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Studying and Modeling Dynamic Adsorption of Lead (II) ion onto Fixed Bed Column of Activated Carbon of Plantain Peels and Bamboo

Uzoije Atulegwu Patrick[1], and Uche Cosmas[2]

[1,2,] Department of Environmental Technology,
Federal University of Technology Owerri Nigeria

Corresponding Author's Email: patuzong@yahoo.com.hk
Phone Number: 08061347535

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Abstract: Lead (Pb(11)) has been widely reported as a major environmental contaminants. Lead is recalcitrant, capable of forming complexes and bio-accumulate in the tissues of plant and animals. A number of adsorbent media and adsorption treatment techniques have been adopted to effect lead removal from wastewater. However, this study reported the decontamination of vegetable oil impacted wastewater containing Pb(II) by dynamic adsorption process using fixed column bed of plantain peel activated carbon(PPAC and bamboo activated carbon(BAC). Pb(II) adsorption Uptake performance of PPAC and BAC bed materials were represented on various breakthrough curves. Uptake capacity of Pb(II) was more phenomenal in PPAC bed material than on BAC. Column Adsorption removal efficiency of various bed materials was assessed at 50% breakthrough with efficiency of removal at 45-50%. In the case of mass transfer, film diffusion played leading roles on the mass transfer of Pb(II) to PPAC bed material. Internal and Kundsen diffusion played significant role on mass transfer of Pb(II) to BAC due to its supposed large pore sizes. Thomas, Yoon - Nelson, Adams-Bohart and Bed depth service time (BDST) models were applied to study the kinetics of adsorption dynamic of Pb(II) onto the bed materials. The models reasonably fitted the data, however the compliance of the models to the data is stated as follows; BDST >Thomas>Yoon-Nelson>Adams-Bohart. Breakthrough and bed service times of PPAC and BAC materials assessed at various saturation levels progressed as percentage of saturation moved from 20% to 90% through 60%. PPAC and BAC reasonably removed Pb(II) ion from vegetable oil polluted wastewater through dynamic adsorption process, hence an enduring approach to sustainable healthy environment.

Keyword: Breakthrough, Bamboo, Dynamic Adsorption, Modeling, and Plantain Peels

1.0 Introduction

Industrial revolution, born out of quest for better standard of living has taken a massive toll on all the environmental media. It is worthy to note that vegetable oil production is one of those industrial processes. Vegetable oil industry has contributed sizable strength to the economy of most African countries. The production is run in both large and small scale with huge revenue derived from it. Nevertheless, huge amount of wastes, ranging from liquid, semi-liquid and solid have been generated from this process (vegetable oil production). In most cases, the wastes are poorly handled as they are disposed on land and surface waters without proper treatment. With the wrong disposal methods, the waste will impact negatively on the lives of plants, animals, humans and even on the esthetics of the environment [1]. Quite a number of reports have been published on this subject matter; virtually all the processing units of a vegetable oil production system produce waste water of high organic matter content [2]. High chemical oxygen demand (COD), oil and grease, heavy metals such as lead (II) ion, dissolved and suspended solids, hexane and low dissolved oxygen of both the wastewater and the receiving body [3] have equally been reported. Excessive oil and grease in a receiving water body of vegetable oil waste impacted water has been widely reported to have impeded adequate aeration of the water body [4]. As a result, micro and macro marine lives die of suffocation occasioned by lack of oxygen and outright trachea blockages by the particles of oil and grease [5,6]. Remnants of Hexane, lead metal and other volatile organic solvents used for extraction during the process find their way into the receiving water body through the wastewater. These elements bio-accumulate in the tissues of the marine lives which translocated to human via food chain [7]. The suspended solids, oil and grease particles in the water body impairs visions of swimmers and prevent other recreationists from achieving fulfilled recreational activity [8]. Efforts to address these problems have been reported. Different wastewater treatment methods which include; precipitation, Flocculation, Sedimentation, Ion-exchange, Biological, reverse osmosis and adsorption have been developed and applied over the years with impressive results [9, 10, 11]. However, most of these techniques have some shortcomings which include; complicated treatment processes and high cost. Moreover, adsorption technique using activated carbon system has proved useful and efficient in waste water treatment due to ease of operation and cost effectiveness [12]. Adsorption onto activated carbon has been considered more superior technique for waste water treatment because of its capability to adsorb a broad range of different types of pollutants (adsorbents). For the purpose of treatment by solid – liquid adsorption, Activated carbon is usually obtained from materials with high carbon content, large pore distributions, large specific surface area and great adsorption capacity [13, 14]

Although pre-analysis of the entire pollutant components of the vegetable oil wastewater will be carried out, but the treatment will be focused on the component(s) of the vegetable oil polluted wastewater with alarming value(s). Dynamic adsorption process having activated carbon of

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bamboo and plantain peels as fixed packed bed column materials will be adopted to study treatment efficiencies on the wastewater. Physical properties, adsorption capacities and behaviors of the two adsorbents through exhaustive, breakthrough times, and breakthrough curves will be presented. Compliance of the fixed bed adsorption models (Adam's – Bohart model, Thomas model and Yoon-Nelson model) with the data obtained, to study the kinetic behavior of the fixed bed adsorption column systems will be tested.

2.0 Materials and Methods

Pre-analysis of wastewater collected from the Vegetable oil Industry was carried out using standard methods to determine values of the constituents before column fixed bed adsorption process. Results of the pre-analysis showed that high value of lead(II) ion was a concern, thus the column adsorption process of this study was directed towards lead(II) removal. The feed-bed column experimental set-up consists of a reservoir connected to a column parked with plantain peel activated carbon (PPAC) through standard process. Underneath the bed is a glass fiber wool meant to support the activated carbon.

Vegetable oil impacted Wastewater from the reservoir was allowed to pass through the packed adsorption column through gravity and a control valve.

