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SORPTION AND DESORPTION KINETICS OF CADMIUM IMMOBILIZATION IN LATERITE SOIL USING ANIMAL BONES

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ABSTRACT

The sorption and desorption kinetics of cadmium immobilization in laterite soil using animal bones were investigated by subjecting the experimental data to various kinetic models. Sorption kinetic was studied with Pseudo first order, pseudo second order and parabolic diffusion while desorption kinetics was investigated with first order, Second order, and Simple Elovich Models. These models were chosen for their significance to understanding and optimizing the kinetics of immobilization processes, since they provide insights into the mechanisms driving sorption and desorption, allowing for the construction of more efficient systems. The SEM analysis of the bones show that they are mainly composed of calcium phosphate in the increasing order of CBA>PBA>HBA. This compound caused the precipitation of metal phosphate, which resulted in decreased mobility because of its low solubility. The physicochemical analysis of the soil shows that it contains iron oxide, which increased the iron and manganese oxide fraction of the soil, which resulted in increased bonding of metal to this fraction thus reducing the amount of metal available for plant uptake. The correlation coefficient R^2 was used to determine the suitability of the model to the kinetic data. For pseudo first order, pseudo second order and parabolic diffusion, R^2 ranges from 0.419-0.8295, 0.9946- 0.9997 and 0.5116- 0.8295 respectively while for desorption process, it ranges from 0.254-0.349, 0.164 – 0.211 and 0.981- 0.990 for first order, second order and Elovich models respectively for all amendments. This resultsimplied that pseudo second order model best suited the sorption process while desorption was best fitted by Elovich kinetic model.

Keywords: Sorption, Desorption, Cadmium, Laterite, Soil, Animal bone, Immobilization

INTRODUCTION

Heavy metals are defined as elements having atomic weights ranging from 63.5 to 2006 and densities more than 4 g/cm3, or five times the density of water. Cadmium, with an atomic weight of 112.4 g and a density of 4.58 g/cm3, is definitely a heavy metal. Cadmium comes from two sources: natural and human activities (Zaman et al., 2022). According to anthropologists, the sources of cadmium are industrial operations such as cement manufacture, mining, electroplating, plastic pigments, fertilizers, and the incineration of sewage and municipal sludge.

Cadmium is the most harmful metal to which a man can be exposed in the workplace or the environment, despite its utility in the production of cadmium-nickel rechargeable batteries, plastic stabilizers, and corrosion protection coatings for iron and steel (Bernard 2008). According to Bernarnd et al. (1992), cadmium accumulates in the kidneys and can cause damage over time. In addition to producing hypertension, cadmium can cause lung cancer and other health issues in people. (Tellez-Plaza and colleagues, 2008).

One of the methods of treating cadmium in the environment is by immobilization process. This is a physical, chemical, and biological process that successfully removes hazardous substances from water or soil, stabilizes them, and prevents their leaching or uptake by plants and organisms (Bolan et al., 2011; Tang et al 2013). Precipitation, adsorption, complexation, encapsulation, pH alteration, and microbial activities are the most common immobilization mechanisms (Chatterjee and Mukherjee 2019; Wuana and Okieimen 2011). Precipitation is the use of chemical reagents (e.g., lime, phosphates) to induce the development of insoluble heavy metal precipitates, such as metal hydroxides, carbonates, or phosphates (equation 1)

$Pb^{2+}+PO4^{3} \rightarrow Pb_3\,(PO4)_2$ (insoluble phosphate) (1)

Adsorption is the binding of heavy metals to the surface of adsorbents such as clay minerals, biochar, activated carbon, or natural zeolites via ion exchange or surface complexation. Complexation is the use of organic or inorganic ligands, such as humic acids or chelating agents, to build stable complexes with heavy metals thus lowering their solubility and mobility. Encapsulation is the process of enclosing polluted soils or garbage in solid matrices, such as cement or polymers, to trap heavy metals and prevent leaching.

pH modification is the process of raising the pH to an alkaline level in order to limit metal solubility. For example, raising the pH with lime decreases the solubility of metals such as lead, cadmium, and zinc by creating hydroxide precipitates.

