Efficacy of low-cost activated carbon in the removal of active compounds in pharmaceutical industrial wastewater

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Abstract: Pharmaceutical wastes are emerging organic contaminants, which are mostly unregulated and in an increasing trend. This study evaluates the potentials of low-cost activated carbons derived from bamboo in remediating Pharmaceutical actives contaminants (PhACs) and compared with Oclansorb. Two species of bamboos were processed into activated carbons using $ZnCl_2$, KCl, and H_3PO_4. Selections of the bamboo adsorbents were based on the porosity and surface area using BET analysis. Batch adsorption process was used with contact time as bench mark for comparison. Carbonized *Bambusa vulgaris* (CBV350^oC H_3PO_4) had the highest surface area (SPAS) of 30.1342 m^2 g^{-1} when compared to other adsorbents while carbonized *Oxythenantera abyssinica* (COA 350^oC KCl) gave the highest pore size (AAPW) 446.4384(Å). CBV (350^oC H_3PO_4) gave type IV isotherm classification which favored mono and multilayer adsorption as compared to other adsorbents that obeyed the type III isotherm classification. COA with KCl activation showed the highest removal efficiency for PhACs (73.3%, paracetamol, 78.1% salbutamol and 86.2% chlorpheniramine), followed by CBA H_3PO_4 (63.9% paracetamol, 66.7% salbutamol and 82.2% chlorpheniramine), and SAC (18.5% paracetamol, 34.3% salbutamol and 51.02% chlorpheniramine). The experimental study showed that adsorptions of PhACs were inconsistent with time but equilibrium was attained at 720 min while 30 min was the optimal time.

Keywords: adsorbent, pharmaceutical actives contaminants, carbonized bamboo species, isotherm

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1 Introduction

Recent environmental contaminations are products of technology advancement resulting from industrialization. Frequent contaminants of water resources especially from industries are organic (polar and non-polar) in nature. With this in mind, water refinement becomes a major critical issue world-wide, for which the World Health Organization has formulated strict laws (Bhomick et al., 2017).

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One of the many industries that generate polar organic wastewater is the Pharmaceutical industry. The products from this industry are ubiquitous because of their daily need and usage, while the pollutants that result from their manufacturing persist (Noelia, 2011). Lately, the production of pharmaceuticals product has increased significantly because of their use in aquaculture and livestock farming in the prevention and cure of fish and livestock diseases (Wang and Wang, 2016; Justyna et al., 2019).

The chemicals or ingredients used in the manufacturing of pharmaceutical products can be active, inactive, additive or preservative. Active chemicals like Paracetamol (acetaminophen), Salbutamol, Amoxicillin, Ibuprofen, Chloramphenicol etc. are referred as Pharmaceutically Active Compounds PhACs (Comerton et al., 2009). Pharmaceutical wastewater forms an emerging organic contaminant (EOCs) that is continually released into the environment mainly from the manufacturing of new products, disposal of unused products or by-products and also from excreta (Chang et al., 2015). Their continuous release and persistent nature pose are threats to the environment because they develop drug-resistant microbial strains (Pal, 2018). Similarly, these EOCs are found in the disruption of the endocrine system-a health condition called endocrine disruptors (Esper, 2009).

Organic contaminants in pharmaceutical wastewater deposited in waterbodies generally have adverse health effects especially when water from such waterbodies is used for domestic purposes. Identification and remediation of the contaminants is necessary to guarantee the availability of portable water and secure the health of humans. One way to reduce water pollution is the installation of conventional Wastewater Treatment Plants (WWTPs). However, this solution does not always prove to be effective especially in the removal of huge class of pollutants such as PhACs, hence further treatments are necessary. A study conducted by Stackelberg et al. (2004) revealed that out of 40 samples of stream and wastewater subjected to treatment, 34 still contained several pharmaceutical compounds after the treatment.

