The Langmuir isotherm adsorption equation: The monolayer approach

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Abstract. Langmuir adsorption isotherm equation is generally derived using the kinetics approach and based on some assumptions. These assumptions are usually not accompanied by an adequate explanation related with the mechanisms and the determination techniques of its adsorption constants. This study presents a simple technique on the determination of the isotherm equation based on the main assumption so called the monolayer assumption. Furthermore, a more precise technique for determining the adsorption constants is also described. This theoretical study is supported with statistical analysis on the experimental data adapted from the literature. Two theories based on adsorption mechanism and based on rate were introduced. Regardless of its limitations, the results show that the technique for the determination is valid.

1. Introduction
Adsorption is the adhesion of an adsorbate such as a fluid, liquid, or gas, by creating a thin layer or film on the surface of an adsorbent whether it is a solid or liquid. The bonding between adsorbate-adsorbent could be physical or chemical bonding. The chemical bonding provides stronger bonding than physical bonding, and the film might be single or multiple layers. Adsorbate can be separated from the adsorbent and the process is called desorption.

There are some factors influencing the adsorption process, but, at the basic level, the most widely studied is the influence of temperature and could be assumed as a monolayer. The adsorption process is usually studied at a given temperature and recognized as an adsorption isotherm. One of them is the Langmuir adsorption isotherm.

Langmuir equation is derived based on the rate of adsorption and desorption [1,2]. If \( S_o \) is the maximum available sites at the adsorbent surface, \( S \) the amount occupied, \( S_1 = S_o - S \) is vacant, and \( C \) is the concentration of the solution, then

\[
\text{the rate of adsorption} = k_1 \, S_1 \, C = k_1 \, [S_o - S] \, C
\]

(1)

The rate of desorption is proportional the number of occupied sites \( S \):

\[
\text{rate of desorption} = k_2 \, S
\]

(2)
$k_1$ and $k_2$, are adsorption and desorption rate constant respectively.

At equilibrium, the rate of adsorption = rate of desorption, or Equation (1) = Equation (2), or

\[
\begin{align*}
  k_1 [ S_o - S] C &= k_2 S \\
  k_1 S_o C &= [ k_2 + k_1 C] S \\
  S &= \frac{k_1 S_o C}{k_2 + k_1 C} \\
  S &= \frac{S_o C}{k_2 / k_1 + C}
\end{align*}
\]

(3)

(4)

If $S_o = K_{ads}$ and $k = (k_2/k_1)$ then

\[
S = \frac{K_{ads} C}{k + C}
\]

(5)

$K_{ads}$ and $k$ are constants with values depend upon the nature of the adsorbent and adsorbate.

There are 3 problems arises with regard to eq. (5). The first is how to relate it with the monolayer assumption. In order to be meaningful, eq. (5) should be derived from the assumption. Second, adsorption phenomena are irreversible processes. Thus, it needs a different reaction mechanism, not just based on the assumption that, at equilibrium, the rate of adsorption = rate of desorption. Third, the curve (of $S$ versus $C$) of eq. (5) is a hyperbolic equation. It is similar with reactions catalyzed by enzyme [3]. Accordingly, it is difficult to determine the value of $K_{ads}$ and $k$ directly from eq. (5). By taking the reciprocal of both sides of eq. (5), however, it is converted into its linear form

\[
\begin{align*}
  \frac{1}{S} &= \frac{1}{K_{ads}} + \frac{k}{K_{ads}} \left( \frac{1}{C} \right) \\
  \frac{1}{S} &= \frac{1}{K_{ads}} \\
  \frac{1}{S} &= \frac{1}{K_{ads}} + \frac{k}{K_{ads}} \left( \frac{1}{C} \right)
\end{align*}
\]

(6)

$K_{ads}$ value is obtained from the intercept and $k$ from the slope of the curve of $(1/S)$ vs. $(1/C)$.

It is interesting to compare eq. (6) with the equation obtained from eq. (5) on the condition of $C$ is relatively higher and lower than $k$.

If $C$ is higher than $k$ and so $C+k \approx C$, eq. (5) becomes

\[
S = K_{ads}
\]

(7)

The reciprocal of eq. [7] is

\[
\begin{align*}
  \frac{1}{S} &= \frac{1}{K_{ads}} \\
  \frac{1}{S} &= \frac{1}{K_{ads}}
\end{align*}
\]

(8)

The equation (8) is different with eq. (6).

Inserting eq. (7) into eq. (6) gives

\[
\begin{align*}
  \frac{1}{K_{ads}} &= \frac{1}{K_{ads}} + \frac{k}{K_{ads}} \left( \frac{1}{C} \right) \\
  k &= 0
\end{align*}
\]

(9)
That is to say, eq. (6) does not apply if \(C\) is relatively higher than \(k\).

