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Batch adsorption of Hg^{2+} and As^{3+} ions in Hospital wastewater using activated carbon from *Balanites aegyptiaca* and *Detarium microcarpum*

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Abstract *Hospital waste water has a great potential to pollute the environment and severely affect public health. In order to remediate the toxic impact of arsenic (III) and mercury (II) ions, activated carbon were prepared from Detarium microcarpum and Balanites aegyptiaca fruit shells using 2M ortho phosphoric acid and 2M zinc chloride as activating agents. The adsorption of Hg^{2+} and As^{3+} were investigated by varying the dosage of the adsorbent, pH and period of contact using batch adsorption experiment. Concentrations of Hg^{2+} and As^{3+} were determined using ICP-AES. The results obtained indicated that all the adsorbent effectively removed As^{3+} and Hg^{2+} at varying experimental conditions. Optimum adsorption capacities were recorded at dosage values of 0.4 and 0.6 g for Hg^{2+} and As^{2+} respectively, pH of 6 and contact time between a0 and 15 minutes. Acitivated carbon synthesised from Detarium microcarpum using H_3PO_4 and $ZnCl_2$ displayed better efficiencies than commercial activated carbon while others exhibited comparative adsorption efficiency. Therefore, the use of local materials can provide the needed technology that can help in the removal of hospital wastewater.*

Key Words: *Hospital wastewater, toxicity, Hg^{2+}, As^{3+}, remediation, adsorption, activated carbon, Balanites aegyptiaca and Detarium microcarpum*

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1.0. Introduction

The hospital sectors have contributed significantly to environmental pollution. According to Tsakona *et* al. (2007), hospital waste management is an imperative environmental and public safety issue, due to the waste's infectious and hazardous character. Faure *et al*. (2003) however stated that hospital waste should be effectively manage because of its toxicity indicating that the management of hospital waste requires scientific approaches.

Hospital wastewater is any water whose quality is adversely adversely affected during provision of healthcare services. The wastewater is mainly liquid waste containing some solids disposed by staff and patients, or during other healthcare-related processes, such as cooking, cleaning or laundry (Emmanuel *et al*., 2011). Hospital waste water maybe sewage water (black water) which is heavily polluted by faecal matter and urine, food residue and toxic chemicals) grey water or spillage (which is low polluted waste water with residues from washing, laboratory processes, bathing, laundry and from rinsing of x-ray films), stormwater (which consist of water collected from hospital roof during rainfall or paved surfaces which may ultimately be used for other purposes within the hospital (Prayitno *et. al*., 2013, Akter *et. al*., 2000; CCLIN, 2010; Deloffre- Bonnamour, 1995; Verlicchi *et. al*., 2010).

Hospital wastewater can be contaminated by organic matters and persistent chemical compounds including pharmaceuticals, detergents, radionuclides, antibiotics, antiseptics, surfactants, solvents, heavy metals and microorganism (Aurelien *et al*., 2013; Ajayi *et al*., 2004; Ekhaise and Omavwoya, 2008; FAO, 2010;

Kamarathilaka *et al*. 2015). Amouei *et al*. (2015, 2010) observed drastically changes in pH, biochemical oxygen demand, chemical oxygen demand and other parameters in hospital wastewater. The changes gave evidence of contamination when their values were compared with permissible limits. Heavy metals contaminants including lead, cadmium, chromium, nickel, cobalt, mercury and copper were also observed. The total coliforms and heterotrophic bacteria were 5.4×10^8 MPN/100 mL and 2.6×10^{10} CFU/mL, respectively. From their results, they concluded that most of the qualitative indices evaluated in wastewater effluent of hospitals were higher than effluent discharge standards. Prayitno *et al*. (2013) also reported the presence of ammonia, orthophosphate, phenol, detergent, chlorine, faecal coliforms and heavy metals in hospital waste water at concentrations that can threaten environmental health.

