



Treatment of landfill leachate using sawdust, rice husk, activated charcoal, and caustic soda

Ajedipe Chrispin^a, Agyei George^b

^aWaste Landfill Company Limited, P Box Dt 1670, Adenta-Accra

^bUniversity of Mines and Technology, Tarkwa

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Abstract

Landfill leachate has organic and inorganic contaminants that produce offensive odour and can migrate to pollute surface and underground water. This paper therefore seeks to utilise sawdust, rice husk, chemical activated charcoal, sand and caustic soda and physicochemical processes to remove contaminants from the leachate to make the water safe to be discharged to the environment. The leachate obtained from Kpone Engineered Landfill was passed through absorbent columns designed from the following: charcoal, sawdust, sand and rice husk. The concentrations of copper, iron, lead, cadmium, manganese, chromium, and zinc were assessed. The pH, total dissolved solids, true colour, apparent colour, turbidity and electrical conductivity were determined. The stratified bed with rice husk at the top followed by sawdust and sand bed showed highest removal efficiency of 89.6 %. The effluent obtained from the multistage adsorption was subjected to chemical precipitation of which 100% of the parameters met WHO Drinking Water Standard.

1. Introduction

The quest for clean environment and good health calls for the dumping of domestic and industrial solid waste in landfills at isolated areas. Decaying waste within landfills create big environmental problems due to the emission of greenhouse gases and the generation of landfill leachate. Leachate is generated when rain water percolates through decomposing waste dumped in landfills, producing dissolved and suspended solids to form noxious effluent. The leachate generated is made up of water, microorganisms, dissolved and suspended substances of wastes [1, 2; 3, 4]. Some of the methods available for treating landfill leachate can be categorised into physical, chemical and biological methods [5].

The leachate at Kpone Engineered Landfill in Ghana has been treated with the biological method (anaerobic) which reduced the concentration of Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD) below permissible discharge limit. However, the concentration of iron, lead, copper, manganese, zinc, cadmium, chromium, colour, turbidity, total dissolved solids, electrical conductivity and pH were found to exceed the WHO (2000) drinking water specifications thereby rendering the effluent unfit to be discharged into water bodies. Additionally, the continuous retention of the leachate in the open ponds results in spillover of the leachate anytime there is heavy downpour affecting downstream community with very offensive odour.

In this study, the combination of physical and chemical methods utilising adsorption and chemical precipitation procedures to complement the existing treatment methods were experimented using bio-adsorbents and caustic soda. The main adsorbents used for pretreatment were sawdust, rice husk and activated charcoal. Sawdust and rice husk materials are made of cellulose, lignin and hemicelluloses with functional groups such as carboxyl, hydroxyl, phenolic and amine groups [6] that combine well with heavy metal ions through different mechanisms to form chelates and lower the pH of the influent for effective adsorption of metals. The rice husk and sawdust are very selective, effective and they can remove all types of soluble heavy metal ions in solution as well as dissolved recalcitrant organic compounds [7]. Due to the high cost of some of the conventional treatment methods and sometimes low effluent quality, it is important to focus attention on the removal of heavy metals and refractive organic substances using low cost and readily available adsorbents. Not much studies have been conducted on the use of the three adsorbents combined, sawdust, rice husk and activated charcoal as well as chemical precipitation for the removal of contaminants from landfill leachate. Most of the studies are limited to composite of sawdust and rice husk [8]. The objective of this research is therefore to reduce the concentrations of the low-quality physicochemical parameters from the leachate at Kpone Engineered Landfill using physicochemical treatment methods and to further develop a general treatment method involving the combination of sawdust, rice husk, activated charcoal, and caustic soda.

2. Materials and Method

2.1 Study area

The study was conducted at Kpone Engineered Landfill with a total area of 95,200m² located at approximately longitude 6°N and latitude 0°E. The study span a period of four months (February-May), 2019. The landfill is located at Kpone-Katamanso District Assembly. The topography of the area is generally flat and forms part of the coastal plains of Ghana. The landfill site is owned by Tema Metropolitan Assembly (TMA) and operated by Waste Landfills Company Limited (WLF). The landfill was constructed between 2011 and 2012 to receive 500 tonnes of solid wastes from Tema metropolis and its environs with three leachate ponds constructed to collect leachate.

