Volume 13, Issue 3, Page 67-72, 2024 https://doi.org/10.46810/tdfd.1344725 Research Article

BİNGÖL UNIVERSITY

Türk Doğa ve Fen Dergisi
Turkish Journal of Nature and Science
www.dergipark.gov.tr/tdfd

Modeling of the Linear Equations of Langmuir Isotherm in the Adsorption of Cd (II) Ion with Siirt Kurtalan Koçpinar Clay

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(Received: 17.08.2024, Accepted: 02.08.2024, Online Publication: 26.09.2024)

Keywords
Cadmium, Langmuir, Linear regression, Adsorption constants, Isotherm, Clay

Abstract: Langmuir isotherm model has been widely used by researchers in adsorption isotherm studies for more than a century. During the process, 6 linear equations have been derived from the Langmuir model, which is a non-linear model. However, the degree of compatibility of these mathematically derived linear equations with experimental data is different. In addition, the constants obtained from the equations are also different.his study focuses on these 6 equations which are rarely given together. Thus, it was possible to compare the degree of fit of the equations with experimental data.

In this study, the performance of clay mineral in the removal of a heavy metal known for its harmful effect, such as cadmium, from solution was measured. The data obtained as a result of adsorption of cadmium (II) ion with clay from Siirt Koçpınar region at a temperature of 298 K were applied to 6 linear equations derived from Langmuir isotherm model. As a result, it was determined that the degree of suitability of the models for adsorption was type 3= type 6 < type 1= type 4 < type 2 = type 5 and the most suitable R^2 values belonged to type 2 and type 5 with values of 0.992. The largest q_m value was found to belong to type 4 with a value of 86.608 gmg^{-1}..

Cd (II) İyonunun Siirt Kurtalan Koçpinar Kili Ile Adsorpsiyonunda Langmuir Izotermine Ait Doğrusal Denklemlerinin Modellenmesi

Anahtar Kelimeler
Kadmiyum, Langmuir, Doğrusal regresyon, Adsorpsiyon sabitleri, İzoterm, Kil

Öz: Langmuir izoterm modeli yüzyılı aşkın bir süredir adsorpsiyon izoterm çalışmalarında araştırmacılar tarafından yaygın olarak kullanılmaktadır. Non-lineer bir model olan Langmuir modelinden süreç içerisinde 6 doğrusal denklem türetilmiştir. Ancak matematiksel olarak türetilen bu doğrusal denklemlerin deneysel veriler ile uyum derecesi farklıdır. Ayrıca denklemlerden elde edilen sabitler de farklı olmaktadır. Bu çalışma çok ender olarak beraber verilen bu 6 denkleme odaklanmıştır. Böylelikle denklemlerin deneysel verilerle uyum derecelerinin karşılaştırılması mümkün olmuştur.

Bu çalışmada, ucuz ve etkin oluşu nedeniyle kil mineralinin kadmiyum gibi zararlı etkisi ile bilinen bir ağır metalin çözeltiden giderimindeki performansı ölçülmüştür. Kadmiyum (II) iyonunun Siirt Koçpınar bölgesinden alınmış kil ile 298 K sıcaklığında adsorpsiyonu sonucunda elde edilen veriler, Langmuir izoterm modelinden türetilmiş 6 doğrusal denkleme uygulanmıştır. Regresyon modeli olarak en küçük kareler yöntemi kullanılarak denklemler karşılaştırılmıştır. Sonuç olarak; modellerin adsorpsiyona uygunluk derecelerinin; tip 3= tip 6 < tip 1= tip 4 < tip 2 = tip 5 olduğu ve en uygun R2 değerleri 0,992 değerleri ile tip 2 ve tip 5'e ait olduğu görülmüştür. En büyük qm değerinin 86,608 mgg-1 değeri ile tip 4'e ait olduğu tespit edilmiştir.

1. INTRODUCTION

Today, environmental pollution has reached a point that threatens the future of the world. Industrial residues play a major role in this. Heavy metals pollute water and land and cause health problems that may even result in death for living things, especially humans.

Cadmium, one of the most toxic elements, has the symbol Cd, atomic mass of 112.411 and atomic number 48 [1]. It is a non-abundant metallic element. Cadmium is often associated with minerals such as ZnS and other zinc ores. One mineral form, greenockite (CdS), is important, but the usual source of Cd is the oxide in industrial slag associated with the refining of Zn or Pb [2].

