Use of agro-waste (*Musa paradisiaca* peels) as a sustainable biosorbent for toxic metal ions removal from contaminated water

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A R T I C L E   I N F O

A B S T R A C T

A study of removal of heavy metal ions from heavy metal contaminated water using agro-waste was carried out with *Musa paradisiaca* peels as test adsorbent. The study was carried by adding known quantities of lead (II) ions and cadmium (II) ions each and respectively into specific volume of water and adding specific dose of the test adsorbent into the heavy metal ion solution, and the mixture was agitated for a specific period of time and then the concentration of the metal ion remaining in the solution was determined with Perkin Elmer Atomic absorption spectrophotometer model 2380. The effect of contact time, initial adsorbate concentration, adsorbent dose, pH and temperature were considered. From the effect of contact time results equilibrium concentration was established at 60minutes. The percentage removal of these metal ions studied, were all above 90%. Adsorption and percentage removal of Pb^{2+} and Cd^{2+} from their aqueous solutions were affected by change in initial metal ion concentration, adsorbent dose pH and temperature. Adsorption isotherm studies confirmed the adsorption of the metal ions on the test adsorbent with good mathematical fits into Langmuir and Freundlich adsorption isotherms. Regression correlation (R^2) values of the isotherm plots are all positive (>0.9), which suggests too, that the adsorption fitted into the isotherms considered.

Capsule Summary: Musa paradisiacal peels is discovered to be an efficient biosorbent for the removal of Pb^{2+} and Cd^{2+} from their aqueous solutions with removal efficiency > 90%.

INTRODUCTION

Heavy metals have hazardous impact on ecosystem including humans, animals and plants health. Therefore, the world health organization (WHO) and Environmental Protection Agency (EPA) have regulated the maximum acceptable discharge level in the environment and thus controlling the pollution level. According to US environmental Protection Agency (EPA), Agency for toxic substances and disease registry (ATSDR) and World Health Organization (WHO), The maximum acceptable concentrations recommended for zinc, copper, chromium, lead, nickel, manganese and iron are 3.0mg/L, 2.0mg/L, 0.05mg/L, 0.1mg/L, 0.02mg/L, 0.05-0.5mg/L and 0.1mg/L respectively (Ahmaruzzaman, 2011; ATSDR, 2012; EPA, 2016).

Most heavy metal ions are reported as priority pollutants, due to their mobility in natural water ecosystem and due to their toxicity (Demirbas, 2008). The problem associated with metal ions pollution is that they are not biodegradable and are highly persistent in the environment. Thus they can be accumulated in living tissues causing various diseases and disorders (Wan-Ngah and Hanafiah, 2008). Heavy metal toxicity can result in damage or reduced mental and control nervous functions, lower energy levels and damage blood composition, lungs, liver, kidney and other vital organs (Ahmaruzzam, 2011). It is thus necessary to remove heavy metal ions from contaminated or waste water before it can be discharged into the environment. In this respect, many physico-chemical methods have been developed for the removal of heavy metal ions from aqueous solution including; precipitation, evaporation, electro-deposition, ion exchange, membrane separation, coagulation etc (Rao and Ikram, 2011).

However, these methods have disadvantages such as secondary pollution, high cost, high energy input, and large quantities of chemical reagents or poor treatment efficiency at low metal concentration (Ding et al., 2012). It can be said that the conventional methods for metal ions removal from contaminated water are limited by technical and economic barriers especially when concentration of heavy metal ions in the water is low (< 100ppm) (Witek-Krowiak et al., 2011). Therefore the search and development of efficient and low-cost metal removal processes are of utmost importance.

The growing trend in biotechnology has resulted to the emergence of bio-adsorption as an alternative and sustainable strategy for treating contaminated water. (Flores-Gamica et al., 2013). Biosorption uses inexpensive biomaterials to sequester environmental pollutants from aqueous solutions by a wide range of physico-chemical mechanism, including ion exchange, chelation, complexation, physical adsorption, and surface micro precipitation. The biomaterials used in these processes are termed bio-adsorbent (Flores-Gamica et al., 2013).

