



Investigation into the Efficiency of Kaolinite Clay for Sorption of Cadmium (II) Ion (Cd^{2+}) from Aqueous Solution: Kinetics and Isotherm Studies

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Abstract The efficiency of raw kaolinite clay without any form of modification or activation for the sorption of heavy metal (Cd^{2+}) ions from aqueous solution was investigated. Purified kaolinite was characterized by X-ray diffraction; surface area and cation exchange capacity (CEC) were also determined. Variations in adsorption parameters such as contact time and adsorbent dosage were employed to examine the sorption process and the result showed that equilibrium was attained after 120 minutes with a corresponding adsorption capacity of 22.0 mg/g. The sorption data were also fitted to few isotherm and kinetic models. Freundlich isotherm and Pseudo second order kinetic models with R^2 values of 0.989 and 0.981 for isotherm and kinetic models respectively gave best fit to the experimental data, thereby verifying the efficiency of natural kaolinite clay for sorption of Cadmium (ii) ions in aqueous solutions.

Keywords Isotherm; Langmuir; Cadmium (ii) ions; adsorption; kaolinite; kinetic model

1. Introduction

The presence of heavy metals in water, soil and plants is a global concern, owing to the wide range of anthropogenic sources of heavy metals polluting the environment with growing industrialization and wide utilization of chemicals [1-4]. Industrial wastewater effluents display wide range of heavy metals in varying concentrations [5]. Heavy metal contaminations are predominant in aqueous waste streams from diverse industries such as metal plating manufacturing, batteries, as well as agricultural sources where the utilization of fertilizers and fungicidal sprays are regularly employed [6]. Cu, Zn, Hg and Cd are hazardous waste materials introduced by industries, posing risks of contamination to groundwater and other water resources [7]. The food chain remains the major medium by which heavy metals enter the human body, causing serious health hazard if present in excess concentration beyond permissible limits [8]. Owing to their high level of persistency, heavy metals can bio accumulate in living organisms, disrupting the metabolic functions and vital organs in humans and animals [9]. Barbooti, [10] and Tiruneh *et al.* [1], reported the negative effects of heavy metals to include a wide range of illnesses such as respiratory problems, renal and organ damage, neurological dysfunction, heart disease, allergy, asthma, cancer as well as cardio vascular effects. However, they are not biodegradable and tend to accumulate in living organisms [11].

Various methods such as membrane filtration (ultra-filtration, reverse osmosis, nano-filtration, electro-dialysis), chemical precipitation, ion exchange and electrochemical method [12] have been employed to remove Cadmium from water. Adsorption on clays has also shown its credibility, as in use of bentonite [13-17] or kaolin [18-20].



Clays are eco-friendly and readily available minerals acting as excellent cation exchangers, which have often been used to adsorb metallic contaminants. They exhibit various adsorption capacities for metal ions, such as mesoporous silica [21] and montmorillonite clay [22-23]. Kaolinite clay ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) is a 1:1 clay mineral including a tiny interlayer structure [22-23]. The sorption properties of this clay are solely determined by the nature of its surface and edges, and it has been used as adsorbent materials for the sorption of various heavy metals [24-26].

Therefore, this study examines the feasibility of using kaolinite clay as an eco-friendly adsorbent for the removal of Cd^{2+} from aqueous solution. The influences of adsorption conditions such as contact time and adsorbent dosage were investigated and various isotherm and kinetic models were employed to study the sorption mechanisms.

2. Materials and Methods

All chemicals used in this study were of analytical grades.

2.1. Adsorbent

The kaolinite clay used in this study was obtained from a natural deposit located at Ubulu-Ukwu, Delta State, Nigeria with stones and other heavy unwanted particles removed from the sample after collection, and was further purified according to the methods of Nourmoradi *et al* [27]. Methylene blue method was used to estimate the surface area and cation exchange capacity (CEC) of the clay, and the purified sample of kaolin powder was further characterized by X-ray diffraction (XRD).

2.2. Preparation of Zinc Solutions

A 1000mg/l of cadmium stock solution was prepared in distilled water by dissolving 1.8158 g of cadmium sulfate (CdSO_4) salt in deionized water. The stock solution was further diluted into different ratios to prepare lower solution concentrations used in the experiments.

2.3. Determination of cadmium concentration in solutions

Atomic absorption spectrometer (BUCK Scientific 230ATS) with hollow cathode lamp and air acetylene flame, was used to determine Cadmium concentrations in water samples at a wave length of $\lambda=228$ nm.