2.2 Preparation of the adsorbents

The sawdust and rice husk were collected from mills at Tema which were composed of particles with various sizes. Adequate quantity of sand was also obtained in a nearby river. The sawdust, rice husk and sand were sieved using 2 mm sieve. The undersize particle sizes of the rice husk, sand and sawdust were taken and washed thoroughly in demineralised water to remove fines and dried in the sun. The charcoal was crushed to smaller particle sizes using the mortar and pestle. It was screened. The undersize granulated charcoal was washed with demineralised water. The particles were further pulverized and washed thoroughly to remove the fines. The powdered charcoal and granulated charcoal were soaked in highly concentrated sodium chloride solution for 24 hours. They were washed several times with de-ionised water while monitoring the Total Dissolved Solids (TDS) concentration to ensure that the ionic concentration due to sodium and chloride ions were removed. A dilute methyl orange solution was passed through the activated and the untreated charcoal to verify the effectiveness of the activation process

2.3 Pretreatment column experiment

The columns as arranged in Figure 1 were washed thoroughly with de-ionised water and allowed to dry. The open bottom of each column was covered with a number 42 Whatman® filter paper and plastic net meshes which were tied using metallic rings. The height of the column was divided

into three equal parts to determine the average mass of each of the adsorbents by filling and compacting the adsorbent to the marked height. The empty column mass was predetermined using an electronic weighing balance and the mass of the materials was obtained by subtracting the mass of the empty column from the gross weight. The measurement was repeated five times for each of the materials and their average masses computed to obtain Adsorbent Height as in Equation (1) as follows:

$$\Delta h = \frac{H}{3} \quad (1)$$

The mass requirement of the various materials is presented in Table 1

Table 1: Mass requirement of adsorbents

Column	Material	Height(cm)	Mass(g) of empty column	Average mass(g) of adsorbent & column	Average mass(g) of adsorbent
Column A	Free board	3			
	Sand bed	10	243	493	250
	Sawdust	11	243	351.6	108.6
	Charcoal	11	243	387	144
Column B	Free board	3			
	Sand bed	10	243	493	250
	Rice husk	11	243	351.6	108.6
	Charcoal	11	243	387	144
Column C	Free board	3			
	Sand bed	10	243	493	250
	Rice husk	5.5	243	297.3	54.3
	Sawdust	5.5	243	297.3	54.3
	Charcoal	11	243	387	144
Column D	Free board	3			
	Sand bed	10	243	493	250
	Rice husk	7.3	243	327.2	84.2
	Sawdust	7.3	243	327.2	84.2
	Charcoal	7.3	243	327.2	84.2
Column E	Free board	3			
	Sand bed	10	243	493	250
	Rice husk	11	243	369.3	126.3
	Sawdust	11	243	369.3	126.3

Each column was packed with a total of 252.6 g of the adsorbents in different combinations and 250 g of sand bed with a free board of 3 cm created on top of the sand bed as shown in Figure 1. The sand bed was not for filtration purposes. It was used to suppress the light weight adsorbents to prevent overboard spillover. Pretrial showed the adsorbents carried overboard when the leachate was introduced into the column beds. The adsorbents in the columns were compacted and mounted on clumps before the leachate was introduced into each column. De-ionised water was passed through each column to condition the adsorbents. The flow rate was set at thirty five (35) drops per minute of influent. Because of the higher adsorptive capacity of the sawdust and rice

husk, it took 186 minutes for the first drop of effluent to be received. A total of 193 minutes was used to collect 50 ml of the effluent and the effluent flow rate was determined at 0.26 ml/minute. The experiment was conducted at room temperature, 25 °C.