If \(C\) is lower than \(k\) and so \(C+k \approx k\), eq. (5) will become

\[
S = \frac{K_{ads}C}{k}
\]  

(10)

The reciprocal of eq. (10) is

\[
\frac{1}{S} = \frac{k}{K_{ads}} \left( \frac{1}{C} \right)
\]  

(11)

which is also differ with eq. (6)

By assumption, \(S_o = K_{ads}\) and so, if all surface occupied (or \(S = S_o\)), then \(S = K_{ads}\) and eq. (10) [or eq. (11)] becomes

\(k = C\)  

(12)

That is to say, eq. (6) also does not apply if \(C\) is relatively lower than \(k\). So, it can be concluded that, theoretically, eq. (6) is ambiguous and therefore, it can not be used to determine, both \(K_{ads}\) and \(k\).

Based on the previously discussion, there are three objectives of this study i.e. to derive the Langmuir adsorption isotherm equation which is based on the monolayer assumption, to derive the Langmuir adsorption isotherm equation which is based on the monolayer assumption and irreversible mechanism, and to find an easier and reliable technique for the determination of \(K_{ads}\) and \(k\).

2. Methodology

This research is a literature study. The first goal is to drive eq. (5) which is based on the monolayer assumption. The second goal is to drive eq. (5) which is based on the monolayer assumption and the reaction mechanism involving the product (irreversible process). The third goal is to find an easier and reliable technique for the determination of \(K_{ads}\) and \(k\). The validity is based on the comparison of the results obtained from of eq. (7) and eq. (10) and with obtained from eq. (6).

This study uses data, not from adsorption experiment, but taken from the bromination of acetone [4], 

\((\text{CH}_3)_2\text{CO} + \text{Br}_2 \rightarrow \text{CH}_3\text{COCH}_2\text{Br} + \text{Br}^- + \text{H}^+\)  

This reaction has the rate law of

\[
-d[P]/dt = \frac{(k_1 k_2 k_3)/k_{-2} [A] [H^+] [\text{Br}_2]}{k_{-2} [H^+] + k_3 [\text{Br}_2]}
\]  

(13)

If in the beginning, \([\text{Br}_2]_o\) is relatively higher than \(k_3[H^+]\), at the early-stage, the reaction is zero-order with respect to \([\text{Br}_2]\) but at the near-end is first-order. During the reaction process, \([\text{Br}_2]\) is decreased while \([H^+]\) increased (released from acetone). But, if in the beginning, \([\text{Br}_2]_o\) is relatively lower than \(k_3[H^+]\), for the same reason, the reaction is always first-order.

As seen in Table 1. and Table 2., the data are not presented in concentration unit but in absorbance of \(\text{Br}_2\) in the solution, measured at \(\lambda = 400\) nm and room temperature. It is merely done for the easier use of Equation (6), Equation (7) and Equation (10). Data in Table 1 is obtained from the experiment where, \([H^+]_o\) is relatively low (and so, \([\text{Br}_2]\) will be higher than \(k_3[H^+]\)). Data in Table 2 is obtained from the experiment where, \([H^+]_o\) is relatively high (and so \([\text{Br}_2]\) is also lower than \(k_3[H^+]\)).
Table 1. Absorbance, A, of Br₂ in Bromination of Acetone: 10 mL Acetone 4.0 M, 5.0 mL HCl 1.0 M, 25 mL H₂O, 10 mL Br₂ 0.05 M [5]

| No. | $t / s$ | Early stage | A  | Near end | A  |
|-----|---------|-------------|----|----------|----|
| 1.  | 10      | 0.483       |    | 590      | 0.073   |
| 2.  | 20      | 0.473       |    | 600      | 0.066   |
| 3.  | 30      | 0.463       |    | 610      | 0.059   |
| 4.  | 40      | 0.454       |    | 620      | 0.053   |
| 5.  | 50      | 0.444       |    | 630      | 0.048   |
| 6.  | 60      | 0.434       |    | 640      | 0.043   |

Table 2. Absorbance, A, of Br₂ in Bromination of Acetone: 10 mL Acetone 4.0 M, 7.5 mL HCl 1.0 M, 22.5 mL H₂O, 10 mL Br₂ 0.05 M [5]

| No. | $t / s$ | Early stage | A  | Near end | A  |
|-----|---------|-------------|----|----------|----|
| 1.  | 10      | 0.493       |    | 330      | 0.103   |
| 2.  | 20      | 0.483       |    | 340      | 0.091   |
| 3.  | 30      | 0.473       |    | 350      | 0.081   |
| 4.  | 40      | 0.464       |    | 360      | 0.072   |
| 5.  | 50      | 0.454       |    | 370      | 0.064   |
| 6.  | 60      | 0.445       |    | 380      | 0.057   |