An alternative treatment method attracting research attention is the use of adsorption mechanism which involves activated carbon. In most industrial processes, activated carbon has been applied for the removal and recovery of organic and inorganic compounds from gaseous and liquid streams, but it has had limited usage for wastewater treatment due to cost implication (Foo and Lee, 2010). Emmanuel and Odigie (2014) studied the adsorption of N-acetyl-P-aminophenol (APAP) otherwise known as acetaminophen, a Pharmaceutical active compound presented in aqueous environment, onto cetyltrime thylammonium bromide (CTAB), hexadecyl-trimethylammonium functionalized kaolinite clay (CTAB-Kao). No significant removal of APAP from aqueous solution was found from the nucleophilicity observed between the adsorbent and the adsorbate. In comparison to KaO which possess a negatively charged edge, APAP removal was not significantly attained due to the same phenomenon.

Adsorbent produced from natural agricultural products have been the popular substitute for commercial and synthetic adsorbents because of their hydrophobic-oleophilic potential which is necessary for the bioremediation processes (Xue et al., 2015). These materials are lignocelluloses in nature with many pores, they exist abundantly and are of low cost. This study investigates the effectiveness of activated carbon made from bamboo species of *Oxytenanthera abyssinaca* and *Bambusa vulgaris*, for the remediation of Pharmaceutical actives contaminants in wastewater. A comparison was made with Oclansorb from peat moss.

2 Materials and methods

2.1 Activated carbon preparation and adsorption experiment

Two species of fresh bamboo culms were cut at about 20 cm above ground level from the South-West, South-East and Central states in Nigeria within the priory of June 2013 to September 2013. They were reduced to 20 cm long and

external materials were removed. The bamboo culms were then dried at room temperature and further reduced to 5cm.Weighted mass of bamboo species were wrapped in double layers of aluminum foil before carbonization to ensure completely deoxygenated condition.

The prepared bamboo species and carbonized were done at the Multidisciplinary Central Research Laboratory (MCRL) of the University of Ibadan from October 2013 to February 2014, using an electric muffle furnace at a temperature of 350^{o}C and for 2hrs. They were thereafter cooled and over dried at 105^{o}C for 360 min. The char samples were granulated, sieved to 1.18m size and stored. Activation was done with Zinc chloride ($ZnCl_2$), Phosphoric acid (H_3PO_4) and Potassium chloride (KCl) as dehydrating agent. About 26.25 w w^{-1} of the activator was used in the activation of the carbon samples (Ijaola et al., 2013). Porosity and surface area of char samples were determined by modeling of N^2-adsorption data with BET isotherm. Surface area and porosity analyzer (ASAP, 2020 micrometrics instrument corps Norcross GA) were used to perform experiment.

Figure 1 shows the adsorbent used for PhACs remediation. Adsorption behavior of PhACs in pharmaceutical effluents onto bamboo activated carbons was studied in batch process. Experiments were carried out in ambient temperature and adsorption capacity of activated carbons from bamboo was compared with Oclansorb from peat moss.

To conduct the experiment, half litre of pharmaceutical effluent was put into conical flasks of 600 ml capacity. Two grammes of each selected bamboo activated carbon and Oclansorb were weighed into the conical flasks to form an adsorbent/solute solution. Solutions were agitated at a stirring speed of 160 rpm to ensure intimate contact of the adsorbent and solute in solution. Each solution was observed for 6 hrs contact time at which it attains dynamic equilibrium. Thereafter, solutions were filtered with 0.45 µm size paper. The filtrate of 300 ml was poured into sample bottles with tie cap sealed with aluminum foils and kept at temperature of 4^{o}C for further analysis of extraction, clean up and Vis-UV. All experimental analysis was duplicated. Each solution was then observed for 30, 120, 360 and 720 min contact time to study the adsorption kinetics.

(a)Bamboo clumps

(b)Carbonized bamboo clump

(c) Oclansorb

Figure 1 the adsorbent used for PhACs remediation

The amount of PhACs (q_e) adsorbed by bamboo activated carbons and Oclansorb, can be expressed mathematically as

$$q_{e\ =\frac{C_O-C_e}{M}}\ \times v \quad (1)$$

The percentage removal is evaluated using

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

Where

V is the volume of PAHs in solution (L)

Co is initial concentrations of PAHs (mg L^{-1})

Ce is equilibrium concentrations of PAHs (mg L^{-1})

M is the mass of the adsorbent (g).

