THE REMOVAL OF CADMIUM (II) IONS FROM AQUEOUS SOLUTION BY THE USE OF "AFUZE" BENTONITE: EQUILIBRIUM, KINETIC AND THERMODYNAMIC STUDIES

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ABSTRACT

Afuze bentonite obtained from Owan east local government area, Edo State, Nigeria was investigated as a low cost adsorbent for the removal of Cadmium (II) ions from aqueous solution. This was determined by the use of batch adsorption methodology to deduce the dependence of solution pH, initial cadmium ion concentration, temperature and contact time on adsorption process. An optimum adsorption pH of 6.0 was obtained. Also, equilibrium removal was achieved within 60 minutes of the process. An increase in adsorption capacity with increase in initial cadmium ion concentration was recorded. Among the two adsorption isotherms tested, the Freundlich isotherm gave a better fit as indicated by the R^2 value (0.9788), than that of the Langmuir isotherm, (0.9356). The value of the regression R^2 obtained from the Pseudo second order model (0.9901) was better than that of the Lagergren first order model, (0.9098) applied in kinetic analysis. Thermodynamic studies indicated a spontaneous adsorption process as negative values of ΔG^0 were obtained at all temperatures. The negative value of ΔH^0, (-14.09KJ/mol) indicated an exothermic and a physisorption process. Also the negative value of ΔS^0, (-28.28J/mol/K) showed that the adsorption process is enthalpy driven. The result of this study suggested the usefulness of Afuze bentonite clay for the removal of cadmium (II) ions from solution.

Keywords: Adsorption, Afuze bentonite, Cadmium, Equilibrium, Heavy metals, Kinetic, Thermodynamic.

1 INTRODUCTION

The continuous increase in the use of metal and their compounds for technological development in many Countries have resulted in an increase in environmental pollution. As a result a lot of efforts have been devoted to minimize the harmful effects of these pollutants to plants animals and humans [1]. Heavy metal pollutants are of major concern to environmentalist due to their high toxicity, are non-biodegradable and can bio-accumulate in living organisms causing health problems [2]. Cadmium is used in most chemical industries for manufacturing pesticides, herbicides and fungicides [3]. Cadmium has been reported to be highly toxic because there does not exist a homeostatic control in the human body for this metal. About 1-2% of ingested cadmium if retained in the body is very harmful to health. It is an enzyme inhibitor and is responsible for kidney tubular impairment, affects calcium metabolism, skeletal calcification and ion regulation. It has been reported to cause diarrhoea, vomiting, a choking sensation, severe abdominal pain and liver damages [4].

Well documented techniques for the treatment of wastewaters contaminated with cadmium and other heavy metals are solvent extraction, ion exchange, membrane filtration, reverse osmosis, precipitation, evaporation, chemical oxidation or reduction, electrochemical treatment and activated carbon adsorption [5]. However, among these techniques, adsorption on activated carbon has been discovered to be the most effective and commercially applicable for the removal of cadmium and other heavy metals from effluents. These techniques are all expensive which limited their application to developing nations. Due to the high cost involved in treating industrial wastewaters by the use of activated carbon, Scientist are presently in search for cheaper alternatives [6]. Adsorption using low-cost materials as adsorbents have been researched extensively by scientist, which include the use of biomass materials [7],[8],[9],[10], soil [11], sawdust [12], clay [13], zeolite [14], kaolinite [15], illite [16], sepiolite [17], montmorillonite [18] and bentonite [19]. However, most studies on adsorption of heavy metals using these materials usually involves chemical modification or other forms of treatment to help improve their adsorption capacity. As a result, the application of these treatments increases the cost of production which makes them expensive to developing nations.

This present study was aimed at the application of Afuze bentonite clay (Nigeria) as a cheap alternative adsorbent for the removal of cadmium(II) from solution. The clay was used without chemical treatment or modification in order to keep the process cost low. Batch adsorption technique was applied and the equilibrium, kinetics and thermodynamic parameters were determined.

