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Research Article

Effect of the Operating Variables on the Adsorption of Lead and Chromium Ions on Plantain (*Musa paradisiaca*) Wastes

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Abstract

Background and Objectives: Industrial effluent often contain heavy metals which bio-accumulate and persist in the environment, thereby constituting health problems. Plantain (*Musa paradisiaca*) wastes, which are nuisance to the environment could be used to produce activated carbon that are of public health importance. This study assessed the effect of the operating variables on the adsorption of lead and cadmium ions on plantain (*Musa paradisiaca*) wastes in effluent from battery recycling plant. **Materials and Methods:** Plantain wastes were collected from a plantation, sun-dried, homogenized, carbonized and activated using phosphoric acid at 400°C to produce the adsorbents. Composite samples of acid-lead battery recycling effluent from the point of discharge into Ogunpa river in Ibadan, southwestern, Nigeria was subjected to physico-chemical (pH, conductivity, Total Dissolved (TDS)), (Lead (Pb) and Chromium (Cr)) analysis using standard methods. Adsorption capacity was studied at varied effects of pH (2-12) and adsorbent doses (0.1-2.0 g) with treatments by activated carbon from plantain ripe peel, plantain stalk while commercial activated carbon served as control. Batch experiment was used in determining the adsorption isotherms of the adsorbents. Data were analyzed using descriptive statistics, Langmuir and Freundlich models and compared with the national standards for industrial effluent. **Results:** Means of pH, conductivity, TDS, Pb and Cr of the effluent sample were: 2.0 ± 0.2 , $2164.7 \pm 0.6 \mu\text{s cm}^{-1}$, 895.0 ± 0.0 , 31.3 ± 0.0 and $13.5 \pm 0.0 \text{ mg L}^{-1}$, respectively. The highest quantities of Pb and Cr were removed at pH¹⁰ for Pb (94.97 and 68.02%). However, optimum dosage occurred at 1.5 g for Pb (96.61%) and 2.0 g for Cr (58.91%). The adsorbents showed satisfactory fits to the adsorption isotherm models. **Conclusion:** Adsorbents had ability to remove the lead and chromium from effluent generated from battery recycling plant with plantain ripe peel having the highest adsorption capacity. Treatment of effluent with plantain wastes should be encouraged in battery recycling plant to promote public health safety and enhance effective waste management.

Key words: Plantain waste, lead and chromium, adsorption capacity, battery recycling effluent

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

The geometric increase in the urbanization coupled with industrialization has brought about the generation of wastes different forms in the environment. These wastes have posed a lot of threat to both man and the environment. Pollution is the release of chemical, physical, biological or radioactive contaminants exceeding the threshold limit which adversely alter natural quality of the environment and consequently constitute deleterious effects on living organisms^{1,2}. Nigeria is urbanizing at an astonishing pace with increasing industrialization. Over 500 major processing industries are concentrated in the urban centres such as Lagos, Port-Harcourt, Sango Ota, Kaduna and Kano. It was observed most of the industries found in Nigeria dump their waste in their own backyard without concern for the environment, hence, the cases of water pollution, which are found in most of the major industrial cities of Nigeria. These wastes and their leachates mixed with hazardous wastes and found their way into the streams and rivers through network of open drains³. Wastewater containing high amount of heavy metals originated from household and industrial wastes leads to the production of the sewage sludge contaminated with the metals of environmental concern where they may contaminate soil, water and eventually also the food chain⁴. Heavy metals are of serious health implications because of their persistence and bio-accumulation potential in the environment and have ultimately had negative effects on health and environment. They do not degrade or nor destroyed, they generally do not breakdown into less harmful constituents. As trace elements, some are essential to maintain the metabolism of human body. However, at higher concentrations they can lead to poisoning⁵. Heavy metals commonly found in wastewater are Lead (Pb), Cadmium (Cd), Chromium (Cr), Copper (Cu), Nickel (Ni) and Zinc (Zn) and the metal concentrations are governed by the nature and the intensity of the industrial activity as well as the type of process employed during the treatment⁶.