Biosorption is a physical chemical process, simply defined as the removal of substance from solution by biological material. This is a property of both living and dead organisms (and their components) and has been heralded as a promising biotechnology because of its simplicity, analogous operation to conventional ion-exchange technology, apparent efficiency and availability of biomass and waste bio-products (Kelly-Vargas et al., 2012). Various waste biomaterials, microorganisms, bacteria, fungi, yeast and algae have been reported for the removal of heavy metal ions from aqueous solution (contaminated water) (Ghaedi et al., 2013). In this regard, besides treatment of contaminated waste water biosorption application proffers a sustainable means of waste management. One of the key sources of bio-adsorbents is Agro waste materials.

Agricultural waste represents a potential source for producing biosorbents as they have no prominent utilization. The use of agro waste to treat contaminated water may add value to agro-waste and eventually reduce the agro waste management problems around the world (Hossain et al., 2012). Agro-materials are composed of lignin and cellulose as major constituents and may also include other polar functional groups of lignin which include alcohol, aldehydes, phenolic and ether groups (Hossain et al., 2012). These groups are behind the chemistry of biosorption or removal of heavy metal ions from contaminated water. During the past few years, several research articles were published reporting the successful use of different kinds of agricultural waste in the removal of toxic metal ions from contaminated water. These led to the interest in this study on the use of *Musa paradisca* (plantain) peels in the removal of heavy metal ions from heavy metal contaminated water.

Botanical description of the test biosorbent (*Musa paradisiaca*)

Plantain plant is a crop plant with green leaves and herbaceous stem. The fruit which is green is typically larger than the common banana and is cherished by many people in Abia state, Nigeria (Mohammed and selena, 2012). Plantain (*Musa paradisiaca*) belongs to the Eumusa of the genus *Musa*, family (Musaceae) closely related to the common banana (*Musa sapientum*) (Andrede et al., 2008). The plantain is a tall plant with a conical false '' trunk'' formed by the leaf sheaths of its spiraling arranged leaves, which are 1.5 3m long and about 0.5m wide. Rodrigues *et al.,* (2005), highlighted that, plantain species are classified either by bunch floral size or size of the pseudostem (false stem). He classified bunch type and floral size based on their characteristics into; French plantain, French horn plantain, horn plantain, false horn plantain and further classified the size of the pseudostem into; giant plantain, medium plantain, and small plantain.

A plantain peel is the outer covering of the plantain fruit. Plantains, whether eaten raw or cooked, are popular fruits consumed worldwide, and its Global production has risen by nearly 60% over the last 30 years to 37 million metric tons. Uganda is by far the largest producer with 9.6 million metric tons compared with the second largest producer, Ghana, with 3.6 million metric tons. Other important producers are Rwanda, Nigeria, Cameroon, and Colombia. Plantain production in Africa has nearly doubled over the last 30 years to 27 million metric tons. There is a significant amount of plantain peel waste being generated as well.

To convert the waste generated from plantain production (in form of plantain peels) into more value added product we suggest that, it can be used as biosorbent in removing toxic metals from heavy metals contaminated water which will yield sustainable and eco-friendly way of treating toxic metal contaminated effluent and waste management. This study investigates the use of *Musa paradisiaca* peels as sustainable biosorbent in toxic metal removal from metal ions solution.

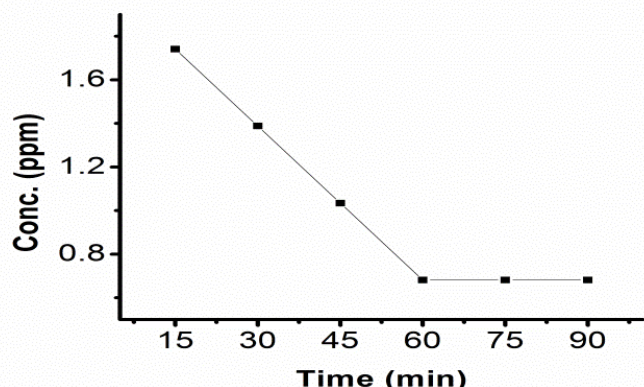## MATERIAL AND METHODS

Sample collection and identification

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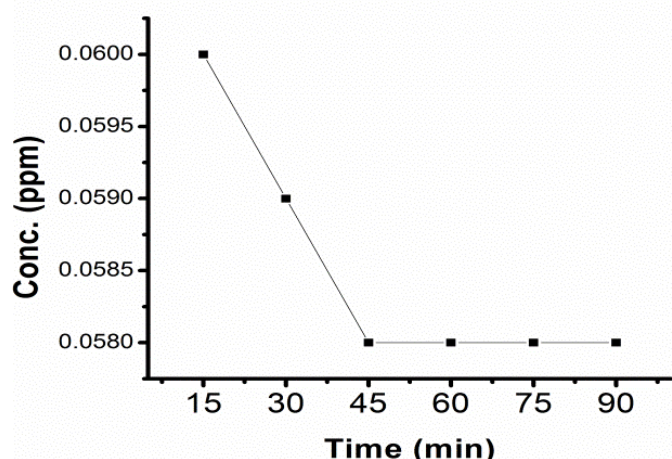**Fig. 1:** Effect of contact time graph on Cd adsorption