Then, to make it more similar to the experimental technique on adsorption, each observation which was taken at the same time interval and the observation resulting at the end of one-time interval being considered as the initial value for a new time interval. Each direct observation is taken as $C_o$ and the following as $C$. The difference between each pair is taken as $S$ (thus, the amount of $S$ is always taken at the same time–interval). The setting of the data from both tables is presented in Table 3. and Table 4.

Table 3. Setting the data of Table 1.

| No. | $t/s$ | Early-stage | Near-end |
|-----|-------|-------------|----------|
|     |       | $C_o$ | $C$ | $C_o - C = S$ | $t/s$ | $C_o$ | $C$ | $C_o - C = S$ |
| 1.  | 10    | 0.483 | 0.473 | 0.010 | 590 | 0.073 | 0.066 | 0.007 |
| 2.  | 20    | 0.473 | 0.463 | 0.010 | 600 | 0.066 | 0.059 | 0.007 |
| 3.  | 30    | 0.463 | 0.454 | 0.009 | 610 | 0.059 | 0.053 | 0.006 |
| 4.  | 40    | 0.454 | 0.444 | 0.010 | 620 | 0.053 | 0.048 | 0.005 |
| 5.  | 50    | 0.444 | 0.434 | 0.010 | 630 | 0.048 | 0.043 | 0.005 |
| 6.  | 60    | 0.434 |       |       | 640 | 0.043 |       |       |

The value of $K_{ads}$ and $k$ is determined using eq. (6) for the conventional and eq. (7) and eq. (10) for the new method.
Table 4. Setting the data of Table 2.

| No. | t/s | Early-stage | Near-end |
|-----|-----|-------------|----------|
|     |     | $C_o$ | $C$ | $C_o - C = S$ | $t/s$ | $C_o$ | $C$ | $C_o - C = S$ |
| 1.  | 10  | 0.493 | 0.483 | 0.010 | 330  | 0.103 | 0.091 | 0.012 |
| 2.  | 20  | 0.483 | 0.473 | 0.010 | 340  | 0.091 | 0.081 | 0.010 |
| 3.  | 30  | 0.473 | 0.464 | 0.009 | 350  | 0.081 | 0.072 | 0.009 |
| 4.  | 40  | 0.464 | 0.454 | 0.010 | 360  | 0.072 | 0.064 | 0.008 |
| 5.  | 50  | 0.454 | 0.445 | 0.009 | 370  | 0.064 | 0.057 | 0.007 |
| 6.  | 60  | 0.445 |        |        | 380  | 0.057 |        |        |

Based on equation (7), the process is of the zero-order with respect to $C$: the value of $K_{ads}$ is obtained from the intercept of the linear curve of $S$ versus $C$. This value is then used to find the value of $k$, by inserting it into the value of the slope of the linear curve of $S$ versus $C$ using eq. (10) (the process is of the first-order with respect to $C$). The results are then compared with that obtained from equation (6).

The process is said to be zero-order if the value of the slope close to 0.00 and first-order if the value of the intercept close to 0.00.

3. Discussions And Implications

3.1. Discussions

3.1.1. Monolayer theory. There are 5 kinds of interaction in adsorption process: between adsorbent-adsorbate, adsorbate-adsorbate, adsorbate-solvent, adsorbent-solvent, and solvent-solvent. In order the adsorption process to occur, the adsorbent-adsorbate interaction must be the strongest. The monolayer is formed if the strength of the bond between adsorbate-adsorbent molecules is stronger than adsorbate-adsorbate, adsorbate-solvent, adsorbent-solvent, and solvent-solvent. The position of the first adsorbed molecules will act as the initiator to the position of the next adsorbed molecule. The second molecule will be tied adjacent to the first molecule, and because of strong adsorbent-adsorbate interaction, besides and not above. Then, the reaction mechanism can be described as equation (14).

$$
S_f + C \xrightarrow{k_{ads}} S_{f-1}C_1^* + \ldots + S_f \rightarrow S_{f-n}C_n^* + S_n \rightarrow \text{Product}
$$

(14)

$S_f$ and $S_n$ are the fraction of $S_i$ and of $S_o$ respectively while $S_nC_n^*$ is the intermediate. Of course, a lot of initiators which be formed but each will increase gradually. But, these intermediates do not accumulate to any significant extent during the reaction. They are in the steady-state condition. Then, there are two possible termination mechanisms: the equilibrium and the irreversible mechanism approach (intermediate $S_nC_n^*$ turned into the product). The first approach would lead to the first goal and the other to the second.

