

# Adsorption Capacity of Low Cost Adsorbent Like Activated Bentonite for the Removal of Cadmium Ions from Wastewater by Adsorption Technique

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**Abstract:** *The present study aims to removal of cadmium ions as a heavy metal ion from aqueous solution by a high inorganic molecular weight compound. This compound is modified clay (activated bentonite) which is used in oil well drilling, and obtained from local company. The experimental results showed that it is an adsorption reaction that follows Langmuir isotherm model. The maximum removal of Cd (II) ions was obtained at low initial concentration, high adsorbent dosage, low temperature. It is clear from the results obtained that the compound within the experimental range investigated cannot be considered as a method for the removal of waste stream within the experimental range investigated as the residual concentration is higher than the limits which are allowed by the environmental limits which are allowed by the low which is 5 ppm. These results may be due to the high initial copper concentration used in this work and low dosage of the compound.*

**Keywords:** Adsorption, heavy metal, wastewater, treatment, Cadmium, Activated bentonite clay

## 1. Introduction

Water pollution by heavy metals through the discharge of industrial effluents, is a worldwide environmental problem. Heavy metals are metals of high relative atomic weight (especially one that is poisonous like mercury or lead) and a specific gravity greater than 5.0 [1]. Heavy metal ion pollution is currently of great concern due to the increased awareness of the potentially hazardous effects of elevated levels of these materials in the environment. With the rapid development of industries such as metal plating facilities, mining operations, fertilizer industries, tanneries, batteries, paper industries and pesticides, etc. Heavy metals in wastewaters are directly or indirectly discharged into the environment increasingly, especially in developing countries. Unlike organic contaminants, heavy metals are not biodegradable and tend to accumulate in living organisms and many heavy metal ions are known to be toxic or carcinogenic [2].

Toxic heavy metals of particular concern in treatment of industrial wastewaters include zinc, copper, nickel, mercury, cadmium, lead, and chromium. Effluent wastewaters from such processes contain toxic substances, metal acids, alkalis, and other substances. High concentrations of metals in the effluents may cause interference with biological treatment processes at sewage treatment plants [3, 4]. Cadmium does essential work in animal metabolism [5]. But the excessive ingestion of Cadmium brings about serious toxicological concerns, such as vomiting, cramps, convulsions, or even death. Many methods that are being used to remove heavy metal ions include chemical precipitation, ion-exchange, adsorption, membrane filtration, electrochemical treatment technologies [6, 7]. These techniques were found not effective due to either being extremely expensive or too inefficient to reduce such high levels of ions from the large volumes of water [8, 9]. Effluent wastewaters from such processes also contain toxic substances, metal acids, alkalis,

and other substances. High concentrations of metals in the effluents may cause interference with biological treatment processes at sewage treatment plants [10]. In addition, these techniques are not affordable in rural communities of developing countries. Therefore, the effective process must be low cost-effective technique and simple to operate. The wastewater purification processes requires the development of new operations based on low-cost raw materials with high pollutant-removal efficiency. Many toxic heavy metals are being discharged into the environment as industrial wastes, causing serious soil and water pollution [11]. Clay is a natural raw material that has been used for various purposes for ages and its dimension is under 2 mm. Montmorillonite is a member of smectite group clay minerals. Bentonites have a high content of montmorillonite and contain fewer amounts of other clay minerals.

Bentonite clay (hydrated aluminum silicate) was shown to be efficient in the removal of many toxic metal ions such as lead, cobalt, nickel, copper, zinc, cadmium and uranium in aqueous solutions [12-16]. Cadmium is a heavy metal posing severe risks to human health. Cadmium intoxication can lead to kidney, bone, Cadmium and pulmonary damages. Cadmium is regularly found in ores together with zinc, copper and lead. Therefore volcanic activity is one natural reason for a temporary increase in environmental cadmium concentrations. Cadmium is widely used in industrial processes, e. g.: as an anticorrosive agent, as a stabilizer in PVC products, as a colour pigment, a neutron-absorber in nuclear power plants, and in the fabrication of nickel-cadmium batteries. Phosphate fertilizers also show a big cadmium load. Although some cadmium-containing products can be recycled, a large share of the general cadmium pollution is caused by dumping and incinerating cadmium-polluted waste [17]. In Scandinavia for example, cadmium concentration in agricultural soil increases by 0.2% per year. Total global emission of cadmium amounts to 7000 t/year [18]. The maximum permissible value for