2 MATERIALS AND METHODS

2.1 Sample Preparation and Characterization

The clay was obtained from Afuze in Owan east local government area of Edo State, Nigeria. It was soaked in excess distilled water in a pre-treated plastic container. Stirred to ensure proper dissolution after which it was filtered to get rid of unwanted plant materials and suspended particles. The filtrate obtained was allowed to settle for 24hrs, excess water was decanted off and the clay was sundried for several days to ensure adequate drying. It was then oven dried at 105^0C for 2hrs, crushed and sieved through a 100µm mesh sieve. The sieved clay was used as the adsorbent.

Chemical composition of the clay was determined by the use of classical method and the help of the Atomic Absorption spectrophotometer (AAS) (Buck scientific model 210VGP).

2.2 Batch Adsorption Method

All the chemicals used in this study were of analytical grade. A standard stock solution of cadmium (II) of concentration 1000mg/l was prepared by dissolving appropriate amount of $Cd(NO_3)_2$ in distilled de-ionized water. Subsequent lower concentrations were prepared from the stock solution by serial dilution. The pH of each solution was adjusted to the desired value by the drop wise addition of 0.1M NaOH or 0.1M HNO_3 by the use of a pH meter to help check the pH. Freshly prepared solution was used for each study. The adsorption experiment was performed using batch method by contacting 2g of the adsorbent with 20mls of a given solution of cadmium (II) ion in a thermostat water bath for temperature regulation. The experiment was conducted to investigate the effect of pH in the range (1-8), initial metal ion concentration, 20-100mg/l, contact time, 10-120 minutes and temperature, 27-45^0C. In each experiment, a parameter was varied while others were kept constant. At the end of a given contact time, the contacted solution was filtered and the filtrate was analyzed for the concentration of cadmium (II) remaining in solution by the use of the AAS. The uptake capacity of the clay for cadmium ions was calculated from the mass balance equation given in (1).

$$qe = v[Co\text{-}Ce]/m \qquad (1)$$

The percentage of cadmium(II) adsorbed by the clay was calculated from (2)

$$\%\ Adsorbed = [Co\text{-}Ce]/Co \times 100 \qquad (2)$$

Where *qe* (mg/g) is the adsorption capacity at equilibrium, *Co* (mg/l) is the initial cadmium concentration in solution, *Ce* (mg/l) is the concentration of cadmium remaining in solution at equilibrium, *v* (litres) is the volume of solution used and *m* (g) is the mass of adsorbent.

3 RESULT AND DISCUSSION

3.1 Chemical Characterization of Afuze Bentonite

The chemical composition of the clay as determined by classical technique is shown in Table 1. It is seen that Silica and Alumina are the major constituents, other elements are present in smaller amounts. It is thus expected that cadmium (II) ions in solution should be removed mainly by SiO_2 and Al_2O_3.

TABLE 1:
Chemical Composition of Afuze Bentonite.

Composition	% by weight
SiO_2	53.12
Al_2O_3	22.81
CaO	4.01
MgO	1.87
Na_2O	1.02
Fe_2O_3	2.65
K_2O	1.27
TiO_2	0.51
MnO	0.42
LOI	12.32

3.2 Influence of Initial Solution pH

The initial pH of a solution is a very important factor to be considered, as the amount of metal ion adsorbed by an adsorbent is strongly dependent on the solution pH [20]. The effect of initial pH of solution on the adsorption of cadmium by Afuze bentonite is shown in fig.1. It was observed that an increase in adsorption with increase in pH up to 6.0 was obtained. Optimum pH of adsorption was recorded at 6.0 after which it became fairly constant up to 8.0. The increase in adsorption with pH can be explained, at low pH values the solution is strongly acidic, a lot of protons are thus present in solution which competes with cadmium ions for the active sites of the clay which resulted in the low uptake recorded. As the pH of the solution increases, the acidity reduces, hence there is a decrease in the number of protons available to compete with cadmium ions for the active sites which resulted in an increase in adsorption. Higher pH values greater than 7.0

in the alkaline region are usually avoided in most adsorption studies due to the possibility of metal precipitation [21].