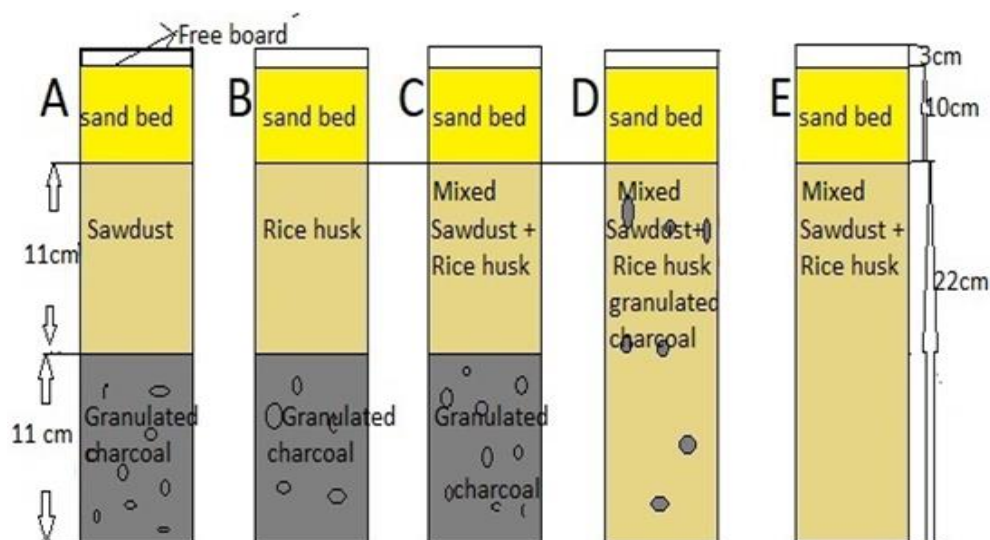


Figure 1: Arrangement of adsorbents in the various columns

The pretreatment was carried out to determine the combination that offered the most efficient contaminant removal. The contaminant concentration of the influent and effluent from each column was determined and removal efficiency for each factor was found. The Percentage Removal Efficiency (R) is expressed in Equation (2) as follows:

$$R(\%) = \frac{C_o - C_e}{C_o} \times 100 \quad (2)$$

The average percentage removal by each column was calculated by adding the percentages of the parameters in a column divided by the total number of parameters measured for that column. The mathematical expression for Average Removal (AR) is,

$$AR = \frac{\sum R\%}{n} \quad (3)$$

2.4 Multistage adsorption column

This stage was designed to offer double adsorption in one column. The multistage bed adsorption column was designed using the height of the columns as described in section 2.3. The mass of each of the bed was calculated using the height of the bed. That is,

$$\text{Mass} = CM - ME \quad (4)$$

The sand bed serves as weighting and separation layer but not for filtration purpose. Contaminants not adsorbed by the first charcoal bed are captured by the bottom charcoal bed which is necessary to prolong the bed life span. The arrangement of the beds are shown in appendix Table A1.

An amount of 160 g of powdered charcoal was packed into a column and 200 g of sand bed was placed on top of it. Another layer of powdered charcoal was formed by measuring 160 g of powdered charcoal which was placed on top of the 200 g of the sand bed. This was followed by additional layer of 200 g of sand bed. A free board of 3 cm was placed on top of the last sand bed.

De-ionised water was passed through the beds first to remove entrained air. The presence of the air could cause channeling when the effluent was introduced which might result in early breakthrough of contaminants. The conditioned beds were given a residence time of 3 hours to ensure that all the water was drained out.

2.5 Chemical precipitation

A 400 ml of the effluent from the multistage bed treatment column was measured and 1ml of 10 M NaOH solution was measured and two drops were added to the 400 ml of effluent leachate to adjust the pH to 10.0. According to the Michigan Department of Environmental Quality Operator Training and Certification Unit, different metals precipitate at different pH, hence depending on the metal of interest the pH can be adjusted to meet the precipitation requirement of that metal. Therefore, to precipitate more than one metal from the same solution, the pH is adjusted to an optimum value for one of the metals with the lowest discharge limit. In this present work, Zinc has the least discharge limit with a precipitation pH of 10. The pH was adjusted to 10 by the addition of sodium hydroxide and stirred. The solution was agitated and precipitate began forming within 10 minutes. The precipitation process lasted for 40 minutes. The mixed supernatant and precipitate were passed through another multistage bed column for filtration and further adsorption. At this stage, the sand bed serves as both weighting and filtration purpose. The effluent was taken for physicochemical analysis.

2.6 Acid digestion of adsorbents

In order to establish whether the adsorbents contributed to the total effluent metals concentration, 0.1g each of the adsorbents were measured and digested in concentrated sulphuric acid. The digested samples were diluted with de-ionised water, filtered using number 42 Whatman® filter paper and taken for metal analysis.

