6)$$

Substituting (6) into (5) yields:

$$\left[\frac{t}{q_t} \right] = \frac{1}{h} + \frac{1}{q_e} (t) \quad (7)$$

The plot of (t/q_t) and t in equation (7) will give a linear relationship whose slope and intercept can be used to determine q_e and k_2 respectively (Edet and Ifelebuegu, 2020).

iii. Elovich model

This model is used to further describe the pseudo second order kinetic on the assumption that the sorbent surface is energetically heterogeneous. Elovich model has also been used for the chemisorptions of gases onto heterogeneous surfaces of adsorbent (Edet and Ifelebuegu, 2020).

The Elovich equation is given as:

$$\frac{dq_t}{dt} = \alpha e^{-\beta q_t} \quad (8)$$

Where:

α is the initial rate of adsorption ($\text{mg.g}^{-1}.\text{min}^{-1}$)

β is the desorption constant ($\text{g}\cdot\text{mg}^{-1}$) during an experiment.

To simplify the equation, it can be assumed that:

$\alpha\beta t \gg t$, and also applying boundary conditions; $q_t = 0$ at $t = 0$ and $q_t = q_t$ at $t = t$, equation 8 becomes (Farouq and Yousef, 2015):

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln(t) \quad (9)$$

If adsorption process fits the equation, then the plot of q_t versus $\ln(t)$ would give a linear relationship with the slope of $(1/\beta)$ and an intercept of $(1/\beta) \ln(\alpha\beta)$.

iv. Bhattacharya and Venkobachar

Bhattacharya and Venkobachar kinetic model is a simplified first-order reversible kinetic model. It is based on solution concentration and is useful in the study of adsorption mechanism and determining characteristic constants of sorption (Venkatesan and Rajagopalan, 2016).

$$\log[U - U(T)] = -\left(\frac{k_B}{2.303}\right)t \quad (10)$$

$$U(T) = \frac{C_0 - C_t}{C_0 - C_e} \quad (11)$$

Where

k_B is a constant (min^{-1}); C_0 is initial concentration (mg/dm^3); C_t and C_e are concentration (mg/dm^3) at time t and at equilibrium. The value of k_B is calculated from the slope of the line obtained from the plot of $\log[1 - (U)T]$ versus t

v. Natarajan and Khalaf

Natarajan and Khalaf's equation depict a relationship between the initial concentration of adsorbate and concentration at any time (Ayash, Elnasr and Soliman, 2019; Venkatesan and Rajagopalan, 2016). The linear form is expressed as:

$$\log\left(\frac{C_0}{C_t}\right) = \left(\frac{k}{2.303}\right)t \quad (12)$$

Where

C_0 and C_t are concentrations (mg/dm^3) at time zero and time t respectively. k is the first order adsorption rate constant (min^{-1}) which is calculated from slope of the plot $\log(C_0/C_t)$ against t .

vi. Intra-particle diffusion

The mechanism of adsorption by adsorbent is described by the mode of diffusion or transportation of adsorbate from the solution onto the solid phase. The intra-particle diffusion model

can be expressed as (Patil, Renukdas and Patel, 2012):

$$R = k_{id}(t)^a \quad (13)$$

Where:

R is the percentage adsorbed

t is the time of contact (h)

a is the linear plots' gradient and depict the mechanism(s) of adsorption

k_{id} is the intra-particle diffusion rate constant (h^{-1}) and could be taken as rate factor (% removed per unit time). Linear form of the equation (13) is:

$$\log qt = \log k_{id} + n \log(t) \quad (14)$$

The values of k_{id} can be calculated from slope of the plot of $\log qt$ against $0.5 \log t$.

Where k_{id} and n are the intraparticle diffusion rate constant and mechanism of sorption respectively

Correlation coefficient (R^2) and sum of square error (SSE) are error functions that can be employed to decide the kinetic model that best describes the interaction of adsorbent and adsorbate.

3. Adsorption Isotherms

Adsorption isotherms describe the interaction of adsorbate with adsorbent and so are important for optimizing the use of any adsorbent. The shape of an isotherm provides information on the stability of the interactions between adsorbent and adsorbate and on the adsorption affinity of molecules. Adsorption isotherms are described in many mathematical forms, some are based on simplified physical description of adsorption, while others are empirical and have to correlate experimental data (Obaid, 2020).

i. Langmuir adsorption isotherm

The basic assumption of the Langmuir theory is that adsorption occurs at specific homogeneous sites within an adsorbent and, once an adsorbate occupies a site, no further adsorption can occur on that site.