Microbial activity is the employment of microorganisms to induce immobilization by changing the redox state of metals or creating biosurfactants that bind metals.

Immobilizing agents include phosphates, biochar, zeolites, lime, fly ash, and cement. (Yutong et al., 2023; Giwa et al., 2013), and animal bone (Eze et al., 2024). However, there is little literature on the research of the immobilization mechanism of the supplements for metal remediation in soils. This study investigated the immobilization principle via adsorption, precipitation, and pH changes under adsorption and desorption with bone ash as an amendment.

The sorption mechanism was investigated using pseudo first order, pseudo second order, and parabolic diffusion, whereas desorption kinetics were addressed using first order, second order, and Simple Elovich Models. These models were chosen for their utility in understanding and optimizing the kinetics of immobilization processes, as they provide insights into the mechanisms driving sorption and desorption and aid in the design of more efficient systems (Yahaya et al., 2024).

Pseudo first-order reaction refers to a chemical reaction that appears to be a first-order reaction due to particular conditions or simplifications, but is actually a higher-order reaction. This is common when the concentration of one or more reactants is high or constant.

The rate law is given as Rate = k [A]m [B]n (2)

Where m and n represent the reaction orders with respect to the reactants A and B.

The pseudo-second-order reaction model is often used to represent second-order reactions that have been reduced to focus on certain reactants or restrictions. The pseudo-second-order model implies that the rate of reaction is proportional to the square of the concentration of the limiting reactant or to the product of two reactant concentrations (eqn 2). However, in pseudo-second-order reactions, one of the reactants is excessive or constant, simplifying the analysis.

The rate law is given as dtdqt = k2 (qe−qt) 2 (3)

qt = amount of adsorbate adsorbed at time t (mg/g),

qe = amount of adsorbate adsorbed at equilibrium (mg/g),

k2 = pseudo-second-order rate constant. (g /(mg·min)

Parabolic diffusion is a type of mass transfer process in which the rate of diffusion reduces over time when concentration gradients decrease or other limiting factors emerge. This behavior is frequently observed in systems where the diffusion mechanism is controlled by physical or chemical restrictions, such as reaction-diffusion processes, porous media, or high-temperature oxidation. Diffusion follows a parabolic rate law

$x^2 = kt$ (4)

Where x is the thickness of the diffused layer. K is the parabolic rate constant, and t represents time.

First-order kinetic model refers to a sort of reaction rate that is proportional to the concentration of just one ingredient. In first-order reactions, the rate of reaction is directly proportional to the concentration of the reactant, implying that as the reactant concentration falls, so does the rate of reaction (Atkins and de Paula, 2010). First-order kinetics is expressed mathematically as follows:

Rate = k [A]. (5)

Where Rate is the reaction rate, k is the rate constant (which is particular to the reaction at a given temperature), and [A] is the reactant concentration.

A second-order kinetic model explains a reaction in which the rate is proportional to the concentration of one or more reactants increased to a power of two. This means that the reaction rate is proportional to either the square of a single reactant's concentration or the product of two reactant concentrations.

Rate = k [A] [B] (6)

The Elovich equation is a semi-empirical kinetic model that is always employed to describe the adsorption of gases and solutes on solid surfaces, especially in heterogeneous chemisorption and adsorption dynamics. It is useful when the adsorption rate gradually declines due to surface saturation or interactions between adsorbed molecules. The equation is

Qt = β1ln (1+αβt) (7)

Where qt is the amount of adsorbate adsorbed at time t,

t is time in minutes,

α is the initial adsorption rate (mg/g·min), and

β is a surface coverage-related desorption constant (g/mg).