Cadmium is mainly used in battery production [3-5], alloys [6-8], solar cells [9-10], as as a plastic stabilizer [11-12] and pigment [13-15] in the industry and its wastes pose pollution and risk for the environment [16-18]. Various studies have been done on the removal of cadmium from solutions. The primary ones of these studies are coagulation method [19], ion exchange method [20], lime softening method [21], reverse osmosis method [22]. However, those conducted by adsorption method [23-25] attract the most attention.

Adsorption can be defined as the accumulation of a substance from a liquid or gas on a solid. If adsorption is in equilibrium at constant temperature, there is adsorption isotherm, if not, it can be mentioned as adsorption kinetics. There are many studies [26-48] on adsorption isotherm, adsorption kinetics and thermodynamics of it. Studying the adsorption isotherm gives insight into the nature of adsorption. A typical isotherm is a curve.

Among adsorption isotherms, Langmuir and Freundlich isotherms are prominent. While Freundlich isotherm model is considered suitable for heterogenous surfaces models [49], the Langmuir is associated with homogeneous surfaces [50], and monolayer adsorption [51]. This is largely due to the fact that Freundlich model anticipates multilayer adsorption while Langmuir isotherm anticipates monolayer adsorption.

1.1. Langmuir Isotherm Model

Irwing Langmuir published a paper in 1916 [52] that laid the foundation for the model that bears his name. Langmuir isotherm model anticipates a monolayer adsorption. Therefore, it is based on a limited adsorption. In chemical adsorption, multilayer adsorption does not occur because the atoms or molecules specifically bind one-to-one with the adsorbent. The Langmuir isotherm equation is essentially a non-linear equation. The equation is expressed as follows:

$$q_e = q_m \frac{K_L C_e}{1+K_L C_e} \quad (1)$$

Where;

q_e ; Amount adsorbed at equilibrium per gram of adsorbent (mgg^{-1})
q_m ; Maximum amount adsorbed at equilibrium per gram of adsorbent (mgg^{-1})
C_e ; Equilibrium concentration (mgL^{-1})
K_L; Langmuir constant (mgL^{-1})

Despite the fact that the Langmuir isotherm equation is nonlinear, six linearized equations derived from eq. 1 have gradually replaced it in the literature. However, 6 equations can hardly be found in the same study. But the sequence number of the equations is given randomly in the literature. These equations can be expressed as follows:

Type 1 $$\frac{1}{q_e} = \frac{1}{K_L q_m}\frac{1}{C_e} + \frac{1}{q_m} \quad (2)$$

Type 2 $$\frac{C_e}{q_e} = \frac{1}{q_m} C_e + \frac{1}{K_L q_m} \quad (3)$$

Type 3 $$q_e = -\frac{1}{K_L}\frac{q_e}{C_e} + q_m \quad (4)$$

Type 4 $$\frac{1}{C_e} = K_L q_m \frac{1}{q_e} - K_L \quad (5)$$

Type 5 $$C_e = q_m \frac{C_e}{q_e} - \frac{1}{K_L} \quad (6)$$

Type 6 $$\frac{q_e}{C_e} = -K_L\, q_e + K_L q_m \quad (7)$$

2. MATERIAL AND METHOD

In this study, mixed type clay from Siirt Koçpınar village was used as adsorbent. As adsorbate Sigma-Aldrich brand $Cd(NO_3)_2.4H_2O$ was used. Adsorption experiment was carried out with Memmert brand WNB 14 model heated shaking water bath at 298 K temperature. Equilibrium concentrations (C_e) were determined by Agilent 400 model atomic absorption spectrophotometer (AAS) Calculations were made as follows:

$$q_e = \frac{(C_i - C_e)V}{m1000} \quad (8)$$

Here;

q_e: The amount of substance adsorbed per gram by the adsorbent at equilibrium (mgg^{-1}),
C_i: Initial concentration (mgL^{-1}),
C_e: Concentration at equilibrium (mgL^{-1}),
V: Solution volume (mL) and
m: Mass of adsorbent (g).

The data were applied to the linearized 6 types of Langmuir isotherm model. Plot of type 1 was obtained by plotting $1/q_e$ versus $1/C_e$, plot of type 2 by plotting C_e/q_e versus C_e, plot of type 3 by plotting qe versus q_e/C_e, plot of type 4 by plot plotting $1/C_e$ versus $1/q_e$, plot of type 5 by plotting C_e versus C_e/q_e, and the plot of type 6 by plotting q_e/C_e versus q_e. For each type of plot equations, K_L, q_m ve R^2 constants were obtained, and these plots were comparized and tabulated. Microsoft Excel 2010 program was used for the operations and the

least squares method was adopted for regression analysis.