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workers according to German law is 15 µg/l. For comparison: Non-smokers show an average cadmium blood concentration of 0.5 µg/l. The major source of inhalative cadmium intoxication is cigarette smoke. The human lung resorbes 40–60% of the cadmium in tobacco smoke [19]. A 50 year-old average non-smoker has a cadmium body burden of 15 mg. While a comparable life-long smoker shows a value of 30 mg. Smokers generally have cadmium blood levels 4–5 times those of non-smokers [20]. cadmium enter the entero-hepatic cycle via secretion into the biliary tract in form of Cadmium-Glutathione conjugates. Enzymatically degraded to cadmium-cysteine complexes in the biliary tree, cadmium re-enters the small intestines [21]. The main organ for long-term cadmium accumulation is the kidney [22]. Here the half-life period for cadmium is approx.10 years. A life-long intake can therefore lead to a cadmium accumulation in the kidney, consequently resulting in tumulus cell necrosis. The blood concentration of cadmium serves as a reliable indicator for a recent exposition, while the urinary concentration reflects past exposure, body burden and renal accumulation [23].

The objective of this study is to remove one hazard heavy metal, which is Cd (II). The main objective of this work is to investigate the affecting parameters on the removal efficiency, such as initial metal ions concentration, adsorbent dosage, stirring speed and temperature.

## 2. Material and Methods

### 2.1. Chemical used for the preparation of solutions

Synthetic metal ion solutions of the desired concentrations were prepared by successive dilution with distilled water. Copper sulfate CdSO<sub>4</sub>. H<sub>2</sub>O was used as a source of Cadmium ions in the adsorption experiments. A stock solution of Cadmium sulfates solution was prepared dissolving an analytical grade of copper sulfate CdSO<sub>4</sub>. H<sub>2</sub>O in distilled water.

### 2.2. Characterization of Adsorbent

Activated Bentonite clay was supplied from the Sphinx milling station Company (Alexandria free zone, Egypt). Bentonite is brown in color; it was characterized by x-ray florescence (XRF) using AXIOS PANalytical2005. The analysis was made in Central Metallurgical Research Institute in Alexandria. Table 1 shows the oxides constituents of the studied bentonite clay. A scan electron microscope with different magnification was done on dry base for a sample of clay to specify the morphological features of bentonite. This was done in laboratory of faculty of science in Alexandria University as shown in Figure 1. Previously, the calculated values and recorded results obtained for Egyptian kaolin and bentonite [24, 25]

**Table 1:** Chemical analysis of Activated bentonite Adsorbent

| Chemical Constituents          | Percentage by Weight |
|--------------------------------|----------------------|
| SiO <sub>2</sub>               | 55.12                |
| Al <sub>2</sub> O <sub>3</sub> | 16.14                |
| Fe <sub>2</sub> O <sub>3</sub> | 8.25                 |
| MgO                            | 2.86                 |
| Na <sub>2</sub> O              | 1.41                 |
| TiO <sub>2</sub>               | 1.18                 |
| CaO                            | 1.17                 |
| K <sub>2</sub> O               | 1.05                 |
| Loss of Ignition (LOI)         | 12.35                |

### 2.3. Adsorption Procedure

The removal technique done in this work was adsorption of Cd (II) on the surface of high molecular weight compound was studied. The required weight of an adsorbent (bentonite) was prepared. Then the stock solution containing 1000 mg/l of metal ions solutions (CdSO<sub>4</sub>. H<sub>2</sub>O) were prepared from their stock solutions and then was mixed with different weight of adsorbents. The solutions were then agitated at room temperature or at other experimental temperature, after mixing for a time specified according to experimental conditions the samples were filtered using Whatmann filter paper and the filtrates were analyzed using EDTA and Titration method. The experiments were conducted using an orbital shaker for a known period of time. Adsorption experiments were carried out in 100ml flasks.