2.2 Visible ultra-violet spectrophotometry analysis

Pure standards of all active ingredients observed with

minimum of 98.5%-99% purity were used for this study. Derivative form of Solid Phase Extraction (SPE) procedure was used for extracting PhACs from sampled solutions. 1cm of moderate packed cotton wool was placed at the bottom of each 10 mm ID. Loup chromatographic column of 250 mm used. Two gram of activated silica gel 10 mL of 1:1 acetonitrile and methanol were prepared and placed into the chromatographic column. Anhydrous sodium of 0.8 cm sulphate was added to the top of the column. The column was rinsed with additional 3mL of acetonitrile followed by 3 mL of methanol and 3 mL acidify ultra-pure milli-Q water to pre-elute the column. Elute was allowed to flow though the column at the rate of about 1 ml min^{-1} until the liquid in the column was just above the sulphate layer and immediately 300ml untreated or treated sampled effluents were transferred into each prepared column.

The sample bottles were rinsed with 1ml methanol eluent and added to the column as well. The eluent was collected into sampling bottles, after which acidified methanol (pH 2.0) was immediately used to extract compounds adsorbed to the silica sorbent at a flow rate of 0.1 mL sec^{-1}. Samples were collected in a graduated cylinder each and allowed to concentrate to 10ml under air vacuum. Following this, the samples were increased to 20 ml by adding 10ml of methanol and kept at 4^{o}C before Vis-UV analysis.

Visible Ultra-violet spectrophotometry, UV-Vis spectrophotometer S/N 18-1901-01-0243 made by PG instrument limited with UV win-Spectrum coupled with T90+UV/vis spectrometer scanner was further used to analyze the pharmaceutical effluents in order to ascertain the concentration of PhACs of interest in the simulated pharmaceutical effluents. The software used in interpreting concentration of pollutant or chromatogram was UV win5 spectrophotometer version 5.2.0.

Extracted concentrated analytes were further diluted with methanol and scanned with visible-UV to determine the wavelength of each PhACs in the samples. The wavelength of paracetamol, salbutamol and chlorpheniramine was set at 257, 278 and 262 nm respectively. The concentrated analytes were transferred into 10 ml cuvette to read each concentration of PhACs with UV win5 spectrophotometer version 5.2.0 at different determined wavelength. Prior to each UV reading, the instrument was blanked with methanol in order to set a new baseline.

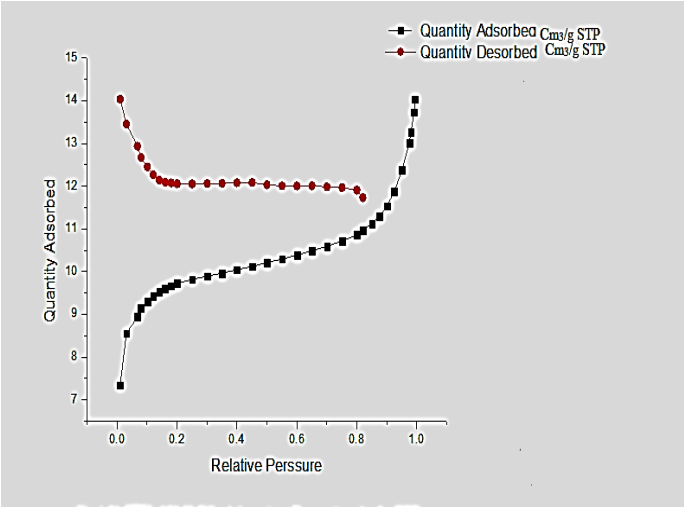
Quantification of PhACs was by an external standard method, which relies on the reproducibility of the standard preparation. The linearity of external calibration was done by preparing different concentrations of paracetamol, salbutamol and Chlorpheniramine standards at different dilution rate of between 0.2 to 3.5.