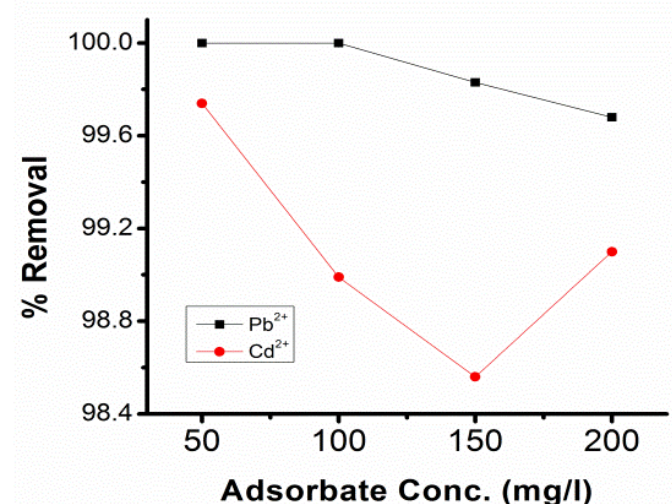**Fig. 2:** Effect of contact time graph on adsorption of Pd

Fig. 3: Plot of percentage removal of Pb^{2+} and Cd^{2+} against adsorbate initial concentration in solution

The test sample *Musa paradisiaca* peels were collected and sorted from west beans around Michael Okpara University of Agriculture Umudike (MOUAU) environs. The collected sample was identified in Forestry Department College of Natural resources and environmental management Michael Okpara University of Agriculture Umudike Abia state Nigeria.

Adsorbent sample preparation

The Agro-waste was brought to chemistry laboratory, Chemistry Department, College Of Physical And Applied Sciences, MOUAU. In the laboratory the sample was spread on laboratory work bench and inspected for extraneous material such as insect larva. Thereafter the sample was washed and cut into pieces to increase the surface area for easy drying. Then the sample was oven dried at 60°C until it became dried and then dry milled to powdery form, followed by sieving through a 0.63 to 1.6mm.

The resultant fine sample was collected and soaked in dilute nitric acid (2%) solution, filtered and washed repeatedly in distilled water and oven dried at 60°C. Thereafter, the sample was rewashed in 1% NaHCO$_3$ to remove any remaining acid, and then washed with distilled water until pH of 6.0 was obtained. Subsequently the washed sample was dried at 60°C for 5 hours and stored for adsorption analysis.

Adsorbate solution preparation

Known qualities of cadmium chloride and lead acetate were separately dissolved in distilled de-ionized water in one liter volumetric flask and made up to mark with distilled de-ionized water to form stock solution. From the resultant metal ion stock solutions test solutions of 50mg\l, 100mg/l, 150mg/l and 200mg/l were obtained using dilution formula in equation 1.

$$C_1 V_1 = C_2 V_2 \qquad (1)$$

Where C$_1$ is initial concentration of the metal ion stock solution, V$_1$ volume taken from the stock solution, C$_2$ is the desired concentration of the metal ion concentration to be studied. C$_2$ is the volume of the desired metal ion concentration to be studied.