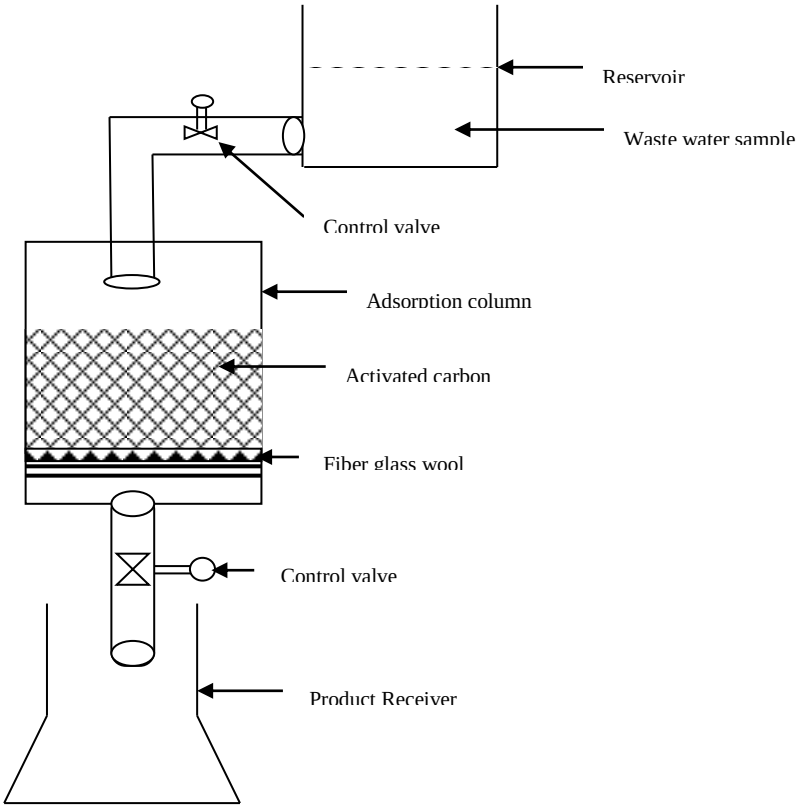

Figure 1: Schematic diagram of experimental set-up of fixed bed adsorption column.

Effluents from the column were collected under three distinct conditions; varying initial concentrations at constant bed height and flow rate, varying bed heights at constant initial concentration and flow rate and varying flow rates with constant initial concentration and bed height. The effluent samples were also collected at specified Breakthrough time intervals till exhausted periods were achieved. Thereafter, analysis of lead (II) ion on each collected sample was carried out. The entire run was repeated with an adsorption system packed with Bamboo activated carbon (BAC).

2.1 Fixed bed adsorption kinetics

Column bed adsorption models were generally developed from the mass balance of solute in a bulk flow through adsorption bed length over a time duration to study the kinetics of solid-liquid phase dynamic adsorption models such as Yoon and Nelson, Thomas, Adam Bohart and Bed Depth service time models are generic in describing the transport of bulk solution containing the pollutant(s) meant to be removed (adsorbate) and mass transfer of the pollutant to the solid phase (adsorbent) ([15].

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These models have been robustly used as tools to model breakthrough curves of an adsorption system, estimate the breakthrough behaviors and effects of various adsorption variables using the experimental data[16]. However, the validity of a models is limited to the range of conditions applied[16]These models were formulated on the basis of different assumptions and as basis on which predictions of vital properties of the column bed adsorption systems are made through the relationship between concentration ratio $\left(\frac{C_0}{C_t}\right)$and the adsorption time (t) [17,*16,* 18]

2.1.1 Yoon and Nelson model:

This model explains the decrease in the possibility of adsorbate adsorbing on the adsorbent as being proportional to the possibility of adsorption of the adsorbate and possibility of its breakthrough from the system [19]. Yoon and Nelson model has the following attributes [20]

- Simple and extremely concise in form
- Requires no detailed data about the properties of the fixed bed adsorption system
- It is convenient to obtain process variables and can easily predict adsorption processes under different conditions.

The linear form of Yoon and Nelson model is represented as follows;

$$ln\left(\frac{C_t}{C_0-C_t}-1\right)=\tau K_Y-K_Y t \quad (1)$$

Where $K_Y(1/min)$ represents the Yoon and Nelson adsorption constant, C_t is the concentration of effluent adsorbate with time , C_0 initial concentration of the adsorbate, $\tau(min)$ is the adsorption time and is the time require for 50% adsorbate breakthrough. I.e $\left(\frac{C_0}{C_t}\right)$ is 0.5 at $t_{1/2}$= . It is also important to note that the fixed bed adsorption system is completely saturated at t = 2 [21]. A plot of $\frac{C_t}{C_0-C_t}$ Vs t produces a linear curve with τ and K_Y as intercept and slope respectively at various bed height, inflow rate, and initial concentration input variables

2.1.2 Adams and Bohart model:

Adams and Bohart proposed this model on the assumption that uptake rate of the adsorbate (contaminant material) is proportional to the concentration of the contaminant in the bulk fluid. Also the model assumes that adsorption is a continuous process and does not get to equilibrium instantly. The model describes the initial adsorption process of adsorbing contaminant material onto the adsorption system. Adams and Bohart is expressed in linear form as

$$In\left(\frac{C_0}{C_t}-1\right)=K_A N_0\frac{Z}{V}-K_A C_0 t \quad (2)$$

K_A and N_0 Represent Adams and Bohart adsorption constant and the maximum adsorption capacity of the adsorbent respectively. Z is the bed height and V becomes the inflow rate of the solution. The aforementioned variables which represent the properties of the adsorption system are determined form the slope and intercept of a linear curve of $\frac{C_t}{C_0}$ Vs t.

2.1.3 Bed depth service time (BDST)

BDST model was derived from Adam – Bohart model by re-arranging equation 2 to solve for time, known and called BDST which is represented as follows;

$$t=\frac{N_0}{C_0 V}Z-\frac{1}{K_B C_0}ln\left(\frac{C_0}{C_t}-1\right) \quad (3)$$

In that regard, it was already presumed that BDST model has conformed with the assumptions of Adam-Bohart model of adsorption rate as being proportional to the residual adsorption capacity of the adsorbent material and the solute concentration of the bulk fluid. The model has been widely applied in a number of adsorption processes to predict the design and performance of dynamic adsorption system (fixed bed adsorption column). For instance, BDST showcases the prediction of relationship between bed depth and service time to optimize vital properties of the adsorption system such as maximum adsorption capacity, Kinetic constant, flow rate etc. [22, 23]. BDST model is analogous to a simple linear model of

$$= . \qquad Y = mx + C \quad (4)$$

Comparing equations 3 and 4, Y represents the bed depth service time(t), while X is the bed depth(Z) The slope, m is related to

$$\frac{N_0}{C_0 V} \quad (5)$$

and the gradient C is likened to

$$. \qquad \frac{1}{K_B C_0}ln\left(\frac{C_0}{C_t}-1\right) \quad (6)$$

By obtaining N_O ,V *and* K_B values, BDST could be estimated by mere knowing the bed height value. Also, design parameters like N_O *and* V could be determined using equation 4. BDST model could be formed at different

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levels of column saturation (20%, 60%, 90% etc) and various design parameters equally determined accordingly [22].

2.1.4 Thomas model:

Thomas model has performed well in describing breakthrough curves and optimizing properties of adsorption systems for many adsorption processes [16, 24,25]. This model seems to be the only model among the ones mentioned, which was proposed by number of assumptions [26, 16,27]. The assumptions include;

- That the fixed bed column must have negligible axial and radial dispersion
- That the column bed material must have constant void fraction
- That the column adsorption system must allow condition of isothermal and isobaric process to thrive
- That there is no change in the physical properties of the adsorption system
- That the model can estimate adsorption processes at negligible external and internal diffusion resistances
- That the adsorption process be described by Pseduo-second order kinetics and Langmuir isotherm.