2.4. Batch adsorption test

The adsorption tests were carried out in batch experiment, by mixing a fixed adsorbent mass of 2 g and 50 mg/L Cd^{2+} ion solution in 50 mL solution, at room temperature (28 ± 2 °C). The mixture was agitated on a magnetic stirrer. After filtration through a 0.5 μm membrane filter, at constant pH, residual cadmium concentration were measured for each filtered sample. The effect of contact times of 30-180 minutes on the cadmium metal ion adsorption efficiency of Kaolinite clay was investigated. The amount of Cd^{2+} ion adsorbed at equilibrium, q_e (mg/g) was estimated according to equation:

$$q_e = \frac{(C_o - C_e)V}{m} \quad (1)$$

Where q_e (mg/g) is the adsorption capacity of the adsorbent, C_o (mg/L) is the initial Cd^{2+} ion concentration, C_e (mg/L) is the equilibrium concentration of the Cd^{2+} ion in the solution, m (g) is the mass of the adsorbent, and V (L) is the volume of the solution.

Also, the effect of various adsorbent dosages (1-6 g) on the adsorption of Cadmium ion was studied at equilibrium time using 100 mg/L Cd^{2+} ion solution in 50 mL solution, at room temperature (28 ± 2 °C) and constant pH.

3. Results and Discussion

3.1. Characterization of kaolinite clay

The chemical composition of the kaolinite clay is presented in Table 1. As seen, the major elements are SiO_2 and Al_2O_3 . The values of 49.02 m^2/g and 16.9 Meq/100 g obtained for Surface area and cation exchange capacity (CEC) respectively are associated with kaolinite clay. From the X-ray diffraction studies (Figure 1), Kaolin show three intense diffraction peaks at 2θ value of 12.2° , 23.3° and 27.2° , less intense peaks at 2θ of 37.3° , 43.1° , 47.2° which



are all associated with kaolinite structures. Diffraction peaks of quartz (Q) could be found at 2θ values of 21.1° and 25.1° , and Illite (I) was also detected at $2\theta = 18.1^\circ$ and 43.2° .

Table 1: Chemical composition of Kaolinite clay used in this study

Chemical composition (%)	Kaolinite
SiO ₂	47.23
Al ₂ O ₃	32.34
Fe ₂ O ₃	1.75
Na ₂ O	0.05
MgO	0.13
Li	16.24
K ₂ O	0.63
CaO	0.04
TiO ₂	0.99
Total	99.4
Others	
CEC (Meq/100g)	16.9
Surface Area (m ² g ⁻¹)	49.02

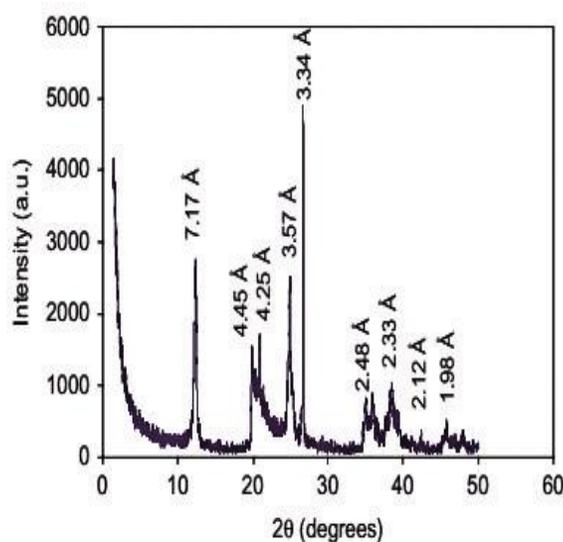


Figure 1: XRD pattern for kaolinite clay

3.2. Effect of contact time and adsorbent dosage

The influence of contact time ranging from 30-180 minutes on the adsorption of cadmium (ii) ions is presented in Figure 2. As seen, the adsorption capacity of cadmium (ii) ions by kaolinite clay varies with the contact time. The adsorption process was rapid in the first 90 minutes, and then slowed down until it attained equilibrium at 120 minutes. At this point, a constant value of 22.0 mg/g for the adsorption capacity was observed, indicating the absence of any further adsorption owing the exhaustion of surface adsorption sites. The rapid rate of adsorption noticed at the initial stage of the sorption was due to the availability of more active vacant sites on the kaolin surface, and the difficulty in occupying the remaining vacant sites after equilibrium time by the cations could be due to repulsive forces between adsorbent present in solid and bulk phases [11, 28]. However, 120 minutes was further used as the optimum contact time for the other experiment.