Heavy metal contaminants in hospital wastewater is one of the most serious contaminants because of its non-biodegradable and tendency to biomagnify or bioaccumulate in the environment (Eddy and Odoemelam, 2009; Ogoko, 2017; Ogoko and Donald, 2018). Heavy metal is defined as those metals whose density is greater than 5 g/cm^3 and whose toxicity is pronounced above certain critical concentration (Wyasu, 2011, 2016). Heavy metals concentration in any part of the environment, if not properly managed, can constitute severe problems to other components of the environment through the food chain and bio-magnification or bioaccumulation tendency (Eddy, 2009). Toxicity of mercury can affect the lung, skin, eye, kidney, digestive and immune system and may even lead to death (Adedirin *et al*., 2011). Mercury can also cause severe disruption of any tissue and exert poisoning effect on neurological and renal disturbances (Eddy and Ekop, 2007; Ekop *et al*., 2007; Odiongenyi *et al*., 2015). On the other hand, symptoms of arsenic toxicity in water include, abdominal pains, swollen skin, lesions on the skin, diarrhoea, abnormal heart rhythm, tingling of fingers and toes, muscle clamps, tingling of fingers and toes (Alau, 2015).

In view of the alarming impact of heavy metals on the environment, successive technologies in combating their presence in water to minimum concentrations have been developed and is still developing (Odoemelam and Eddy, 2009). Among these technologies, adsorption is one of the best options (Wyasu, 2016). Adsorption involves selective removing of materials (adsorbates) of interest by allowing them to stick to a surface called adsorbent through physisosorption or chemisorption or both mechanism (Eddy and Odiongenyi, 2010). However, most activated carbon that is common in Nigeria are imported therefore, the need for local production has received welcome research promotion (Wyasu, 2016; Okibe *et al*., 2013)

Adverse effect of wastewater generated from hospital may include, eutrophication (which is algal bloom and biomass caused by nutrients enrichment of the receiving water body), alteration of photosynthesis mechanism in the water (due to reduce penetration of light), endocrine disruption (through pharmaceuticals), development of antibiotic resistant pathogens, heavy metal poisoning and outbreak of water borne diseases such as hepatitis A and E, schistosomiasis, typhoid fever, dengue fever, malaria and roundworms (Wyasu, 2011). Therefore, it is necessary to purify hospital wastewater before it is discharged to the environment in order to prevent public health. Therefore, the present study is aimed at employing batch adsorption process to remove heavy metal ions from hospital wastewater using activated carbon from *Balanites aegyptiaca* and *Detarium microcarpum* as adsorbents.

2.0 Materials and Methods.

2.1 Sample collection and pre-treatment

Balanite aegyptiaca fruits and *Detarium macrocarpum* fruits (Precursors for the activated carbon) were purchased from Dutsin-ma market in Dutsin-ma Local Government area of Katsina State-Nigeria. The edible parts of *Balanites aegyptiaca* and *Detarium microcarpum* fruits were removed leaving behind the epicarp shell, which was thoroughly washed with distilled water to remove impurities. The samples were sun-dried to a constant weight, washed again with distilled water and then dried in a thermostatic oven at 105 °C for 48 hours which facilitated easy crushing and grinding. The dried samples were pulverised using mortar and pestle and sieved with a mechanical shaker into a particle size of 1180 µm. The fine sieved particles were stored in a clean air tight plastic container ready for further treatment (Alau, 2016).

2.2 Preparation of 2M H_3PO_4 and 2M $ZnCl_2$ as activating agents

The 2M solution of H_3PO_4 was prepared by diluted 136 cm^3 of H_3PO_4 with distilled water and

make up to 1000 cm^3, while 272g of zinc chloride was mixed with distilled water and made up to 1dm^3 of the solution to prepared 2M zinc chloride (APHA, 1998).

A portion (100g) of *Balanites aegyptiaca* and *Detarium microcarpum* (precursors) each were mixed with 100cm^3 of the activating agents in a crucible for five hours to enhanced proper mixing (homogeneity). The mixture is then heated in an Oven at a temperature of 105°C for one hour to dry the sample. The precursors in a porcelain crucible was heated using a muffle furnace at optimal temperature temperatures of 600°C and an optimal residence time of 30 minutes. The product obtained was washed with deionized water to a neutral pH before drying at 105°C for 5 hours (Kagbu *et al*., 2010). The dried product was stored in a clean air tight plastic container for further analysis.

