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The Efficiency and Effectiveness of Combined Coagulation and Adsorption Process in Treatment of Landfill Leachate

Evbuomwan B. O.[1], Obidike E. O.[2]
[1,2]Department of Chemical Engineering, University of Port Harcourt, Port Harcourt, Nigeria
([1]benson.evbuomwan@uniport.edu.ng)

Abstract-Leachate is the effluent liquid generated from landfill. Treatment of leachate is important in sanitary landfilling to reduce the effect to the ecosystem. The leachate treatment methods can be physical, chemical or biological and combined methods is adopted for effective treatment. This experimental study was performed to determine the efficiency and effectiveness of combined coagulation and activated bone char absorption in landfill leachate treatment. The Coagulation experiment was carried out with aluminum sulphate and result shows maximum COD removal of 49.3% at coagulants dose of 2.5g/l. The supernatant liquid at maximum COD removal from coagulation was subjected to Adsorption. Animal bones were carbonized with muffle furnace, crushed and activated with sodium hydroxide. The activated bone char was characterized for principal physiochemical parameters and used as an adsorbent in adsorption process. The results of combined treatment shows increase in the removal efficiencies as the adsorbent concentration was increased and maximum COD of 80.3% was obtained. The experimental results were correlated with both Freundlich and Langmuir models to determine the equilibrium behavior of adsorption. The correlated results show regression coefficient of 0.9522 and 0.9588 respectively. Freundlich Isotherm Models give an adsorption capacity of $2.04mg^{-0.1}g^{-1}$ and measure of deviation from linearity is 0.91. The Langmuir adsorption isotherm model also gives an adsorption capacity of $2x10^{-3}$ and a maximum amount of adsorption obtained as 5000 mg/g.

Keywords- *Coagulation, Adsorption Leachate, Bonechar, Isotherm, Landfill*

I. INTRODUCTION

A common practice of burying Municipal Solid waste in landfill is adopted in many places of the world. The increasing landfill in most urban cities of the world is as a result of rapid growth in population and industrialization and also due to availability of infrastructure. In Nigeria, the discovery of Crude oil and its exploration has contributed greatly to the increasing population of urban cities. In addition, the country is richly blessed with many national resources and this continues to gather people from all over the world to exploit the huge national heritage of the country.

Landfill is of different categories ranges from a road site garbage or refuse dump, dump site, engineered landfill, sanitary landfill to even a more sophisticated system. Oftentimes, municipal solid waste and domestic refuse is dropped in an open dump. Waste burning is commonly adopted to bury the waste, there is minimal or no attention towards sanitary landfilling practices (Klinck B.A and Stuart M.E, 2017). Leachate is the liquid waste generated by excess rainwater generated from the waste layers in the landfill or by inherent nature of waste themselves. The increase in the number of landfill has contributed to the increased leachate generated in the urban cities (Peter Kjeldsen et al., 2002).

Waste disposal and management is often put on the list of priorities with little or no proper regulations in place to control disposal of waste to the environment or even to persecute the offenders. The topmost environmental problem all over the world is disposal of the increasing solid waste due to high rate of waste generation with the increasing population growth and the increase in per capita (Al-Yaqout et al., 2005). An indiscriminate discharge of untreated leachate poses danger to the environment; the neighboring residents, contaminants the soil, underground and surface water. Leachate generated from landfill or dumpsite is of important concern due to its potential to penetrate into water resources systems (Sartaj et al., 2010).

There are two major cause of pollution from landfilling practice that result to migration into the natural environment. Leachate, contaminants the soil and ground water and biogas which pollute the atmospheric environment by causing air pollution and produce as a result of gases release during the fermentation of organic matter. In recent time, improvement in technology has pave way to the use of modern landfills, highly engineered facilities, designed to minimize the adverse effect of the waste on the surrounding environment. However, the use of modern landfills is limited in practice in most developing countries, the polluted leachate generated remains consequence of the existing solid waste disposal practice and the future landfills (Abdulhussain et al., 2009).

The composition of Leachate depends largely on the age of landfills and the types of solid waste in a landfill site. Other factors that affect the quality of leachates also include precipitation, seasonal weather variation and composition of solid waste. The leachate composition is determined in terms of COD, BOD, and ratio of COD/BOD, colour, pH, alkalinity, oxidation-reduction potential and heavy metals and also used to determine the treatment method to adopt for treatment of leachate (Shabumam M.A et al, 2011).