The effect of variations in the amount of kaolin from 1-6 grams (Figure 3) on the adsorption of Cd²⁺ ions indicated that an increase in the amount of kaolin resulted to a decrease in the adsorption capacity. when 1 and 2 grams of the



adsorbent were separately applied, their adsorption capacities were found to be 16.6 mg/g and 12.7 mg/g respectively, but a further increase in the adsorbent dosage resulted in a drastic decrease in the adsorption capacity. This could be allocated to the fact that as the adsorbent dosage in gram is increased, the total available surface area for cadmium (ii) ion sorption decreases, occasioned by overlapping or aggregation of sorption sites [11, 29].

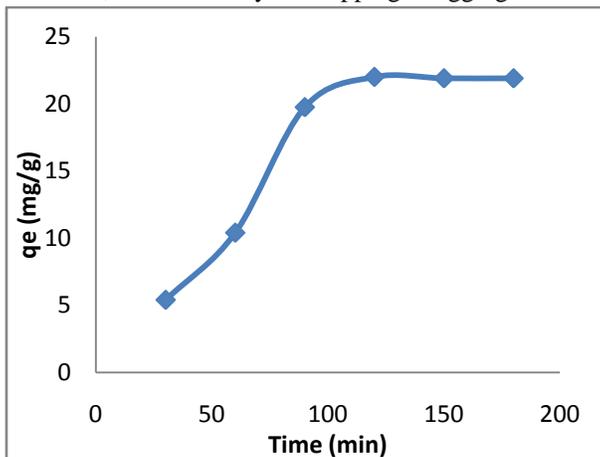


Figure 2: Effect of contact time on the adsorption of Cd^{2+} by Kaolinite clay (Initial cadmium ion concentration = 100 mg/L, mass of adsorbent = 2g, pH = 8.8, at 28 °C)

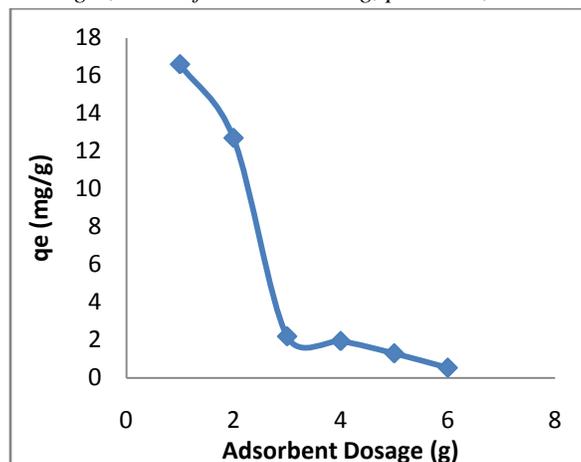


Figure 3: Effect of kaoline dosage on the adsorption of Cd^{2+} ion (Initial cadmium ion concentration = 100 mg/L, contact time = 120 minutes, pH = 8.8, at 28 °C)

3.3. Adsorption isotherm Modeling

(a) Langmuir Isotherm model

This is a mathematical isotherm model, which is gotten from theoretical approach or non experimental analysis and also has a constant referred to as Langmuir constant. The Langmuir isotherm model assumes that the adsorption of adsorbate molecule can only occur at a homogeneous site of the adsorbent and when this happen, no other adsorption will take place on the adsorbent surface [27, 30, 31, 32].

This isotherm model [27] in linear form is given as:

$$\frac{C_e}{q_e} = \frac{C_e}{Q_m} + \frac{1}{bQ_m} \quad (2)$$

Where C_e (mg/L) and q_e (mg/g) are the concentration of adsorbate and the adsorption capacity of the adsorbent at the equilibrium time, respectively. b (L/mg) is the Langmuir constant and Q_m (mg/g) is maximum adsorbent capacity. Q_m and b are attained by the slope and intercept of C_e/q_e versus C_e , respectively.