3. Results and Discussion

3.1 Pretreatment

The pretreatment results showed the concentration of the metals was reduced to meet the WHO, 2000 water specification permissible discharge limits. Lead (Pb) and Cadmium (Cd) were not detected in the effluent leachate. Iron (Fe) and chromium (Cr) reduced. However, the concentration of these metals in the effluent exceeded the tolerable limits. The Zinc concentration in the effluent leachate (Zn) reduced to meet the WHO standard. On the contrary, the concentrations of Manganese in the effluents of columns B to E were above the values of the WHO standard (Table 2). According to [9] Manganese and Iron are mobile in acidic medium and also [10] suggested Mn has various oxidation (Mn^{2+} and Mn^{4+}) state which precipitate at pH ranging from 9 to 9.5 and complete removal at pH of 10.5. It is possible the Mn precipitated in the influent which has a pH of 10.12 and when the pH was reduced below 7, more mobile Mn ions were available which were not adsorbed by the adsorbents. This could account for the high concentrations of Mn in the effluent from each of the columns.

Table 2: Results for heavy metals in the influent and effluent

Adsorbent Materials	Heavy metals in effluent						
	Cu	Fe	Pb	Cd	Mn	Cr	Zn
	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l
Influent leachate	0.055	6.700	0.534	0.016	0.151	2.689	0.572

Column A	0.013	2.974	Nd	Nd	0.013	0.102	0.200
Column B	0.044	0.914	Nd	Nd	5.821	0.132	0.292
Column C	0.003	0.236	Nd	Nd	2.109	0.028	0.059
Column D	0.039	0.498	Nd	Nd	3.702	0.434	0.51
Column E	0.047	0.805	Nd	Nd	1.569	0.016	0.043
WHO guideline,2000	2.0	0.3	0.01	0.005	0.05	0.05	5.0
nd= no detection							

There was a reduction in the pH by 35.7%,36.9%, 36.1%, 35.4%, 37.1% for Column A, Column B, Column C, Column D and Column E respectively. The generation of organic acids by the sawdust and rice husk which contain carboxyl, phenolic, and hydroxyl groups contributed to the reduction in pH [6]. Because of the gradual percolation of the influent through the beds, the dissolved gases in the leachate causing the odour were liberated into the surrounding environment and some were possibly adsorbed by the adsorbents. The other parameters such as TDS, turbidity, apparent colour, true colour and electrical conductivity (EC) were reduced in the effluent, however, they did not meet the WHO, 2000 permissible discharge limits as indicated in Table 3. The higher concentration of these parameters beyond the WHO permitted discharge limit values shows the effluent requires further treatment before discharging into water bodies. The colour observed was yellowish.

Table 3: Analytical results of physicochemical parameters in the influent and effluent leachate

Adsorbent Materials In column	Physicochemical parameters							
	pH unit		TDS	True Colour	Turbidity	App colour	EC	Odour
			mg/l	Pt.co	NTU	Pt.co	µS/cm	
Influent leachate	10.12		4865	6080	224	37050	7943	Very offensive
	%change							
Column A	6.51	35.7%	2244	181	22	1089	1681	No offensive odour
Column B	6.39	36.9%	3244	167	9	1203	5817	No offensive odour
Column C	6.47	36.1%	1863	198	22	1056	2661	No offensive odour
Column D	6.54	35.4%	3008	232	14	1280	5283	No offensive odour
Column E	6.37	37.1%	2972	164	9	1890	3043	No offensive odour
WHO guideline,2000	6.5-9.5		1000	20	4	20	2500	

3.2 Multistage adsorption bed

The concentration of 2.109 mg/l Mn detected in Column C whose effluent was passed through the multistage bed adsorption column was reduced to WHO standard. The Cu, Pb and Cd were not detected after passing through this bed. The Zn concentration was reduced to meet the WHO standard. However, Fe and Cr concentrations (shown in Table 4) exceeded the WHO permissible discharge limit.

The pH, TDS and EC were observed to conform to the WHO standard. Although the apparent colour, true colour and turbidity reduced their concentrations in the effluent, the values were higher than the recommended concentrations of 20 pt.co, 20 pt.co and 4 NTU respectively for

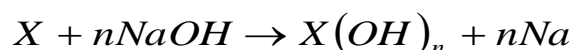
WHO standard shown in Table 4. A colourless effluent was obtained after the multistage column adsorption. The alternate arrangement of the charcoal and sand beds contributed to the high performance of the column.

