Differences between Chemical Reaction Kinetics and Adsorption Kinetics: Fundamentals and Discussion

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ABSTRACT
Adsorption kinetics is an essential part in adsorption studies. The pseudo-first-order (PFO) and pseudo-second-order (PSO) models are frequently used to model the experimental dataset of time-dependent adsorption. The differential equations (based on reaction rate and rate law) of the PFO and PSO models are similar to those of chemical reactions (i.e., first and second order-kinetic reactions). The adsorption kinetics is illustrated through the plot of $q_t$ (the amount of adsorbate adsorbed by adsorbent at time t) vs. time. This plot includes two important regions (kinetic and equilibrium). The adsorption rate constant ($k_1$(PFO) or $k_2$(PSO), respectively) of the PFO or PSO models needs to be calculated from two regions. The appropriate selection of initial adsorbate concentrations for studying adsorption kinetics should be based on adsorption isotherm to ensure that adsorption sites in adsorbent (material) are highly (nearly fully) covered by adsorbate (solute). Only in this case, the rate constant of the adsorption is accurately obtained.

KEYWORDS
Chemical reaction kinetic; Adsorption kinetics; Pseudo-first-order model; Pseudo-second-order model; General kinetic model.

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1. Introduction

Kinetics is an important component in many studies (i.e., chemical reaction and adsorption). In adsorption process, adsorption and desorption occurs simultaneously until adsorption equilibrium reached. Recently, many scholars commented many problems and mistakes related to modelling, presenting, and explaining the results of adsorption kinetics [1, 2]. In essence, adsorption kinetics has some difference to chemical reaction kinetics. Figure 1 shows a brief summary on such similarity and difference. This study mainly focuses on the derivation and discussion on adsorption kinetics. In addition, some technical terms are also defined and corrected herein.

![Figure 1. Brief summary on similarity and difference between chemical reaction kinetics and adsorption kinetics](image)

Before discussing some common reaction models used in adsorption kinetics, it is necessary to explain basic information on the “reaction rate”, “rate law”, and “rate constant” of reactions. Several relevant integration formulas that are used in this paper are given in Equations 1–4 [3]. In those equations, A and C are constants. Equation 5 is an integral example between the limits $x_0$ and $x_i$ ($x_i > x_0$).
\[
\int dx = x + C \\
\int A \, dx = Ax + C \\
\int x^a \, dx = \frac{x^{a+1}}{a+1} + C \\
\int \frac{1}{x} \, dx = \ln(x) + C \\
\int_{x_0}^{x_1} dx = (x)^{x_1}_{x_0} = x_1 - x_0
\]

2. Chemical reaction kinetics

2.1. Reaction rate

In essence, the measure of the speed (or progress) of a chemical reaction is defined as reaction rate \([4]\). For a chemical reaction taken place in batch reactors with a fixed volume of solutions, reaction rate or the rate of reaction [denoted as the rate; M/s, M/min, or M/h] is defined as the change of concentrations (moles per liter; molarity; M) of a reactant or a product with time \(t\) (second, minute, or hour). Notably, the rate of reaction is a positive quantity \([5]\).

Taking an irreversible homogeneous reaction (Equation 6) as a simple example, the molar concentrations of reactant \([A]\) (i.e., \([A]_2 > [A]_1\); Equation 7) decrease within increasing reaction times \(t\) \((t_2 > t_1)\); therefore, the rate of below stoichiometrically simple reaction is expressed as Equation 8 by introducing a minus sign \([5]\).

\[
\text{Rate} = -\frac{\Delta[A]}{\Delta t} = -\frac{\text{Change in concentrations}}{\text{Change in times}} \\
\text{Rate} = -\frac{d[A]}{dt}
\]

The rate (Equation 6) can be also expressed in the term of the appearance of product \(B\). In contrast to the change in the concentrations of reactant \([A]\), the molar concentrations of product \(B\) (i.e., \([B]_2 > [B]_1\)) increase within increasing reaction time \(t\) \((t_2 > t_1)\), so its rate is positive quantity (Equation 10).

\[
\text{Rate} = \frac{d[B]}{dt}
\]

In general, for a stoichiometrically complicated reaction (Equation 11), the instantaneous rate of consumption of each reactant \((A\ or\ B)\) at a given time is defined as Equation 12. Meanwhile, the rate of formation of each product \((C\ or\ D)\) at a given time is expressed as Equation 13 \([7]\).

\[
aA + bB \rightarrow cC + dD \\
\text{Rate} = -\frac{1}{a} \times \frac{\Delta[A]}{\Delta t} = -\frac{1}{b} \times \frac{\Delta[B]}{\Delta t} \\
\text{Rate} = \frac{1}{c} \times \frac{\Delta[C]}{\Delta t} = \frac{1}{d} \times \frac{\Delta[D]}{\Delta t}
\]

2.2. Rate law and rate constant

The rate law is an expression of empirical differential equation that is used to describe the mathematical relationship between the rate of a reaction and the concentrations of reactant (Equation 14).

\[
\text{Rate} = k[A]^x[B]^y
\]

where \(k\) (dimensional) is defined as the rate constant (or proportionality constant); \(x\) and \(y\) are the partial order of reaction with respects to \(A\) and \(B\), respectively; and the overall order of the reaction is obtained
by the combination of the partial orders of reaction \((x + y)\). The order of reactions can be positive, negative, zero (when the concentration does not affect the rate), or fraction (i.e., \(1/2\)).

2.3. Determination of the order of reaction

Although some methods (i.e., integration, differential, half-life, and isolation methods) have been applied to determine the order of reactions [5], the integration method is the most popularly used in the literature, especially in the field of adsorption science and technology. Therefore, this study only focuses on the method based on the integrated rate laws.