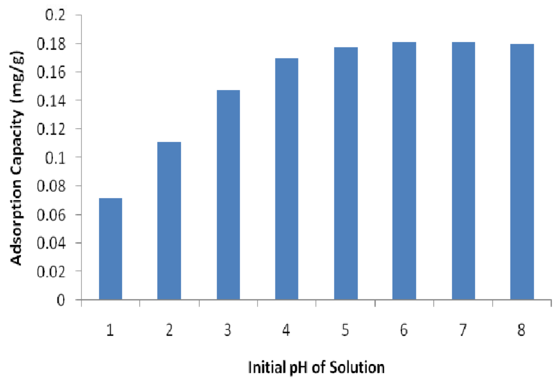

Fig.1: Effect of pH on the removal of Cadmium (II) ions by Afuze Bentonite Clay (20mg/l, 2hrs, 300K)

3.3 Influence of Initial Cadmium (II) ion concentration

The initial concentration of metal ion in solution determines the amount of metal ions adsorbed by an adsorbent in the presence of available active sites [22]. This can be seen in fig.2, which shows the effect of initial cadmium ion concentration on its adsorption by Afuze bentonite clay. An increase in adsorption capacity with increase in cadmium concentration was obtained. This is simply due to the presence of more metal ions in solution available for binding to the active sites. As metal concentration increases there is an increase in collision between the metal ions and adsorbent, as a result, a greater driving force is generated which leads to the effective utilization of binding sites [23].

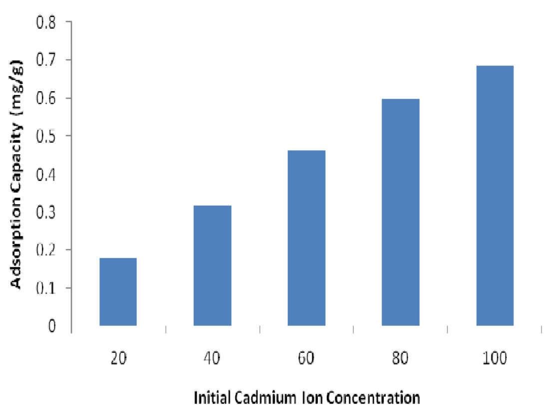

Fig.2:Effect of initial Cadmium ion concentration on its adsorption by Afuze bentonite clay (pH, 6.0, 2hrs, 300K).

3.4 Influence of Contact time

The effect of contact time is important in adsorption studies as it helps to determine the rate of adsorption and the time required for equilibrium sorption to be established. The result on the effect of contact time on the removal of cadmium(II) ions by Afuze bentonite is shown in fig.3. The adsorption rate was rapid initially and then gradually diminished to attain an equilibrium time beyond which there was no significant change in the rate of removal. Equilibrium was established within 60 minutes of the process. Batch experiments performed in this study were conducted at a contact time of 2hrs, hence we ensured equilibrium was attained. The fast adsorption rate at the initial stage is due to the availability of abundant active sites on the surface of the clay which became saturated with time. The insignificant adsorption observed at the later stages is simply as a result of saturation of actives sites responsible for sorption [24].

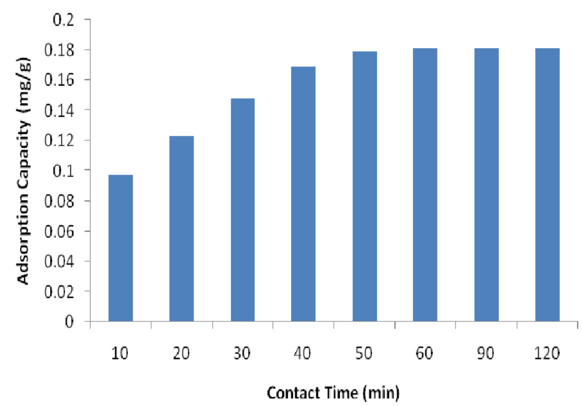

Fig.3: Effect of Contact time on the removal of Cadmium (II) ions by Afuze bentonite (pH, 6.0, 20mg/l, 300K)

3.5 Influence of Temperature

The effect of temperature on the adsorption of metal ions by various adsorbents have been known to present different types of behaviours. An increase in adsorption with temperature, decrease in adsorption, temperature independent effect and irregular behaviours have all been reported [25]. The effect of solution temperature on the removal of cadmium by Afuze bentonite is shown in fig.4. A slight decrease in the adsorption capacity with increase in temperature was observed. This implies that the adsorption process is exothermic in nature, that is lower temperatures will favour the sorption process. Similar result have been reported by other researchers [11],[26].