Heavy metal contamination of the environment is a major problem especially for the growing cities in developing countries primarily due to uncontrolled pollution levels driven by causative factors like industrial growth and heavy increase in traffic using petroleum fuels⁷. Increasing industrialisation and urbanisation have been accompanied by the extraction and distribution of mineral substances from their natural deposits. Many of these are concentrated and undergo chemical changes through technical processes and finally pass, by way of effluent, sewage, dumps and dust, into the water, the earth and the air and thus into the food chain. The

dramatic increase in the demand for metals led to high anthropogenic emissions of heavy metals into the biosphere⁸. These metals become an environmental concern when their concentrations begin to affect adversely human health and the environment. The heavy metals essentially become contaminants in the environments because their rates of generation via anthropogenic cycles are more rapid relative to natural ones, the concentrations of the metals in discarded products are relatively high compared to those in the receiving environment and the chemical form (species) in which a metal is found in the receiving environmental system may render it more bioavailable^{6,5}.

Various methods such as ion exchange, adsorption and phytoremediation had been applied in the adsorption of heavy metals from waste water⁹. Adsorption technology is being used extensively for the removal of heavy metals from aqueous solutions because it is a cleaner, more efficient and cheap technology. Various agricultural wastes had been studied as an adsorbent for heavy metals in polluted water which include rice husk¹⁰, sugar bagasse¹¹ and kaolinite clay¹². These agricultural wastes which are generated in large quantities and which in some cases might become difficult to dispose have proved very effective in the adsorption of heavy metals in water under varying abiotic conditions. This study assessed the effect of the operating variables on the adsorption of lead and cadmium ions on plantain (*Musa paradisiaca*) wastes in effluent from battery recycling plant, in Ibadan, Oyo state, Nigeria.

MATERIALS AND METHODS

Collection of substrate and analysis: Acid-Lead battery recycling plant effluent from the point of discharge into Ogunpa Stream was collected and analyzed using the standard methods¹³. The concentration of the heavy metals in the effluent was determined using Atomic Absorption Spectrophotometer (Hanna C-100). Plantain wastes (ripe peel and fruit stalk) were collected in Oje market Of Ibadan in Oyo state, Nigeria, homogenised and analyzed for physicochemical parameters. The study was carried out in Ibadan, Oyo state, Nigeria between July and September, 2017.

Carbonization and activation of plantain wastes: The plantain waste samples were carbonized and activated by the two steps method^{14,15} in which 50.00 g of each raw ground plantain waste samples was prepared by drying in temperature of 400°C for 1 h under a closed system in a porcelain crucible and then cooled to room temperature. The

charcoal was subjected to H₃PO₄ activation and heated to optimize temperatures of 400°C and maintained at a constant temperature for 1 h before cooling. Ripe peel plantain activated carbon (PAC) and plantain fruit stalk activated carbon (SAC) were prepared. The commercial activated carbon (CAC) served as the control.

Adsorption studies: A series of batch experiment was carried to determine the adsorption isotherms of lead and chromium on the adsorbents. Batch sorption experiments were performed in which 50 mL of effluent was measured into each 250 mL conical flask and 1.0 g of each prepared activated carbon was added into each flask and agitated thoroughly at 200 rpm with an electric shaker for 90 min to attain equilibrium. The operating variables such as pH (2-12), contact time (30-150 min) and adsorbent dose (0.1-2.0 g) were used. The treated effluent was filtered through Whatman No. 1 filter paper and concentrations of lead and chromium were determined by Atomic Absorption Spectroscopy.

Adsorption isotherms and models: A series of batch experiments was carried out to determine the adsorption isotherms of lead and chromium ions on the adsorbents¹⁶. The amount of metal ion adsorbed (Q_e) was determined using a mass balance equation:

$$Q = \frac{(c_v - c_f) \times V}{M}$$

where, Q is the metal uptake (mg g⁻¹), C_v and C_f are the initial and final metal equilibrium concentration in the effluent sample (mg L⁻¹), respectively, M is the mass of the adsorbent (g) and V is the volume of the effluent sample (l).