Leachate can change greatly in quantity and composition from one location to another and time to time. Different methods (physical, chemical or biological) are available and can be adopted for treatment of leachate. The method of treatment is a function of leachate characteristics and its tolerance against changes in leachate quality and such determine treatment applicability in any landfill area. The success of leachate treatments also depends on the characteristics of the leachate and age of the landfill. Selection of the best leachate treatment process should be on effluent discharge alternatives and limitations, residual treatment process, permit requirements and cost-effective treatment process.

Effective treatment method for wastewater is not yet available, oftentimes combine treatments are adopted based on international regulations to safeguard the environment, farmers and other key players should be practical and offer possible risk management options.

Coagulation-flocculation is used in water and wastewater treatment to clump small destabilized particles together into larger aggregates that are more readily separated from water. Coagulation is brought about or activated by different substances commonly known as coagulants. In the treatment of landfill leachate using Coagulation, treatment should aimed at selection of most appropriate coagulant, determination of experimental conditions, the effect of pH and flocculants addition.

Adsorption on the other side is a phenomenon that occurs by accumulation of large number of molecular species at the surface of liquid or solid phase in comparison to the bulk. There are two components in adsorption: adsorbent, the substance on the surface at which adsorption takes place and adsorbate, the substance being adsorbed on the surface of adsorbent.

The difficulties encounter in treating leachate is due to bacteria activities, chemical, organic and non-organic pollutants, heavy metals, dissolved and colloidal solids and many other contaminants that constitute the components of landfill. Leachate is found to have high concentration of BOD and COD, heavy metal and ammonia nitrogen concentration (Maleki et al., 2009). When landfill is poorly managed and treated, the leachate produced penetrates into soil and contaminants the ground water and surface water especially where there is a permeable layer of soil or landfill built without sheeting layer and even failure of the sheeting layer. According to a statistics, 71.4 % of local authorities are faced with serious ground water pollution, 57.2% have problem in dealing with leachate management (Nasir et al., 1999).

The dissolved organic and inorganic salts in leachate pose significant pollution potential to ground and surface water, trace element is limited and therefore contributes to groundwater pollution. This is as a result of its strong attenuation. Leachate can be source of pollution to groundwater where drinking and irrigation water is sourced from groundwater or leached hydraulically to the receiving surface water.

There is need for effective and efficient treatment of leachate before discharge to the surrounding environment. There are several methods available for leachate treatment which can be adopted based on the age and composition of landfill. Coagulation-flocculation is commonly used for wastewater treatment due to ease of operation and process implementation. (Rivas et al, 2004).

Often times, treatment methods are combine to improve the efficiency of the treated wastewater before discharge to the environment due to different composition of leachate, one treatment method alone cannot be sufficient to achieve ideal treatment (Sha Liu, 2013). An integrated treatment processes are used together for a better performance, hence, this research work seek to investigate the use of combined Coagulation and Adsorption processes.

II. MATERIALS AND METHODS

A. *Collection of Leachate Samples*

Leachate samples were collected from Igwurita-Eneka dumpsite located in Ikwere Local Government Area of Rivers State, Nigeria. The dumpsite which is about eight years old measures 200m length by 425m width (85,000m^2) covering up to 8.5 hectares of land. The leachate sample were taken to laboratory and stored in a refrigerator at 4^{0}C temperature. The leachate sample was characterized for some important parameters.

B. *Preparation of Adsorbent*

Animal bones (typically of femur) were collected from slaughter house Trans-Amadi Port Harcourt, Rivers State. The bones were cut into small parts and washed with double distill water to remove fresh and dirt. The washed bones were dried at temperature of 120^{0}C for 24 hours using Tray Drier. The dried bones were carbonized on a muffle furnace at temperature of 600^{0}C for an hour. The bone char activation was carried out with the crushed bone char soaked into solution of sodium hydroxide (NaOH) for a period of an hour. The Bone char physiochemical/structural properties were analysed using standard methods.