MATERIALS AND METHODS

Collection and preparation of Soil and Bone Samples

The lateritic soil was collected from Emene. Lat. 6.488203 and long. 7.57282 in Enugu East local Government Area of Enugu State. A soil auger was used to gather the soil at a depth of 0-20 cm. it was air dried and passed through a 2mm (10 mesh) stainless steel sieve and then stored in a sealed container for analysis and future experiments,

The three immobilizing agents were collected as follows:

The cow and pig bone were collected at Oye, Emene Central Abattoir in Enugu East L.G.A while the horse bone was collected at Obollo -Afor main market Abattoir in Udenu L.G.A. of Enugu State .In order eliminate surface contaminants, it was rinsed with water multiple times, sliced into pieces with a cutlass, and then rewashed again and again. The bones were dried in an oven set at 80 degrees Celsius after being cleaned with de-ionized water. After being ground using a motorized crusher, the dry bones were moved to a furnace and heated to 700 ºC for five (5) hours, according to Abdulrahman et al. (2015).

Analysis of the bone sample

A Scanning Electron microscope (SEM) and an Atomic Absorption Spectrophotometer (AAS) 340 N were used to determine the surface morphology, and metal concentration, respectively.

Sorption Kinetic Experiment

Ping et al. (2016), Aishat et al. (2017), and Choko (2017) detailed the steps used to conduct the sorption kinetics. 3% and 5% of the bone samples were used in the experiment. The following solutions were added: 25 ml of DTPA-TEA (conc. 0.001molL-1) and 50 mg/L of Cd. For one hour, the mixture was stirred and allowed to acclimate to ambient temperature (27 degrees Celsius). Whatman 42 filter paper was used for the separation process. The concentration of heavy metal cations in each filtrate was measured using an Atomic Absorption Spectrophotometer (AAS). Soil and adsorbent were thought to have soaked up the difference between the starting and ending concentrations of the metal in solution during the reaction time. 1,2,3,4,5,6,8,24,72,168,336, 504, and 672 hours were the intervals between repetitions of the experiment. It was from this relation that the amount sobbed was determined.

$$q_{sorbbed} = \frac{(C_{o-}\ C_{1)}\ V}{W_1} \qquad (8)$$

Where :

V = Volume of the electrolyte (ml),

C_o = Initial metal concentration (mg/L).

C_t = Concentration of the metal at time (hrs.),

W_1 = Weight of soil plus amendment (g)

Desorption Kinetic Experiment

Aisha et al. (2017), and Choko (2017) detailed the process of heavy metal desorption from soils using an adsorbent. The 25 ml of DTPA-TEA solution with a concentration of 0.001molL-1 was added to the heavy metal enriched soil residue that had been separated from the supernatant solution containing the greatest concentration of Cd (50 mg/L) during the sorption experiment. At 27 degrees Celsius, the suspension was shook for one hour. The amount of Cd that was absorbed into the solution was measured using an atomic absorption spectrophotometer, specifically a Model AA430 N, after centrifugation at 800 rpm for 15 minutes. At 2, 4, 6, 8, 24, 72, 168, 504, 672, and 1344 hours of contact time, the desorption procedure was repeated (Choko, 2017).

The amount of metal that is adsorbed is provided by

$$q_{des} = \frac{C_1\ x\ V}{w_1} \qquad (9)$$

$$\%\ Desorbed = \frac{q_{desored}}{Q_e absorb}\ x\ \frac{100}{1} \qquad (10)$$

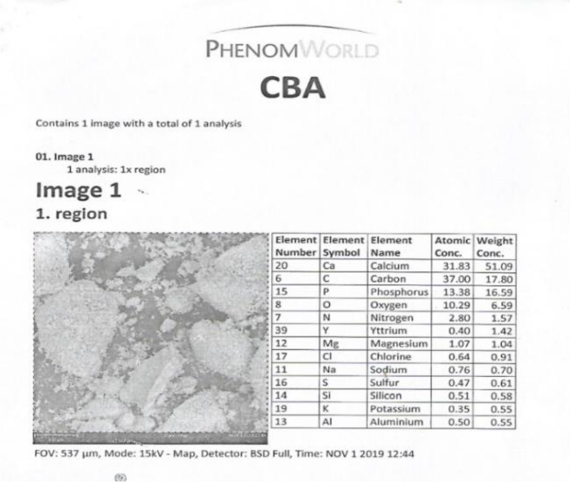
Where C_1 =final conc. in mg/L;