3 Results and discussion

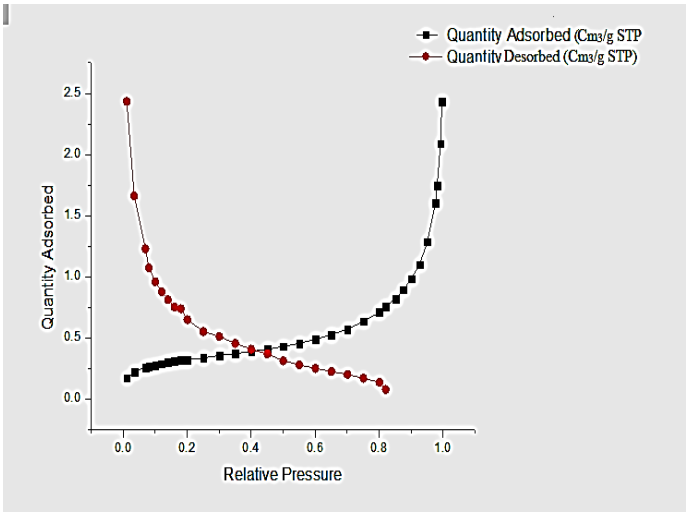
Table 1 showed the surface areas, pore volumes and pore sizes of the modified bamboo species. The data shows that all adsorbents have a good BET surface area, pore volume and pore size. Carbonized *Bambusa vulgaris* with H_3PO_4 at 350^{o}C (CBV 350^{o}C H_3PO_4) exhibited better surface area and pore volume parameters in comparison with other adsorbents. Adsorption isotherm curve specifies the quantity of material adsorbed per unit mass of adsorbent as a function of the equilibrium concentration of the adsorbate. To deduce the efficacy of the bamboo activated carbons in terms of strong or weak adsorption qualities, the quantity of adsorbates adsorbed in cm^3 g^{-1} STP were plotted against the relative pressure (P/Po) to reflect the adsorption-desorption isotherms shapes as established by Brunauer et al. (1938). These are shown in Figures 2 and 3 for CBV and COA respectively.

As observed in the isotherm plots, all bamboo adsorbent activated with $ZnCl_2$ and KCl comform to type III isotherm while those activated with H_3PO_4 conform to type IV isotherm. The quantity of N_2 adsorbed was directly proportional to the relative pressure (P/Po) applied. The initial uptake of N_2 was slow but the rate of adsorption increased with time. The desorption process was similar but in reverse order for all isotherms. Thereafter, adsorption rate was proportional up to a point on the curve and to pressure applied until there was no further adsorption at

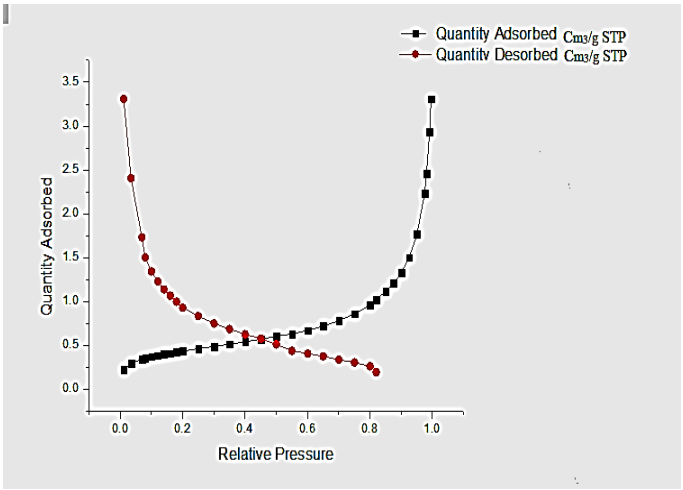
P/Po value of one. The above observation could be due to the differences in porosity, surface area and pore structure of activated carbons studied.