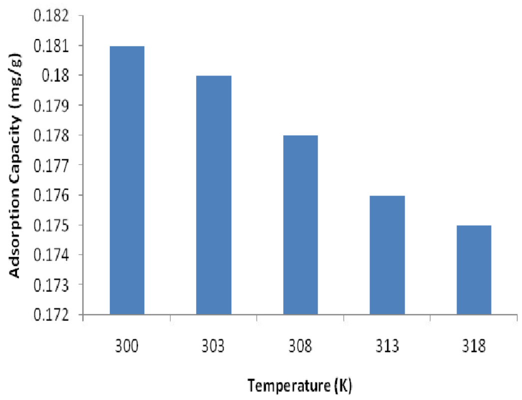

Fig.4: Effect of temperature on the removal of Cadmium (II) ions by Afuze bentonite (pH, 6.0, 20mg/l, 2hrs)

3.6 Adsorption Isotherms

An adsorption isotherm helps establish the equilibrium relationship between the adsorbate concentration in the liquid phase and that on the adsorbent surface at a given temperature. The two most widely used isotherms namely the Langmuir and Freundlich adsorption isotherms were applied to the experimental data.

TABLE 2:
Langmuir and Freundlich Isotherm Parameters

Langmuir Isotherm Model			
qm	**K_L**	**R^2**	
0.917	0.086	0.9356	
Freundlich Isotherm Model			
1/n	**n**	**Kf**	**R^2**
0.493	2.029	0.127	0.9788

3.6.1 Langmuir Isotherm Model

The Langmuir isotherm model was applied to determine the maximum adsorption capacity corresponding to a complete monolayer coverage. This isotherm describes monolayer sorption on a surface containing a finite number of non-interactive identical binding sites [27]. The linear form of the Langmuir isotherm model is given in (3)

$$Ce/qe = Ce/qm + 1/qmK_L \qquad (3)$$

Where *qe* (mg/g) is the equilibrium cadmium concentration on the bentonite surface, *qm* (mg/g) is the maximum adsorption capacity for a complete monolayer coverage and K_L (L/mg) is the Langmuir isotherm constant. The constants *qm* and K_L were obtained from the slope and intercept of the plot of *Ce/qe* against *Ce* shown in fig.5. The Langmuir isotherm parameters are presented in Table 2. The low K_L value obtained indicated a favourable adsorption between the adsorbent and adsorbate species [28].

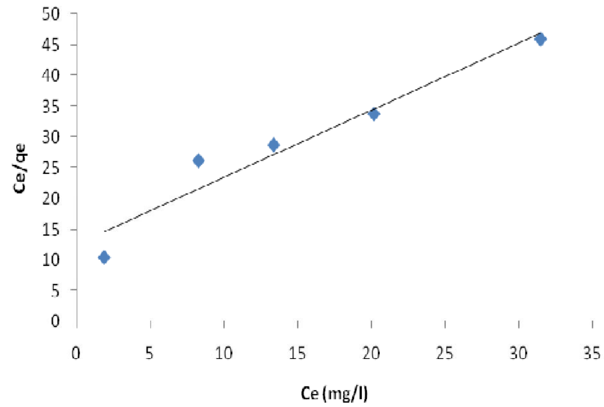

Fig.5: Langmuir isotherm plot for the removal of Cadmium (II) ions by Afuze bentonite (pH, 6.0, 2hrs, 300K)

3.6.2 Freundlich Isotherm Model

The Freundlich isotherm was used to deduce the adsorption intensity of the adsorbent towards the adsorbate. The isotherm assumes a multilayer adsorption unto a heterogeneous surface [29]. The linear form of this isotherm equation is given in (4)

$$lnqe = [1/n]lnCe + lnKf \qquad (4)$$

where *Kf* (L/g) and *n* (dimensionless) are the Freundlich isotherm constants describing the adsorption capacity and intensity respectively. The linear plot of *lnqe* against *lnCe* is shown in fig.6 and the constants *n* and *Kf* were obtained from the slope and intercept respectively. The Freundlich isotherm parameters are recorded in Table 2. Looking at the linear regression coefficient (R^2), it is seen that the Freundlich isotherm is more applicable to the sorption process than the Langmuir isotherm model. Values of *n* between 1 and 10 represents a high affinity of the adsorbent for the adsorbate [30].