Adsorption models: The isotherm data obtained was fitted using the Langmuir and Freundlich isotherms to describe equilibria^{17,18}:

- **Langmuir equations:** The linearized equation is described by:

$$Q_e = \frac{1}{Q_e} = \frac{1}{Q^\circ b C_e} + \frac{1}{Q^\circ}$$

where, Q_e is the amount of metal ion adsorbed per unit mass of adsorbent (mg g⁻¹), C_e is the equilibrium concentration of adsorbate (mg L⁻¹), Q[°] and b are the Langmuir constants and are the significance of adsorption capacity (mg g⁻¹) and energy of adsorption (L mg⁻¹), respectively.

- **Freundlich equations:** A linear form of this expression is given as:

$$\text{Log } Q_e = \log k + \frac{1}{n} \log C_e$$

where, k and n are the Freundlich constants and represent the significance of adsorption capacity and intensity of adsorption, respectively. Values of k and n were calculated from the intercept and slope of the plot log Q_e and log C_e.

A comparison of isotherm parameters was done to determine the effectiveness of the adsorbents. The coefficients of determination (r²) and the isotherm constants were obtained. The high values of r² (>95%) for both isotherms show that the adsorption of Pb(II) and Cr(VI) could be well described by both the Langmuir and Freundlich isotherms.

- **Characteristics of Langmuir isotherm:** The characteristics of a Langmuir isotherm can be expressed in terms of a dimensionless separation factor, R_L, which describes the type of isotherm and is defined by¹⁹:

$$R_L = \frac{1}{1 + K_L C_0}$$

where, R_L is a dimensionless separation factor, C₀ is initial Pb²⁺ and Cr⁶⁺ (mg L⁻¹) and K_L is a Langmuir constant (L mg⁻¹).

RESULTS AND DISCUSSION

Physico-chemical characteristic of the effluent: The mean pH value of the effluent indicated that the effluent was highly acidic than the NESREA's recommended limits for battery factory effluents which had adverse effect on the recreational function of the water and on the aquatic life (Table 1). Temperature plays an important role for its effect on other qualities of wastewater. The mean temperature value of the effluent is within the range of NESREA limit. The mean concentration of total dissolved solids (TDS) was lower than the NESREA standard of 2000 mg L⁻¹ for the discharged of wastewater into surface water. Conductivity of water is a measure of the total concentration of ionic species or salt content. The mean conductivity value of the effluent is very high. The high conductivity value (i.e., high salt concentration) in the effluents can increase the salinity of the receiving river, which may result in adverse ecological effects on the aquatic biota and such high salt concentrations hold potential health hazard²⁰.

Table 1: Physico-chemical characteristic of the effluent

Parameters	Mean value	NESREA standards
pH	2.0±0.15	6-9
Temperature (°C)	30.0±1.50	<40
TDS (mg L ⁻¹)	895.0±0.00	2000
Conductivity (µS cm ⁻³)	2164.7±0.60	1000
Lead (Pb) (mg L ⁻¹)	31.3±0.00	0.01
Chromium (Cr) (mg L ⁻¹)	13.1±0.00	<1

All data were mean ± standard deviation of triplicate determinations

Table 2: Characteristic of the prepared activated carbons

Parameters	PAC	SAC	CAC
pH _{pzc}	7.0±0.01	6.5±0.11	7.1±0.10
Ash (%)	5.2±0.01	6.2±0.01	5.4±0.01
Bulk density (kg m ⁻³)	0.8±0.01	0.8±0.01	0.6±0.01
Porosity	0.6±0.01	0.7±0.01	0.8±0.01
Surface area (m ² g ⁻¹)	625.6±0.01	530.7±0.01	200.4±0.01
Carboxylic	0.1±0.01	0.1±0.01	0.1±0.01
Phenolic	0.1±0.0	0.3±0.01	0.3±0.01
Lactones	0.6±0.01	0.6±0.01	0.6±0.01
Acidic	0.8±0.01	10.0±0.01	10.0±0.01
Basic	0.6±0.01	0.6±0.01	0.5±0.01