2.4 Collection of samples

The wastewater sampling area is the Ahmadu Bello University Teaching hospital Wastewater Treatment Plant located at Shika-Zaria. The hospital consists of several Departments such as laundry, Laboratories and mortuary centre where they discharged and channelled wastewater into the aerated vessel of the treatment plant. Samples were picked at the reservoir of the aerated vessel.

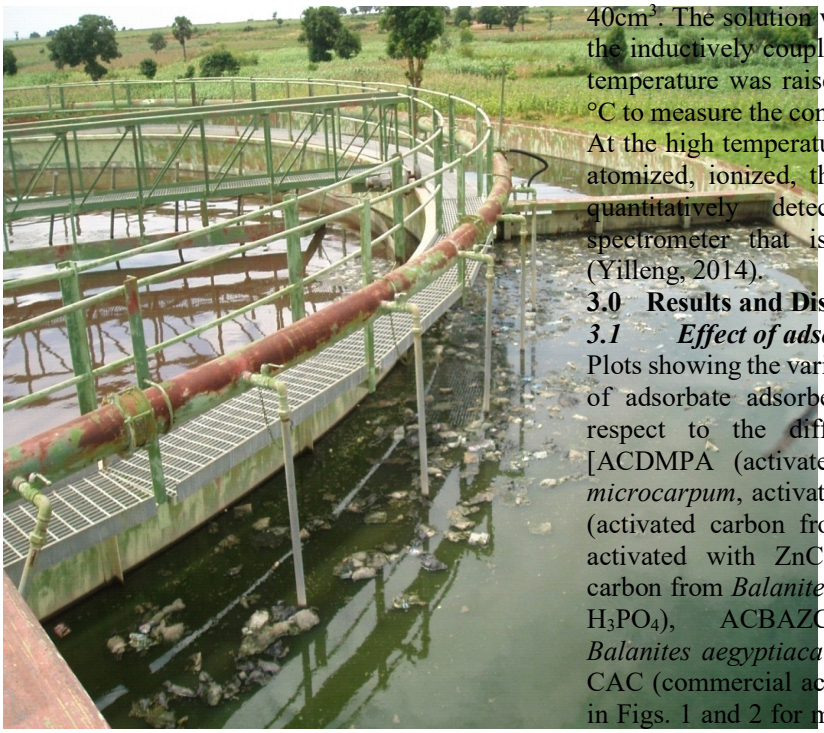

Plate 1: Wastewater at the aerated vessel (sampling points)

Plate 2.1 above indicate where the wastewater samples were collected randomly at five different locations within a depth of 100 cm using a water sampler, in which composite samples were prepared (Wyasu, 2016). Samples of hospital wastewater were collected at 2.00pm in cleaned sterile containers.

2.5 Batch adsorption experiment

The activated carbon produced from the epicarp of *Balanites aegyptiaca* and *Detarium micropcarpum* seed shell were used for the treatment of the wastewater. Samples of the wastewater (10 cm^3) were mixed with 0.1g of carbon in 250cm^3 Elemeyer flasks. The mixture was consequentially stirred with a magnetic stirrer at 15 minutes, 30 minutes, 45 minutes and 60 minutes respectively. The procedure was repeated for all adsorbent with dosages (0.2g, 0.4g, 0.6g, 0.8g and 1.0g respectively). Similar experimental procedures were also designed and implemented for different pH (2, 4, 6 and 8 respectively). After each experiment, the solution was filtered using Whatman filter paper No 1, stored and preserved for mercury and arsenic ions preserved.

2.6 Determination of heavy metals using inductive coupling plasma (ICP-AES)

Each analyte was digested with a mixture of concentrated hydrochloric acid and trioxonitrate (V) acid, in a ratio of 3:1 to make up a volume of 40cm^3. The solution was sprayed into the core of the inductively coupled argon plasma, where the temperature was raised to approximately 976.85 °C to measure the concentration of Hg^{2+} and As^{3+}. At the high temperature, all analyte species were atomized, ionized, thermally excited, and were quantitatively detected with an emission spectrometer that is couples to the recorder (Yilleng, 2014).