2.3.1. \(n^{th}\)-order-kinetic reaction

Considering a simple chemical reaction (Equation 15), the rate of this reaction is summarized as Equations 16 and 17. By combining two equations, a general rate equation is obtained as Equation 18. In this equation, the exponent \(n\) (without any unit; dimensionless) is defined as the overall order of reaction. Equation 18 is also known as the \(n^{th}\)-order-kinetic reaction or the general-order-kinetic reaction.

\[
A \rightarrow B \quad \text{(reactant)} \rightarrow B \quad \text{(product)} \quad (15)
\]

\[
\text{Rate} = \frac{d[A]}{dt} \quad (16)
\]

\[
\text{Rate} = k[A]^n \quad (17)
\]

\[
\frac{d[A]}{dt} = k[A]^n \quad (18)
\]

After taking integration for both sides of Equation 18, Equation 19 or 20 is obtained, with the limits of integration being \([A]_o\) (the molar concentration of reactant A at beginning) at \(t_0\) and \([A]_t\) (the molar concentration of reactant A at any time) at \(t\).

\[
\int_{[A]_o}^{[A]_t} \frac{d[A]}{[A]^n} = - \int_{t_0}^{t} kdt \quad (19)
\]

\[
\int_{[A]_o}^{[A]_t} [A]^{-n}d[A] = - \int_{t_0}^{t} kdt \quad (20)
\]

After integral calculator, Equation 20 becomes:

\[
\left(\frac{[A]^{-n+1}}{-n+1}\right)_{[A]_o}^{[A]_t} = -(kt)_{t_0}^{t_1} \quad (21)
\]

\[
\left(\frac{1}{1-n}\right)(([A]_t^{1-n} - [A]_o^{1-n})) = -kt \quad (22)
\]

Setting \(t_0 = 0\) at the beginning of most reactions and \(t = t_1\), Equation 22 is rewritten as follows:

\[
\left(\frac{1}{1-n}\right)(([A]_t^{1-n} - [A]_o^{1-n})) = -kt \quad (23)
\]

After re-arrangement, Equation 23 becomes Equation 24 that is called as the linear form (or integrated form) of the \(n^{th}\)-order-kinetic reaction.

\[
[A]_t^{1-n} = (n-1)kt + [A]_o^{1-n} \quad (24)
\]

In essence, the half-life of a reaction \((t_{1/2})\) is an important parameter in kinetic aspects. Notably, the half-life of a reactant is significantly different to the half-life of a reaction. The half-life of a reactant is the time required for its concentration decreasing to a half of its initial concentration (Equation 25) [5].

\[
[A]_t = \frac{1}{2}[A]_o \quad (25)
\]

For the \(n^{th}\)-order-kinetic reaction, the concentration of reactant A decreases to a half of its initial concentration is defined in Equation 26.

\[
[A]^n = \left[\frac{1}{2}[A]_o\right]_t^{1-n} = 2^{n-1}[A]_o^{1-n} \quad (26)
\]
Submitting Equation 26 into Equation 24 and replacing \( t_{1/2} = t \), Equation 24 becomes Equation 27. The half-life of a reactant of the \( n^{th} \)-order kinetic reaction is obtained as Equation 29 (with \( n \neq 1 \)) after re-arranging Equation 28.

\[
2^{n-1}[A]_0^{-n} = (n - 1)k t_{1/2} + [A]_0^{-n} \tag{27}
\]

\[
(n - 1)k t_{1/2} = [A]_0^{1-n} (2^{n-1} - 1) \tag{28}
\]

\[
t_{1/2} = \frac{(2^{n-1} - 1)}{(n - 1)k[A]_0^{1-n}} \tag{29}
\]

In essence, the unit of \( k \) is correctly obtained from the half-life of reactions. By submitting the unit of \([A]\) (molarity; M) and \( t \) (second; s) into Equation 29, the unit of \( k \) in the \( n^{th} \)-order kinetic reaction is \(1/(M^{n-1} \times s)\) or \(M^{1-n}/s\).

### 2.3.2. Zero-order-kinetic reaction

The rate law for a zero-order-kinetic reaction is expressed as Equation 30 when considering \( n = 0 \) (Equation 18). This means that the rate \( k_o \) of the zero-order kinetic reaction (Equation 30) is not dependent on the concentration of reactant \( A \).

\[
-\frac{d[A]}{dt} = k[A]^n = k_o \tag{30}
\]

After rearranging Equation 30 and then taking integration for both its sites, Equation 31 is obtained as follows:

\[
\int_{[A]_0}^{[A]_t} d[A] = \int_{t_0}^{t} k_o dt \tag{31}
\]

Integration of Equation 31 within the following conditions: \( t_0 = 0 \) and \( t = t \) at \([A]_0\) and \([A]_t\) (see Equation 32), the equation for the zero-order kinetic reaction is introduced as Equation 33.

\[
[A]_t - [A]_0 = -k_o(t - 0) \tag{32}
\]

\[
[A]_0 - [A]_t = k_o t \tag{33}
\]

Simply, the equation of the zero-order kinetic reaction (Equation 35) can be directly obtained from the \( n^{th} \)-order kinetic reaction (Equation 24) when considering \( n = 0 \) (Equation 34). In order words, the zero-order kinetic reaction is a special case of the \( n^{th} \)-order kinetic reaction in Equation 24 when \( n = 0 \) and \( k = k_o \).

\[
[A]_t^{1-o} = (0 - 1)k_o t + [A]_0^{1-o} \tag{34}
\]

\[
[A]_t = -k_o t + [A]_0 \tag{35}
\]

The half-life of a reactant of the zero-order kinetic reaction is given as Equation 36 when submitting Equation 25 into Equation 35. The unit of the rate constant \( k \) of this reaction that is directly obtained from Equation 36 is \( M/s \).

\[
t_{1/2} = \frac{[A]_0}{2k_o} \tag{36}
\]

Submitting \( n = 0 \) into Equation 29 to verify the general half-life equation, an equation similar to Equation 36 is obtained as follows:

\[
t_{1/2} = \frac{(2^{0-1} - 1)}{(0 - 1)k_o[A]_0^{0-1}} = \frac{[A]_0}{2k_o} \tag{37}
\]