V = Vol. of the dissolved liquid (ml);

W_1 = Weight of the soil mixture (g);

q_{des} = quality desorbed

RESULTS AND DISCUSSION



CBA

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01. Image 1
1 analysis: 1x region

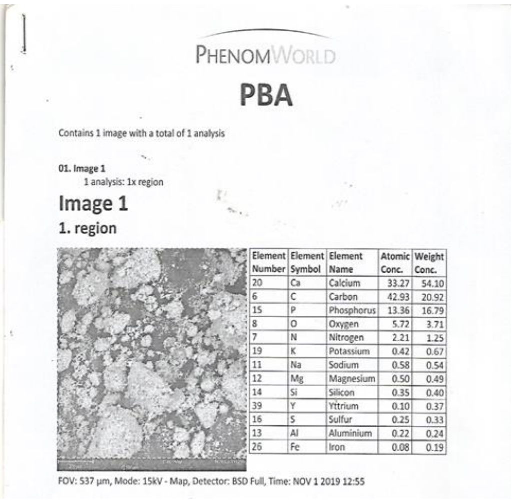
Image 1

1. region

Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
20	Ca	Calcium	31.83	51.09
6	C	Carbon	37.00	17.80
15	P	Phosphorus	13.38	16.59
8	O	Oxygen	10.29	6.59
7	N	Nitrogen	2.80	1.57
39	Y	Yttrium	0.40	1.42
12	Mg	Magnesium	1.07	1.04
17	Cl	Chlorine	0.64	0.91
11	Na	Sodium	0.76	0.70
16	S	Sulfur	0.47	0.61
14	Si	Silicon	0.51	0.58
19	K	Potassium	0.35	0.55
13	Al	Aluminium	0.50	0.55

FOV: 537 µm, Mode: 15kV - Map, Detector: BSD Full, Time: NOV 1 2019 12:44

Figure 1: SEM Image of CBA



PBA

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01. Image 1
1 analysis: 1x region

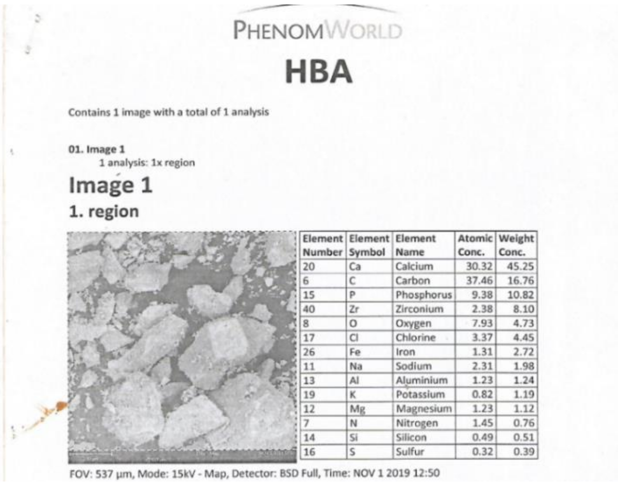
Image 1

1. region

Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
20	Ca	Calcium	33.27	54.10
6	C	Carbon	42.93	20.92
15	P	Phosphorus	13.36	16.79
8	O	Oxygen	5.72	3.71
7	N	Nitrogen	2.21	1.25
19	K	Potassium	0.42	0.67
11	Na	Sodium	0.58	0.54
12	Mg	Magnesium	0.50	0.49
14	Si	Silicon	0.35	0.40
39	Y	Yttrium	0.10	0.37
16	S	Sulfur	0.25	0.33
13	Al	Aluminium	0.22	0.24
26	Fe	Iron	0.08	0.19