3.1.2. First goal. By steady-state approximation,. The addition of these equations gives equation (15).

$$
dS_1C_{tot}^*/dt = 0
$$

(15)

where $S_1C_{tot}^* = \sum_{n=0}^{\infty} S_nC_n^*$. 

5
In each step, one intermediate is consumed and one is produced (by undergoing further adsorption or back to the original by desorption). Hence, these steps do not affect \( S_i C_{tot}^* \) and \( dS_i C_{tot}^*/dt \). Therefore, it is only the rate at equilibrium that need to be considered.

To make it easier, it is assumed, the intermediate reactivities are independent of size and all have the same rate constant and for adsorption, \( k_{ads-1} = k_{ads-2} = \ldots = k_i \) and for desorption, \( k_{des-1} = k_{des-2} = \ldots = k_2 \). These assumptions are not exaggerated, because in a reversible reaction, the constants \( k_i \) (and \( k_2 \)) always has the same value even if determined at different concentration [6].

At equilibrium,

\[
k_i [ S_o - S] C = k_2 S
\]

which, on solving, gives eq.

\[ (3) \]

### 3.1.3. Second goal.

Under the second approach, the mechanism of the reaction is assumed to take place as eq. (16)

\[
S + C \xrightleftharpoons[k_{i-1}]{k_i} [SC^*] \xrightarrow{k_2} P
\]

\[ (16) \]

\([SC^*]\) is an intermediate and \( P \) is the product.
The first step is reversible that stay close to equilibrium and the second is irreversible. The latter is the rate-determining step or

\[
dP/dt = -dC/dt = k_2[SC^*]
\]

Applying the steady state approximation on \([SC]^*\) gives

\[
d[SC^*]/dt = 0 = k_i SC - k_{i-1}[SC^*] - k_2[SC^*]
\]

\[
k_i SC = k_{i-1}[SC^*] - k_2[SC^*]
\]

\[ (18) \]

If the maximum available sites are \( S_o \) then \( S = S_o - [SC]^* \) and eq. [18] becomes

\[
k_i (S_o - [SC^*])C = k_{i-1}[SC^*] + k_2[SC^*]
\]

\[
k_i S_o C = (k_i + k_2)[SC^* + k_i C[SC^*]]
\]

\[
[SC^*] = \frac{k_i S_o C}{(k_i + k_2) + k_i C}
\]

\[ (19) \]

Substitution of eq. [19] into eq. [17] gives

\[
-dC/dt = \frac{k_i k_2 S_o C}{(k_i + k_2) + k_i C}
\]

\[
-dC/dt = \frac{k_2 S_o C}{(k_{i-1} + k_2) / k_i + C}
\]

\[ (20) \]

\[
(k_i + k_2)/k_i = k
\]

\[ (21) \]
And

\[ k_2 S_0 = K_{ads} \]  \hspace{1cm} (22)

then eq. [15] becomes

\[- \frac{dC}{dt} = \frac{K_{ads}C}{k + C} \]  \hspace{1cm} (23)

Because the rate is proportional to the amount adsorbed at a certain time, then eq. (23) becomes

\[ S = \frac{K_{ads}C}{k + C} \]  \hspace{1cm} (6)

3.1.4. **Third goal.** The results of the calculation are presented in Table 5. and Table 6.

| Method          | Data Source   | A      | B      | r      | A      | B      | r      |
|-----------------|---------------|--------|--------|--------|--------|--------|--------|
|                 | Early-stage   | 0.0100 | -0.0004| 0.0146 | 106.5711 | -1.9708 | -0.00297|
|                 | Near-end      | 0.0004 | 0.1040 | 0.9404 | 9.3918  | 8.4730 | 0.9411 |

Table 5. Calculation Results of Table 3.

Based on Table 5., the new method confirms that at the early stage, the process is zero-order (slope B = -0.0004; the curve is linear with x-axis) and at the near-end is first-order (intercept A = 0.0004). The eq. (6) is not valid at the early-stage (negative slope) but seems valid at the near-end. However, the curve (rate versus concentration) of the first-order reaction cross the x-axis at x = 0 and the y-axis at y = 0.

| Method          | Data Source   | A      | B      | r      | A      | B      | r      |
|-----------------|---------------|--------|--------|--------|--------|--------|--------|
|                 | Early-stage   | 0.0000 | 0.0206 | 0.5651 | -1.6749 | 49.1769 | 0.5650 |
|                 | Near-end      | -0.0012 | 0.1419 | 0.9937 | -10.9633 | 8.7668 | 0.9969 |

Table 6. Calculation Results of Table 4.