### 2.3.3. First-order kinetic reaction

The rate law for a first-order-kinetic reaction is expressed as Equation 38 when \( n = 1 \) (Equation 18).

\[
-\frac{d[A]}{dt} = k_1 [A]^1 \tag{38}
\]

After re-arranging Equation 38 and then taking integration for both its sites, Equation 39 is achieved as follows:

\[
\frac{\int_{[A]_0}^{[A]_t} d[A]}{[A]_0} = -\int_{t_0}^{t} k_1 dt \tag{39}
\]
The equation of the first-order-kinetic reaction is given as Equation 40 (or commonly as Equation 41) by integrating Equation 39 within the following conditions: $t_0 = 0$ and $t = t$ at $[A]_0$ and $[A]_o$.

$$\ln [A]_t - \ln [A]_0 = -k_1t \quad (40)$$

$$\ln [A]_t = -k_1t \quad (41)$$

The half-life of a reactant is given as Equation 42 when submitting Equation 25 into Equation 41. Clearly, the value of $t_{1/2}$ is independent on the concentration of any reactant that is a special property of this reaction as well as inversely proportional to its first-order-rate constant ($k_1$). This conclusion is consistent with the literature [5]. Therefore, only this case, the half-life of a reactant is also called as the half-life of a reaction. The unit of the rate constant $k_1$ of this reaction that is directly obtained from Equation 42 is 1/s.

$$t_{1/2} = \frac{\ln 2}{k_1} \quad (42)$$

The equation for the first-order-kinetic reaction (Equation 42) is popularly used in the literature for describe the experimental data of some advanced oxidation processes. They includes UV/chlorine advanced oxidation process [8], photocatalysis [9], wet oxidation [10], and Fenton-like reactions [11]. In this reaction, the rate of disappearance of a reactant (i.e., pollutant in water like paracetamol) is frequently expressed as Equation 43.

$$\ln \frac{C_t}{C_o} = -k_{obs}t \quad (43)$$

where $C_o$ and $C_t$ are the concentration (i.e., M) of reactant (i.e., paracetamol) at beginning ($t = 0$) and any time $t$ (i.e., s); and $k_{obs}$ is the observed rate constant of the first-order equation (1/s).

Figure 2a gives an example of the common plot ($C_t/C_o$ vs. time) of an advanced oxidation process using the combination of UV and chlorine [8]. The kinetic region can be identified in Figure 2a. The constant $k_{obs}$ that is calculated based on Figure 2b (when only considering the kinetic region) is 0.003 s.
2.3.4. Second-order-kinetic reaction

The rate law for a second-order-kinetic reaction is listed as Equation 44 when \( n = 2 \) (in Equation 18).

\[
- \frac{d[A]}{dt} = k_2[A]^2
\]  

(44)

Equation 45 is reached after re-arranging Equation 44 and then taking integration for both its sites.

\[
\int_{[A]_0}^{[A]_t} \frac{d[A]}{[A]^2} = - \int_{t_0}^{t} k_2 \, dt
\]  

(45)

The equation for the second-order-kinetic reaction is given as Equation 46 (or Equation 47) by integrating Equation 45 within the following conditions: \( t_0 = 0 \) and \( t = t \) at \([A]_0\) and \([A]_t\).

\[
- \frac{1}{[A]_t} + \frac{1}{[A]_0} = - k_2 t
\]  

(46)

\[
\frac{1}{[A]_t} = k_2 t + \frac{1}{[A]_0}
\]  

(47)

The equation for the second-order-kinetic reaction is also given as Equation 49 when considering \( n = 2 \) (Equation 24).

\[
[A]^{-2} = (2 - 1)k_2 t + [A]_0^{-n}
\]  

(48)

\[
\frac{1}{[A]_t} = k_2 t + \frac{1}{[A]_0}
\]  

(49)

The half-life of a reactant is given as Equation 50 or 51 when submitting Equation 25 into Equation 49 or submitting \( n = 2 \) into Equation 29, respectively. The unit of the rate constant \( k_2 \) of this reaction that is directly obtained from Equation 50 is \( 1/(M\times s) \).

\[
t_{1/2} = \frac{1}{k_2[A]_0} \left( \frac{2}{2^{2-1} - 1} \right)
\]  

(50)

\[
t_{1/2} = \frac{1}{(2 - 1)k_2[A]_0^{2-1}}
\]  

(51)

2.3.5. Third-order-kinetic reaction

The rate law for a second-order-kinetic reaction is given in Equation 52 when \( n = 3 \) (Equation 18).

\[
- \frac{d[A]}{dt} = k_3[A]^3
\]  

(52)

After re-arranging Equation 52 and then taking integration for both its sites, Equation 53 is introduced as follows:

\[
\int_{[A]_0}^{[A]_t} \frac{d[A]}{[A]^3} = - \int_{t_0}^{t} k_3 dt
\]  

(53)

After integral calculator, Equation 53 becomes:

\[
\left( \frac{[A]^{-3+1}}{-3+1} \right)_{[A]_0}^{[A]_t} = -(k_3 t)|_{t_0}^{t_1}
\]  

(54)

The equation for the third-order-kinetic reaction is given as Equation 56 by considering conditions: \( t_0 = 0 \) and \( t = t \) at \([A]_0\) and \([A]_t\).

\[
- \frac{1}{2} \left( \frac{1}{[A]_t^2} - \frac{1}{[A]_0^2} \right) = - k_3 t
\]  

(55)

\[
\frac{1}{[A]_t^2} = 2k_3 t + \frac{1}{[A]_0^2}
\]  

(56)
The equation for the third-order-kinetic reaction is also given as Equation 57 or 58 when \( n = 3 \) in Equation 24.

\[
[A]_t^{3-n} = (3 - 1)k_3t + [A]_0^{1-n}
\]

The half-life of a reactant is given as Equation 59 or 60 when submitting Equation 25 into Equation 58 or submitting \( n = 3 \) into Equation 29, respectively. The unit of the rate constant \( k_3 \) of this reaction that is directly obtained from Equation 59 is \( 1/(\text{M}^2\times\text{s}) \).

\[
t_{1/2} = \frac{3}{2k_3[A]_0^2} \frac{1}{(2^{3-1} - 1)} = \frac{3}{2k_3[A]_0^2} \frac{1}{(3 - 1)k_3[A]_0^{3-1}}
\]

In summary, the relevant information the rate equations (\( n^\text{th} \)-order-kinetic, zero-order-kinetic, first-order-kinetic, second-order-kinetic, and third-order-kinetic reactions), their integrated forms (linear forms), the unit of their rate constants, and their half-life of a reactant \( A \) is provided in Table 1.