3.0 Results and Discussion

3.1 Effect of adsorbent dosage

Plots showing the variation of equilibrium amount of adsorbate adsorbed by the adsorbents with respect to the different adsorbents dosages [ACDMPA (activated carbon from *Detarium microcarpum*, activated with H_3PO_4), ACDMZC (activated carbon from *Detarium microcarpum* activated with $ZnCl_2$), ACBAPA (activated carbon from *Balanites aegyptiaca*, activated with H_3PO_4), ACBAZC (activated carbon from *Balanites aegyptiaca*, activated with $ZnCl_2$) and CAC (commercial activated carbon)] are shown in Figs. 1 and 2 for mercury (II) and arsenic (III) respectively.

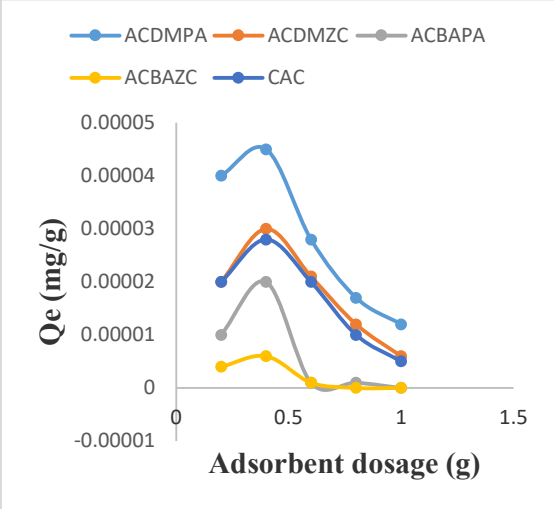

Fig. 1: Variation of equilibrium concentration of Hg^{2+} adsorbed with adsorbent dosage

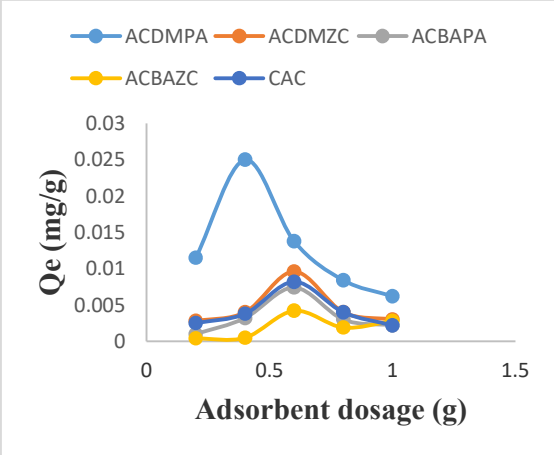

Fig. 2: Variation of equilibrium concentration of As^{3+} adsorbed with adsorbent dosage

3.2. Effect of Contact time

Fig. 1 presents plots for the variation of equilibrium concentration of Hg^{2+} adsorbed with adsorbent dosage, while Fig. 2 shows similar plots for the adsorption of As^{3+}. From the plots, the amount of As^{3+} and Hg^{2+} adsorbed by all the adsorbents was observed at adsorbent dosage of 0.4 g for Hg^{2+} and 0.6 g for As^{3+}, indicating that the optimum dosage for adsorption of mercury ion from the hospital wastewater by the synthesised activated carbon. Also, the trend for the variation of extent of adsorption for both metal ions was ACDMPA > ACDMZC > CAC > ACBAPA >ACBAZC, which indicated that ACDMPA and ACDMZC has better adsorption capacity than the commercial activated carbon. It was observed that adsorption capabilities of all the adsorbents decreases when the adsorbent dosage exceeded 0.4g, for Hg^{2+} and 0.6g for As^{3+} which may be attributed to overlapped of adsorbent sites, due to overcrowding of adsorbate particles (Nazar *et al*., 2013). According to Odoemelam *et al.* (2018), each adsorbent has maximum adsorption sites and within the adsorption sites, only the activated sites can be occupied, which represent the fraction of the original available sites. Activation of the sites can be initiated by adsorption dosage, period of contact, pH and other factors. Once the active sites have been occupied, further diffusion of the adsorbate onto the adsorbent may lead to desorption. Therefore, the observed trend for the variation of Q_e with adsorbate dosage can be explained in terms of factors that altered adsorption-desorption equilibrium, which include adsorbent dosage and diffusion rate in this case.