### 2.4. Adsorption Calculations

$$N_{\text{EDTA}} V_{\text{EDTA}} = N_{\text{Cd (II)}} V_{\text{Cd (II)}}$$

$$= \text{eq Cd (II) (If V is given in Litre)}$$

$$= m \text{ eq Cd (II) (If V is given in ml)}$$

The mass of Cd equals (eq Cd (II)) x (equivalent mass of Cd (II)) and

$$\text{Percentage Cd} = \frac{\text{Mass of Cd(II)}}{\text{Mass of Cd(II) salt}} \times 100$$

The concentration of Cd (II) in aqueous solutions before and after adsorption were determined as following.

$$\text{Original Cd(II)} = \frac{\text{Initial volume}}{100} \times 112.4$$

$$\text{Residual Cd(II)} = \frac{\text{Remaining volume}}{100} \times 112.4$$

$$\text{Adsorbed Cd (II) (qt)} = [\text{original Cd (II)-residual Cd (II)}] \times 10^4$$

$$[\text{mg Cd (II) adsorbed/gm adsorbent}]$$

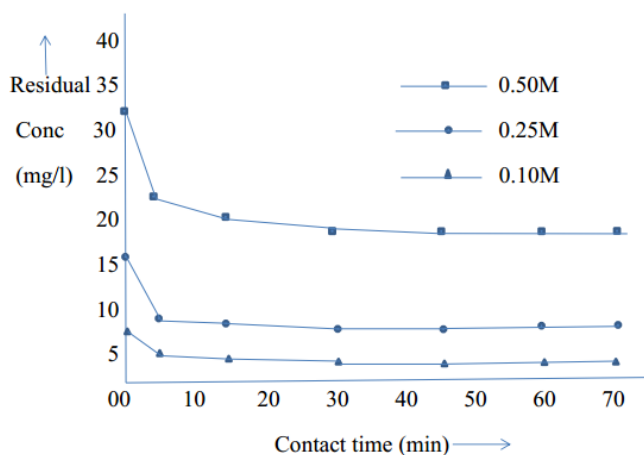
$$\text{Percentage Removal} = \left[ \frac{C_0 - C_t}{C_0} \right] \times 100$$

Where,  $C_0$  and  $C_t$  are the concentrations of Cd (II) initially and after  $t$  time respectively.

### 3. Results and Discussion

#### 3.1 Effect of Initial Metal Ion Concentration

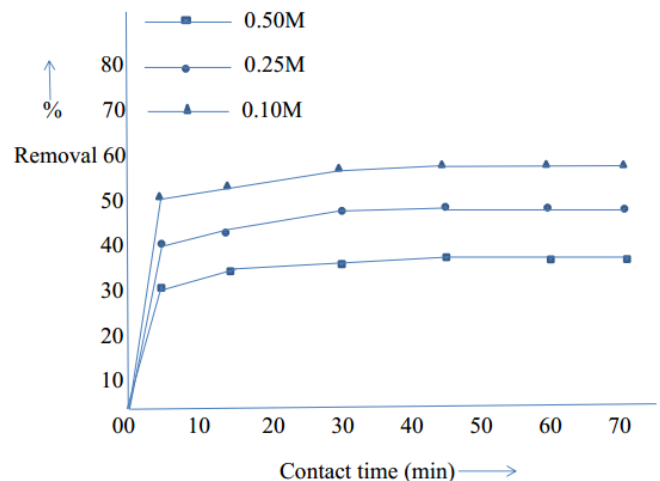
Figure 1 and 2 indicate that the effect of initial concentration of the Cd (II) ions on the residual concentration and percentage removal respectively. It was found that as the initial concentration of Cd (II) increases the residual concentration increases and the percentage removal decreases. It is also clear that at high concentration, the available sites of adsorption become fewer. This behavior is connected with the competitive diffusion process of the Cd (II) ions through the micro-channel and pores in the modified clay. That adsorption of metallic ions will lock the inlet of channel on the surface and prevent the metal ions from passing deeply inside the modified clay. The adsorption occurs on the surface only. Similar results in previous studies using natural zeolite [10] and olive cake [11], in addition to what have been found by karthikeyan et al. [12] confirm this point of view.