All data were mean ± standard deviation of triplicate determinations

Lead is a prime pollutant in a battery recycling effluent because it is a major raw material in the manufacture of lead acid accumulated batteries. The Pb level was exceeded in the effluent being studied (Table 1). Lead at very low concentration is toxic and hazardous to most forms of life and its chronic effect of Pb on man includes neurological disorders, especially in the foetus and in children, behavioural change and impaired performance in IQ ability^{11,21}. Also, the level of chromium in the effluent sample exceeded the NESREA standards. Ingestion of chromium in low value is carcinogenic. When inhaled, hexavalent chromium damages the lining of the nose and throat and irritates the lungs as well as the gastrointestinal tract. When swallowed, it can upset the stomach and damage the liver and kidneys and allergic skin reaction on contact^{15,22}. The direct use of water from this receiving river (Ogunpa) for domestic purposes without treatment could be detrimental especially to young children in the vicinity of the catchment.

Physical and surface chemistry of the adsorbents: The findings of the study showed a neutral pH (pzc-point of zero charge) which was found to be acceptable since that for most applications²³. The acidic pH_{pzc} shown by the plantain wastes adsorbents is consistent with the Boehm titration result that presented dominance of this acidic group of the surface of the plantain. This is in accordance with Unuabonah *et al.*²⁴, who observed the same trend on activated carbon from defatted *Carica papaya* seeds. The bulk density plays a major role in determining the amount of carbon that can be filtered out and the retention of treated effluent by its cake²⁵. Activated

carbon with high bulk density has capacity to filter more effluent/water before its available cake space is filled. The prepared adsorbents have a low ash content which is good for its efficiency (Table 2). It should be noted that the ash content reduced the overall activity of activated carbon and the efficiency of reactivation.

Iodine number is used to characterize activated carbon performance by measuring surface area. The micro-pores are majorly responsible for the large surface area of activated carbon particles and are created during the activation process. The surface areas of PAC and SAC are higher when compared to CAC (Table 2). The surface chemistry of carbon materials is basically determined by the acidity and basicity of their surface. The type and concentration of surface functional groups of activated carbons was determined by the Boehm titration technique. It should be noted that the adsorbents consisting of the amount of acidic and basic functional groups. The surface titration method stipulates that only strongly acidic carboxylic groups are neutralized by sodium bicarbonates and those neutralized by sodium carbonate are considered to be lactonic and carboxylic group. The weakly acidic phenolic groups only react with sodium hydroxide. Neutralization with hydrochloric acid (HCl) characterizes the amount of surface basic group (pyrones and chromenes) that are present in the activated carbon^{15,26}.

Operating conditions for adsorption capacity

Effect of pH: The result showed that the adsorption capacity (Q_e) of Pb(II) and Cr(VI) increased at a steady rate as pH increased up to 10, attaining a maximum value of 1.98 and 0.71, respectively (Fig. 1). The adsorption of the metal ions into PAC and SAC was largely influenced by pH. The pH affects the solubility of the metal ions, concentration of the counter ions on the functional groups of the adsorbent and the degree of ionization of the adsorbate during reaction. The decrease in the adsorption was observed after pH of 10, which is due to the formation of soluble hydroxyl complexes. At low pH values, the surface of the adsorbent would be closely associated with hydroxonium ions (H₃O⁺) by repulsive forces, to the surface functional groups, thereby lowering the percentage removal of metal²⁷. As the solution pH increases, there is onset of adsorption therefore which occurred before the hydrolysis and precipitation²⁸. When the pH of the adsorbing medium was increased from 2-10, there was a corresponding increase in deprotonation of the adsorbent surface, leading to a decrease in H⁺ ion on the adsorbent surface. This creates more negative charges on the adsorbent surface, which favours adsorption of positively charge species and the positive sites on the adsorbent surface¹⁴.