Figs. 3 and 4 are plots showing the variation of equilibrium concentration of metal ions adsorbed with period of contact time for Hg^{2+} and As^{3+} respectively.

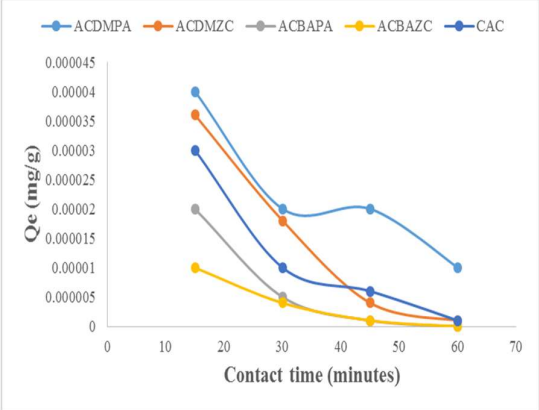

Fig. 3 : Variation of equilibrium concentration of Hg^{2+} adsorbed with period of contact time

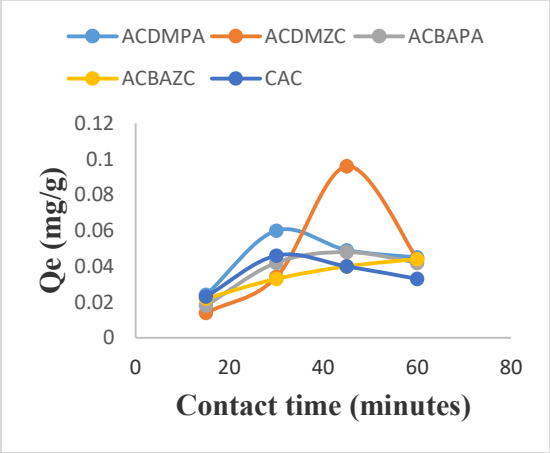

Fig..4: Variation of equilibrium concentration of As^{3+} adsorbed with period of contact time

The rate of adsorption of mercury ions was rapid in the first 15 minutes as presented in Fig. 3 due to sufficient adsorption sites, which resembles results obtained by Okibe, 2014, when using activated carbon for the removal of Cr^{6+} and Pb^{2+} from synthetic wastewater. The adsorption of mercury ions by all the adsorbents decreased as the contact time exceeded 15 minutes, which can be attributed to deactivation of the adsorption sites by the mercury ions. However, adsorption trend for arsenic ion increases comprises of three section, the first second and third sections represent initial increase, optimum adsorption and decrease of adsorption with time. Such trends have been reported for other adsorbent with respect to arsenic and other heavy metal ions. According to Sharma *et al*. (2008), the removal of Hg^{2+} and As^{3+} in wastewater by fly ash was observed to decrease with increase in the period of contact because mercury level was extremely low, and desorption occur as a result of overcrowded of adsorbate particles (Nazar *et al*., 2013). Fig, 4 also indicated that ACDMZC showed optimum adsorption for As^{3+} among all the adsorbents with respect to time indicating that the respond of the adsorption tendency with respect to time by this adsorbent is more favourable than others. As stated before, the adsorption sites can be deactivated if the period of contact is not set at optimum (Ekop and Eddy, 2009)

3.3 Effect of pH

Figs. 5 and 6 respectively shows the variation of equilibrium concentration of Hg^{2+} and As^{3+} adsorbed with pH. Both plots reveal trends for adsorption capacity of the adsorbents to follow the order, ACDMPA > ACDMZC>CAC>ACBAPA>ACBAZC, indicating that ACDMPA and ACDMZC have better adsorption capacities for Hg^{2+} and As^{3+} than the commercial activated carbon and that those of ACBAPA and ACBAZC are lower than that of the commercial activated carbon, i.e CAC.