The model is useful in predicting the performance of different adsorbent. A K_L value (related to the energy of sorption) that is indicating high adsorption affinity and q_m representing maximum adsorption capacity (i.e monolayer saturation) is known to decrease with increasing temperature (Sampranpiboon et al., 2014).

From his discovery of surface chemistry, Langmuir formulated a relationship between the amount of gas adsorbed onto a surface and gas pressure, which is now known as the Langmuir adsorption isotherm. In his model, he assumed that:

- There are fixed adsorption sites on the surface of each adsorbent and a fraction of these sites could be occupied by adsorbates at a given temperature and pressure
- Each adsorption site on the surface of adsorbent can accommodate one entry
- Heat of adsorption for each adsorption site is the same and independent of the fraction of sites occupied by adsorbate.
- Interaction does not exist between adsorbates occupying different adsorption sites (Musah, Yisa, Suleiman, Mann and Shaba, 2018; Meera and Ganesan, 2015).

From the assumptions above, the Langmuir adsorption isotherm is valid for only monomolecular adsorption and is expressed as (Islam, Chowdhury, Mozumder and Uddin, 2021; Ademiluyi and Nze, 2016):

$$q_e = \frac{bCQ^o}{1 + bC} \tag{15}$$

where:

q_e is the amount of adsorbate removed per unit weight of adsorbent

Q^o is a constant relating to single layer adsorption capacity

b is the constant which measure the surface energy of the adsorption process

C is the concentration of adsorbate in solution at equilibrium

The linear forms of the Langmuir equations are depicted in equations (16) and (17) respectively.

$$\frac{C}{q_e} = \frac{1}{bQ^o} + \frac{C}{Q^o} \tag{16}$$

$$\frac{1}{q_e} = \frac{1}{Q^o} + \left[\frac{1}{bQ^o} \right] \left[\frac{1}{C} \right] \tag{17}$$

The plot of $1/q_e$ against $1/C$ would give a straight line with slope $1/bQ^o$ and intercept of $1/Q^o$

The Langmuir isotherm model can be used in describing the equilibrium conditions for adsorption and to provide parameters (Q^o and b) which can be used to quantitatively compare

adsorption behaviours in different adsorbate-adsorbent system (Obaid, 2020).

Langmuir adsorption isotherm can also be expressed using the equilibrium parameter, R_L a dimensionless constant separation factor expressed as:

$$R_L = \frac{1}{1 + bC_o} \tag{18}$$

Where, C_o (mg/dm^3) is the highest initial concentration and b (dm^3/mg) is the Langmuir constant (Prasat *et al.*, 2014).

The parameter, R_L indicates the nature of the isotherm shape as interpreted in Table 1.

Table 1: Parameters of Langmuir Isotherm

R_L Value	Type of Isotherm
$R_L > 1$	Unfavourable
$R_L = 1$	Linear
$R_L = 0$	Irreversible
$0 < R_L < 1$	Favourable

ii Freundlich adsorption isotherm

The Freundlich adsorption isotherm is usually employed to quantify the equilibrium relationship between the amount of adsorbate removed per unit weight of the carbon and the adsorbate concentration still remaining in the solution (Sampranpiboon, Charnkeitkong and Feng, 2014). Freundlich adsorption isotherm is expressed as:

$$q_e = K_f C^{1/n} \tag{19}$$

Where:

n is a constant related to efficiency of sorption and sorption energy.

K_f is a constant measuring adsorption capacity

q_e is the amount of adsorbate removed per unit weight of carbon

C is the equilibrium concentration of the adsorbate in solution (Mohamed, Abdelkarim, Ziat and Mohamed, 2016),

The Freundlich equation is basically empirical, but it is sometimes used as a means for data description. This is due to the adequacy of the equation in describing non-linear adsorption process in a range of adsorbate concentrations. Its mathematical simplicity allows its usage easily, and its ability to describe adsorption process on surface adsorption sites that are energetically heterogeneous. Freundlich equation can be used in linear form by obtaining the logarithm of both sides of the equation:

$$\log q_e = \log K_f + \frac{1}{n} \log C \tag{20}$$

The plot of $\log q_e$ versus $\log C$ will give a straight line with $1/n$ and $\log K_f$ as slope and intercept respectively. The K_f value increases as the capacity of the activated carbon to remove adsorbate also increases (Paudel and Shrestha, 2020). Freundlich isotherm, though correctly establish the relation between adsorption and pressures at low values but does not predict the value of adsorption when pressure is high.