Biosorption experiment

Biosorption experiment with the processed sample was conducted to investigate the parametric effects of adsorbate concentration, pH, temperature and adsorbent dose on the adsorption and removal of Cd^{2+} and Pb^{2+} from metal ion contaminated water. 50ml of each metal ion concentration was taken and put into 100ml crew cap conical flask, and specific dose of adsorbent was added at constant pH of 6.0. The mixture was agitated at a speed of 200rmin^{-1} in a thermostatic shaker bath at 30°C for a specific period of contact time. The resultant solution was filtered through 0.45μm membrane filter paper. The experiment was repeated at various contact time to establish equilibrium point.
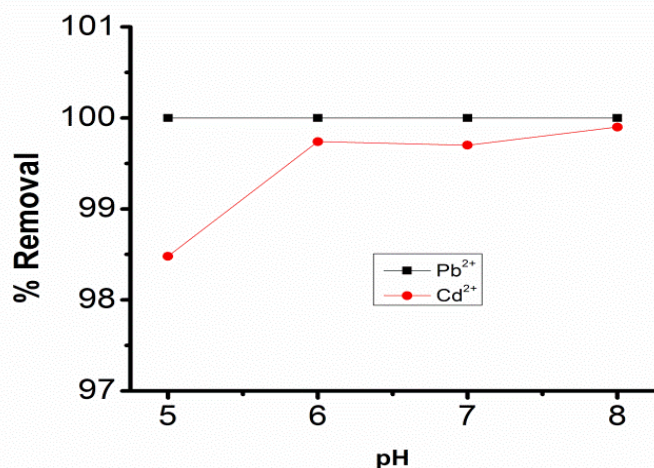
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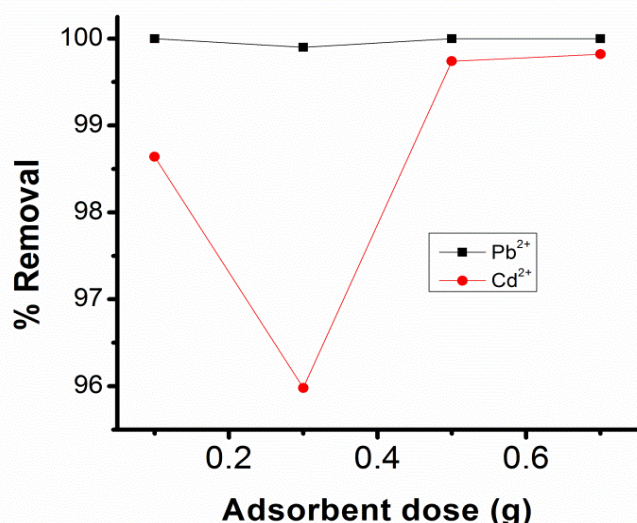
Fig. 4: Plot of Percentage removal of Pb^{2+} and Cd^{2+} against change in pH

Fig. 5: Plot of percentage removal of Pd^{2+} and Cd^{2+} against adsorbent dose
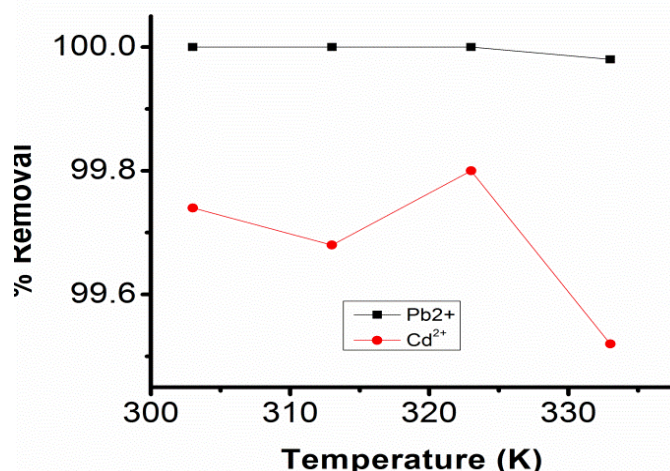

Fig. 6: Plot of percentage removal of Pb^{2+} and Cd^{2+} against temperature

The amount of metal ions in the aqueous phase was determined by Perkin Elmer Atomic Adsorption Spectrophotometer model 2380. The amount of each metal ion adsorbed was determined by subtracting the final concentration from the initial concentration. The percentage removal of the metal ions in solution by the test adsorbent was calculated using the relation in equation 2.

$$Rem(\%) = \left(\frac{C_0 - C_1}{C_0}\right) X\ 100 \qquad (2)$$

Where, Rem (%) is percentage removal, C_0 is initial concentration of the metal ion before treatment and C_1 is the final concentration of the solution after treatment. The experiment was repeated severally varying pH from range of 5.0 to 8.0 while other parameters were kept constant, varying temperature from 303K to 333K while other parameters were kept constant, varying adsorbent dose from 0.1g to 0.7g while other parameters were kept constant, and varying adsorbate initial concentration from 50mg/l to 200mg/l, while other parameter were kept constant.