Thomas has the mathematical expression of dynamic adsorption column as follows;

$$ln\left(\frac{C_0}{C_t}-1\right)=\frac{K_0 q_0 M}{Q}-K_0 C_0 t \qquad 7$$

Where $C_{0\,and}\,C_t$ are concentrations at inflow and outflow respectively, $K_{0,}q_0, M\ and\ Q$ represent Thomas adsorption constant (ml/min.mg), maximum adsorption capacity (mg/g), adsorbent mass (g) inflow velocity (ml/min) respectively and t is the flow time (min). From the slope and intercept of the linear plot of $ln\left(\frac{C_0}{C_t}-1\right)\ Vs$ t properties of the adsorption system such as $K_{0\ and}\,q_0$ will be determined to predict the column bed performance

2.1.5 Column data Analysis;

Analysis of column data (parameters) is essential for enduring bed design. The information keeps the designer abreast with the size and types of bed material (adsorbent)by computing the total quantity of the solute adsorbed in the column and adsorption system efficiency as follows; the total quantity adsorbed on the column is generally presented as the product of the area under the breakthrough curve of concentration ratio $\frac{C_0}{C_t}$ Vs time and the flow rate expressed as

$$.q_{Total}=\frac{F\,A}{1000}=\frac{F}{1000}\int_{t=0}^{t=t_{max}} C_{ad\ dt}$$
6

Where $C_{ad\ dt}$ represents the area under the curve.

Solute uptake capacity of the adsorbent (Q) is expressed as the ratio of the total quantity of the solute sorbed on the column and mass of the adsorbent(M) expressed as ;

$$Q=\frac{q_{Total}}{M} \qquad 7$$

Total amount of solute (adsorbate) sent to the column or adsorption system can be expressed as

$$W_{Total}=\frac{C_0 F t_e}{1000} \qquad 8$$

Where t_e represents the column exhaustion time.

Column Adsorption removal efficiency is expressed as

$$C_{ARE}=\frac{q_{total}}{W_{total}}\times 100 \qquad 9$$

Other column parameters are the sorption zone Δt and length of mass transfer zone L_M are calculated as follows;

$$\Delta t=t_e-t_b \qquad 10$$

Where t_b represents breakthrough time.

$$L_M=L\left(1-\frac{t_e}{t_b}\right) \qquad 11$$

L is length of the column.

3.0 Results and Discussions

3.1 Characterization of activated adsorbents;

Results of the characterization of activated carbon (BAC for BA and PPAC for PL were used interchangeably especially on the diagrams) used as adsorbents in this work are presented in table1. There were glaring differences in the values of the analyzed parameters between the two adsorbents. This disparity could be attributed to natural structural make-up peculiar to each adsorbent. Bulk density property of BAC was observed to be higher than that of PPAC. Previous studies observed that low bulk density adsorbents have strong affinity for adsorbates in aqueous solution due to the perceived large number of pore distributions that characterize low bulk density adsorbents [28, 29, 30]. Given the bulk density values shown on table 1 adsorbates may show stronger affinity to PPAC than BAC adsorption system.

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Table 1 Physical Characteristic of BAC and PPAC

Parameters	BAC	PPAC
pH	6.51	6.74
Bulk density (kg/m^3)	360	197
Porosity	1.96	1.18
Pore volume (Cm^3/g)	0.098	0.059
Ash content %	3.69	1.60
Particle size mm	1.90	1.70
Moisture content %	2.72	2.15
Surface Area m^2/g	738	826
Iodine number %	21.1	25.3

The Ash contents were observed to be higher in BAC than PPAC. Ash content residue in activated carbon media has been considered an impurity hence used as criteria to rank the quality of activated carbons[31] . Thus, high ash content value gives an impression of low quality adsorbent [32]. In that light, high ash content residue may affect adsorption capacity of BAC adsorbent. Other properties such as porosity, surface area, particle size and pH play determinant roles in adsorption process. Depending on the molecular size of the adsorbing substance, high porosity and particle size values place the adsorbent on a pedestal of high adsorption capacity [33,34] . However, large molecular sized adsorbate can experience adsorption difficulty on a small porous and particle sized adsorbent due to inaccessibility of large adsorbate molecules hence poor adsorption [35, 28]. In this study, BAC has higher porosity value than PPAC whereas the reverse is true for the particle size. This implies that the small molecular sized adsorbate will easily adsorbed on PPAC adsorbent. The results also show remarkable differences in the surface areas of the adsorbents. Large external surface was observed on PPAC adsorbent. In line with the previous studies, one will expect higher adsorbate uptake on PPAC than on BAC due to availability of more adsorption sites [36, 22]. There was slight variation in pH values of BAC and PPAC adsorbents. pH value of BAC is lower than that of PPAC . Lower pH value usually reduces uptake drive of most adsorbents [37, 38]. Although the difference is not remarkable, BAC was expected to have lower uptake drive than PPAC going by the reported observations.

3.2 Characterization of the Adsorbate

The adsorbate (vegetable oil polluted wastewater) used for this work was analyzed based on its active ingredients. The results of the analysis are presented on table 1. Values of Conductivity, turbidity, salinity and oil and grease were in alignment with WHO recommended values for waste water and therefore not considered in subsequent discussions. However high presence of lead (Pb^{2+}) in analyzed wastewater was high and a source of concern viz-a-viz its negative environmental implications. Lead is known for its recalcitrant nature especially in soil environment [39, 40, 41]. It has a profound ability to form complexes with other compounds at slightest change in pH environment [42]. Health issues such as mental retardation, reduction in heamoglobin and interference with normal cellular metabolism have been widely reported as being caused by lead [40]. In that regard, discussions of results will be focus on the high presence and removal of lead (II) ions by column adsorption systems.

3.3 Fixed-Bed Column Adsorption Study

Breakthrough Time is a key factor in evaluating the dynamic behavior of a column adsorption process. Adsorption behavior of a solute substance onto the fixed packed bed is studied by concentration ratio $\frac{C_0}{C_t}$ with respect to time. The curve produced by this relationship is termed the breakthrough curve. As the solute solution is introduced into the packed bed, three distinct zones are formed. These zones are the saturate adsorbent zone, Mass transfer zone and fresh unsaturated adsorbent zone [33]. The saturated zone is an area where no adsorption takes place because the entire pores in the zone are completely occupied following previous adsorptions. In Mass transfer zone, adsorption process is usually ongoing because adsorbent pores in the zone are not completely saturated. On fresh unsaturated zone of the packed bed, no adsorption process takes place because the wave front of the solute solution is yet to get to the zone. As adsorption time progresses, the wave front moves forward across the packed bed leaving behind large portion of saturated zone. At the point the wave front covers the entire packed bed that is when the saturated zone has moved to the end of the bed column, the entire adsorption system is said to be at equilibrium. In that case effluent concentration of the solute (C_t) becomes equal to the initial concentration(C_0). There are two basic contact times in column adsorption process. These are; the breakthrough time t_b and the exhaustive time t_e . At

exhaustive time, the adsorbent is completely saturated and at this point the adsorbent medium is unable to take up more solutes. in this study, the breakthrough curves which represent the adsorption behavior of lead(II) ion in the vegetable oil polluted wastewater, onto adsorption systems of BAC and PPAC were expressed for a given adsorbent mass, initial concentration and flow rate, and are shown on figure 2. It was observed that shapes of the breakthrough curves vary with the adsorbent media and other input factors such as adsorbent height, initial concentrations and inflow rates of the solute solution. The effects on the varying input factors will be discussed on the subsequent headings. The breakthrough curves which described adsorption behavior of lead molecules onto PPAC adsorption systems are less steep than the curves of BAC systems due to delayed breakthrough and exhaustive times.