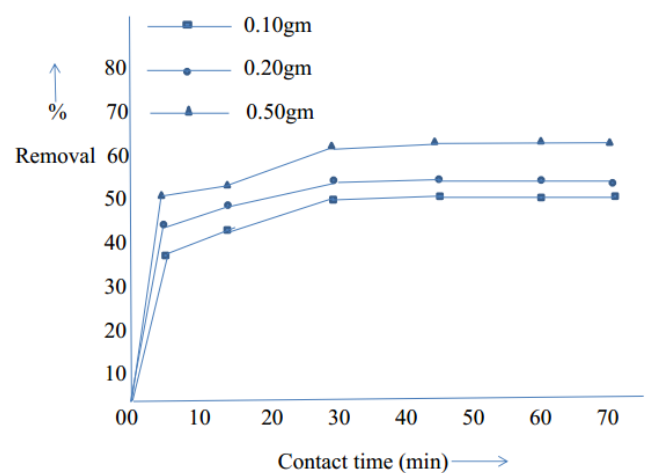
**Figure 1:** Effect of initial metal ion concentration on the residual concentration of Cd (II). pH = 4.6, agitation speed = 140 rpm, adsorbent = 0.1 gm, and Temperature = 25°C.

#### 3.2 Effect of Adsorbent Dosages-

The effluence of modified activated bentonite clay dosages on the percentage removal of Cd (II) is shown in Figure 3. This figure showed that as the amount of modified clay increases the percentage removal of Cd (II) increases. It is may be due to those higher amounts of modified clay means availability of a larger surface area or a larger number for adsorption sites [26-29] and Therefore, higher capacity for adsorption.



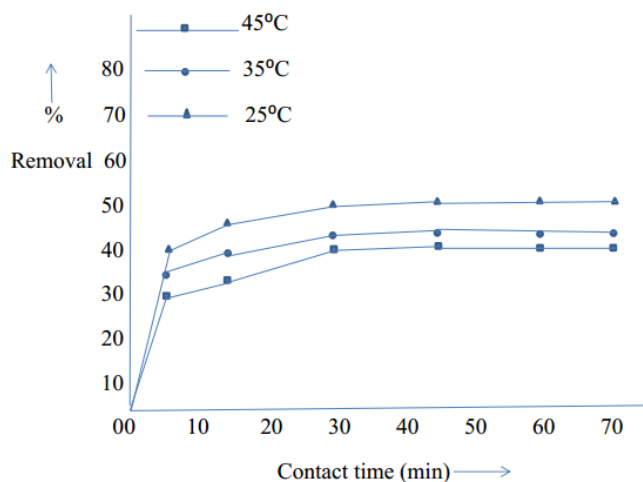
**Figure 2:** Effect of initial metal ion concentration on the percentage removal of Cd (II). pH = 4.6, agitation speed = 140 rpm, adsorbent = 0.1 gm, and Temperature = 25°C.



**Figure 3:** Effect of amount of adsorbent dosages on the percentage removal of Cd (II). pH = 4.6, agitation speed = 140 rpm, Concentration = 0.25M, and Temperature = 25°C.

#### 3.3 Effect of Temperature

Figure 4 indicates the effect of temperature on the percentage removal on Cd (II). It was found that as the temperature increases the percentage removal decreases. This was because with increasing temperature, the attractive forces between the adsorbent surface and metal ions are weakened and the desorption decreases. At high temperature, the thickness of the boundary layer decreases, due to the increased tendency of the metal ion to escape from the biomass surface to the solution phase, which results in a decrease in adsorption as temperature increases [18]. As a whole, it is clearly seen that as the temperature increases the loading capacity for the same initial adsorbent concentration decreases. This means that the rate of desorption was more significant than the rate of adsorption, which implies that adsorption is exothermic in nature [19-21].