Table 4: Physicochemical parameters from the multistage bed

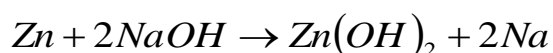
Parameter	Influent	Effluent	WHO guideline,2000
Cu, (mg/l)	0.055	Nd	2.0
Fe,(mg/l)	6.700	0.396	0.3
Pb, (mg/l)	0.534	Nd	0.01
Cd, (mg/l)	0.016	Nd	0.005
Mn, (mg/l)	0.151	0.012	0.05
Cr, (mg/l)	2.689	0.063	0.05
Zn, (mg/l)	0.572	0.350	5.0
pH unit	10.12	6.53	6.5-9.5
Turbidity, NTU	224	8	4
App.colour(pt.co)	37050	103	20
True colour, (pt.co)	6080	96	20
TDS,mg/l	4865	1108	1500
EC, μ S/cm	7943	1750	2500

3.3 Chemical precipitation

The concentration of the physicochemical parameters measured met the WHO permissible level as shown in Table 5. The achievement of cleaned effluent at the final stage was attributed to the following reasons. For instance, the general chemical reaction occurring at this stage is;



Where X is cation of a metal and n is the valency of the cation. Example, the reaction between zinc and sodium hydroxide can be expressed as:



The amorphous solid zinc hydroxide formed ($Zn(OH)_2$) is the precipitate with adhesive properties attracting many contaminants to the loose structure; not only the metals but including dissolved refractive organics. The continuous attachment of many ionic species (contaminants) to the structure increased the bulk mass of the new formed precipitate causing it to settle to the bottom leaving supernatant. The formation of the precipitate was assumed likely not to remove all the targeted parameters because of the variations in the precipitation pH of the various contaminants. Due to the difference in precipitation pH and solubility pH of the various contaminants and the resolubilisation of some of the precipitated metals, the mixed sludge was passed through a second multistage sand and charcoal beds for filtration of the sludge as well as adsorption of the solubilized contaminants. This resulted in the removal of the physicochemical parameters to the desirable standard as presented in Table 5.

Table 5: Concentration of contaminants in the effluent- Chemical treatment process.

Parameter	Influent	Effluent	WHO guideline 2000
	mg/l	mg/l	mg/l
Cu	0.055	0.017	2.0
Fe	6.700	0.193	0.3
Pb	0.534	Nd	0.01
Cd	0.016	Nd	0.005
Mn	0.151	Nd	0.05
Cr	2.689	0.003	0.05
Zn	0.572	0.172	5.0
Turbidity (NTU)	224	3.8	4
Apparent colour (pt.co)	37050	19.1	20
True colour (pt.co)	6080	18.7	20
TDS	4865	989	1500
EC	7943	1452	2500
pH unit	10.12	Adjusted to 8	6.5-9.5

3.4 Analysis of Acid digestion results

The heavy metals of interest were not detected in the sawdust, charcoal and the rice husk after the analysis. Therefore, the adsorbents had no impact on the overall heavy metals concentration in the effluent. The concentrations observed were only due to the leachate. The heavy metals and corresponding concentration levels are presented in Table 6

Table 6: Concentration of heavy metals in the adsorbents-acid digestion process

Parameter	Concentration (mg/l)
Cu	Nd
Fe	Nd
Pb	Nd
Cd	Nd
Mn	Nd
Cr	Nd
Zn	Nd
nd= no detection	

3.5 Pretreatment efficiency-column experiment.

3.5.1 Heavy metals removal

All the columns gave 100% removal efficiency for Pb and Cd. Column E showed the least removal for Cu shown in Table 7.

The sawdust and rice husk as bio-sorbent are made of polymeric materials such as lignin, tannin or cellulose containing carboxyl, hydroxyl, phenolic and amine groups [11]. These materials generate organic acids from the carboxyl, hydroxyl and amino groups [6] which serve as chelating agents whose ligands bind with the metal ions to keep the metallic species in solution. The enhanced surface area and the increased number of binding sites with the combination of the adsorbents in different ratios resulted in the high removal efficiency of all the metals.