**Table 1.** Summation of the rate equations (\( n^\text{th} \)-order-kinetic, zero-order-kinetic, first-order-kinetic, second-order-kinetic, and third-order-kinetic reactions), their integrated forms (linear forms), the unit of their rate constants, and their half-life of a reactant \( A \)

| Reaction order \((n)\) | Differential form (Rate equation) | Integrated form \((y = ax + b)\) | Unit of rate constant \((k)\) | Half-life \((t_{1/2})\) |
|------------------------|----------------------------------|----------------------------------|----------------------------|--------------------------|
| 0                      | \(-\frac{d[A]}{dt} = k_0[A]^n\) | \([A]_t = -k_0t + [A]_0\)        | M/s                        | \(\frac{[A]_0}{2k_0}\)  |
| 1                      | \(-\frac{d[A]}{dt} = k_1[A]^1\) | \(\ln\frac{[A]_t}{[A]_0} = -k_1t\) | 1/s                        | \(\frac{\ln 2}{k_1}\)  |
| 2                      | \(-\frac{d[A]}{dt} = k_2[A]^2\) | \(\frac{1}{[A]_t} = k_2t + \frac{1}{[A]_0}\) | 1/(M\times s)             | \(\frac{1}{k_2[A]_0}\)  |
| 3                      | \(-\frac{d[A]}{dt} = k_3[A]^3\) | \(\frac{1}{[A]_t} = 2k_3t + \frac{1}{[A]_0}\) | 1/(M\times s)             | \(\frac{3}{2k_3[A]_0^2}\) |

Note: The units of the concentrations of reactant \(([A])\) and \([A]_0\) and time \(t\) are molarity (M) and second (s); chemical reaction is considered herein as: \( A \) (reactant) \( \rightarrow \) \( B \) (product); and \( n \) is the general order of a reaction \((n \neq 1\) for its integrated form and half-life).

### 3. Adsorption kinetics

In the field of adsorption in solid–liquid phases, two adsorption models (pseudo-first-order and pseudo-second-order models) are frequently used to model the experimental data of time-dependent adsorption. Some authors disagree to use the term “pseudo”, while others suggest using this word. In fact, in the plot of \( q_t \) vs. \( t \), its y-axis is expressed as the amounts of adsorbate adsorbed by an adsorbent at any time \( t \) (\( q_t \); mol/kg; Equation 61) instead of the concentration of adsorbate (mol/L) in solution at any time \( t \) (\( C_t \); mg/L) [12]. The amount of adsorbate adsorbed by an adsorbent at equilibrium \((q_e, \text{ mol/kg})\) is expressed as Equation 62.

\[
q_t = \frac{C_0 - C_t}{m} \times V
\]

(61)

\[
q_e = \frac{C_0 - C_e}{m} \times V
\]

(62)

where \( C_0, C_e, \) and \( C_t \) (mol/L) are the concentration of adsorbate in solution at beginning, equilibrium, and time \( t \) (i.e., min), respectively; \( m \) (kg) is dried mass of adsorbent; and \( V \) (L) is the working volume of adsorbate.

In order words, in the adsorption kinetics, the adsorption rate (different to the adsorption rate constant) is defined as the change of the amount of adsorbate adsorbed by adsorbent with time \( t \) (not the
change of the concentration of adsorbate with time \( t \). Therefore, the prefix of "pseudo" (unreal) is used to describe the rate law and rate constant for adsorption processes \([2]\).

Earlier, Ho suggested using the term of the pseudo-first-order model and pseudo-second-order model rather than that of the first-order and second-order models. This is because the former kinetic equations based on the adsorption capacity of adsorbent \( (q_o; \text{mol/kg}) \) while the latter based on the concentration of adsorbate \( (C_i; \text{mol/L}) \) \([13]\). Therefore, the terms of the pseudo-first-order model (PFO) and pseudo-second-order model (PSO) are used in this paper.

### 3.1. Pseudo-first-order (PFO) model

According to previous discussion \([2, 14, 15]\), the PFO model (Equation 63) was firstly developed by Lagergren \([16]\) for applying charcoal to adsorb oxalic acid and malonic acid in water. The differential equation of the PFO model \([16]\) is commonly expressed as Equation 63. Because \( q_i \) values often increase within increasing contact time. Therefore, the adsorption rate in Equation 63 is always positive. However, as discussed in Section 2.1, the differential equation of this model is correctly given as Equation 64 rather than Equation 63.

\[
\frac{dq_t}{dt} = k_{1(PFO)}(q_{e(PFO)} - q_t)^1
\] (63)

where \( k_{1(PFO)} \) is the rate constant of the PFO model (an un-known parameter; 1/min or 1/h); and \( q_{e(PFO)} \) (an un-known parameter; \text{mol/kg}) and \( q_t \) (obtained from time-dependent experiments; \text{mol/kg}) are the amounts of adsorbate adsorbed by adsorbent at equilibrium and any time \( t \) (min or h), respectively.

\[
\frac{dq_t}{dt} = k_{1(PFO)}(q_o - q_t)^1
\] (64)