**Figure 4:** Effect of temperature on the percentage removal of Cd (II). pH =4.6, Adsorbent = 0.1 gm, concentration=0.25M and agitation speed =140 rpm

### 3.4 Adsorption Isotherm Studies

In general, the isotherm studies for solid-liquid systems are carried out by changing Cadmium concentration and keeping other conditions, such as: volume of solution, adsorbent size, temperature, solution pH, and speed as constant. The performance of adsorbent is usually gauged by its uptake. Adsorbents can be compared based on their respective maximum uptake value, which can be calculated by fitting different isotherm models to the actual experiment data if it fits. The adsorption isotherm molecules distribute between liquid and solid phase when the adsorption process reaches an equilibrium state [30, 31]. The analysis of the isotherm data by fitting model that can be used for design purposes. Adsorption isotherm study was carried out on two isotherm models: Langmuir, and Freundlich isotherm models. The applicability of the isotherm models to the adsorption study done was compared by judging the correlation coefficient,  $R^2$  values.

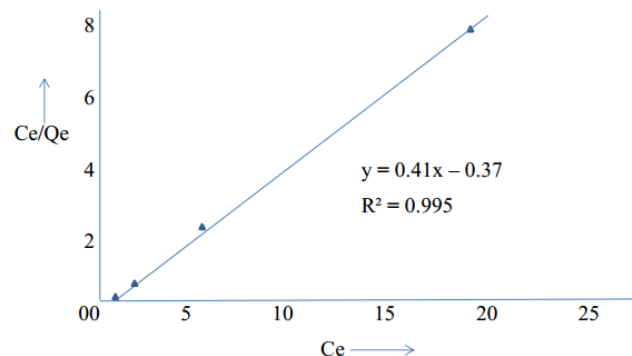
#### 3.4.1 Langmuir isotherm model

Langmuir's isotherm model suggests that the uptake occurs on homogeneous surface by monolayer sorption without interaction between adsorbed ions. The linear form of Langmuir isotherm equation is represented by the following equation [32, 33]:

$$\frac{C_e}{Q_e} = \frac{1}{Q^0 b} + \frac{1}{Q^0} C_e \dots\dots\dots(1)$$

Where  $Q^0$  is the maximum Cd (II) ions uptake per unit mass of adsorbent ( $\text{mg g}^{-1}$ ) related to adsorption capacity and  $b$  is Langmuir constant ( $\text{L. mol}^{-1}$ ) related to energy of adsorption. Therefore, a plot of  $(C_e/Q_e)$  vs  $C_e$ , gives a straight line of slope  $1/Q^0$  and intercept  $1/(Q^0 b)$ . Figure 5 shows the experimental data that were fitted by the linear form of Langmuir model,  $(C_e/Q_e)$  vs  $C_e$ . The values of  $Q^0$  and  $b$  were evaluated from the slope and intercept respectively. These values of  $Q^0$  and  $b$  are listed in Table 2 with their uncertainty and their determination coefficient ( $R^2$ ). The results obtained from the Langmuir model for the removal of Cd (II) onto modified bentonite clay in Table 2

which showed strong positive evidence on the adsorption of Cd (II) onto modified clay follows the Langmuir isotherm. The applicability of linear forms of Langmuir model modified clay was proved by the high correlation coefficients  $R^2 > 0.99$ . This suggests that the Langmuir isotherm provides a good model of the adsorption system.



**Figure 5:**  $C_e/Q_e$  vs  $C_e$  [Langmuir isotherm model]

#### 3.4.2 Freundlich Isotherm Model

Freundlich isotherm describes the adsorption equation for non-ideal adsorption that involves heterogeneous adsorption. This empirical isotherm is expressed by the following equation [32, 33]:

$$Q_e = K_F C_e^{1/n} \dots\dots(2)$$

The equation is conveniently used in the linear form by taking the logarithm of both sides as:

$$\log Q_e = \log K_F + \frac{1}{n} \log C_e \dots\dots\dots(3)$$

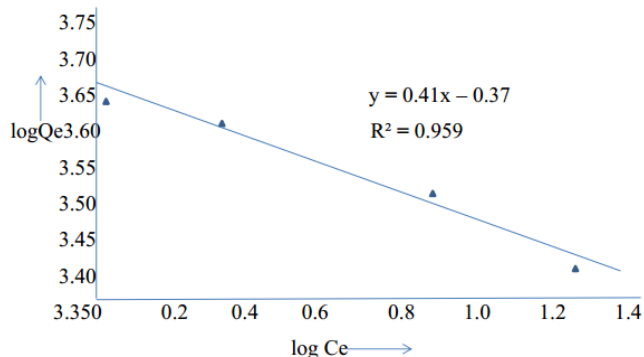
Freundlich constants,  $K_F$  and  $1/n$ , are related to adsorption capacity and intensity of adsorption, respectively. The value of  $n$  and  $K_F$  can be calculated from the slope and intercept of the plot of  $\log Q_e$  vs  $\log C_e$  derived from Eq. (3). The magnitude of the exponent  $1/n$  gives an indication of the favorability of adsorption, where values in the range of 2-10 represents good adsorption 1-2 gives moderate adsorption and less than 1 means that the adsorption has poor characteristics [34]. The applicability of the Freundlich adsorption isotherm was also analysed, using the same set of experimental data, by plotting  $\log Q_e$  vs  $\log C_e$ . The data obtained from linear Freundlich isotherm plot for the adsorption of the Cd (II) onto adsorbent. The correlation coefficients reported in Figure 6.

The equilibrium experimental results of adsorbed modified clay at various initial concentrations of Cd (II) were compared by using the adsorption isotherm equations, namely Langmuir, and Freundlich models. The temperature was constant throughout the experiments at 25°C. The experimental results were fitted to Langmuir and Freundlich models were done by linear regression using the statistical analysis package Excel. Figure 5 and 6 show the fitted equilibrium data of Langmuir and Freundlich respectively. For model comparison accordingly to  $R^2$  which are shown in Table 2 we concluded that Langmuir

isotherm is the best model to fit the data.

**Table 2:** Values of  $Q^0$ ,  $b$ , and  $R^2$  for Langmuir and Freundlich isotherm models

| Values of constant | Langmuir isotherm | Freundlich isotherm |
|--------------------|-------------------|---------------------|
| $b$                | -0.373            | 3.656               |
| $Q^0$              | 0.406             | -0.185              |
| $R^2$              | 0.995             | 0.959               |



**Figure 6:**  $\log Q_e$  vs  $\log C_e$  [Freundlich isotherm model]

According to  $R^2$  values, the higher the value of  $R^2$  the better model, from Table 2, it was concluded that Freundlich model has the least  $R^2$  value (0.959) while Langmuir isotherm model has the highest value of  $R^2$  which is (0.995). Therefore, Langmuir isotherm model is the best model to describe the Cd (II) adsorption system on modified clay. For Freundlich model we found that  $1/n = 0.1846$ , so that  $n = 5.417$ , therefore it lies in the interval 2-10 and applied good adsorption. The Langmuir model fit the data for metal ions ( $R^2 = 0.995$ ). Thus, it represents a monolayer adsorption process as chemical adsorption. Freundlich model fits the data for metal ions also ( $R^2 = 0.959$ ). Thus, it describes a heterogeneous system characterized by physical adsorption.

### 3.5 Kinetic Models Applied on Adsorption Process

Kinetics of adsorption is one of the most important characteristics to be responsible for the efficiency of adsorption. The adsorbate can be transferred from the solution phase to the surface of the adsorbent in several steps and one or any combination of which can be the rate-controlling mechanism [34]:

- (i) Mass transfer across the external boundary layer film of liquid surrounding the outside of the particle.
- (ii) Diffusion of the adsorbate molecules to an adsorption site either by a pore diffusion process through the liquid filled pores or by a solid surface diffusion mechanism.
- (iii) Adsorption (physical or chemical) at a site on the surface (internal or external) and this step is often assumed to be extremely rapid.