3. RESULTS

The isotherm plot giving the relationship between the independent variable C_e and the dependent variable q_e obtained as a result of the experiment and the calculations made afterwards is given in Figure 1:

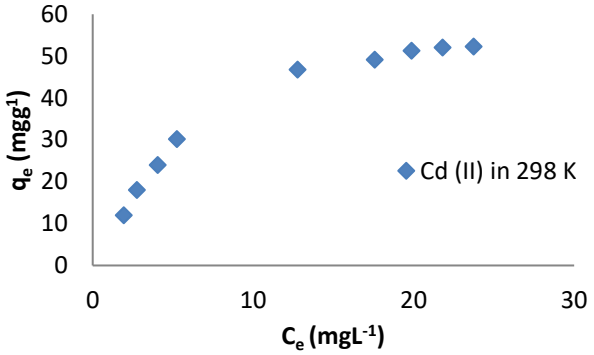

Figure.1. Isotherm curve of Cd (II) ion adsorption with Siirt Koçpınar clay at 298 K temperature

The plots generated by fitting the experimental data to the linear equations of the Langmuir model are given in Figures 2-7:

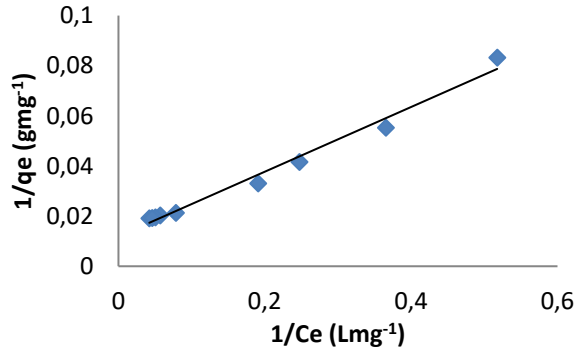

Figure.2. Linear plot of Langmuir type 1 linear equation for the adsorption of Cd (II) ion with Siirt Koçpınar clay at 298 K

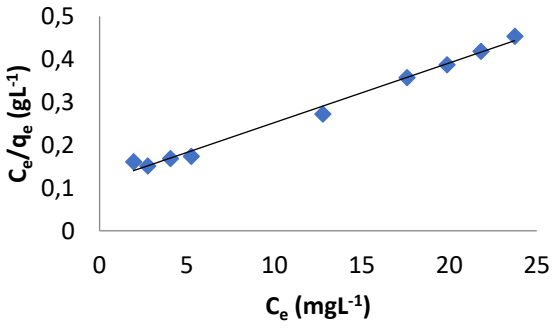

Figure.3. Linear plot of Langmuir type 2 linear equation for the adsorption of Cd (II) ion with Siirt Koçpınar clay at 298 K temperature

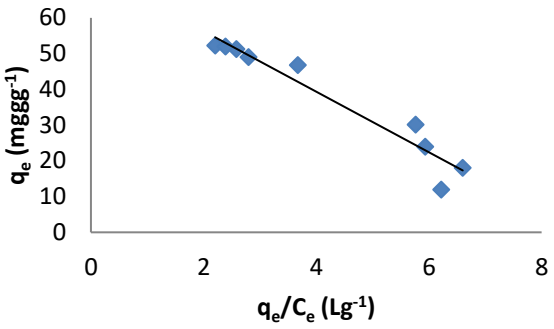

Figure.4. Linear plot of Langmuir type 3 linear equation for the adsorption of Cd (II) ion with Siirt Koçpınar clay at 298 K temperature

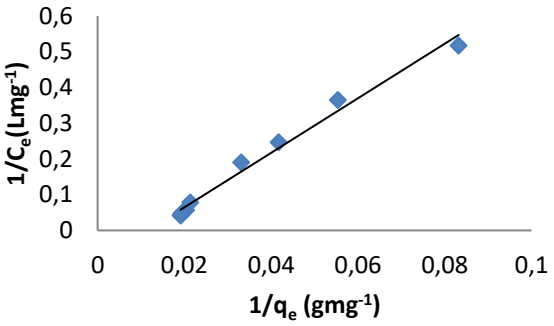

Figure.5. Linear plot of Langmuir type 4 linear equation for the adsorption of Cd (II) ion with Siirt Koçpınar clay at 298 K temperature