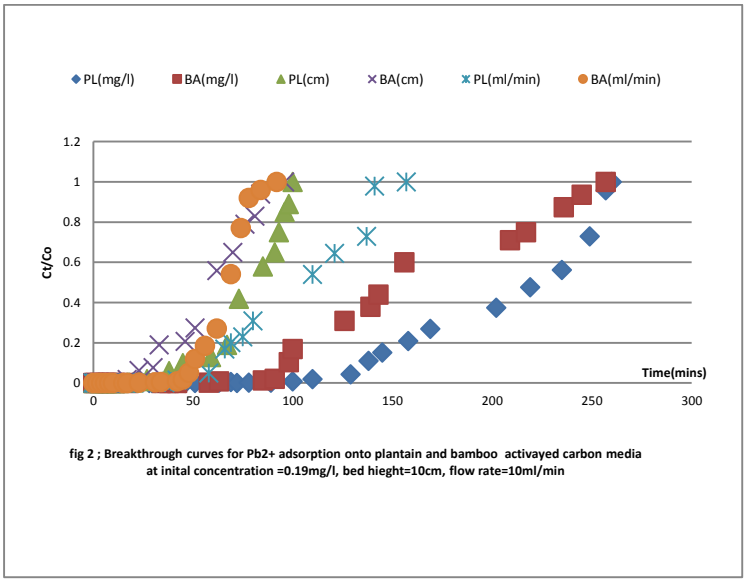

fig 2 ; Breakthrough curves for Pb2+ adsorption onto plantain and bamboo activayed carbon media at inital concentration =0.19mg/l, bed hieght=10cm, flow rate=10ml/min

The difference in shape of the breakthrough curves could be attributed to variations in breakthrough and exhaustive times of the adsorbents due to differences in the physical structures of the adsorbing media [30, 43]. Considering the huge role large surface area and large small particle size distribution play in driving adsorption process, the apparent flat shapes of breakthrough curves observed for PPAC adsorption system was expected because prolonged interaction periods between the Pb(II) solute with the adsorbent due to large surface area and high small particle sizes distribution[resulted to delay in Pb(II) solute breakthrough or increase in breakthrough and exhaustive times with consequent flat breakthrough curves and large solute uptake44, 45] . On the other hand, breakthrough curves for BAC adsorption systems are steeper in shape, suggesting that the lead molecules experienced faster breakthrough because the BAC adsorption medium is predominantly of large particle sizes and smaller surface area. The same reasons were adduced for low adsorption of Cu(II) ions on, pinapple peel and commercial activated carbons[43]. This observation also corroborated with that of where large surface media achieved high capacity for solute adsorption [46, 47]**.** It was expected that BAC would have high uptake capacity for the lead (II) given its large porosity and the perceived large molecular size of the adsorbate (lead molecule), but the study showed contrary results. The prevailing relationship could be predicated on the fact that external adsorption(film diffusion or adsorption at the exterior surface of the adsorbent) by PPAC system took upper hand due to its small particle and pore sizes and large surface area dispositions, whereas in BAC adsorption system, internal and Kundsen diffusion played significant role because the pore sizes of the of BAC are presumably larger than the diameter of the lead molecules thereby making external adsorption relatively difficult[48, 30, 49].

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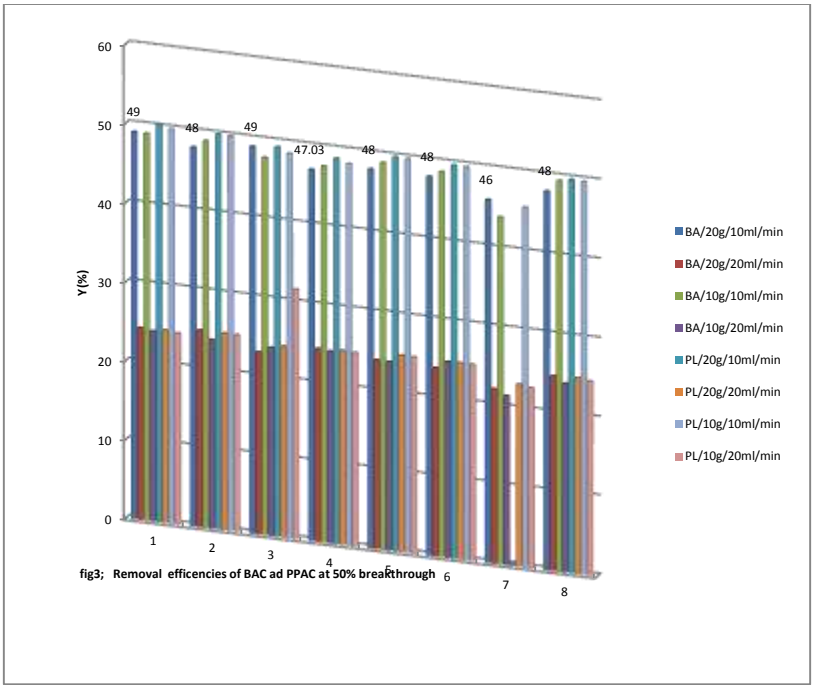

fig3; Removal efficencies of BAC ad PPAC at 50% breakthrough

It also presupposes that high ash content and low pH attributes of BAC adsorption system have taken a toll on the integrity of its structure and adsorption capacity as poor Pb(II) uptake was apparent on the system[37, C1] removal efficiencies of PPAC (PL) and BAC (BA) at various adsorptions were showcased on figure 3. Removal efficiencies of the two adsorbent were computed at 50% breakthrough and their efficiency values were between 40- 50%. The figure also showed higher removal efficiency of Pb(II) ion for PPAC adsorption medium at high bed height and low inflow rate than that of BAC. This confirmed high Pb(II) uptake capacity attributed to PPAC adsorption system. In industrial scale application, adsorbents at 50% breakthrough point can still operate until at 90% breakthrough point, therefore the prevailing efficiency value is quite encouraging as the adsorbents have the potentials to operate to maximum efficiency[50].