RESULTS AND DISCUSSION

Effect of contact time

Influence of contact time on adsorption of heavy metal ions (Pb^{2+} and Cd^{2+}) on test adsorbent (*Musa paradisiaca*) as graphically represented in Figs. 1 and 2, were investigated in the time range of 15 to 90 minutes. The maximum rate of removal occurred within 45 minutes for Pb, and 60 minutes for cadmium. Above these times for each of these metals adsorption, the adsorption rate was observed to remain unchanged, which established that the system has reached equilibrium point.

Effect of initial adsorbate concentration

The initial metal concentration is an important factor to be considered in adsorption experiments. The rate of adsorption of an adsorbent for a metal ion dependent on the concentration of metal ion present in solution. The effect of initial Pb^{2+} and Cd^{2+} concentration on their adsorption/removal by *Musa paradisiaca* peels from their aqueous solutions is graphically represented Fig. 3.

Observations from the graphical representations in the figure 3 reveal, that increase in adsorbate concentration from 50mg/l to 100mg/l did not have a significant change on the percentage removal of Pb^{2+}, but when the metal concentration in solution was increase to 150 mg/l and 200mg/l the percentage removal of Pb^{2+} from its aqueous solution decreased instead of increasing as expected. In the case of Cd^{2+} its percentage removal from aqueous solution by the test adsorbent decreased as initial concentration of cadmium in the solution increase from 50 mg/l through 150 mg/l, but when the metal concentration increased from 150 mg/l to 200 mg/l the percentage removal increased. As a rule, increasing the initial metal concentration results in an increase in the biosorption capacity because it provides a driving force to overcome mass transfer resistance between
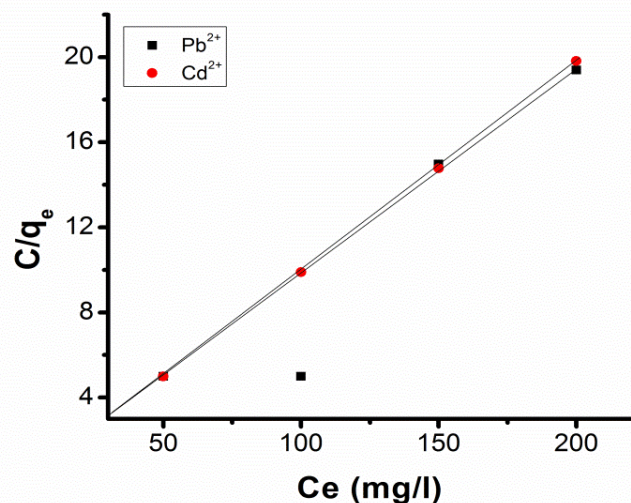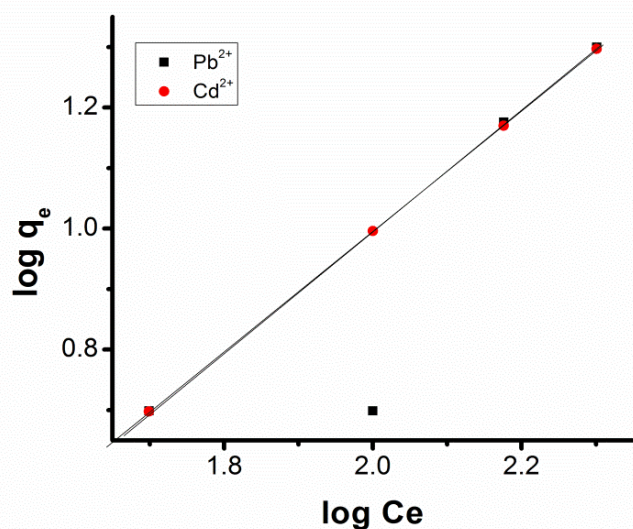
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Fig. 7: Langmuir adsorption isotherm

Fig. 8: Freundlich adsorption isotherm

the biosorbent and biosorption medium.[17]. It is generally agreed that the biosorption capacity increases as the initial metal ion concentration in the solution increases, whereas the metal removal percentage decreases by increasing the metal ion initial concentration (Pahlavanzadeh et al., 2010; Abdel-Ghani et al., 2013).