FOV: 537 µm, Mode: 15kV - Map, Detector: BSD Full, Time: NOV 1 2019 12:55

Figure 2: SEM Image of PBA



HBA

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01. Image 1
1 analysis: 1x region

Image 1

1. region

Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
20	Ca	Calcium	30.32	45.25
6	C	Carbon	37.46	16.76
15	P	Phosphorus	9.38	10.82
40	Zr	Zirconium	2.38	8.10
8	O	Oxygen	7.93	4.73
17	Cl	Chlorine	3.37	4.45
26	Fe	Iron	1.31	2.72
11	Na	Sodium	2.31	1.98
13	Al	Aluminium	1.23	1.24
19	K	Potassium	0.82	1.19
12	Mg	Magnesium	1.23	1.12
7	N	Nitrogen	1.45	0.76
14	Si	Silicon	0.49	0.51
16	S	Sulfur	0.32	0.39

FOV: 537 µm, Mode: 15kV - Map, Detector: BSD Full, Time: NOV 1 2019 12:50

Figure 3: SEM Image of HBA

The morphologies of the amendments - cow bone ash (CBA), pig bone ash (PBA), and horse bone ash (HBA) was studied using scanning electron microscope (Figs 1-3). In CBA, the content of calcium and phosphorus is 31.83% and 11.38%, in PBA, it is 30.32 % and 11.38%, and in HBA, it is 11.36% and 9.38%. Therefore, it is quite evident that calcium phosphate is the main component of all the changes.

The immobilization of cadmium in soils containing phosphate-bearing minerals has been demonstrated in studies conducted by Hodson et al., (2000), and Joonki (2005) found that the amount of phosphorous determines the degree of immobilization, and the micrograph shows that cow bone ash has the highest quantity of phosphorous, followed by pig bone ash while the lowest is horse bone ash. Consequently, the immobilization of cadmium will follow that order.

Sorption Studies

Fig. 4 represents the sorption of cadmium by animal bones from contaminated laterite soil. The amount of Cd adsorbed by the amendments – CBA: 3%, 5%; PBA: 3%, 5% and HBA: 3%, 5% in the soil vary with contact time. In all sorption experiment studied, the quantity of Cd sobbed with 5 % of the amendments was higher than that with 3 %. This is because the higher the amount of animal bones (calcium phosphate) which is insoluble in water, the more the propensity to create additional binding sites for the cadmium metal to adsorb thus the lower the metal mobility and bioavailability (Hechelski et al., 2019). It was observed that the quantity of cadmium sobbed increased sharply during the early part of the experiment. This could be due to the increased binding sites but later slowed down. This could be as a result of the binding sites being saturated. This two-stage time dependent sorption behavior is similar to the results obtained by (Chokor, 2017).