C. *Pretreatment of Leachate by Coagulation*

Coagulation studies were performed using conventional jar test experiment by filling a beaker with 1 litre volume of leachate at room temperature. The coagulation experiment was carried out with Alum (aluminum sulphate) coagulants at different doses (1.5g to 3.5g) without prior adjustment to pH. The Coagulation experiment was done following three subsequent stages. The initial rapid mixing stage was performed at 160 rpm for 3 minutes. The slow mixing stage at 40rpm for 20 minutes and the settling stage for an hour. Mixing was terminated at settling stage and flocs allowed to settle. The supernatant solution was separated and analyzed for residual COD concentration and pH according to standard method. The experiment was repeated for the different coagulant dose.

D. *Analysis of Leachate Sample*

All analysis was performed in accordance with standard methods. The total dissolved solids, Total suspended solids, Biochemical oxygen demand, Chemical oxygen demand, Total hardness, Nitrogen, Fluoride, Nitrate were performed with APHA Standards for water and wastewater. The pH was determined with HACH pH meter and Heavy metals determined by AAS. All experimental procedures followed the EPA (2010) methods of chemical analysis.

E. Adsorption Equilibrium Studies

All the experiments were carried out at ambient temperature of 29±1^{0}C in batch mode. The experiment was first conducted at different contact time (5min, 10min, 15min, 20min, 25min) with 20mg/l of bone char adsorbent in 100ml of pretreated leachate sample to determine the optimum time of adsorption. The magnetic stirrer was on and leachate samples with the adsorbent shaken at an agitation rate of 150 rpm for the different contact time using 250ml flask. The samples were removed from the stirrer and poured into the adsorption set-up. The supernatant liquids were separated and analyzed for COD.

The experiment was repeated with varying adsorbent (bone char) concentrations from 5 to 25mg/l in the same 100ml of pretreated leachate samples at the optimum adsorption time.

III. RESULT AND DISCUSSIONS

TABLE I. CHARACTERIZATION OF LEACHATE SAMPLE AND BONECHAR ANALYSIS

S/N	PARAMETERS	VALUE
1	pH	7.9
2	Total Dissolved Solids (mg/l)	1776.0
3	Turbidity (ntu)	63.2
4	Total Suspended Solids (mg/l)	15.64
5	Total Hardness (mg/l)	1050.0
6	Nitrogen (mg/l)	18.62
7	Fluoride (mg/l)	<0.1
8	Sulphate (mg/l)	66.14
9	Nitrate (mg/l)	13.14
10	Phosphate (mg/l)	26.21
11	Ammonium ion (mg/l)	6.76
12	Biological Oxygen Demand (mg/l)	8.20
13	Chemical Oxygen Demand (mg/l)	192.0
14	BOD_5/COD	0.04
Metals		
15	Cobalt (mg/l)	0.014
16	Copper (mg/l)	0.004
17	Total Chromium (mg/l)	<0.001
18	Silver (mg/l)	<0.001
19	Lead (mg/l)	<0.001
20	Mercury (mg/l)	<0.001
21	Cadmium (mg/l)	0.028
Physicochemical Properties of Bone Char		
22	Moisture (%)	10.70
23	Ash (%)	81.40
24	Total Organic Carbon (%)	18.60
25	Chloride (Mg/G)	7000.00
26	Chloride (Mg/G)	7000.00
27	Calcium (Mg/Kg)	7711.00

The results of the fresh leachate when compared to the available standard range of landfill shows some significant quantities in concentration of parameters. The BOD and COD of the fresh leachate were very low and this also gives rise to low BOD/COD ratio. The fluoride concentration was considered initially because of its important in using bone char as an adsorbent for adsorption experiment. The result of the fresh leachate sample analysis for fluoride concentration shows negligible value of fluoride in the leachate. The result also shows very low value of Heavy metals in the leachate and hence COD was considered more than other parameters initially adopted for determination of the effectiveness of treatment process. The COD analysis was also adopted because of its relationship with other parameters such as biological oxygen demand, dissolved oxygen, turbidity etc.

The results of coagulation of leachate using alum (aluminum sulphate) coagulant at varying concentration were shown in fig. 1 below. The results show maximum removal of 49.3% from the initial BOD at concentration of 250mg/l and pH of 6. Hence, the effluent of 250mg/l concentration of alum from coagulation was subjected to adsorption. The effects of alum coagulants on the percentage COD shows gradual increase from initial dose of 50mg/l to its maximum at 250mg/l and decreased slightly from maximum to 350mg/l dose.