(a) CBV-350°C H_3PO_4

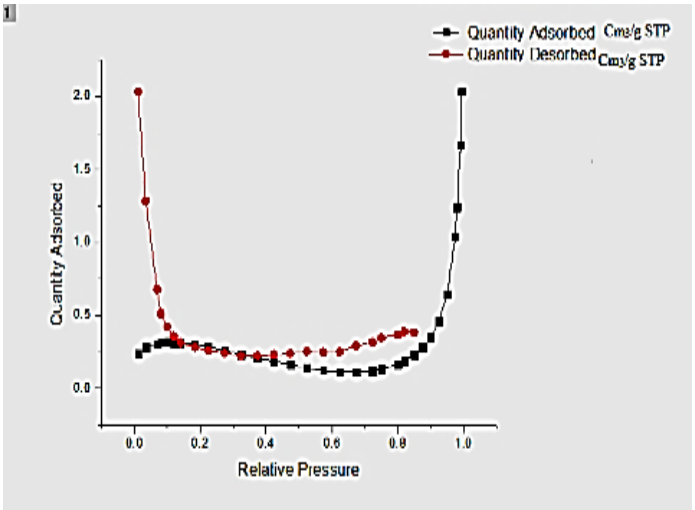


(b) CBV-350°C $ZnCl_2$

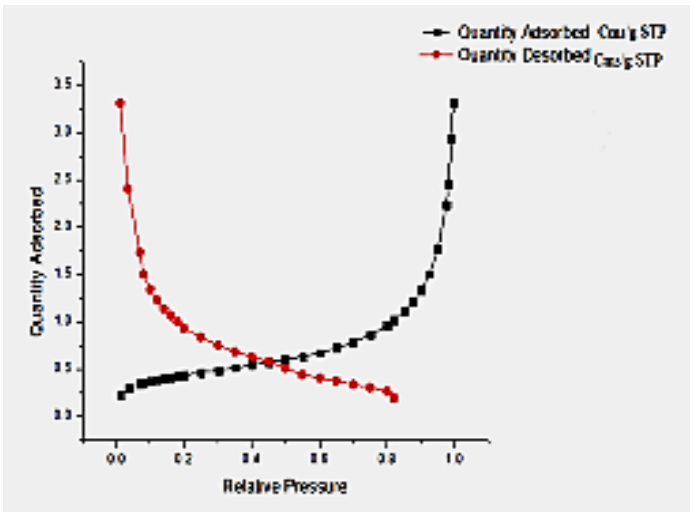


(c) CBV-350°C KCl

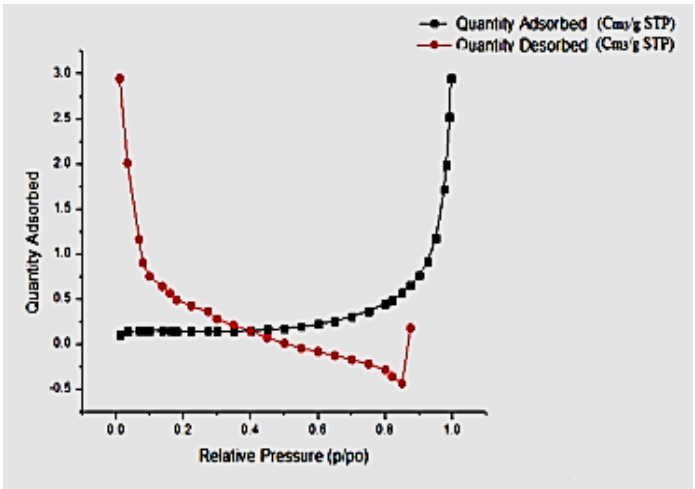
Figure 2 Adsorption- desorption isotherms of Bambusa vulgaris