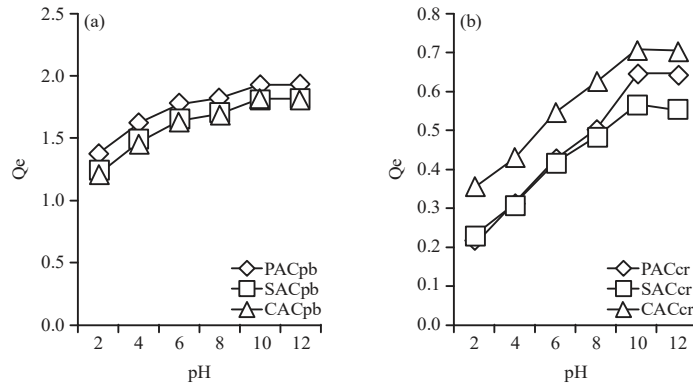


Fig. 1(a-b): Adsorption capacity of Pb²⁺ and (b) Cr⁶⁺ with pH

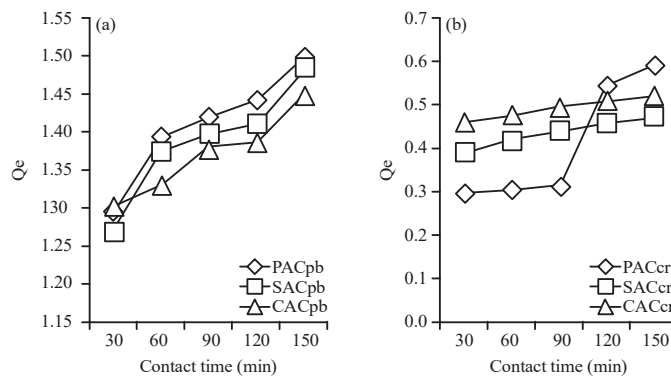


Fig. 2(a-b): Adsorption capacity of Pb²⁺ and (b) Cr⁶⁺ with time

Another factor that contributes to enhancing metal ion adsorption is the increasing pH which encourages metal ion precipitation from the solution in the form of hydroxides. The hydrolysis of cations by the replacement of metal ligands in the inner coordination sphere with the hydroxyl groups, this replacement occurs after the removal of the outer hydration sphere of metal cations. In addition, increasing pH decreases the concentration of H⁺, therefore reducing the competition between metal ions and protons for adsorption sites on the particle surface.

Effect of contact time: This is the removal of lead and chromium ion from water sample using PAC and SAC as a function of time. The result showed that the adsorption rate was rapid within the first 30 min, sharply increased for 60 min and gradually increased between 90 and 150 min for Pb(II) for the adsorbents. The adsorption capacity (q_e) of Pb(II) and Cr(VI) increased at a steady rate as contact time increased up to 150 min attaining a maximum value of 1.50 and 0.59, respectively (Fig. 2). The initial faster rate was due to the availability of the uncovered surface area of the adsorbents, since the adsorption kinetics depends on the

surface area of the adsorbents. The lead and chromium adsorption takes place at the more reactive sites. As these sites are progressively filled the more difficult the sorption becomes as the sorption process tends to be more unfavourable^{11,16}.

Effect of adsorbent dose: The values were generated by varying the adsorbent doses (0.1-2.0 g) at room temperature with the adsorbents. The adsorption capacity (q_e) of Pb(II) and Cr(VI) decreased at a steady rate as adsorbent dose increased up to about 1.5 g for Pb(II) and 2.0 g for Cr(VI) attaining a minimum value of 0.60 and 0.15, respectively (Fig. 3). The result suggested that after a certain dose of adsorbent the maximum adsorption sets in and hence the amount of ions bound to the adsorbent and the amount of free ions remain when additional dose of adsorbent is added. The increased percentage adsorption by adsorbent was as a result of increased surface area and increased adsorption site occasioned by increased adsorbent dose. The same trend has been observed by Adebowale *et al.*²⁹. The observed decrease in adsorption capacity is due to decrease in liquid-solid ratio which direct resulted in this trend since amount adsorbed, q_e ,

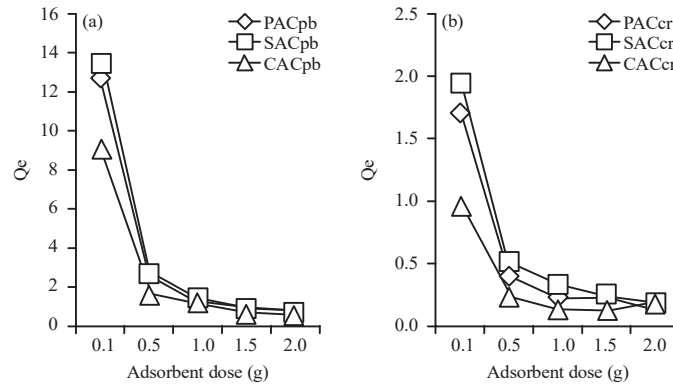


Fig. 3(a-b): Adsorption capacity of Pb²⁺ and (b) Cr⁶⁺ with adsorbent dose

Table 3: Adsorption isotherm constants and coefficient of determination (r²) of adsorbent for Pb²⁺