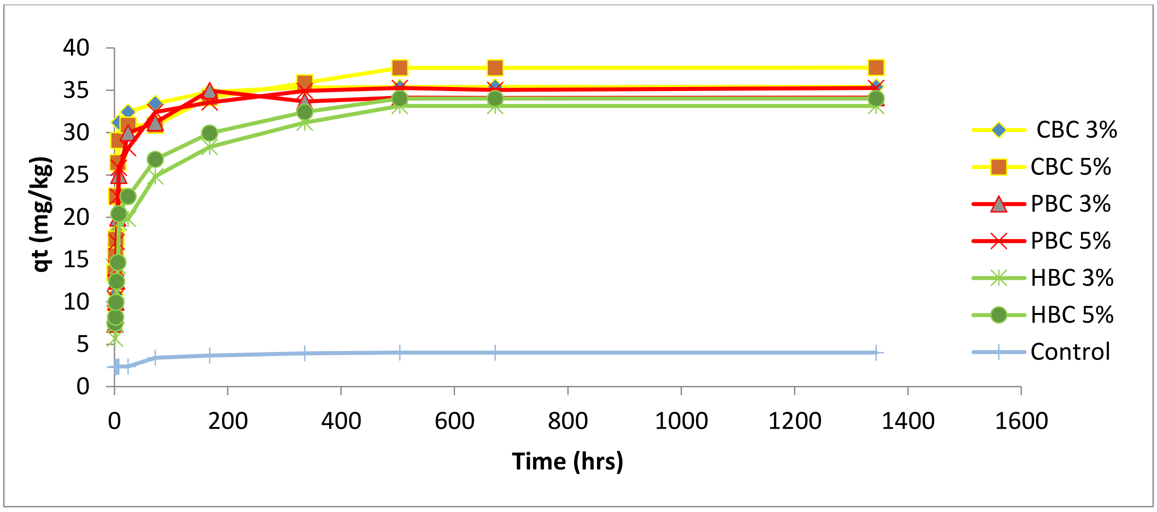

Figure 4: Kinetics of cadmium adsorption in Lateritic soil

Sorption Kinetic Models

The pseudo First Order, pseudo second order, and parabolic diffusion models were demonstrated for the sorption of Cd from laterite soil using bone amendments and the kinetic parameters were evaluated as presented in Table 1, 2 and three respectively.

Table 1: Summary table for the pseudo first order sorption of Cd

Soil	Model parameters	Control	3% CBA	5% CBA	3% PBA	5% PBA	3% HBA	5% HBA
Laterite	qm/kg	0.9949	0.9980	0.9913	0.9927	0.9987	0.9986	0.9961
	K_1/hr^{-1}	0.0051	0.0040	0.0087	0.0073	0.0013	0.0014	0.0039
	R^2	0.7135	0.722	0.8295	0.7214	0.5714	0.4619	0.7107

Table 2: Summary table for the pseudo second order sorption of Cd

Soil	Model parameters	Control	3% CBA	5% CBA	3% PBA	5% PBA	3% HBA	5% HBA
Laterite	qm/kg	2.529	30.487	32.573	29.1545	30.395	27.027	28.490
	K_2/hr^{-1}	0.003	0.0134	0.0106	0.0183	0.0113	0.0399	0.0336
	R^2	1	0.9997	0.9995	0.9994	0.9998	0.9948	0.9946

Table 3: Summary table for the parabolic diffusion of Cd

Soil	Model parameters	Control	3% CBA	5% CBA	3% PBA	5% PBA	3% HBA	5% HBA
Laterite	Kid	0.1016	1.4048	1.3496	1.3929	1.772	0.7873	0.8526
	R^2	0.4546	0.6871	0.6728	0.655	0.6936	0.5116	0.5728
	C	2.8722	57.525	65.741	53.214	54.637	55.701	58.289

The pseudo-first order rate constant k_1 ranges from 001 – 0087 hr^{-1}, this is quite low. The maximum quantity sobbed is also low. Mohammed and Mohammed, (2010) pointed out that the values of the rate constant and the quantity sobbed must be high for the sorption process to follow the kinetic model. The R^2 values, which indicates how close the data are to the fitted regression, are less than 0.81which shows that the model did not explain large proportion of the variance. Yahaya et al., (2024) pointed out that R^2 close to 1 is an indication of a good model. Therefore, the data for the sorption of Cd from Lateritic soils by Cow Bone Ash, (CBA), Pig Bone Ash (PBA) and Horse Born Ash (HBA) did not fit into the pseudo first order kinetic model. Some works that followed this trend include Cheng and Hseu (2002); Kumar and Bandyopadhyay (2006); Nwabunne and Igbokwe (2008).

Pseudo Second-Order Kinetic Model

The pseudo-second order kinetic model was illustrated by plotting t/qe. Vs. t. The results of the calculations for the rate constants K_2, qm, and R^2 are shown in Table 2. As can be seen, the equilibrium sorption values for (qm) and the coefficient of correlation R^2, are also quite high, showing that HBA, PBA, and CBA may be satisfactorily explained by the pseudo second order model.