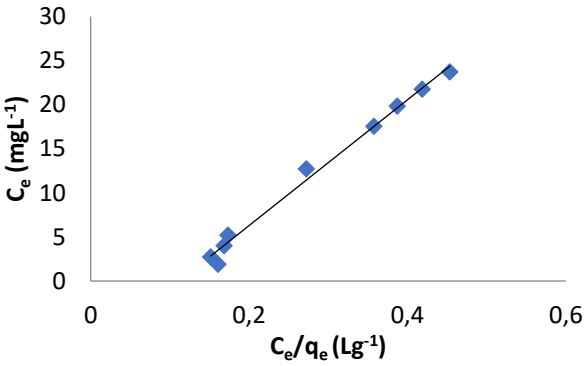

Figure.6. Linear plot of Langmuir type 5 linear equation for the adsorption of Cd (II) ion with Siirt Koçpınar clay at 298 K temperature

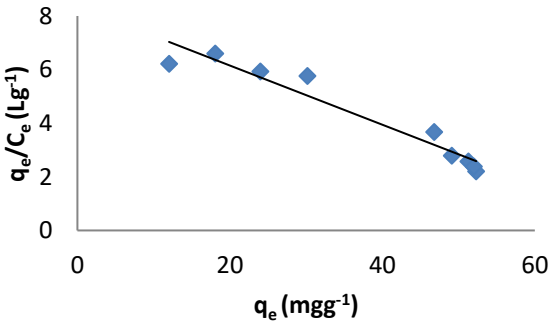

Figure.7. Linear plot of Langmuir type 6 linear equation for the adsorption of Cd (II) ion with Siirt Koçpınar clay at 298 K temperature

The constants obtained from the above 6 plots are given in table 1:

Table.1. Constants of the linear type equations of the Langmuir isotherm model

Linear Types	K_L (Lmg^{-1})	q_m (mgg^{-1})	R^2
Type 1	0.092	84.034	0.985
Type 2	0.123	71.942	0.992
Type 3	0.118	73.246	0.936
Type 4	0.088	86.608	0.985
Type 5	0.125	71.303	0.992
Type 6	0.110	75.715	0.936

Results that given table. 1 were showed that the degree of suitability of the models for adsorption; type 3 = type 6 < type 1 = type 4 < type 2 = type 5. The most suitable R^2 values belong to type 2 and type 5 with values of 0.992. q_m values; type 5< type 2 < type 3 < type 6 < type 1 < type 4 and K_L values were found to be type 4 < type 1 < type 6 < type 3 < type 2 < type 5. It was determined that the largest q_m value was belong to type 4 with a value of 86.608 gmg^{-1}. It is no coincidence that the smallest K_L value which is 0.088 Lmg^{-1} belongs to the same linear type. Low K_L values indicate high affinity, high q_m values indicate high adsorption capacity in accordance with affinity, and high R^2 values indicate a measure of the fit of the experimental data to the linear equation. As a result, the experimental data were found to be in high agreement with the theory.

4. DISCUSSION AND CONCLUSION

Recently, environmental pollution has become a global issue and has received much attention. One of the main causes of environmental pollution is heavy metal pollution. Due to the harm and widespread use of cadmium, a separate parenthesis can be opened here.
In this adsorption study carried out to remove cadmium pollution, 6 linearized equations of the Langmuir isotherm equation, which are rarely given together in the literature, constituted the main core of the study. Each of these 6 equations, which are almost never encountered in thesis studies and scientific articles in our country, means a new possibility for the Langmuir model. Because each equation will give different K_L and q_m values. Also, the degree of linearity of each equation will be different. As the adsorbent, adsorbed and other conditions affecting adsorption vary, the degree and sequence of concordance of each equation with the experimental data changes.

In this study, mixed type clay taken from Koçpınar region of Siirt Kurtalan district was used as adsorbent. As for adsorbed material, Cd (II) ion was used. The raw data obtained in the experiment carried out in a water bath with shaker at 298 K temperature were processed and applied to 6 linear equations of the Langmuir isotherm model. Least squares method was used as regression model for the comparison of the equations. As a result of the regression analysis, it was determined that the degree of suitability of the models for adsorption; type 3 = type 6 < type 1 = type 4 < type 2 = type 5. The most suitable R^2 values belong to type 2 and type 5 with values of 0.992. K_L values, a measure of affinity, were found to be be type 4 < type 1 < type 6 < type 3 < type 2 < type 5 and q_m values were found to be type 5 < type 2 < type 3 < type 6 < type 1 < type 4. It was determined that the largest q_m value was belong to type 4 with a value of 86.608. It is no coincidence that the smallest K_L value which is 0.088 belongs to the same linear type. Because low K_L value indicates high affinity, in this context, maximum adsorption is inevitable. The same is true for the vice versa. Namely, K_L value of type 5 was found to be the highest and q_m value was found to be the lowest. As a result, the experimental data were found to be in high agreement with the theory.

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