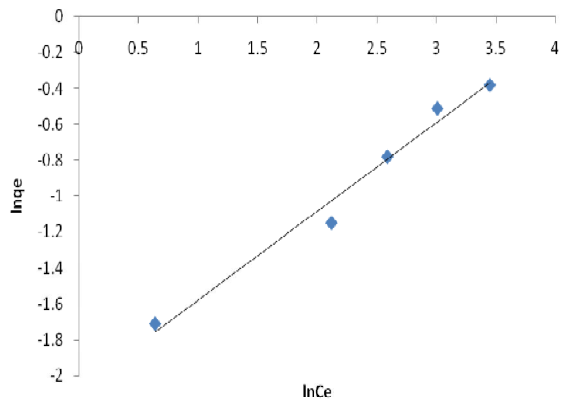

Fig.6: Freundlich Isotherm plot for the removal of Cadmium (II) ions by Afuze bentonite (pH, 6.0, 2hrs, 300K)

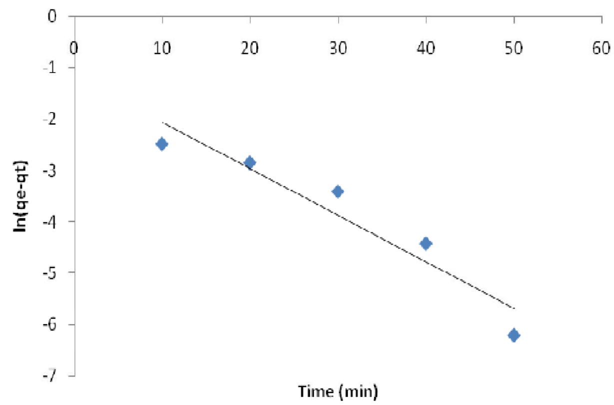

Fig.7: Lagergren first order plot on the removal of cadmium (II) ions by Afuze bentonite (pH, 6.0, 20mg/l, 300K)

3.7 Kinetics of Adsorption

Kinetics of adsorption is one of the most important parameters responsible for understanding the rate of adsorption, adsorbent design, reactor dimensions and the residence time of an adsorbate. Both the Lagergren first order and the Pseudo second order kinetic models were applied to the experimental data to predict kinetic mechanism.

TABLE 3:
Kinetic Model Parameters

Lagergren First Order Model		
qe	**K_i**	**R^2**
0.312	0.0903	0.9098
Pseudo Second Order Model		
qe	**K_2**	**R^2**
0.235	0.265	0.9901

3.7.1 Lagergren First order model

The Lagergren first order model equation [31] was adopted and is given in its linear form in (5).

$$ln(qe-qt) = lnqe - Kit \qquad (5)$$

where *qe* (mg/g) and *qt* (mg/g) are the amounts of cadmium(II) ions adsorbed by the bentonite at equilibrium and time *t* respectively. *Ki* (min^{-1}) is the Pseudo first order rate constant of sorption. This model was applied by a linear plot of *ln(qe-qt)* against *t* as shown in fig.7 and the constants *Ki* and *qe* were calculated from the slope and intercept respectively. The Lagergren first order kinetic parameters are given in Table 3.

3.7.2 Pseudo Second Order Model

The Pseudo second order equation for the sorption of divalent metal ions assumes that adsorption follows a second order mechanism and the rate of occupation of adsorption sites is proportional to the square of the number of unoccupied sites [32]. The linear form of the Pseudo-second order equation is given in (6)

$$t/qt = 1/K_2qe^2 + t/qt \qquad (6)$$

K_2 (g/mg/min) is the equilibrium rate constant of pseudo second order adsorption. The values of *qe* and K_2 were obtained from the slope and intercept of the plot of *t/qt* against *t*. The model parameters obtained are presented in Table 3. The values of the linear regression coefficient (R^2) obtained showed that the pseudo second order model gave a better fit to the experimental data than the pseudo first order model. This model assumes that the metal removal from solution is due to physisochemical interactions between the adsorbate in solution and adsorbent surface [26].