In Equation 64, \( q_o \) is the theory maximum (or total) adsorption capacity of adsorbent by its dry mass. For example, if the adsorption process of \( \text{NH}_4^+ \) ions in solution by zeolite adsorbents is primarily regarded as ion-exchange mechanism, the \( q_o \) value (\text{meq/g}) of zeolite will be nearly equal to its capacity exchange capacity (CEC; meq/g). When adsorption process reaches equilibrium, \( q_e \) value will be equal to \( q_o \) value (or the ratio of \( q_e/q_o \approx q_o/\text{CEC} \approx 1.0 \)). Therefore, if adsorption processes are conducted at very low initial concentrations of adsorbate \( (C_0 \rightarrow 0) \), \( q_e \) value will be significantly lower than \( q_o \) value \( (q_e \ll q_o) \). As a result, Equation 64 cannot reduce to Equation 63. This problem is often ignored by many researchers and recently discussed by authors \([17]\).

In order words, the differential equations of the pseudo-first-order, pseudo-second-order, and pseudo-general-order models are only valid when adsorption process reach a “true equilibrium” under high initial adsorbate concentrations. Only in this case, \( q_e \) will be equal to \( q_o \) (an un-known parameter).

After separating the variables in Equation 63, Equation 64 is obtained as follows:

\[
\frac{dq_t}{(q_{e(PFO)} - q_t)} = k_{1(PFO)}dt
\] (64)

Equation 65 is achieved by taking integration for both sites of Equation 64.

\[
\int_0^{q_t} \frac{dq_t}{(q_{e(PFO)} - q_t)} = \int_0^\infty k_{1(PFO)}dt
\] (65)

Equation 65 can be also rewritten as follows:

\[
\int_0^{q_t} \frac{dq_t}{(q_{e(PFO)} - q_t)} = \int_0^\infty k_{1(PFO)}dt
\] (66)

\[
- \int_0^{q_t} \frac{dq_t(q_{e(PFO)} - q_t)}{q_{e(PFO)} - q_t} = \int_0^\infty k_{1(PFO)}dt
\] (67)

After applying the boundary conditions \( (q_t = 0 \text{ at } t = 0, \text{ and } q_t = q_i \text{ at } t = t) \), the linear form of the PFO model is achieved as Equation 70.

\[
-ln(q_{e(PFO)} - q_t)\big|_0^{q_t} = k_{1(PFO)}t
\] (68)

\[
-ln(q_{e(PFO)} - q_t) + ln(q_{e(PFO)}) + c_{\text{constant}} = k_{1(PFO)}t + c_{\text{constant}}
\] (69)

\[
ln(q_{e(PFO)} - q_t) = -k_{1(PFO)}t + ln(q_{e(PFO)})
\] (70)
Equation 70 is the linear form of the PFO model that is often used in the literature [14, 18, 19]. After re-arrangement, Equation 73 is common the non-linear form of the PFO model that is an exponential function.

\[
q_e(\text{PFO}) - q_t = e^{-k_{1(\text{PFO})}t + \ln(q_e(\text{PFO}))} \quad \text{or} \quad q_e(\text{PFO}) - q_t = e^{-k_{1(\text{PFO})}t} \times e^{\ln(q_e(\text{PFO}))}
\]

\[
q_t = q_e(\text{PFO})(1 - e^{-k_{1(\text{PFO})}t}) \quad \text{or} \quad e^{-k_{1(\text{PFO})}t} = \frac{q_e(\text{PFO}) - q_t}{q_e(\text{PFO})}
\]

If submitting Equation 61 into Equation 73, another expression of the non-linear form of the PFO (Equation 75) can be obtained as suggested by authors [20].

\[
\frac{C_o - C_t}{m} = q_e(\text{PFO})(1 - e^{-k_{1(\text{PFO})}t})
\]

\[
\frac{C_t}{C_o} = 1 - \left[\frac{q_e(\text{PFO})}{C_o} \times \frac{m}{V} \times (1 - e^{-k_{1(\text{PFO})}t})\right]
\]

It is necessary to calculate the half-life of an adsorption process \((t_{1/2};\) minute or hour). The half-life is defined as the time required to decrease the amount of adsorbate in solution adsorbed by adsorbent to half of its theory maximum adsorption capacity \((q_o).\)

\[
q_t = \frac{1}{2} q_o = \frac{1}{2} q_e
\]

After taking the logarithm of both sides of Equation 72, Equation 78 is obtained. The half-life of adsorption process is given as Equation 79 by submitting Equation 76 into Equation 78. This equation is similar to the literature [18]. Similar to the half-life of a reaction (Section 2.3), the half-life of adsorption process is independent on the amount of adsorbate adsorbed by adsorbent at any time \((q_t)\) and inversely proportional to its rate constant \((k_{1(\text{PFO})})\). The unit of \(k_{1(\text{PFO})} (1/\text{min})\) can be obtained from Equation 79 when the unit \(t\) is minute.

\[
\ln(e^{-k_{1(\text{PFO})}t}) = \ln\left(\frac{q_e(\text{PFO}) - q_t}{q_e(\text{PFO})}\right) \quad \text{or} \quad -k_{1(\text{PFO})}t = \ln\left(\frac{q_e(\text{PFO}) - q_t}{q_e(\text{PFO})}\right)
\]

\[
t = -\frac{\ln(q_e(\text{PFO}) - q_t)}{-k_{1(\text{PFO})}}
\]

\[
t_{1/2} = -\frac{\ln(1/2)}{k_{1(\text{PFO})}} = \frac{\ln(2)}{k_{1(\text{PFO})}} = 0.693
\]