Adsorbent	Langmuir isotherm constants			Freundlich isotherm constants		
	Q ^o (mg g ⁻¹)	b (Lg ⁻¹)	R ²	k (mg g ⁻¹)	1/n	r ²
PAC	1.9566	0.0648	0.9779*	1825.58	0.2552	0.8682
SAC	3.5855	0.0598	0.6894	49.8884	0.3928	0.8356
CAC	0.0524	0.0524	0.5607	38.3354	0.4188	0.7794

*: Fit at r² (>95%)

Table 4: Adsorption isotherm constants and coefficient of determination (r²) of adsorbent for Cr⁶⁺

Adsorbent	Langmuir isotherm constants			Freundlich isotherm constants		
	Q ^o (mg g ⁻¹)	b (Lg ⁻¹)	r ²	k (Lg ⁻¹)	1/n	r ²
PAC	12.8866	0.0165	0.9992*	13.9766	0.6506	0.9920*
SAC	1.5903	0.0366	0.8215	51416.20	0.2367	0.9604*
CAC	18.9036	0.0122	0.9917*	9.2939	0.7280	0.9713*

*: Fit at r² (>95)

has an inverse proportionality function to weight of biosorbent but a direct proportionality function to percentage adsorbed²⁴.

Adsorption isotherms: Adsorption data of lead ions on PAC only showed satisfactory fits to Langmuir model and other adsorbents showed poor fit to both models (Table 3). For chromium ions, all the adsorbents showed satisfactory fits to both models except SAC which showed poor fit (Table 4). This confirmed that the adsorption of Pb²⁺ and Cr⁶⁺ into plantain waste adsorbents is favourable for higher initial ion concentrations with respect to lower ones. The values of 1/n were also found to be less than 1, again that the adsorption is favourable²⁴. However, from the correlation coefficient (r²), there is strong indication that the adsorption data obtained fits the Freundlich better than Langmuir model. The PAC and SAC have potentials for removing lead and chromium from effluent generated from battery recycling plant with PAC having highest adsorption capacity. Also, the value of R_L for each adsorbent obtained were found to be less than 1 and decreased with increasing initial concentration of Pb²⁺ and Cr⁶⁺.

CONCLUSION

The study confirmed that the discharge of untreated effluent from battery recycling plant containing heavy metals and hazardous materials like lead and chromium over a long period of time into the receiving water body is potential source of contamination. The study revealed that the background level of lead and chromium in the effluent were above the recommended limits of NESREA. From the analysis of the result, it is evident that the plantain wastes particularly ripe peel has great potential for the uptake of lead and chromium from the industrial effluent. It has also been observed that the sorption capacity of plantain wastes depends on conditions such as pH, initial metal ion concentration, adsorption dose and contact time. Considering the efficiency of the plantain wastes for adsorption in addition to the fact that it is readily available in bulk quantities, constitute nuisance to the environment as well as the shortcoming of conventional treatments such as poor recovery, require a lot of skill and technical constraints. It is evident that the use of plantain wastes and other natural sorption materials is a better option to the conventional

treatment options. It is therefore recommended that the use of plantain wastes as well as other agricultural wastes as adsorbent for heavy metals in polluted water should be given priority over the conventional methods as it helps to clean up the environment among the several advantages it offers.

SIGNIFICANCE STATEMENT

"This study discovered that plantain waste especially ripe peel represents a good sorption material for lead and chromium in aqueous solution. Converting the plantain wastes into activated carbon has greatly helped to reduce its menace in the environment and enhance effective waste management". This study will help the researchers to uncover the critical areas of waste recovery and remediative technology that many researchers were not able to explore.

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