3.4 Effects Of Bed Heights On Adsorption.

The column Adsorption process of Pb(II) was further studied by varying the bed heights from 10cm to 20cm for BAC and PPAC at constant flow rate of 20ml/min and constant initial concentrations of 0.48mg/l of lead (II) ion in the wastewater. Figure 4 shows the breakthrough curves obtained from the process of adjusting the column bed height at each run. From The figure, breakthrough curves represented by adsorption systems with higher bed height are less steep, meaning that the slope of the breakthrough curve decreased with increase in bed height.

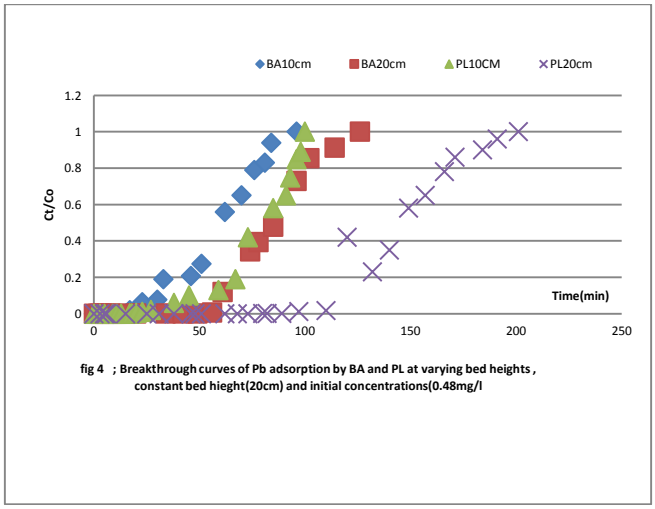

fig 4 ; Breakthrough curves of Pb adsorption by BA and PL at varying bed heights , constant bed hieght(20cm) and initial concentrations(0.48mg/l

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With less steep curves observed, increase in bed height of PPAC and BAC adsorption systems has made available specific surface areas and active sites for robust adsorption of Pb(II) and consequent increase in adsorption zone, breakthrough and exhaustive periods. Also effluent concentrations of each wastewater constituents from the adsorption systems of high bed height (20cm) were low, meaning that large adsorption uptake of (Pb(II)) took place on the adsorbents with small amount of the Pb(II) pollutant at the effluent . The observed variations in the uptake capacities of the adsorbents could be linked to the earlier discussed mass transfer zones. it is apparent that the values of mass transfer zone of high adsorbent is large due to high bed column, in so doing providing high opportunity for (Pb(II)) to move through large adsorption zone and to make good contacts with granular activated carbon of the adsorbents[51, 52]. Effects of variations of column bed height were evident on various levels of adsorption system performance predicted by the following evaluated parameters of the breakthrough curve and exhaustive and breakthrough times as shown on table 2. Values of the evaluated parameters were higher at 20cm bed height than at 10cm bed height because high bed height provided wide surface area for large to thrive. It also increases the adsorption and mass transfer zones of column. With higher bed height, sufficient active or binding sites were made available for prolonged interaction of the lead(II) ion with the bed material, thereby delaying breakthrough and exhaustive periods of the bed. Reduction in values of the evaluated parameters accounts for inadequate active or binding sites occasioned by low bed height of 10cm. insufficient active sites reduced the chances of adsorption lead molecules on the receiving bed material. This led to quick breakthrough and exhaustive periods and reduction in values of other evaluated parameters.

Table 2. Computed Column Bed Parameters

Adsorbents	**Bed heights(cm)**	$q_o(\frac{mg}{g})$	W_{total}	C_{ARE} %	$t_{e(\min)}$	$t_{b(min)}$	(Δt)	L_M
BAC	20	0.15	0.22	66	213	20	193	18.12
	10	0.10	0.19	52.6	120	12	108	9.0
PPAC	20	0.6	0.85	70.56	241	45	196	17.08
	10	0.36	0.61	57.03	225	20	205	9.2
	Flow rate(ml/min)	$q_o(\frac{mg}{g})$	W_{total}	C_{ARE} %	$t_{e(\min)}$	$t_{b(min)}$	(Δt)	L_M
	10	0.2	0.96	21	220	30	190	17.27
	20	0.16	0.92	17.4	96	20	76	15.83
BAC	20	0.12	0.20	60	80	8	72	18
PPAC	10	0.89	1.2	74	275	60	215	21.8
	Initial concentrations	$q_o(\frac{mg}{g})$	W_{total}	C_{ARE} %	$t_{e(\min)}$	$t_{b(min)}$	(Δt)	L_M
	0.48	0.720	1.9030	45.56	230	25	205	17.82
	0.19	0.108	0.421	25.65	61	17	44	14.43
BAC	0.49	1.536	3.07	50.23	360	44	361	17.22
PPAC	0.19	0.475	1.06	44.8	280	10	270	19.30

Axial dispersion, usually occurs at relatively low bed height and high inlet adsorbate solution flow rate because of inadequacy of binding site created by low bed height [50,53] . Rather than dispersing the Pb(II)ion evenly in the case of wide surface area of the bed matrix, it concentrated on small surface provided by low bed height. The axial dispersion effects immobilize factors which drive effective adsorption/diffusion such as contact time and even distribution of solute solution [54, 55]. In the present study, even distribution and adequate contact time of lead molecules were stifled at reduced bed height due to small surface area. low lead adsorption capacity of the system having low bed height of 10cm, could also be attributed to large external film diffusion resistance due to

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reduced exterior surface of the bed material [56, 57]. In other words, the film resistance impeded external film diffusion of the lead (II) on the bed materials.

3.5 Effect of Feed Flow Rate on Adsorption

Effects of varying the inlet flow rate of wastewater into the column fixed bed from 10ml/min to 20ml/min was also studied at constant adsorbent dosage of 20 cm and initial concentration of 0.48mg/l. Figure 5 illustrates various breakthrough curves for BAC and PPAC adsorption systems at inlet flow rates of 10ml/min and 20ml/min. The figures show different levels of breakthrough positions for each flow rate into BAC and PPAC adsorption systems. Gradients of Curves for high flow rates are higher i.e. they are steeper than that of low flow rates. The high gradient curves portray fast column exhaustion and saturation because it took short contact time between the lead pollutant and BAC and PPAC adsorbents with consequent reduction in adsorption, unlike in a low flow rate where the wastewater constituents had adequate contact time with adsorbents.