(a) CBV-350°C H_3PO_4



(b) CBV-350°C $ZnCl_2$



(c) CBV-350°C KCl

Figure 3 Adsorption- desorption isotherms of Oxytenanthera *abyssinaca*

The BET analysis distinctly clarifies activated carbons from bamboo *sp*. in terms of surface areas, pore volumes and pore sizes. Table 1 show that CBV (350°C H_3PO_4) has the highest surface area (SPAS) of 30.1342 m^2 g^{-1} among the adsorbents, while COA (350°C KCl) gave the highest pore size (AAPW) 446.4384(Å). CBV and COA (350°C $Zncl_2$) showed lower pore size of 133.3656(Å) and 133.9970 (Å). All tested activated carbons show similar pore volume as indicated in Table 1. Since COA (350°C KCl) has the highest pore size, it is expected to be the best activated carbon for adsorption. However, the N_2 adsorption-desorption curves in Figures 2 and 3 revealed that CBV (350°CCH_3PO_4) had better adsorption properties as it obeyed the type IV isotherm classification which favored mono and multilayer adsorption. All the three stages of adsorption mechanism are obeyed by CBV (350°CCH_3PO_4 as compared to CBV and COA (350°C $Zncl_2$) and CBV and COA (350°C KCl) that obeys the type III isotherm classification which are adsorbed by weak forces of interactions between the adsorbent and adsorbate. COA (350°C H_3PO_4) obeys the type II isotherm as compared to others which are seen as better adsorbent. On the basis of better adsorption properties and highest pore size, CBV (350°C H_3PO_4) and COA (350°C KCl) were selected for the adsorption experiment along Oclansorb from peat moss (SAC).

Table 1 Some characteristics of the chemically activated Bambusa *vulgaris* and Oxytenanthera *abyssinaca*

Porous Characterization of Activated Carbon	Bambusa *vulgaris*				Oxytenanthera *abyssinaca*			
	CBV	CBV (350°C) (KCl)	CBV (350°C) ($ZnCl_2$)	CBV (350°C) (H_3PO_4)	COA	COA (350°C) (KCl)	COA (350°C) ($ZnCl_2$)	COA (350°C) (H_3PO_4)
Surface Area								
SPAS	1.29	1.09	1.49	30.13	1.13	0.42	1.49	0.69
BSA	1.31	1.11	1.53	29.76	1.12	0.41	1.56	0.68
MA at t-plot	0.36	0.16	0.12	21.02	0.35	0.53	0.12	1.37
ESA at t-plot	0.96	0.94	1.41	8.74	0.77	0.12	1.41	0.69
ACSAP	1.12	1.14	1.52	7.19	1.35	0.65	1.52	0.26
DCSP	1.36	1.10	1.54	3.48	1.92	0.97	1.54	0.37
Pore volume								
SAPVP × 10^{-3}	7.256	3.766	5.116	21.71	6.413	4.552	5.116	3.147
MV at t-plot ×10^{-4}	2.07	0.86	0.59	11.2	1.62	2.80	0.59	7.51
ACVP× 10^{-3}	6.423	3.821	5.145	10.035	6.411	4.561	5.145	2.784
DCVP× 10^{-3}	7.141	3.642	4.975	4.797	6.415	4.552	4.975	3.086
Pore size								
AAPW	22.1	136.4	134.0	29.2	229.1	446.4	134.0	185.1
AAPD	229.7	133.6	135.6	55.8	190.7	283.0	135.6	435.1
DAPD	210.3	132.5	128.9	55.1	133.8	187.4	128.9	332.3

Note: Surface area parameters are in (m^2 g^{-1}), Pore volume parameters are in (cm^3 g^{-1}) and Pore size parameters are in (Å) Single point surface area at P/Po = 0.300387927: (SPAS); BET Surface Area (BSA), Pore volume is also expressed in terms of Single point adsorption total pore volume of pores less than 4765.507 Å diameter at P/Po = 0.995944128 (SAPVP) ; t-Plotmicropore volume (MV at t-polt), Adsorption average pore width 4V A^{-1} by BET (AAPW), Adsorption average pore diameter 4V A^{-1} (AAPD)