Table 7: Percentage removal of heavy metals by each column

Column	Percentage (%) Removal Efficiency						
	Cu (%)	Fe (%)	Pb (%)	Cd (%)	Mn (%)	Zn (%)	Cr (%)
COL A	96.4	55.6	100	100	91	65	96.2
COL B	20	86.4	100	100	-3755	49	95.1
COL C	94.5	96.5	100	100	-1297	89.7	99
COL D	29.1	92.6	100	100	-2352	10.8	83.9
COL E	14.5	88	100	100	-939	92.5	99.4

3.5.2 Physicochemical parameters

The removal efficiency of contaminants was relatively high (Table 8). The low EC values in the columns may be explained by the hydrogen ions and ionic metal species present in the effluent which may not have been completely removed by the adsorbents. The retention time of the influent within the adsorbent beds contributed to the high adsorption observed for both the metals and the physicochemical properties. The retention time of 36 minutes allowed for adequate interaction between the ions or molecules of the contaminants and the surface of the adsorbents. This may have contributed to the high removal of true colour, apparent colour and turbidity in all the columns.

Table 8: Percentage removal of physicochemical parameters by each column

Column	Percentage (%) Removal Efficiency					
	pH unit	TDS (%)	True colour (%)	Apparent colour (%)	Turbidity (%)	EC (%)
COL A	35.7%	53.9	80.6	97.1	90.2	78.8
COL B	36.9%	33.3	97.3	96.8	96	26.8
COL C	36.1%	61.7	96.7	97.1	90.2	66.5
COL D	35.4%	38.2	96.2	96.5	93.8	33.5
COL E	37.1%	20.7	97.1	94.9	96	51.3

3.6. Heavy metals and physicochemical parameters removal

The efficiency of each combination was calculated by finding the Average of the Percentage Removal of parameters measured for a particular column. The removal efficiency of the various columns using heavy metals were as follows: column C had the highest, 96.6% followed by, columns E, 82.2%; A, 82.5% and B, 75.1% and column D, 69.4%. The high removal in column C could be attributed to the combined three adsorbents offering larger surface area and many binding sites for adsorption of metals

3.7 Parameters meeting the WHO standard in each treatment method

A total of 14 parameters (Apparent colour, True colour, conductivity, TDS, Turbidity, pH, Odour, Cadmium, copper, Lead, Zinc, Manganese, Chromium, Iron) were analysed. The concentrations of these parameters in the influent were found to be high. After the first columns treatment, 43% of the parameters were within WHO permissible limit in Columns B and D, 50% in Column E, 57% in Columns A and C, 64% in the multistage bed adsorption column and 100% in the chemical precipitation method. At the end of the treatment processes, a general treatment model was developed out of the various treatment processes. The flowchart of the arrangement of the procedure is shown in Figure 2.