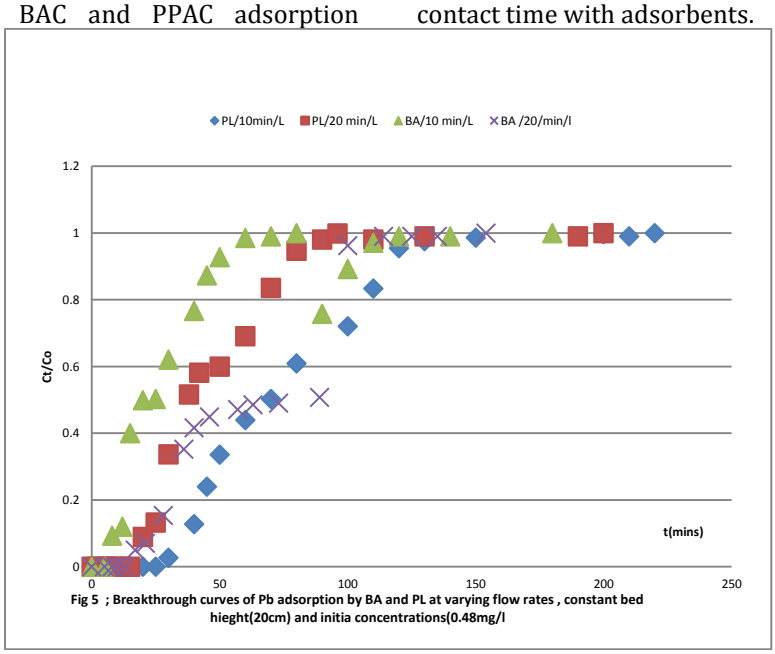

Fig 5 ; Breakthrough curves of Pb adsorption by BA and PL at varying flow rates , constant bed hieght(20cm) and initia concentrations(0.48mg/l

With high flow rate, Pb(II) made little contact with the adsorbents and vise verse. The little contact period reduced the adsorption of lead (Pb (II) thereby, quickening exhaustion and breakthrough times of the adsorption systems with steepness of the breakthrough curves [58, 59]. On the other hand, low inflow rate of the adsorbate (wastewater) provided adequate contact of the adsorbate (Pb(II)) with PPAC and BAC adsorption systems, with Consequent reduction in exhaustion and breakthrough periods, broad adsorption zone and flat breakthrough curves. This equally gave account of low effluent concentrations of the Pb(II) measured as relative concentrations (Ct/Co) at low flow rates. Variation of inflow rates of the adsorbates also influenced other evaluated parameters as shown on table 2. Values of Total quantity of solute (lead molecule) sorbed by various adsorbents (BAC and PPAC), adsorption removal efficiency and mass transfer zone increased at low inlet flow rate and vise verse. Whereas the prevailing value increase of the parameters could be attributed to sufficient solute (lead molecules) contact with the adsorbents (BAC and PPAC) at low inflow rate, the reduced values were obtained at high inflow rate. The present observation aligned with that of the previous ones [58, 50, 22]. In terms of mass transfer, high inflow rate does not favor both external and internal particle diffusion because at high flow rate, the solute molecule builds insufficient film thickness on the adsorbent and also intra-particle diffusion (internal diffusion) is usually impaired due to lack of adequate interactions of the solute molecules with adsorbents and vise verse[60, 29]. Decrease and increase in values of and at inflow rates of 20ml/min and 10ml/min respectively, implied that there were larger lead film thicknesses at low inflow rate(10ml/min) than at the flow rate of 20ml/min. this observation also corroborated the studies of Hameed et al 2009[60] and Maiyalagan and Karthikeyan 2013 [61]on the film- pore diffusion modeling for sorption of Azo dye exfoliated graphics on Nanoplatelets.

3.6 Effects of Initial Concentrations;

Variations of initial concentrations from 10mg/l to 20mg/l at constant inflow rate (20ml/min) and bed height (20cm) was adopted to study the adsorption dynamic behavior of BAC and PPAC systems. Adsorption behaviors of each system observed at different initial concentrations are represented by the breakthrough curves shown on figure 6. Observed from the figure were flat breakthrough curves for initial concentration of 20mg/l whereas the steep curves showed the adsorption behavior of a process carried out at 10mg/l initial concentration.

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The breakthrough curves were flat in various adsorption systems because of increase in adsorption time due to apparent availability of more adsorption sites provided by high inlet of initial concentration [57, 62].

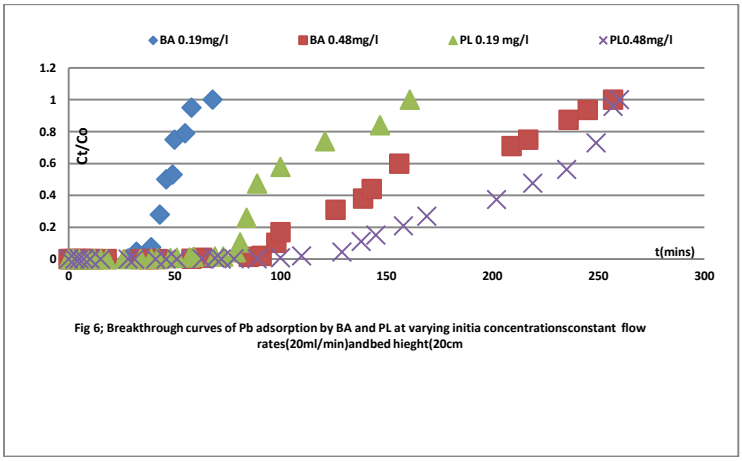

Fig 6; Breakthrough curves of Pb adsorption by BA and PL at varying initia concentrationsconstant flow rates(20ml/min)andbed hieght(20cm

Also, high initial concentration of 20mg/l created a driving force for mass transfer of the Pb(II) , by so doing increasing breakthrough and exhaustive times, broadening the mass transfer zone with high lead (II) adsorption and adsorption rate efficiency [55, 51] . These factors were determined and presented on table 2. Inlet of the adsorbate (lead II) at lower initial concentration of 10mg/l consequently provided reduced amount of adsorption sites with quick adsorption process, breakthrough and exhaustive times and decrease in the total amount of lead molecules adsorbed, narrowing of mass transfer zone and reduction of adsorption rate efficiencies of various adsorbents [50].

3.7 Dynamic adsorption models;

In dynamic adsorption processes for water purification, kinetic models are usually applied to determine the principal design values for adsorption systems. In this study, Yoon and Nelson, Thomas, Adams-Bohart and BDST models were applied to ascertain their levels of compliance with adsorptions of Pb(II) ion onto PPAC and BAC adsorption systems.

3.7.1 Yoon and Nelson

Yoon and Nelson, was applied to analyze the behavior of various adsorption systems at different adsorption conditions. Adsorption data obtained from various adsorption systems were fitted into Yoon and Nelson model to determine the time for 50% adsorbate breakthrough and the rate constant by computing the slopes and intercepts respectively of the following linear relationship; $ln\frac{C_t}{C_0-C_t}$ Vs t. see figure 7.

Table 3 ;parameters and Yoon-Nelson Model values

	Bed heights(cm)	$Q\left(ml/_{min}\right)$	C_o	τ	K_y	R^2
	Yoon-Nelson Model					
BA	20	20	0.48	32.97	0.16	0.91
	20	10	0.48	64.01	0.15	0.97
PL	20	20	0.48	91.48	0.103	0.97
	20	10	0.48	92.29	0.069	0.69
PL	10	20	0.48	78.16	0.096	0.92
	20	20	0.48	150.06	0.089	0.97
BA	20	20	0.48	88.39	0.1189	0.98
	10	20	0.48	60.52	0.1189	0.85
PL	20	20	0.48	205.49	0.048	0.93
	20	20	0.19	109.89	0.0907	0.89
BA	20	20	0.48	171.64	0.046	0.87
	20	20	0.19	60.20	0.156	0.89

The computed values are summarized on table 3. There were increase in values of at bed heights increase and at inflow rate decrease.