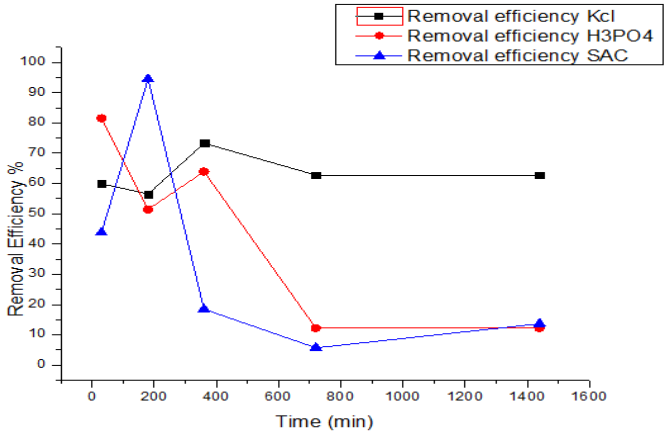


Figure 4 Effect of contact time on adsorption rate of Paracetamol

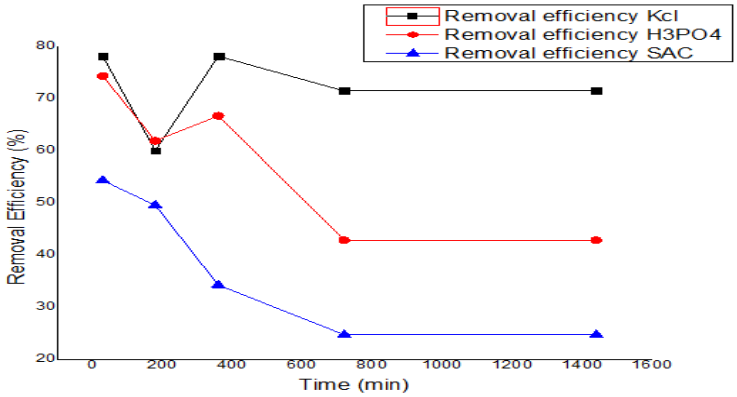


Figure 5 Effect of contact time on adsorption rate of Salbutamol

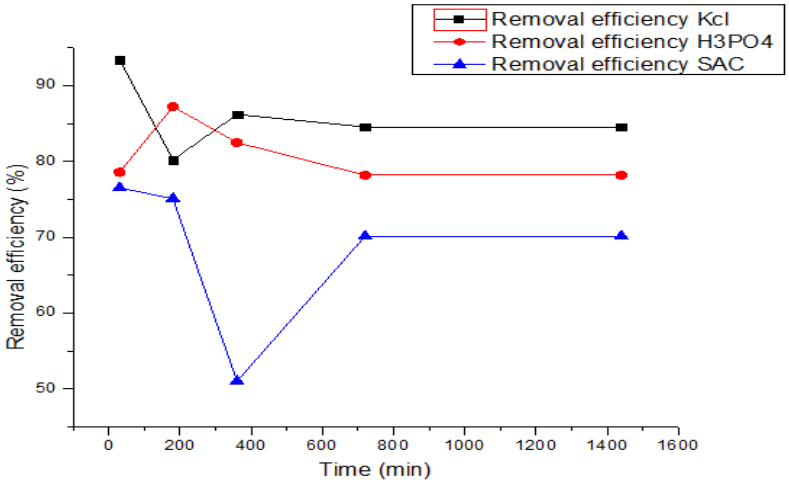


Figure 6 Effect of contact time on adsorption rate of Chlorpheniramine

Figures 4-6 showed the adsorption rate and removal efficiency of PhACs (Paracetamol, Salbutamol, and Chlorpheniramine) by COA (350^{o}C KCl) CBV (350^{o}C $H_3PO_{4)}$ and SAC at varying contact time of 30, 180, 360, 720 and 1440 mins.

In Figure 4, the removal efficiency of COA 350^{o}C KCl for Paracetamol was found inconsistent with 59.9% adsorbed at 30 min, 56.5% at 180 min and 73.3% at 360 min which was the optimal contact time since the highest removal efficiency was recorded at that time. Equilibrium was attained at 720-1440 min contact time with adsorption efficiency of 62.78%. For CBV (350^{o}C H_3PO_4) the optimal contact time was attained at 30 min with adsorption efficiency of 81.5%, after which there was a reduction in adsorption efficiency to 51.4% at 180 min, and 63.9% at 360 min, while equilibrium was attained at 12.2% within 720-1440 min contact time. Adsorption rate for paracetamol with SAC is not also consistent. There were variations in removal efficiency as 43.8% of the analyte was removed at 30min and increased to 94.5% at contact time of 180min. Thereafter, a drastic reduction of adsorption rate was recorded at contact time of 360, 720 and 1440 min to about 18.5%, 5.7% and 13.6% respectively.