(b) Freundlich Isotherm Model

The Freundlich isotherm model is an empirical adsorption isotherm model which explains the equilibrium relationships existing between the adsorbate and the adsorbent molecules and assumes multilayer adsorption on the adsorbent heterogeneous site. The Freundlich isotherm model [31-33] is given as;

$$\ln q_e = \ln k_f + \frac{1}{n} \ln C_e \quad (3)$$

q_e and C_e are the adsorption capacity (mg/g) and the concentration (mg/L) of the adsorbate at equilibrium. The values of $1/n$ and k_f are gotten from the linear plot of $\ln q_e$ against $\ln C_e$. $1/n$ is the slope of the graph while k_f is the intercept on y-axis, which are the Freundlich constant relating to heterogeneity of the adsorbent surface and adsorption capacity respectively [32].

Table 2: Freundlich and Langmuir adsorption isotherm parameters of Cd^{2+} ion adsorbed onto kaolinite clay

Freundlich isotherm parameters			Langmuir isotherm parameters		
K_f (mol g ⁻¹)	1/n	R ²	q_{\max} (mol g ⁻¹)	R_L (l g ⁻¹)	R ²
1.68	1.35	0.989	2.70	0.05	0.903

From figures 4 and 5, it can be seen that the sorption data fitted appropriately to both the Langmuir and Freundlich isotherm models. The value of $1/n$ for the Freundlich isotherm was found to be above unity (1.35), suggesting the existence of a strong adsorption bond between the adsorbent and the adsorbate [27]. Also, the value of the Langmuir constant (R_L) was found to be 0.05, which is less than 1 (Table 2.), implying favorability of the isotherm model to the sorption data. Judging from the values of their correlation coefficient R^2 as seen in Table 2, the Freundlich Isotherm displayed a higher value of 0.989 compared to that of Langmuir found to be 0.903, thereby confirming Freundlich isotherm the most appropriate fit to the sorption data. Various studies [1, 20, 26] have also reported similar results.

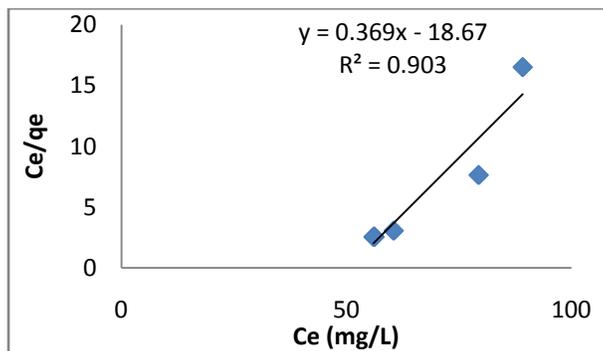


Figure 4: Langmuir isotherm plot for adsorption of Cd^{2+} ions by kaolinite clay

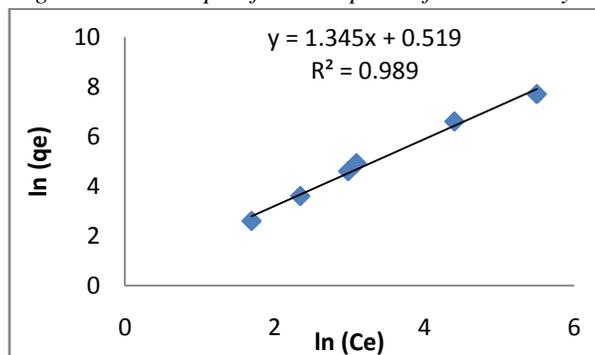


Figure 5: Freundlich isotherm plot for adsorption of Cd^{2+} ions by kaolinite clay



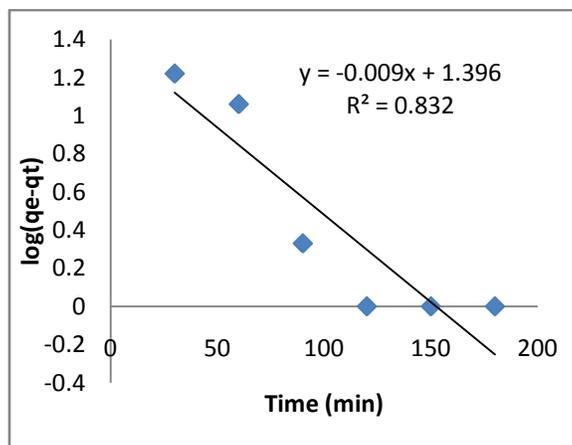


Figure 6: Pseudo first order kinetic plot for adsorption of Cd^{2+} ions by kaolinite clay