iii Radlich-Peterson adsorption isotherm

It is an intermediate adsorption isotherm that combines both features of Freundlich and Langmuir adsorption isotherms in a single equation. This isotherm model gives a more realistic representation of an adsorption system that operates over a range of concentrations (Musah *et al.*, 2018b; Sheba and Nandini, 2016). The general Radlich-Peterson isotherm equation is given as:

$$q_e = \frac{KRC_e}{1 + aRC_e} \quad (21)$$

The equation is expressed in linear form as:

$$\ln \frac{C_e}{q_e} = g \ln C_e - \ln KR \quad (22)$$

Where, aR (dm^3/mg) and K_R (dm^3/g) represents constants of Redlich-Peterson isotherm while g represents exponent between 0 and 1. A plot of $\ln \frac{C_e}{q_e}$ versus $\ln C_e$ is used to obtain values of values g and $\ln KR$ (Musah *et al.*, 2018b; Sampranpiboon *et al.*, 2014)

iv Temkin adsorption isotherm

According to the Temkin isotherm, the heat of sorption should decrease linearly with sorption coverage on the adsorbent due to interactions of adsorbent and adsorbate (Obaid, 2020; Sampranpiboon *et al.*, 2014). Temkin isotherm equation is expressed as:

$$q_e = \frac{R_T}{b_T} \ln(k_t C_e) C \quad (23)$$

Equation 23 can be presented in the linear form as:

$$q_e = \frac{R_T}{b} \ln k_t + \frac{R_T}{b} \ln C_e \quad (24)$$

Where:

K_t (dm^3/g) is equilibrium binding energy which corresponds to the optimum binding energy.

b_T (J/mol) is a constant which is related to the heat of adsorption.

R is the ideal gas constant; and

T is the absolute temperature (Edet and Ifelebuegu, 2020; Javadian, Ghorbani, Tayebi and Hosseini 2015).

v Dubinin-Radushkevich (D-R) adsorption isotherm

This isotherm assumes that the porosity of adsorbent is related to the characteristics of adsorption curve (Sampranpiboon *et al.*, 2014). It is useful in determining the nature of what occurred, whether it is chemisorption or physisorption and to determine the apparent energy of adsorption. D-R does not assume that the surface of adsorbent is homogeneous (Edet and Ifelebuegu, 2020). The linear form of the model expressed as:

$$\ln q_e = \ln Q_D - B_D + [RT \ln(1 + \frac{1}{C_e})]^2 \quad (25)$$

where Q_D (mol/g) represents the maximum adsorption capacity and B_D (mol^2/KJ^2) are D-R constants derived from the slope and intercept of the plot of $\ln q_e$ versus $RT \ln(1 + 1/C_e)$. The mean free energy of adsorption, E (KJ/mol); the required energy for transfer of one mole of adsorbate from solution to the surface of adsorbent is determined using the expression:

$$E = \frac{1}{\sqrt{2B_D}} \quad (26)$$

B_D (mol^2/KJ^2) is the free energy of sorption per mole of the adsorbate as it moves towards the surface of the adsorbent and Q_D (mol/g) is related to the extent of adsorption of adsorbate (Edet and Ifelebuegu, 2020).

4. Analysis of Error

Residual root mean square error (RMSE) and chi-square test can be used to evaluate the fit of adsorption isotherm equation from experimental data in determining the model parameters.

The Residual root mean square error is expressed as:

$$\text{RMSE} = \sqrt{\frac{1}{n-2} \sum_{n=1}^n (q_{e,\text{exp}} - q_{e,\text{cal}})^2} \quad (27)$$

Where:

Subscripts 'exp' and 'cal' are the experimental and theoretical values of q_e

n is the number of observations in the experiment. Small value of residual mean square error (RMSE) indicates better fit for the adsorption process.

The chi-square test is given as:

$$X^2 = \frac{(q_{e,exp} - q_{e,cal})^2}{q_{e,exp}} \quad (28)$$

A small X^2 value is an indication of a good fit of a model to an adsorption process (Musah *et al.*, 2018a; Samprapiboon *et al.*, 2014).

5. Conclusion

This article discussed adsorption kinetics and isotherms, their mathematical expressions, defined parameters in the expressions and information that could be obtained. Parameters in adsorption kinetics provide information on adsorption capacity of adsorbent, rate of adsorption and amount of adsorbate removed at equilibrium and at any given time. Adsorption provide insight into the type of layer formed during adsorption, whether the process is favourable or not, physical or chemical. Adsorption kinetics and isotherms can be applied to adsorption processes especially those that involved variations in time of interaction and concentration of adsorbate.

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Conflict of interest

The authors declare no conflict of interest.

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