Figure 5 also showed the ability of COA (350^{o}C KCl), CBV (350^{o}C H_3PO_4) and SAC at various contact time in the removal of Salbutamol. For COA (350^{o}C KCl) removal efficiency fell within 60.0%-78.1% for all contact time tested. Optimal contact time was found at 30 min with removal efficiency of 78.1% although 360 min had same removal efficiency. Other percentage removal efficiencies ranged between 60.0% and 71.0%. Equilibrium was

attained at 71.4% at contact time of 720 and 1440 mins. CBV (350^{o}C H_3PO_4) and SAC showed a progressive reduction in removal efficiency of salbutamol in that, the higher the contact time the lower the removal efficiency. The 30 min contact time has the highest removal efficiency of 74.3% for CBV (350^{o}C H_3PO_4) while equilibrium been attained with 42.9% removal for 720 min contact time. SAC had the highest removal efficiency of 54.3% at 30 min while equilibrium was attained between 720-1440 min.

Chlorpheniramine shows a good affinity for all adsorbent used in remediation as indicated in Figure 6. COA (350^{o}C KCl) shows better adsorption efficiency when compared with CBV (350^{o}C $H_3PO_{4)}$ and SAC. COA (350^{o}C KCl) had the highest removal efficiency at 93.4% at contact time of 30 min, attaining equilibrium at contact time of 720 min with removal efficiency of 84.6%. This is followed by CBV H_3PO_4 with 87.2% at 180 min contact time while equilibrium was reached at 720 min with removal efficiency of 78.2%. SAC shows removal efficiency that is lower when compared with COA (350^{o}C KCl) and CBV (350^{o}C $H_3PO_{4)}$. Highest removal efficiency was 76.54% at 30 min while equilibrium was attained at 720 min with 70.2% removal.

The trend of adsorption in Figure 4 revealed a slight reduction of adsorption rate before an increase after which equilibrium was observed for COA (350^{o}C KCl). Adsorption trend of CBV (350^{o}C $H_3PO_{4)}$ revealed a sharp reduction to a level and thereafter an increase was observed before equilibrium point was reached. For SAC, there was an initial increase then reduction before equilibrium was attained. The same trends were observed for the adsorption of salbutamol and chlorpheniramine in Figures 5 and 6. These observations are consistent with Vergilli and Barlas (2009), Meenakshisundaram et al. (2011) and Lateefa et al. (2014) findings. It can be deduced that absorption of PhACs with COA (350^{o}C KCl), CBV (350^{o}C H_3PO_4) and SAC occurred in two different stages. The first stage is 30-360 mins contact time, with high number of active binding sites on the adsorbents surfaces. Adsorption rate is rapid in this stage and points to adsorption being controlled by diffusion processes of paracetamol, salbutamol and chlorpheniramine molecule from the bulk phase to the adsorbent surface. The second stage of adsorption is an attachment-controlled process which is due to decrease in the number of active sites available for paracetamol, salbutamol and chlorpheniramine molecule onto the adsorbents surfaces. Slow uptake of adsorbate and establishment of equilibrium over a longer period indicate strong chemical binding of adsorbate with adsorbent.

4 Conclusion

This study shows that COA (350^{o}C KCl) and CBA (350^{o}C H_3PO_4) have better properties in term of porosity and surface area when compared to other bamboo activated carbons on the basis of BET analysis.

Adsorption comparison of paracetamol, salbutamol and chlorpheniramin from pharmaceutical wastewater by COA (350^{o}C KCl) , CBV (350^{o}C H_3PO_4)and SAC with respect to contact time revealed that COA(KCl) showed better removal efficiency of 93.4% when compared to CBV (H_3PO_4) and SAC with 87.2% and 76.54% respectively at optimal contact time while CBV (H_3PO_4) had better removal efficiency when compared to SAC.

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