Effect of pH

A Study of the effect of pH experiments were carried out in the pH range of 5-8 for pb^{2+} and Cd^{2+} with the test adsorbent

Musa paradisica peels. The results of effect of pH on the percentage removal of the metal ions studied are graphically represented in Fig. 4. Observations from the graph, show that the removal of lead (II) ion were not significantly affected by change in pH, but cadmium (II) ion removal rate increased with increase in initial pH of metal ion solution and maximum value was reached at pH of 8. The increase in percentage removal and adsorption as the solution becomes more alkaline can be explained based on the competition between Cd^{2+} and H$_3$O$^+$ on adsorption sites on *Musa paradisiaca* peels.

At low pH, the surface of biosorbent would also be surrounded by excess H$_3$O$^+$ which decreased the Cd^{2+} interaction with biding sites of *Musa paradisiaca* peel by greater repulsive forces, resulting in low level of adsorbed Cd^{2+}. With an increase in pH, more ligands with negative charges can be expected to be exposed, and this would attract more positively charged Cd^{2+} for binding. (Li et al., 2012).

Effect of adsorbent dose

The results of the percentage removal of metal ion at different doses of *Musa paradisiaca* peels as graphically represented in figure 5 show that, as the *Musa paradisiaca* peels powder concentration increased from 0.1g to 0.7g, the removal efficiency of Pb^{2+} increased slightly, but removal efficiency of Cd^{2+} at first decreased as the adsorbent dose increased from 0.1g to 0.3g, then increased as the adsorbent dose was further increased from 0.3g to 0.5g and 0.7g. The increased in percentage removal as the adsorbent dose increases is due to the greater availability of active sites on the adsorbent at higher concentration of the adsorbent, thus making easier penetration of the metal ions to the sorption sites. (Sary et al., 2011).

Effect of temperature

The temperature effects on the percentage removal of heavy metal ions by the absorbent studied are graphically represented in Fig. 6. Biosorption and removal of heavy metal ions were carried out in different temperatures such as 303k, 313k, 323k and 333k, while other parameters were kept constant. Observations reveal that percentage removal increased with rise in temperature from 313k to 323k for cadmium adsorption. As the temperature increases, the rate of adsorbate diffusion across the external boundary layer and in the internal pores of the adsorbate particles which decreases the liquid viscosity with increase in temperature resulting to more adsorption of the absorbate molecules on the adsorbent surface (Kautukeyan et al., 2007; Hossain et al., 2012).

On the other hand, change in temperature from 303K to 313K and 323K to 333K for cadmium and 323K to 333K for lead there were decreases in percentage removal or absorption rate. This may be attributed to increase in temperature which increases the kinetic energy of the metal ions and thus, weakening the forces of attraction between the metal ions and the adsorbent (Anuear et al., 2010).

Adsorption isotherms

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Table 1: Estimated equilibrium parameters of Langmuir isotherm

Metal ion	R_L (1/L)	B (L/mg)	q_m (mg/g)	R^2
Pb^{2+}	0.01	2	10	0.95
Cd^{2+}	0.01	2	10	0.99

Table 2: Freundlich isotherms for the adsorption of Cd^{2+} and Pb^{2+} sorption onto *Musa paradisiaca*

Metal ions	n_f (L/g)	K_f (mg/g)	R^2
Pb^{2+}	1	0.1	0.88
Cd^{2+}	1	0.1	1

The equilibrium relationships between adsorbent and adsorbate are best explained by sorption isotherms (Munagapati et al., 2010). The biosorption isotherms describe the relationship between the mass of the adsorbed component per biosorbent mass and the concentration of this component in the solution. Determination of equilibrium parameters provides important information that allows for future design of adsorption systems (Witek-Krowiak et al., 2011). Biosorption isotherms are characterized by definite parameters, which express the surface properties and affinity of the biosorbent and can also be used to compare the biosorptive capacities of the biosorbent for different pollutants (Tuzen and Sary, 2010; Bulgariu and Bulgariu, 2012).

Several equilibrium isotherm models can be employed to fit the biosorption experimental data in order to study the nature of adsorption process. These include two parameters isotherms such as the Langmuir, Freundlich, Temkin, Dubinin-Radushkevich and three parameters isotherms such as the Sips and Redlich.Peterson. Two isotherm models were employed in this study to fit the adsorption data, the isotherm models are Langmuir and Freundlich adsorption isotherms.