3.8 Thermodynamics

The thermodynamics of adsorption was applied to obtain important parameters which helps provide a better understanding of the adsorption process. These parameters include free energy change ΔG^0, enthalpy change ΔH^0 and the entropy change ΔS^0. The parameters were calculated from (7)-(9).

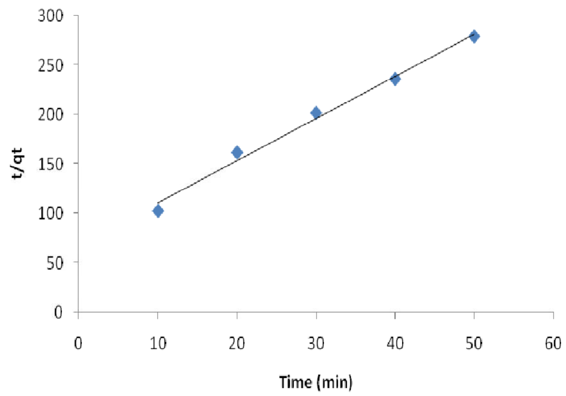

Fig.8: Pseudo second order plot for the removal of Cadmium (II) ions by Afuze bentonite (pH, 6.0, 20mg/l, 300K)

Kad = Cad/Ce (7)

$\Delta G^0 = -RTlnKad$ (8)

$\Delta G^0 = \Delta H^0 - T\Delta S^0$ (9)

Where *Kad* is the equilibrium constant of adsorption, *Cad* (mg/l) and *Ce* (mg/l) are the equilibrium concentration of cadmium (II) ions on the adsorbent and in solution respectively, *R* is the universal gas constant (8.314$Jmol^{-1}K^{-1}$) and *T*(K) is the absolute temperature. The thermodynamics was applied by a linear plot of ΔG^0 against *T* as shown in fig.9. The parameters ΔS^0 andΔH^0 were obtained from the slope and intercept. Thermodynamic parameters obtained are presented in Table 4. The value of R^2 obtained showed a good agreement between free energy changes and temperature. A spontaneous adsorption process was indicated by the negative values of ΔG^0 obtained at all temperatures. The upper limit for the change of enthalpy for physisorption is generally 80KJ/mol and for chemisorptions is 80-420KJ/mol [33]. The magnitude of the negative values of enthalpy change (-14.09KJ/mol) obtained indicated an exothermic physisorption process. Also, the negative value ofΔS^0 obtained suggested that the adsorption of cadmium (II) ions unto Afuze bentonite clay is enthalpy driven.

TABLE 4:
Thermodynamic Parameters

T(K)	ΔG^0 (KJ/mol)	ΔH^0 (KJ/mol)	ΔS^0 (J/mol/K)	R^2
300	-5.62	-14.09	-28.28	0.9629
303	-5.53			
308	-5.35			
313	-5.18			
318	-5.41			

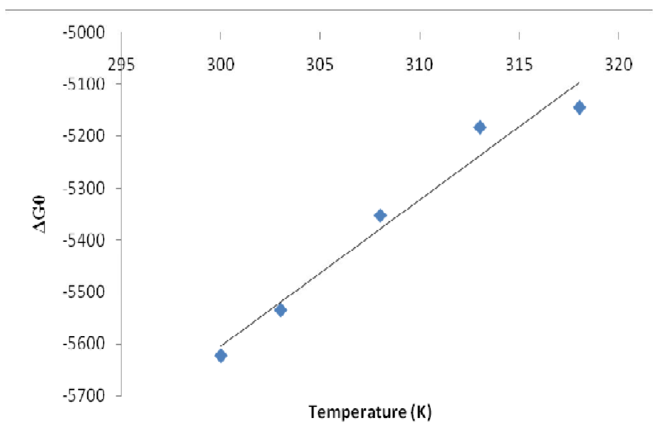

Fig.9: Thermodynamic plot for the removal of Cadmium (II) ion by Afuze bentonite (pH,6.0, 20mg/l, 2hrs)

4 CONCLUSION

The results obtained in this study revealed the usefulness of Afuze bentonite as a low-cost adsorbent for the removal of cadmium(II) ions from solution. This clay material can be applied in the treatment of industrial wastewaters in Nigeria and other developing Nations.

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