The plots (Figs. 5 and 6) further revealed that the adsorption of the ions (Hg^{2+} and As^{3+}) increase with increase in pH up to pH of 6, beyond which further increase in pH led to decreasing adsorption. pH depends on the concentration of hydrogen and hydroxyl ions that are free at that instance. At low pH, hydrogen ion dominates and been a proton with an empty orbital (i.e H^{+}), adsorption can be easily enhanced through charge transfer or electron sharing (Abdel-Salam *et al*., 2011, Cho *et al*., 2005). However, increase in pH deprotonate the system and hence the adsorption equilibrium, which could lead to decrease in extent of adsorption as observed in this study (Essien and Eddy, 2015). The observed trend is similar to the one reported by Oladunni *et* al. (2012) who used activated carbon produce from

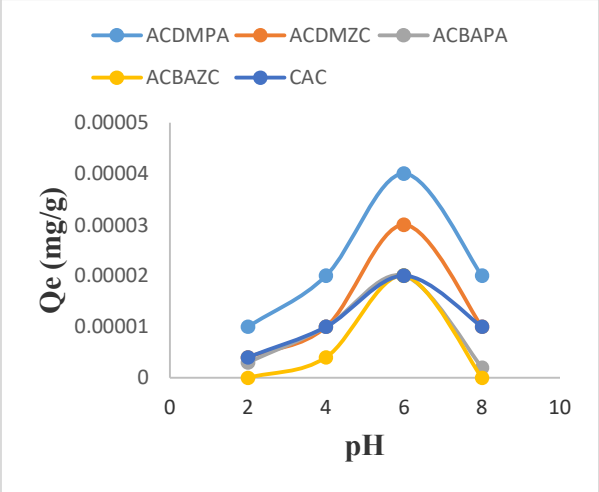

Fig. 5: Variation of equilibrium concentration of Hg^{2+} adsorbed with pH

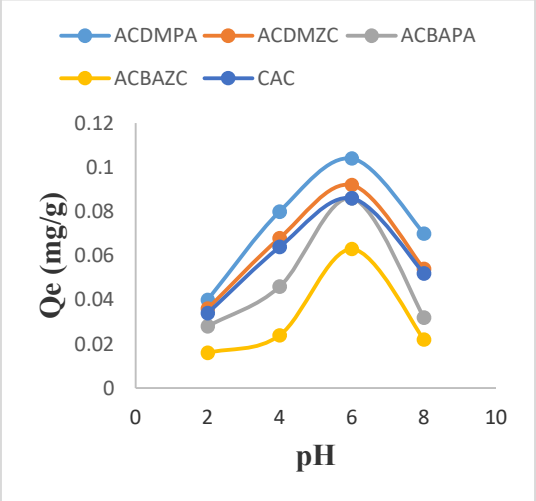

Fig. 6: Variation of equilibrium concentration of As^{3+} adsorbed with pH

locust beans husk to remove Cr^{6+} from aqueous solution and observed optimum pH for best adsorption to be 6

4.0 Conclusion

The results and findings of this study indicated that ACDMPA (activated carbon from *Detarium microcarpum*, activated with H_3PO_4), ACDMZC (activated carbon from *Detarium microcarpum* activated with $ZnCl_2$), ACBAPA (activated carbon from *Balanites aegyptiaca*, activated with H_3PO_4), ACBAZC (activated carbon from *Balanites aegyptiaca* are good adsorbent for the removal of Hg^{2+} and As^{3+} from hospital

wastewater. The adsorption capacities of ACDMPA (activated carbon from *Detarium microcarpum*, activated with H_3PO_4) and ACDMZC (activated carbon from *Detarium microcarpum* activated with $ZnCl_2$), are better than that of CAC while those of other are comparable to that of CAC. The efficiency of the adsorbents depends on adsorbent dosage, period of contact and pH. Therefore, for optimum absorption, these variables should be optimised.

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