3.2. Pseudo-second-order (PSO) model

In the literature, there is a strong argument on the derivation of the PSO model. A series of comment works has been made by the authors [21-23] to argue who is the original authors propose this model. Ho strongly concluded that the PSO model was initially reported in the author’s doctoral thesis for describing the adsorption process of divalent metal ions onto sphagnum moss peat. Therefore, he suggested that other researchers need to “cite Ho’s original pseudo-second-order kinetic expression paper” [21-23]. However, the PSO model is not originally proposed by Ho. More information on this conclusion has been summarized by a critical review [14]. In fact, the PSO model was early introduced by Blanchard et al. [24] to model time-dependent experiment data of the adsorption process of potentially toxic metals onto an ion exchanger derived from natural zeolite clinoptilolite. Differential equation of the PSO model (Equation 80) is commonly given in the literature. Similar to the case of the PFO, the model of PSO is valid when \(q_e\) equal to \(q_o\). Only this case, Equation 81 reduce to Equation 80.

\[
\frac{dq_t}{dt} = k_{2(\text{PSO})}(q_e(\text{PSO}) - q_t)^2
\]

\[
\frac{dq_t}{dt} = k_{2(\text{PSO})}(q_o - q_t)^2
\]

where \(k_{2(\text{PSO})}\) the rate constant of the PSO model [kg/(molxmin)] or kg/(molxh); \(q_e(\text{PSO})\) an un-known parameter; and \(q_o(\text{PSO})\) (an un-known parameter; mol/kg) and \(q_t\) (obtained from time-dependent experiments; mol/kg) are the amounts of adsorbate adsorbed by adsorbent at equilibrium and any time \(t\) (min or h), respectively.

After separation of the variables, Equation 81 is rephrased as follows:
\[
\frac{dq_t}{(q_e(PSO) - q_t)^2} = k_{2(PSO)}dt 
\] (82)

Taking integration for both sites, Equation 83 is obtained as follows:

\[
\int_0^{q_t} \frac{dq_t}{(q_e(PSO) - q_t)^2} = \int_0^t k_{2(PSO)}dt 
\] (83)

Equation 83 can be also rewritten as follows:

\[
\int_0^{q_t} \frac{dq_t}{(q_e(PSO) - q_t)^2} = \int_0^t k_{2(PSO)}dt 
\] (84)

\[
- \int_0^{q_t} \frac{dq_t}{(q_e(PSO) - q_t)^2} = \int_0^t k_{2(PSO)}dt 
\] (85)

Applying the boundary conditions \((q_t = 0 \text{ at } t = 0, \text{ and } q_t = q_i \text{ at } t = t)\), Equation 85 becomes

\[
\frac{1}{(q_e(PSO) - q_0)} \int_0^{q_t} \frac{dq_t}{(q_e(PSO) - q_t)^2} = k_{2(PSO)}t 
\] (86)

\[
\frac{1}{(q_e(PSO) - q_0)} \int_0^{q_t} \frac{dq_t}{(q_e(PSO) - q_t)^2} = k_{2(PSO)}t 
\] (87)

After arrangement, the nonlinear form of the PSO model is obtained as Equation 88 [19].

\[
q_t = \frac{q_e^2(PSO)k_{2(PSO)}}{1 + q_e(PSO)k_{2(PSO)}}t 
\] (88)

If submitting Equation 61 into Equation 88, another expression of the non-linear form of the PSO model (Eq. (4.100)) can be yielded as suggested by authors [20].

\[
\frac{C_o - C_t}{m} = q_e^2(PSO)k_{2(PSO)}t 
\] (89)

\[
\frac{C_t}{C_o} = 1 - \frac{q_e(PSO)m}{C_o} \left( q_e^2(PSO)k_{2(PSO)}t \right) 
\] (90)

Six types of the linear forms of the PSO model (Equations 91–96) are found in the literature [14, 20, 25, 26]. Although Type 5 and Type 6 have been reported by some authors [20, 25], they are not commonly applied in the literature compared to Type 1.

\[
\frac{t}{q_t} = \frac{1}{q_e(PSO)} + \frac{1}{k_{2(PSO)}q_e^2(PSO)} 
\] (Type 1: the plot of \(t/q_t \text{ vs. } t\)) (91)

\[
\frac{1}{q_t} = \frac{k_{2(PSO)}q_e^2(PSO)}{q_e(PSO)} - \frac{1}{q_e(PSO)} 
\] (Type 2: the plot of \(1/q_t \text{ vs. } 1/t\)) (92)

\[
q_t = \frac{k_{2(PSO)}q_e^2(PSO)}{q_e(PSO)} + \frac{q_e(PSO)}{t} 
\] (Type 3: the plot of \(q_t \text{ vs. } q(t)\)) (93)

\[
\frac{q_t}{t} = - \frac{k_{2(PSO)}q_e(PSO)}{q_e^2(PSO)} + \frac{k_{2(PSO)}q_e^2(PSO)}{q_e(PSO)} 
\] (Type 4: the plot of \(q(t)/t \text{ vs. } q(t)\)) (94)

\[
\frac{1}{t} = \frac{k_{2(PSO)}q_e^2(PSO)}{q_e(PSO)} - \frac{1}{q_e(PSO)} 
\] (Type 5: the plot of \(1/t \text{ vs. } 1/q_t\)) (95)

\[
\frac{1}{q_e(PSO) - q_t} = k_{2(PSO)}t + \frac{1}{q_e(PSO)} 
\] (Type 6: the plot of \(1/(q_e(PSO) - q_t) \text{ vs. } t\)) (96)

Because the PSO model does not include the parameter of initial adsorption rate, Ho introduced a simple definition to calculate this rate from the Type 1 of this model. The initial adsorption rate \(h_o\) \([\text{mg/(g×min)} \text{ or mg/(g×h)}]\) is expressed as Equations 97.

\[
h_o = q_e^2k_{2(PSO)} = q_e^2q_{PSO}k_{2(PSO)} 
\] (97)

The half-life of the amount of adsorbate adsorbed by adsorbent (not the half-life of adsorption process as the case of PFO model) is obtained as Equations 98 by submitting Equations 76 into Equations 88. This equation has been reported elsewhere [18, 27].
\[ t_{1/2} = \frac{1}{q_e k_{2(PSO)}} \]  