Langmuir isotherm

The Langmuir isotherm model assumes a surface with homogeneous binding sites, equivalent sorption energies, and no interaction between adsorbed species. (Gupta et al., 2010). In this model once a site is filled, no further sorption can take place at that site. As such, the surface will eventually reach a saturation point where the maximum adsorption of the surface will be achieved (Farhan and Al-Dujaili, 2013). The Langmuir equation developed by Irving Langmuir in 1916 is represented in equation 3.

$$\frac{C_{eq}}{Q_{eq}} = \frac{1}{bQ_{max}} + \frac{C_{eq}}{Q_{max}} \tag{3}$$

where Q_{max} (mg/g) is the maximum amount of metal ion per unit weight of biosorbent to form a complete monolayer on the surface bound at high C_{eq}, and b (l/ mg) is the Langmuir constant related to the energy of adsorption. The Q_{max} and b can be determined from the linear plot of C_{eq}/Q_{eq} versus C_{eq}

(Abdel-Ghani et al., 2009). Figure 7 is a plot of C_{eq}/Q_{eq} against Ceq with intercept of $1/bQ_{max}$ and a slope of $1/Q_{max}$. Observation shows a linear relation which indicates that the adsorptions of Pb^{2+} and Cd^{2+} fitted into Langmuir isotherm.

The essential characteristic of the Langmuir isotherm maybe expressed in terms of dimensionless separation parameter R_L expressed in equation 4, which is indicative of the isotherm shape that predicts whether an adsorption system is favourable or unfavourable (Abdel-Ghani and El-Chaghaby, 2014).

$$R_L = \frac{1}{1+bC_o} \tag{4}$$

The adsorption process as a function of R_L may be described as: Unfavorable when $R_L > 1$; Linear when $R_L = 1$; Favorable when $0 < R_L < 1$; Irreversible when $R_L = 0$ (Abdel-Ghani and El-Chaghaby, 2014).

The results of R_L of Pb^{2+} and Cd^{2+} adsorption by the test adsorbent are presented in Table 1, and from the results the adsorption of the metal ions to the test adsorbent is deduced to be favourable. The regression correlation coefficient (R^2) values are > 0.9 which indicates a good mathematical fit.

Freundlich isotherm

The Freundlich isotherm model applies to adsorption on heterogeneous surfaces with the interaction between adsorbed molecules. The applications of the Freundlich equation also suggest that, sorption energy exponentially decreases on completion of the sorption centers of an adsorbent (Farhan and Al-Dujaili, 2013). The empirical Freundlich equation (Freundlich, 1906), based on sorption on a heterogeneous surface is given as shown in equation 5.

$$logQ = logK_f + \frac{logCe}{n} \tag{5}$$

Where, K_f and n are the Freundlich constants characteristic of the system and are indicators of adsorption capacity and adsorption intensity, respectively (Abdel-Ghani et al., 2009). The Freundlich equilibrium constants are usually determined from the plots of log Qeq versus log Ceq. The n value indicates the degree of nonlinearity between solution concentration and adsorption as follows: if n = 1, then

adsorption is linear; if n < 1, then adsorption is a chemical process; if n >1, then adsorption is a physical process (Farhan and Al-Dujaili, 2013). The experimental result of n for both Pb^{2+} and Cd^{2+} are 1.0 which indicates linearity of the plot, as seen in figure 8. From the regression correlation coefficient (R^2) values that are regarded as a measure of the goodness of fit of experimental data on the isotherm's model (Table 2), the R^2values are 0.88 for lead and 1.0 for cadmium, indicating a very good mathematical fit.

CONCLUSIONS

From the results of the study it can be concluded that, the test adsorbent (*Musa paradisica* peels) efficiently and effectively removed lead (II) and cadmium (II) ions from aqueous solutions containing these metal ions each and respectively. The equilibrium concentration of these metal ions was established at 60minutes of adsorption. Percentage removal of Pb^{2+} and Cd^{2+} each and respectively were influenced by changes in parameters like, initial adsorbate concentration, adsorbent dose, pH and temperature of the system. Adsorption isotherm studies confirmed the adsorption of the metal ions on the test adsorbent with good mathematical fits into Langmuir and Freundlich adsorption isotherms. Regression correlation (R^2) values of the isotherm plots are all positive (>0.9), which suggests too, that the adsorption fitted into the isotherms considered.

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