Based on Table 6., the new method, confirms that at the early-stage (intercept A = 0.0000) and at the near-end (intercept A = -0.0012), the process is always first-order. There are two slopes, but it is the slope at the near-end that should be used. This slope has higher \( r \) (= 0.9997).

From a kinetic point of view, in eq. (5) the reaction is always first-order to \( S_0 \), either if \( C \) is much lower or higher than \( k \). This means, doubling \( S_0 \) will double the rate (and so as \( K_{ads} \)). This theory is very much in accordance with the fact found from the value of the slopes obtained from Table 5. and Table 6. The slope from Table 6. is, somewhat, 1.5 times higher than that from Table 5. This is because, the concentration of HCl used in Table 6. is also 1.5 times higher. Then, if the concentration of HCl is used for calculation, approximately, the same value of \( k \) will be obtained by using eq. (10). This is not the case in the use of eq. (6); both give the same slope value. So, as the conclusion, the value of \( K_{ads} \) and \( k \) must determine separately, using eq. (7) and eq. (10).

3.2. **Implications**

There are two cases that will be discussed further as the possible implication of this study. First, literature sources state that Freundlich adsorption isotherm [7], can be derived from the Langmuir
equation [8]. It is also implicit on the above discussion; the Freundlich equation is the equation of Langmuir on the condition of $C$ is relatively lower than $k$. The problem is, the $b$ constant in the Freundlich equation,

$$S = K C^b$$  \hspace{1cm} (23)

still need a further discussion, especially when related to reaction mechanism concept.

In many textbooks Freundlich isotherm is presented as an empirical equation with no or limited explanation on the physical significance of both $K$ and $b$ [9], and [10] [11]. Eq. (21) can be readily derived by kinetics approach [12]. However, it is not yet fully associated with the concept of reaction order and also not associated with the monolayer assumption. The reaction orders are usually integers or half-integers (1/2, 3/2, ...), positive or negative [13]. The value of $b$ in the adsorption of soluble organic nitrogen (SON) by granular activated carbon (GAC) is 0.746 [14], glycolic acid and acetic acid by Amberlite IRA-67 are 0.124 and 0.3174 respectively [15], and Procion Red MX-5B by filter paper is 0.57 [16]. In general, the parameter $b$ is dimensionless and typically has a value of $b < 1$. The parameter is a measure of heterogeneity of a system and a more homogeneous system would have a $b$ value approaching unity a more heterogeneous system would have a $b$ value approaching zero [17].

The fractal Langmuir equation has also been introduced [18], but still not clearly related to the reaction order concept and the monolayer assumption.

The next problem is the technique of experiment for the determination of the constants. As has been discussed above, the values of the constants are determined from $S$, using the concentrations data obtained at equilibrium condition. In facts, sometimes, equilibrium condition need too long times. However, $S = C_o - C$ and rate are proportional to the amount adsorbed at a certain time. Moreover, the rate constant of reversible reaction could be determined by initial rate method and will give the same value at any stages of a reaction. There is no need to wait for the equilibrium achieved [19].

The experiment can also be done from an experiment on the condition of $C$ is higher and lower than $k$. The observation is just done at the very beginning of the reaction on the condition of the constant time interval. This is done by using the physical properties resulting at the end of the one-time interval is being considered as the initial value for a new time interval. Because the amount absorbed is directly proportional to the rate, the constants can be determined by way of rate concept. Basically, there are 2 methods, differential and integral. The differential method gives the true order directly but not the exact value of the rate constant. The integral method gives the exact value. It is also easier to use because using $C$ data directly [20].

4. Conclusion and Recommendation

4.1. Conclusion

We present a simple technique on the determination of the isotherm equation based on the monolayer assumption and a product-based mechanism. In addition, precise technique for the adsorption constants determination is also explained. $K_{ads}$ and $k$ should be determined separately i.e. $K_{ads}$ from the intercept of the linear plot of $S$ versus (relatively high) $C$ and $k$ from the calculated value of $K_{ads}$ and the slope of the linear plot of $S$ versus (relatively low) $C$. The results show that this technique is more valid.

4.2. Recommendation

We proposed two suggestions. First is to derive the fractal Langmuir isotherm adsorption equation from monolayer assumption and relate it with the fractal Freundlich isotherm equation. Second is to determine $K_{ads}$, $k$, and $b$ by way of initial rate or better integral method.
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