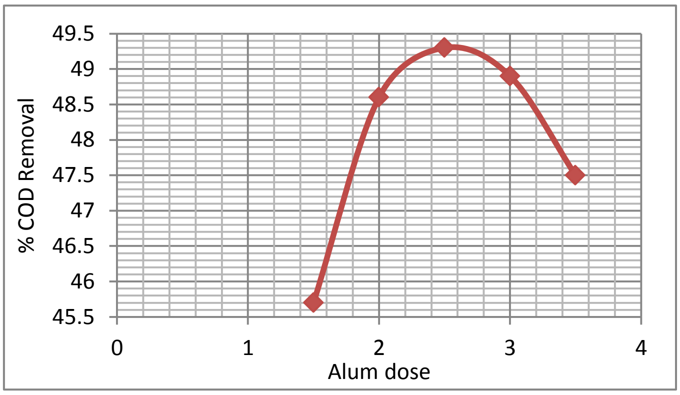

Figure 1. Plot of Alum Dose against % COD Removal

A. Coagulation and Adsorption

Adsorption a physical-chemical surface phenomenon is a process where multi components mixture is attracted to each other to forms attachments. Animal bones contain high composition of calcium, phosphate and magnesium minerals. The results of the physiochemical properties show high content of calcium and chloride. Although, phosphate and magnesium were not characterized, their presence in the activated bone char is not negligible. The presence of these ions generates positive and negative ions that can absorb dissolved pollutants in the leachate. The cations can eliminate effectively some inorganic non-metallic pollutants such as fluorides, sulphate etc. if present in the leachate. In addition, the coagulants (Alum) used for coagulation introduced some ion into the treated leachate. The effect is a reduction in the pH of leachate after coagulation which was found to be 4.7. The low pH indicates acidic media which is not favourable for absorption experiment. The pH is an important parameter in absorption experiment. The pH of 6 adopted for the adsorption process was to balance the effect of predominate negative or positive ion in the leachate. Negative charges predominate above the zero point and positive charges predominate below zero point. (Vimoses, 2011).

B. Effect of Contact time on Percentage COD

The result of % COD removal against contact time was shown in fig. 2. The results show an increase in % COD removal from 5mins to 10mins and decrease from 10mins to 15mins contact time. The % COD moved to its optimum of 86.7mg/l at 20mins contact time of the experiment and then decrease again after 25mins. Subsequent experiment were carried out at the optimum COD at 20mins contact time.

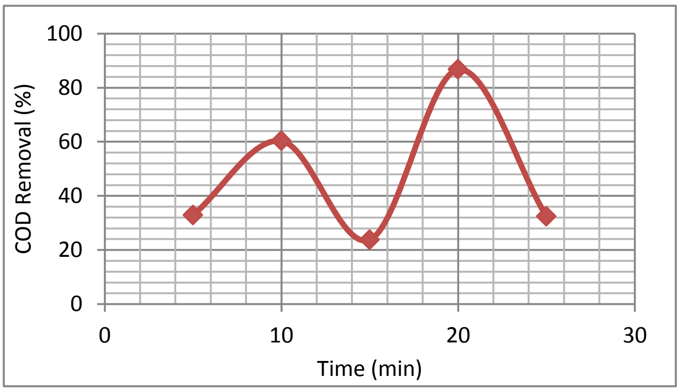

Figure 2. Effect of Contact time on % COD Removal

C. Effect of Concentration on Percentage COD Removal

Fig. 3 shows effect of concentration on COD removal efficiency on concentration at contact time of 20mins. The results show sharp increase in percentage COD of the combined coagulation and adsorption process from the concentration of 5g/l to 15g/l and slight increase as it tends to maximum. The results show good COD removal after treatment with adsorption. There was increase in COD as the adsorbent dose was increased.

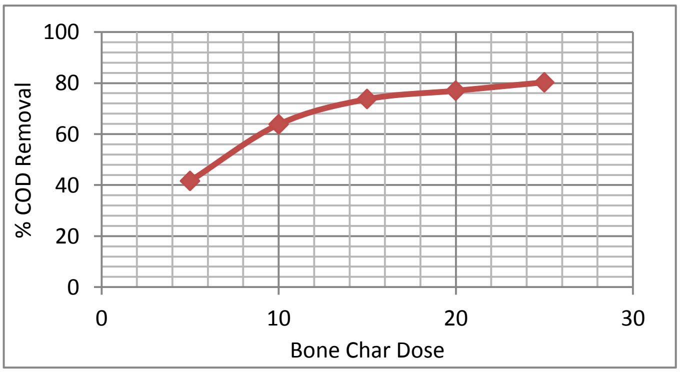

Figure 3. Plot of COD Removal against Bone Char Dose

D. Adsorption Isotherm Models

Freundlich and Langmuir adsorption isotherm are the two most common models to describe the equilibrium behavior of adsorption and were used to correlate the experimental data obtain from adsorption experiment.