The overall adsorption can occur through one or more steps. In order to determine the mechanism of process and potential rate controlling steps, the kinetics of Cd (II) adsorption onto modified clay were analyzed using pseudo-first order [35], pseudo-second-order [36]. The conformity between experimental data and the model predicted values was expressed by the correlation coefficients ( $R^2$  values close or equal to 1). The relatively higher value is the more

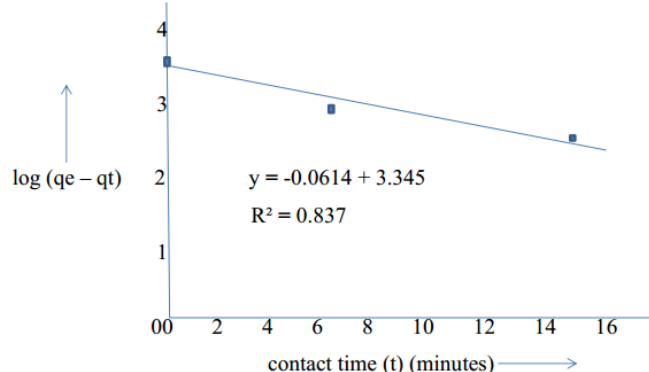
applicable model to the kinetics of Cd (II) adsorption onto adsorbent.

#### 3.5.1. Pseudo-first-order model

The adsorption kinetic data were described by the pseudo-first-order model which the earliest known equation is describing the adsorption rate based on the adsorption capacity. The linear form equation is generally expresses as follows [35]:

$$\log (q_e - q_t) = \log q_e - \frac{k_1}{2.303} .t \dots\dots(4)$$

the value of  $k_1$  and  $q_e$  can be determined from the slope and intercept of the plot  $\log (q_e - q_t)$  vs  $t$  (Figure 7). The variation in rate should be proportional to the first power of concentration for strict surface adsorption. However, the relationship between initial solute concentration and rate of adsorption will not be linear when pore diffusion limits the adsorption process.



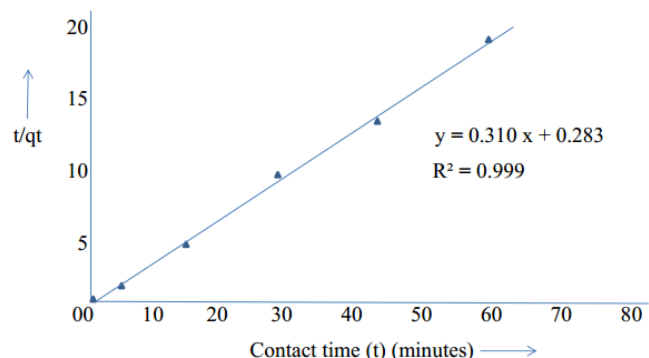
**Figure 7:**  $\log (q_e - q_t)$  vs  $t$  (Pseudo-first-order model)

#### 3.5.2. Pseudo-second-order model

The pseudo-second-order model may describe the adsorption Kinetic. The linear equation given as follows [36]:

$$\frac{t}{qt} = \frac{1}{K_2} \frac{1}{q^2e} + \frac{1}{qe} t \dots\dots(5)$$

If the second-order kinetics is applicable, then the plot of  $t/qt$  vs  $t$  should show a linear relationship. Values of  $K_2$  and equilibrium adsorption capacity  $q_e$  were calculated from the intercept and slope of the plots of  $t/qt$  vs  $t$ .



**Figure 8:**  $\log (t/qt)$  vs  $t$  (Pseudo-Second-order model)

**Table 3:** Values of R<sup>2</sup> for pseudo-first-order and pseudo-second-order models

| Kinetic Modal  | Pseudo-first-order | Pseudo-Second-order |
|----------------|--------------------|---------------------|
| R <sup>2</sup> | 0.837              | 0.999               |

According to table3, R<sup>2</sup> value was higher for pseudo-second order model than pseudo-first-order model; it confirms that pseudo-first-order model is the best model to fit the data.

#### 4. Conclusions

The removal Cd (II) ion from aqueous solution by modified activated bentonite clay has been investigated. The following conclusions can be drawn based on this investigation:

Modified clay remove Cd (II) from solutions by adsorption, it follows Langmuir isotherm model and Pseudo second order kinetic model. Maximum percentage removal takes place at low initial metal ion concentration, low temperature and high amount of adsorbent dosage. The results show that the use of modified clay cannot be considered as a waste treatment method for the removal of Cadmium from solution within the experimental range investigated.

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