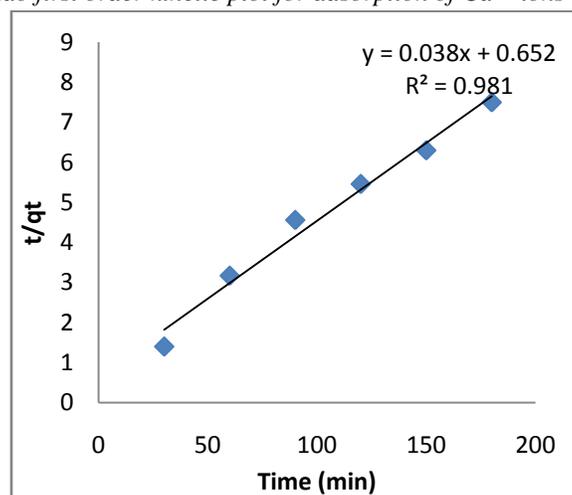


Figure 7: Pseudo second order kinetic plot for adsorption of Cd^{2+} ions by kaolinite clay

3.4. Adsorption kinetic modeling

Adsorption kinetics is one of the most vital parameters for determining the adsorption mechanism and also to investigate the efficacy of adsorbent for the removal of pollutants [27]. In this study, two kinetic models; pseudo-first-order and pseudo-second-order were used to predict the sorption behavior of the data. The pseudo-first-order kinetic model known as the Lagergren equation [34] is defined as:

$$\log(q_e - q_t) = \log q_e - \left(\frac{k_1}{2.303}\right)t \quad (4)$$

Where q_e is the amount of Cd^{2+} ions adsorbed at equilibrium (mg/g), q_t amount of Cd^{2+} ions adsorbed at any given time t (mg/g), k is the rate constant for the pseudo-first-order model. A plot of $(q_e - q_t)$ against t gives a linear slope (Figure 6) from which the values of k_1 and q_e can be determined from the slope and intercept.

The pseudo-second-order model was also fitted to the sorption data using the following equation [35]:

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

where q_e and q_t are the same as for the pseudo-first-order parameters and k_2 (g/mg²h) is the rate constant of pseudo-second order. k_2 and q_e values are obtained from the intercept and slope of the linear plot of t/q_t against t , respectively.

Table 3: Pseudo first and second order kinetic model parameters of Cd^{2+} ion adsorbed onto kaolinite clay



Pseudo first order model			Pseudo second order model			
k_1 (min^{-1})	$q_{e\text{Cal}}$ (mg/g)	R^2	K_2 (g/mg·h)	$q_{e\text{Cal}}$ (mg/g)	$q_{e\text{Exp}}$ (mg/g)	R^2
0.021	24.89	0.832	0.058	22.16	22.0	0.981

From the correlation coefficient presented in table 3, the adsorption of Cd^{2+} ion onto kaolinite clay was best fitted by the Pseudo second order kinetic model. This was confirmed by comparing the similarities between the $q_{e\text{Experimental}}$ and $q_{e\text{Calculated}}$ for both Pseudo first and Pseudo second order kinetic models. From the comparison, $q_{e\text{Calculated}}$ values of 24.89 and 22.16 for both models respectively confirms suitability of the Pseudo second order kinetic model to the sorption data. Many researchers [26,36,37,38] have reported non suitability of the Pseudo first order equation to the initial stages of adsorption processes. However, the suitability of the Pseudo second order kinetic model to the sorption data implies that the mechanism of the sorption process depends on the adsorbate and adsorbent interactions [39-40].

4. Conclusion

In the present study, the efficacy of kaolin clay as an eco-friendly, readily available and low-cost adsorbent for the adsorption of cadmium (ii) ion from aqueous solution has been investigated. From the results obtained, it is evident that kaolinite clay without any form of modification or activation can sorb a substantial amount of cadmium (ii) ions from aqueous solution. Also the investigation of some sorption parameters, such as contacts time and adsorbent dosage on the adsorption of cadmium (ii) ions varied with adsorption capacity. The adsorption of cadmium (ii) ion from aqueous solution can be appropriately described by both the langmuir and Freundlich isotherm models. Nevertheless, the Freundlich isotherm model may be considered the most appropriate fit to the sorption data owing to highest R^2 value compared to that obtained from the Langmuir isotherm. The sorption data were also fitted more appropriately by the Pseudo second order kinetic model. Thus, authenticating the suitability of kaolinite clay for sorption of heavy metals like Cadmium (ii) ions from aqueous solutions.

Conflict of Interest

Authors declare no conflict of interest.

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