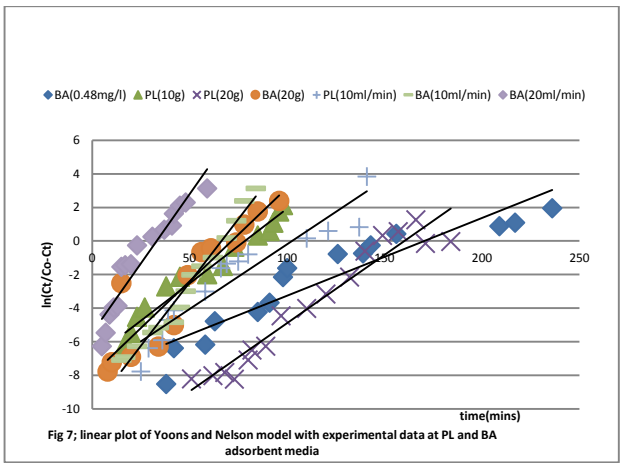

Fig 7; linear plot of Yoons and Nelson model with experimental data at PL and BA adsorbent media

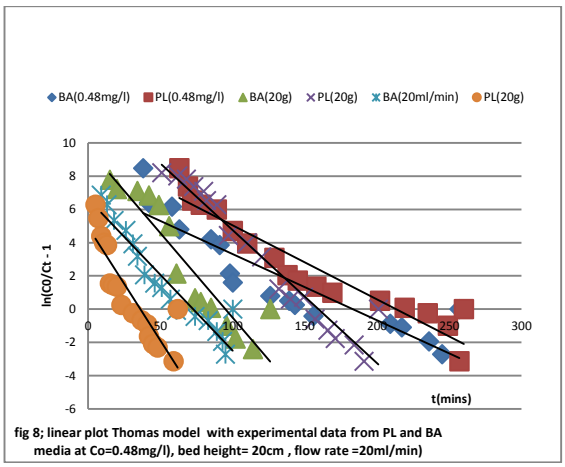

fig 8; linear plot Thomas model with experimental data from PL and BA media at Co=0.48mg/l), bed height= 20cm , flow rate =20ml/min)

Rate constant values were observed to decrease at low inflow rate and high bed height. This could be attributed to long lead – adsorbent contact period occasioned by low inflow rate and availability of surface contact area due to increase in bed height [63, 31]. At high initial concentration, values $\boldsymbol{\tau}$ of were also high in adsorption systems of PPAC and low in BAC because PPAC contains more active sites for interactions with the lead ions, hence the delayed adsorbate breakthrough [35, 64]. The correlation coefficients observed for various factors and adsorption systems showed good data fit to the model.

3.7.2Thomas model

The maximum adsorption capacity and adsorption constants of the systems at various conditions were further determined by applying Thomas model to the adsorption data in which a linear relationship of $ln\left(\frac{C_0}{C_t}-1\right)$ Vs t was obtained and represented on figure 8. The determined values shown on table 3 varied with changes in the input variables. There were increase and decrease at increase in bed height.

Table 3; Values of Input and Thomas Model Parameters

	Thomas models					
	Bed heights(cm)	$Q\left(ml/\right.$	C_o	q_o	K_o	R^2
B A P L	20 20 20 20	20 10 20 10	0.48 0.48 0.48 0.48	15. 59 15. 94 31. 94 54. 43	0.3 3 0.3 11 0.2 2 0.1 63	0.9 04 0.9 7 0.9 6 0.9 0
B A P L	10 20 20 10	20 20 20 20	0.48 0.48 0.48 0.48	54. 80 62. 41 79. 30 76.	0.2 5 0.2 5 0.1 88 0.2	0.8 5 0.9 4 0.9 0 0.9

				89	06	1
B	20	20	0.48	78.	0.1	0.9
A	20	20	0.19	50	0	0
				49.	1.3	0.9
P	20	20	0.48	33	0	6
L	29	20	0.19			
				98.	0.1	0.9
				74	0.5	2
				19.	1	0.9
				60		2

Availability of active and binding sites resulting to more Pb (II) contact and interaction time with the adsorbent as a result of increase in bed height, gave rise to the glaring adsorption capacity increase of the adsorption system [48]. Also, increase in bed height resulted to broadening of mass transfer zone and consequent high adsorption capacity of the system [54]. High adsorption capacity of the bed column could be attributed to negligible axial and radial dispersion and negligible external and internal diffusion [5, 64, 65, 17].

In a similar manner, change of initial concentrations from 10ml/l to 20ml/l raised and lowered the capacity of the adsorption system and Thomas constant values respectively. The observed adsorption capacity increase is predicated on the established fact that high concentration gradient between adsorbing Pb(II) ion and the bulk solution, occasioned by increase in initial concentration drives the adsorption processes[28, 55, 37]. On the contrary, inflow rate variations of the adsorbate solution (Pb(II)) from 10 to 20ml/min reduced adsorption capacities of the system and increased the values of the rate constant. The fast movement of the solution out of the system before column saturation was established, consequently led to a decrease in the adsorption capacity of the system and quick movement of adsorption zone [17, 52, 22]. High values of observed at various conditions and adsorption systems showed substantial compliance of adsorption data with model.

3.7.3 Adams Bohart model:

Adams-Bohart model was applied on the experimental data to test its compliance with adsorption process of lead (II) ions onto PPAC and BAC adsorption systems. In the process, linear relationships of $\frac{C_t}{C_0}$ Vs t were obtained, with adsorption capacity and Adams-Bohart constants values computed at various varying adsorption system conditions. The curves of the linear relationships are shown on figure 9 whereas the computed values are shown on table 4

Table 4: Input Values and Adams Bohart Model Parameters

	Adams Bohart Model					
	Bed heights(cm)	$Q\left(ml/min\right)$	$C_o\ \left(mg/l\right)$	N_o	K_A	R^2
	20	20	0.48	112.38	0.079	0.75
BA	20	20	0.19	11.42	0.66	0.78
PL	20	20	0.48	113.06	0.08	0.84
	20	20	0.19	25.44	0.30	0.82
BA	20	20	0.48	74.78	0.18	0.87
	10	20	0.48	50.35	0.183	0.75
	20	20	0.48	88.29	0.139	0.90
PL	10	20	0.48	83.72	0.143	0.89
BA	20	10	0.48	19.18	0.221	0.95
	20	20	0.48	23.0	0.2	0.72
PL	20	10	0.48	31.26	0.107	0.78
	20	20	0.48	41.37	0.148	0.87

At initial concentration increase, there was corresponding increase in maximum adsorption capacity of Pb(II) ion and decrease in values of both adsorbents .similar trend was observed when the bed height was decreased from 20cm to10cm. It can be seen from the table that there was decrease in maximum adsorption capacity of the system as inflow rate of solute solution into the adsorption systems (BAC and PPAC) increased.