(98)

### 3.3. Pseudo-\(n^{th}\)-order (PNO) model or general-order-kinetic (GOK) model

In some cases, it is hard to determine the order of adsorption process. This means that the experience data of time dependence are well fitted to both PSO and PFO models. Therefore, the pseudo-\(n^{th}\)-order (PNO) model or the general order kinetic (GOK) model is additionally used to determine the overall order of adsorption process appropriately. Differential equation of the PNO model is expressed as Equations 99.

\[
\frac{dq_t}{dt} = k_n(q_e(PNO) - q_t)^n
\]

(99)

After separating of the variables and taking integration for both sites, Equations 100 is obtained.

\[
\int_0^{q_t} \frac{dq_t}{(q_e(PNO) - q_t)^n} = \int_0^t k_n dt
\]

(100)

Equations 100 can be also rewritten as follows:

\[
\int_0^{q_t} \frac{d(q_t - q_e(PNO))}{(q_e(PNO) - q_t)^n} = \int_0^t k_n dt
\]

(101)

\[
- \int_0^{q_t} \frac{d(q_e(PNO) - q_t)}{(q_e(PNO) - q_t)^n} = \int_0^t k_n dt
\]

(102)

Applying the boundary conditions \(q_t = 0\) at \(t = 0\), and \(q_t = q_e\) at \(t = t\), Equation 102 becomes Equation 102 and then Equation 104. Equation 105 is the linear form of this model \([19]\).

\[
\frac{(q_e(PNO) - q_t)^{1-n}}{n+1} q_t^{1-n} = -k_n t_0^{1-n}
\]

(103)

\[
\frac{1}{1-n} (q_e(PNO) - q_t)^{1-n} - (q_e(PNO))^{1-n} = -k_n t
\]

(104)

\[
(q_e(PNO) - q_t)^{1-n} = (n-1)k_n t + (q_e(PNO))^{1-n}
\]

(105)

Setting \(AB = (n-1)k_n t + (q_e(PNO))^{1-n}\) and then logarithm of both sides of Equation 105, Equation 106 and then Equations 107–111 are attained.

\[
ln(q_e(PNO) - q_t)^{1-n} = lnAB
\]

(106)

\[
ln(q_e(PNO) - q_t) = \frac{1}{1-n} lnAB
\]

(107)

\[
q_e(PNO) - q_t = e^{\left(\frac{1}{1-n} lnAB\right)}
\]

(108)

\[
q_t = q_e(PNO) - e^{lnAB\left(\frac{1}{1-n}\right)}
\]

(109)

\[
q_t = q_e(PNO) - AB\left(\frac{1}{1-n}\right)
\]

(110)

\[
q_t = q_e(PNO) - \left(\frac{1}{AB}\right)^{\frac{1}{n-1}}
\]

(111)

Submitting \(AB = (n-1)k_n t + (q_e(PNO))^{1-n}\) into Equations 111, Equations 112 is obtained.

\[
q_t = q_e(PNO) - \left(\frac{1}{(n-1)k_n t + (q_e(PNO))^{1-n}}\right)^{\frac{1}{n-1}}
\]

(112)

After arranging Equations 112, Equations 113–115 are attained. Equations 115 is the nonlinear form of the PNO or GOK model and has been reported elsewhere \([18]\).

\[
q_t = q_e(PNO) - \left((q_e(PNO))^{1-n} \left((n-1)k_n t(q_e(PNO))^{n-1} + 1\right)\right)^{\frac{1}{n-1}}
\]

(113)

\[
q_t = q_e(PNO) - \left((q_e(PNO))^{n-1} \left(1 + (n-1)k_n t(q_e(PNO))^{n-1}\right)\right)^{\frac{1}{n-1}}
\]

(114)
or

\[ q_t = q_e(PNO) - q_e \left( \frac{1}{1 + (n-1)k_n t (q_e(PNO))^{n-1}} \right)^{\frac{1}{n-1}} \]

\[ q_t = q_e(PNO) \left( 1 - \left[ \frac{1}{1 + (n-1)k_n (q_e(PNO))^{n-1}} \right]^{\frac{1}{n-1}} \right) \]

The nonlinear form of the PNO model (Equations 116) is reported by Lima et al. [19, 28] after re-arranging Equations 114.

\[ q_t = \frac{q_e(PNO)}{[(n-1)k_n q_e(PNO)t + 1]^{\frac{1}{n-1}}} \]  

(116)

The half-life of the amount of adsorbate adsorbed by adsorbent is obtained by submitting Equations 76 into Equations 105. This half-life equation (Equations 121) has been reported elsewhere [18]. The unit of rate constant of PNO model that is \((kg/(mol)^{1-n})\times(1/min)\) (or \(kg^{-1}mol^{1-n}min^{-1}\)) obtained by submitting the units of \(q_e\) (mol/kg), \(C_o\) (mol/L), and \(t\) (min) into Equation 105 or 121.

\[
\begin{align*}
(q_e(PNO) - \frac{1}{2}q_e(PNO))^{1-n} &= (n-1)k_n t_{1/2} + (q_e(PNO))^{1-n} \\
\left(\frac{1}{2}\right)^{1-n}(q_e(PNO))^{1-n} &= (n-1)k_n t_{1/2} + (q_e(PNO))^{1-n} \\
2^{n-1}(q_e(PNO))^{1-n} - (q_e(PNO))^{1-n} &= (n-1)k_n t_{1/2} \\
(q_e(PNO))^{1-n}(2^{n-1} - 1) &= (n-1)k_n q_e(PNO) \\
t_{1/2} &= \frac{2^{n-1} - 1}{(n-1)k_n} \\
\end{align*}
\]

(117) (118) (119) (120) (121)

In summary, the relevant information the adsorption kinetics models, their integrated forms (linear forms), the unit of their rate constants, and their half-life is provided in Table 2.