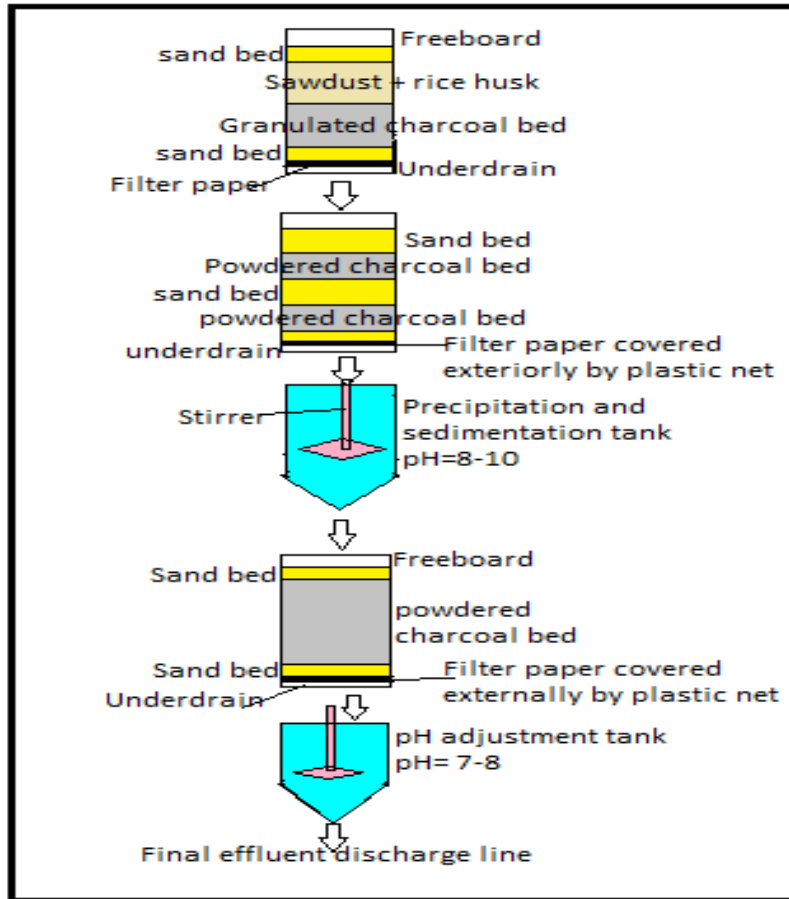


Figure 2: flow chart illustrating the arrangement of the treatment process

4. Conclusion

The average removal efficiency of the various combinations were calculated for each column as; Col A, 81.2%; Col B, 72.6%; Col C, 89.6%; Col D, 70.5%, and Col E, 80.3%. Column C showed highest removal efficiency and the effluent was passed through a multistage adsorption column in which 64% of the parameters measured met the WHO standard.

The effluent obtained from the multistage adsorption was subjected to chemical precipitation of which 100% of the parameters met WHO standard.

By integrating all the treatment processes, a general treatment method was developed using Column C, multistage adsorption bed column, chemical precipitation method and second multistage adsorption bed column. The final multi stage adsorption column was introduced to remove the precipitate and adsorption of resolubilised metals.

4.1 Recommendation

The following recommendations were made;

- Further research should be carried out by extracting the ligno-cellulose component of the sawdust and rice husk for the adsorption process to determine the possibility of minimising the addition of colour to the effluent.
- Further research should be carried out to determine the possibility of regenerating the adsorbents when their adsorptive capacity is attained for reuse to minimise the requirement for fresh adsorbents

- c. One of the setbacks of this research work was its inability to determine the adsorptive capacities of the combined adsorbents. Further research should be carried out to quantify this property.

Nomenclature





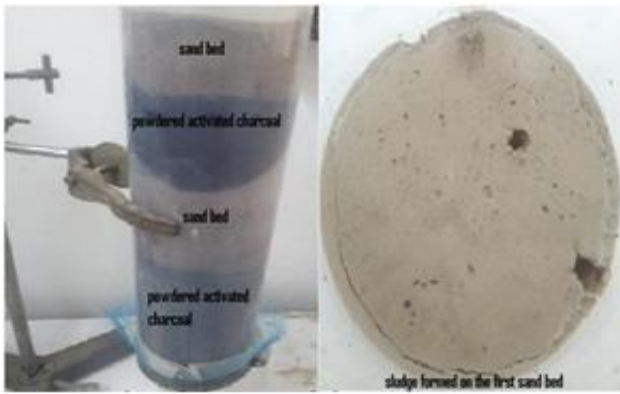
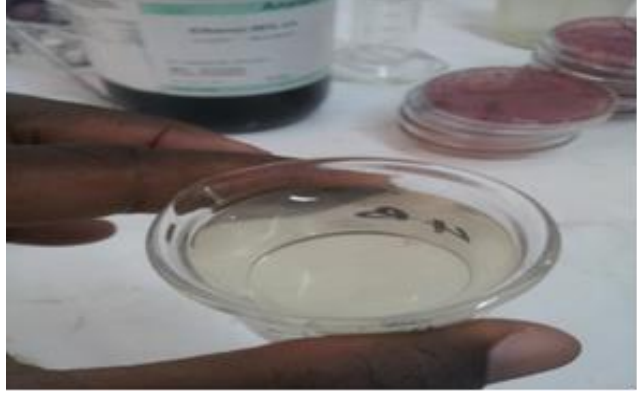
H	height of column (cm)
Δh	adsorbent height in columns (cm)
R	removal efficiency (%)
C_o	influent concentration (initial) (mg/l)
C_e	effluent concentration (final) (mg/l)
AR	average Removal (%)
n	total number of parameters measured in a particular column
CM	the combined mass of the column and the adsorbent (g)
ME	mass of the empty column (g)

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Appendix

Table A1. Summary of the output of the various treatment processes

	
<p>Colour of leachate after passing through sawdust and rice husk bed</p>	<p>Arrangement of multistage adsorption bed</p>
	
<p>Multistage adsorption bed column</p>	<p>Colour remaining after passing through multistage adsorption bed</p>
	
<p>Chemical precipitate repassed through multistage adsorption bed and the sludge formed on the first sand bed</p>	<p>Final colour of leachate after chemical precipitation and re-passing through multistage adsorption bed</p>