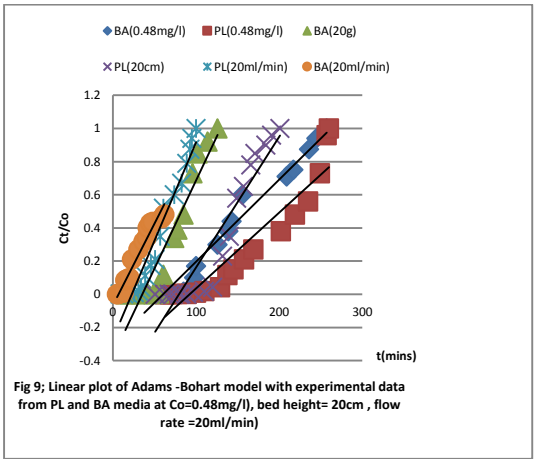

Fig 9; Linear plot of Adams -Bohart model with experimental data from PL and BA media at Co=0.48mg/l), bed height= 20cm , flow rate =20ml/min)

These observations are in agreement with the work on Sorption of Mn(II) Ions from Wastewater by Using Mangostana garcinia Peel-Based Granular-Activated Carbon [35] and Removal and recovery of heavy metals from aqueous solution using papaya wood as a new biosorbent [66]. Increase in values of the maximum adsorption capacity at increased bed height and initial concentration, confirmed the external mass transfer of lead(II) ions onto various adsorbents[52, 44,66, 67, 68].

3.7.4 BDST model:

Bed depth service time (BDST) model is essentially to predict the characteristic parameters of the fixed bed column for optimal design of adsorption system ([22, 35]. Plot of Equation 4 produced linear relationships of bed depth service time and bed depth for PPAC and BAC adsorption systems at 20, 60 and 90 % column saturation. The process which was carried out at constant inflow rate and initial concentration is represented on figure 10.

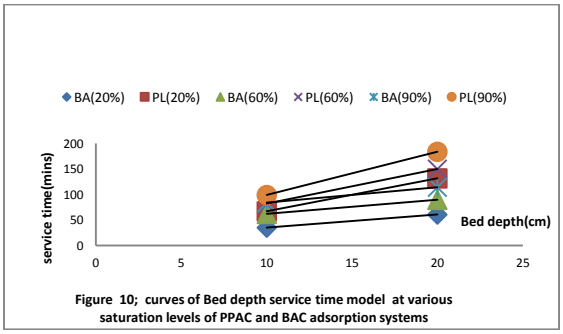

Figure 10; curves of Bed depth service time model at various saturation levels of PPAC and BAC adsorption systems

Vital design parameters; maximum adsorption capacity of the systems and kinetic constants at various column saturations were obtained from the slopes and intercepts in relation to Equations 5 and 6 respectively. The computed values are presented on table 5. Variations in the adsorption capacities and rate constants of various adsorption systems at different bed column saturations were once more show cased. At column saturation levels of 20, 60 and 90%, PPAC system showed high adsorption capacities of 624, 652.9 and 816 respectively whereas adsorption capacities of 249.6, 268.8 and 288 were obtained in column saturations of BAC system at various saturation levels.

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Table 5: Breakthrough Times In Relation To % Column Saturation and Bed Heights

% of Column Saturation	Adsorbent type	N_o	K_B	R^2	Adsorbent/ %Column saturation	Bed heights (cm)	Breakthrough time(mins)	Regression equations
20	BA	249.6	0.0015	0.98	BA(20%)	10	40	
	PL	624	0.0002	0.99		20	60	y = 2.6x + 9
					BA(60%)	10	62	
						20	90	y = 2.8x + 34
60	BA	268.8	0.0011	0.99				
	PL	652.8	0.00009	0.99	PL(20%)	10	69	y = 6.5x + 2
						20	130	
90	BA	288	0.001	0.97	BA(90%)	10	82	y = 3x + 54
	PL	819	0.0005	0.99		20	115	
					PL(60%)	10	80	y = 6.8x + 14
						20	152	
					PL(90%)	10	100	y = 8.5x + 14
						20	183	

The table also showed a decrease in values of the rate constants as column saturation levels increased. The same trend of observations hold true for previous findings on modeling and optimization of removal of Cu(II) ion by thylamine modified chitosan carbonized rice husk composite beads [69] and adsorption of malachite green dye from wastewater using acid activated sawdust[64]. Results from the table also showed variations in breakthrough times for various PPAC and BAC adsorption systems with bed heights and percentage column saturation. The breakthrough times were low at 20% saturation and increased through 60% to the highest value at 90% saturation level. Saturation time was also observed to be lower in BAC adsorption systems than in PPAC systems perhaps due to apparent large adsorption capacity potentials earlier ascribed to PPAC adsorbent. Equations of various saturation lines are also presented on table-5. Under the same experimental conditions, the equations can predict values of service times at corresponding bed height values for each column saturation level. Values of the design parameters obtained could be used to step-up adsorption process of the fixed bed columns under study. High values close to unity suggested that the model is an appropriate predictor of the breakthrough curves and the design parameters.

4.0 Conclusion

Dynamic Adsorption process of Pb(II) onto fixed-beds of PPAC and BAC materials to treat vegetable oil polluted wastewater has been investigated. There were glaring variations in the physical properties of the bed materials. Low ash content, low bulk density and Large surface area and small pore size distribution favoured high adsorption of Pb(II) on PPAC. Adsorption capacities of PPAC and BAC bed materials were also investigated under the conditions of varying bed height, initial concentrations and inflow rates. Capacity to adsorb Pb(II) by PPAC and BAC bed materials increased with increase in initial concentration and bed height but decreased at inflow rate increase and initial concentration and bed height decrease. Efficiency of various bed materials were assess at 50% breakthrough, the uptake efficiency of Pb(II) by PPAC and BAC bed materials was good.

However, uptake efficiency of Pb(II) was more phenomenal on PPAC bed material that on BAC. Film diffusion played leading roles on the mass transfer of Pb(II) to PPAC bed material. internal and Kundsen diffusion played significant role on mass transfer of Pb(II) to BAC due to its supposed large pore sizes . The obtained data were fitted into Thomas, Adams Bohart, Yoons and Nelson dynamic adsorption models to determine the extent of compatibility. Thomas model was most Complied followed by Yoon-Nelson model whereas the least complied model was Adams-Bohart. Pb(II) Breakthrough times and bed service time of PPAC and BAC materials assessed at various saturation levels progressed as saturation levels moved from 20% to 90% through 60%. PPAC and BAC reasonably removed Pb(II) ion from vegetable oil polluted wastewater through dynamic adsorption process, hence an enduring approach to sustainable healthy environment.

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