Parabolic Diffusion Dynamic model

Lateritic soil's Cd sorption mechanism toward CBA, PBA, and HBA was studied by means of intra-particle diffusion calculations. Adsorption of Cd by the amendments as proposed by this model is expected to vary proportionally with the square root of the contact period. This model is depicted by the graph of qt against t0.5, where kid is the slope and C is the constant. Table 4 displayed the model's parameters. From the table, the correlation coefficient values of R^2 and Kid are low indicating that the data does not conform to the parabolic diffusion model.

Desorption Kinetic Studies

A plot of qt against t was used to study the desorption mechanism of heavy metals in the soils (Figure 5). CBA, PBA and HBA desorbed cadmium from 50mg/kg to 4.815 mg/kg, 5.905 mg/kg and 7.895 mg/kg, respectively, after applications of the amendments at 3% and 5% rates as against the control sample desorption of 27.76 mg/kg. The reduction in the desorption of the metal by the animal bones is lower than the control because calcium phosphate forms stable metal complexes that prevents metals from being easily released back to the soil matrix (Tang and Wang 2013). It was observed that the desorption rate increased sharply at the early part of the experiment and later reduced as was the case during the sorption. In addition, the presence of iron oxide in lateritic soil increased the amount Fe – Mn bound and this in turn increased the retention of heavy metals (Jalili and Rosteri 2011, McNear et al. 2007, Kumpiene et al. 2008, and Chokor 2017).

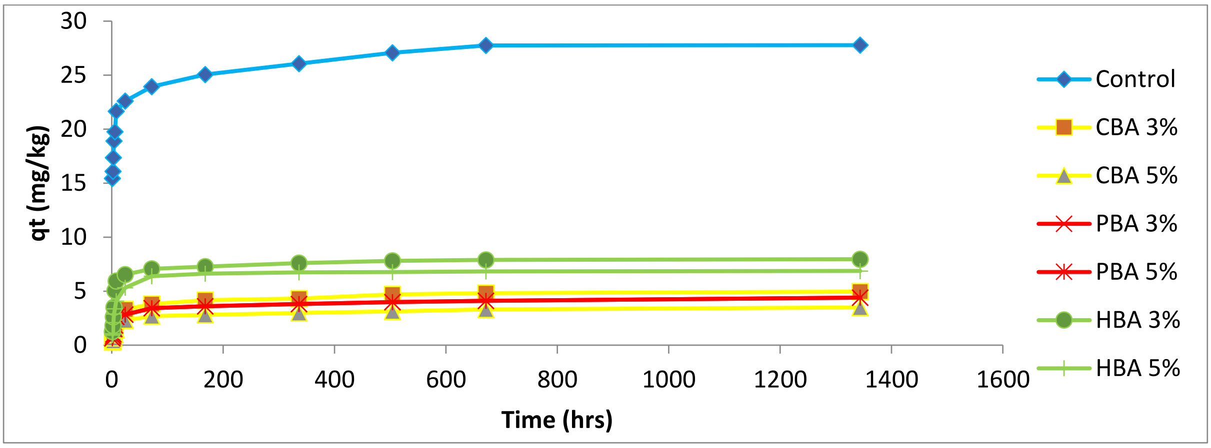

Figure 5: Kinetics of Cd Desorption in Laterite Soil

First order kinetic model

From the plot of Ln qt. against t, the first-order kinetic model for desorption of Cadmium by bio-sorbent materials was derived. The slope and correlation coefficient values were used to estimate the rate constants, while the intercept was used to obtain the amount of metal desorbed. The coefficient of determination and the standard error (SE) are less than 0.5 and more than 175 respectively (table 4). Thus, it can be inferred that the first order kinetic model was not satisfied by the Cd desorption via CBA, PBA, and HBA. One possible explanation is that the desorbing surfaces are not uniform. According to Fan et al. (2013), first-order reactions are characterized by weak bonding, such as the sort found on homogeneous desorbing surfaces. Here is the formula for calculating the standard error:

$$SE = \left[\frac{\sum(E - \mathrm{E1})^2}{n-2}\right]^{0.5} \qquad (6)$$

Second order kinetic Model

Where E and E^1 are the experimented and predicted or calculated amounts of metal released in soil at time it, respectively and n are the number of measurement