| Differential form | Integrated form (linear form) | Integrated equation (nonlinear form) | Unit of rate constant (k) | Half-life \((t_{1/2})\) |
|-------------------|-------------------------------|--------------------------------------|---------------------------|--------------------------|
| PNO \((n \neq 1)\) | \(\frac{dq_t}{dt} = k_n (q_e - q_t)^n\) | \(\frac{dq_e}{dt} = \frac{ln(q_e - q_t)}{k_1 t} + \frac{ln(q_e)}{t}\) | \(kg/(mol)^{1-n}\times(1/min)\) | \(\frac{2^{n-1} - 1}{(n-1)k_n q_e^{n-1}}\) |
| PFO | \(\frac{dq_t}{dt} = k_1 (q_e - q_t)^2\) | \(\frac{pq_e}{t} = \frac{1}{q_e + k_2 t}\) | 1/min | \(\frac{ln2}{k_1}\) |
| PSO | \(\frac{dq_t}{dt} = k_2 (q_e - q_t)^2\) | \(\frac{pq_e}{t} = \frac{1}{q_e + k_2 t}\) | kg/(mol)x(min) | \(\frac{1}{q_e k_2}\) |

Note: the units of \(t = min\), \(q_e\) and \(q_t = mol/kg\), and \(C_o\) and \(C_e = mol/L\).

### 3.4 Discussion

As early discussed, the term \(q_o\) (the theory maximum adsorption capacity of adsorbent) needs to be used instead of \(q_e\). Only the case \(q_o \approx q_e\), the differential equations of the PFO, PFO, and PNO models are obtained. Figure 3a shows the adsorption kinetics of naproxen onto biosorbent generated from peanut shell under two initial naproxen concentrations (i.e., \(C_o = 97\) and \(498\) mg/L). Clearly, under a low \(C_o\) (97 mg/L), the adsorption process reaches very rapidly equilibrium, with a plateau point being reached after 15 min. The kinetic region is not identified in this case. Under low \(C_o\) the adsorption, the adsorption sites available in the biosorbent are not fully occupied by naproxen molecules (see Figure 3b). Figure 3b shows three regions (i.e., un-saturation, near saturation, and saturation) in the adsorption isotherm of
naproxen by the biosorbent. Its maximum adsorption pacity of that is calculated based on the Langmuir model and experiment is 55.1 mg/g and 48.5 mg/g, respectively.

In contrast, under a higher \( C_0 \) (498 mg/L), the adsorption sites in the biosorbent are fully covered by naproxen molecules (Figure 3b). This is because \( q_e (48.2 \text{ mg/g}; q_t \approx q_e \text{ when the adsorption reaches equilibrium}) \) is nearly equal to \( q_e (48.8 \text{ mg/g}) \). As a result, the kinetic region and equilibrium region are obtained (Figure 3a). Table 3 presents a comparison on the parameters of the PSO and PFO models obtained from full data (kinetic region and equilibrium region), kinetic region, and only equilibrium region. The result indicated that the parameters of the PSO and PFO models calculated based onto the kinetic and equilibrium regions are highly reliable.

![Figure 3. (a) Adsorption kinetic of naproxen by the peanut shell-derived biosorbent and (b) its adsorption isotherm [29] (Image)](image)

**Table 3. Parameters of the PSO and PFO models of the adsorption process of naproxen by the biosorbent**

| Region | PFO model | PSO model | PFO model | PSO model | PFO model | PSO model |
|--------|-----------|-----------|-----------|-----------|-----------|-----------|
| 1. Kinetic region and equilibrium region | \( q_e \) | 47.9 | 49.9 | | | |
| | \( k_{1,\text{PFO}} \) or \( k_{2,\text{PSO}} \) | 0.0239 | 0.00163 | | | |
| 2. Kinetic region | \( q_e \) | 61.5 | 100 | | | |
| | \( k_{1,\text{PFO}} \) or \( k_{2,\text{PSO}} \) | 0.0123 | 7.73E-5 | | | |
| 3. Equilibrium region | \( q_e \) | 49.1 | 59.1 | | | |
| | \( k_{1,\text{PFO}} \) or \( k_{2,\text{PSO}} \) | 0.018 | 3.28E-4 | | | |

Note: the unit of \( q_e \) (mg/g), \( k_{1,\text{PFO}} \) (1/min), and \( k_{2,\text{PSO}} \) [g/(mg×min)]; the \( q_e \) value (from the experiment) of 48.2 mg/g; the raw data extracted from the literature [29].

4. Conclusions

The order of chemical reaction kinetics is different to that of adsorption kinetics. The chemical reaction kinetics is described by the change of concentrations (in mol/L) of reactant (i.e., paracetamol) within reaction time. Meanwhile, the adsorption kinetics is described by the change of the amount of reactant (commonly called adsorbate, i.e., paracetamol) absorbed by adsorbent (\( q_t \); mol/kg or mg/g).
within adsorption time. Taking the same order of reaction \( (n = 1) \), the constant \( k_1 \) of chemical reaction kinetics is often calculated based on the kinetic region. In contrast, the constant \( k_{1(PFO)} \) of adsorption kinetics is calculated based on both kinetic region and equilibrium region. Some incorrect cases for adsorption kinetics are provided in Section 3.4. The first common mistake is the rate constant \( (k_{1(PFO)} \) and \( k_{2(PSO)} \)) is calculated from equilibrium region (the unsuitable selection of initial interval time). The adsorption occurs very fast; therefore, it is necessary to repeat the adsorption kinetics by taking shorter period-times (i.e., 1 min, 3 min, 5 min, etc.). The second common mistake is the unsuitable selection (of initial adsorbate concentrations (i.e., low or dilute concentration). Under this low initial adsorbate concentration, the adsorption sites in the material are not fully covered by adsorbate. This means that the \( q_e \) value will not be equal to \( q_0 \). As a result, the parameters of the adsorption kinetic models (especially the relevant rate constants) obtained are not acceptable.

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