In Freundlich isotherm models of fig 4, $K_f$the adsorption capacity of the bone char adsorbent was determined to be 2.04 $mg^{-0.1}g^{-1}$. The constant, n, is measure of deviation from linearity was found to be less than 1 which is true for adsorption as a chemical process. n a dimensionless constant found to be 0.91. A high regression of 0.9522 was obtained from the result and shows that the experimental data fitted the freundlich models.

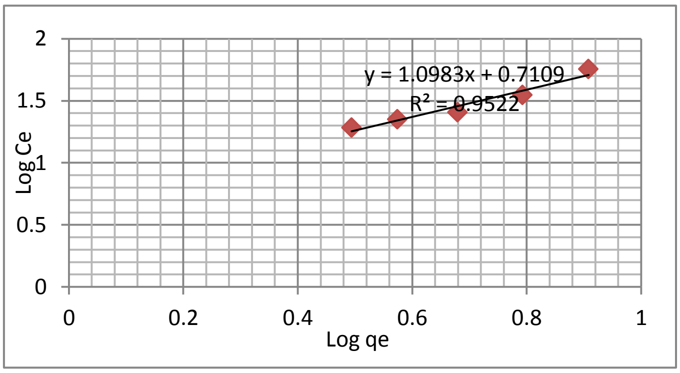

Figure 4. Freundlich adsorption isotherm model

Fig. 5 shows the results of Langmuir adsorption isotherm also correlated to obtain the adsorption capacity K_f and the maximum amount of adsorption (qmax). The adsorption capacity was found to be 1.2 x 10^{-3} and maximum amount of adsorption as 5000 mg/g respectively. A high regression coefficient was also obtained from data which shows that the experimental data was well fitted with the Langmuir isotherm model. A regression coefficient of 0.9588 was obtained from the relation.

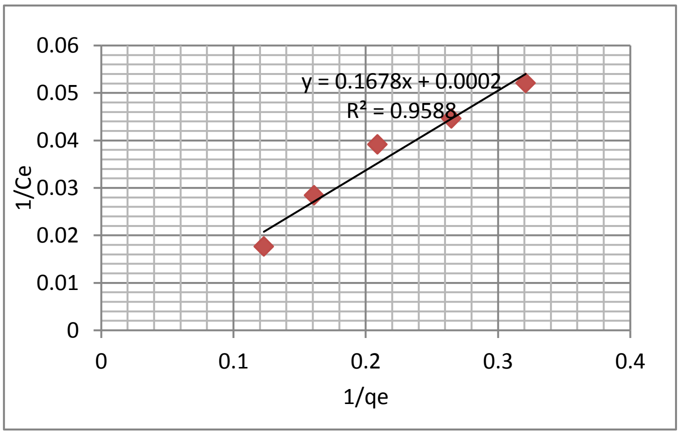

Figure 5. Freundlich adsorption isotherm model

IV. CONCLUSION

The combined treatment (Coagulation and Bone Char Adsorption) improved the efficiency of treatment. The removal efficiency was found to increase as the bone char dose was increased. The maximum COD removal of 80.3% was found at bone char concentration of 25mg/l.

The experimental data were fitted with Langmuir and Freundlich isotherm models and regression coefficients of 0.9588 and 0.9522 were obtained from the correlated data. Thus, the Langmuir and Freundlich isotherm model fit the experimental data. However, the regression coefficient was not good enough; experimental errors could have affected the value. Langmuir constant (k_i) and maximum adsorption (qmax) were$1.2x10^{-3}$ (1/mg) and 5000mg/g respectively. Adsorption capacity and measure of deviation, n, were 2.04 $mg^{-0.1}g^{-1}$and 0.91 for Freundlich isotherm model.

In general, the combined Coagulation (Alum) and Bone char Adsorption yields high efficiency and can be adopted for treatment of Igwurita-Eneka landfill leachate.

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