Table 4: Summary Table for the first order desorption of Cd

Soil	Model parameters	Control	3% CBA	5% CBA	3% PBA	5% PBA	3% HBA	5% HBA
Lateritic	Qdes	31.64	7.83	6.64	9.86	8.34	11.84	10.80
	K_1	0.000358	0.0001224	0.001406	0.00123	0.000995	0.000784	0.000934
	R^2	0.4090	0.34530	0.34948	0.38009	0.3514	0.2543	0.27187
	S.E	106.302	125.326	126.723	125.608	126.727	182.336	168.24

By plotting 1/q against t, the parameters of the second order kinetic model was obtained and the result presented in table 5. All of the amendments have extremely low coefficients of determination (R^2), ranging from 0.32 to 0.41, for desorption of Cd in the soil. The standard error (SE), however, is likewise quite significant. Consequently, Cd desorption did not follow a second-order mechanism, as seen by the high standard error and low regression coefficient values. Ghasemi-Fashei (2006) also found the same thing. There are numerous processes that influence desorption of metals in soils, including adsorption, desorption, complexation, precipitation, and ion exchange, which could explain why the desorption mechanism does not fit into the second order kinetic model (Saffari et al., 2015).

Table 5: Summary Table for the Second order desorption of Cd

Soil	Model parameters	Control	3% CBA	5% CBA	3% PBA	5% PBA	3% HBA	5% HBA
Lateritic	Qdes	19.734	1.249	0.690	1.6893	1.4577	3.2745	2.373
	K_2	1.623×10^{-5}	0.00068	0.00133	0.00050	0.00051	0.00020	0.00032
	R^2	0.413	0.211	0.217	0.263	0.225	0.164	0.168
	S.E	115.36	153.416	154.720	152.321	153.361	180.225	180.225

The simple Elovich Kinetic Model

Plotting qt. against ln t for the examination of data for Cd desorption into the soil matrix by CBA, PBA, and HBA showed that this model was applicable. The model's analysis was conducted using the constants α, β, R2, and SE, which are listed in table 6. Gupta and Babu (2006) state that the basic Elovich equation presupposes that the sorbent is active surfaces are heterogeneous, leading to varying chemisorption activation energies. Due to its low standard error (SE) and high R^2 value, the simple Elovich model is the best fit for describing the desorption process of these metals, as shown in the table.

Table 6: Summary Table for the Simple Elovich Kinetic Desorption of Cd

Soil	Model parameters	Control	3% CBA	5% CBA	3% PBA	5% PBA	3% HBA	5% HBA
Lateritic	$\propto$	38.286	21.426	19.266	25.36	23.142	28.027	26.440
	β	0.5826	0.8432	0.8650	0.7920	0.782	0.742	0.7033
	R^2	0.974	0.987	0.989	0.985	0.981	0.989	0.990
	S.E	26.05	4.111	4.362	4.521	4.444	4.322	4.732

CONCLUSION

Bones mostly consist of calcium phosphate, as shown by SEM studies, with CBA, PBA, and HBA being the order of increasing composition. Soil physicochemical testing has revealed the presence of iron oxide, which raises the soil's iron and manganese oxide fractions. Because of this, less of the metal that plants can absorb will be mobile. The precipitation of metal phosphate, caused by the calcium phosphate in the bone sample, reduces mobility due to the solubility of the compound. The heavy metal concentration of Laterite soil was cleaned up by applying animal bones to it, which effectively lowered the bioavailable Cd. Cow, pig, and horse bone ash, were found to be effective in immobilizing and remediating lateritic soil that was contaminated with Cd, according to the data analyzed in this experiment. In terms of immobilizing agent efficiency, CBA>PBA>HBA is the preferred order. The sorption and desorption processes which controls immobilization was best fitted by pseudo second order and Elovich kinetic models respectively.

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