Evaluation of the adsorption isotherm of activated charcoal used in pharmaceutical medicine from some Nigerian plant parts, corn cobs, the wooden parts of *Mangifera indica* and *Azandirachta indica*

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ABSTRACT

Activated charcoal is variously referred to as charcoal, carbon, Norit and medicoal. It is generally derived from many carbonaceous vegetable materials and in this study from Nigerian wooden plants as well as farm wastes. The uses include, detoxification, decontamination and in flatulence. The aim of this research is to explore cheaper and more effective activated charcoal alternatives than those that are currently commercially available. Corn cobs and the wooden parts of *Mangifera indica* and *Azandirachta indica as raw* materials were carbonized in a muffler furnace at 600 to 700°C for 30 min. The carbonized residue so obtained were pulverized, sieved and activated in an oven at 120°C for 2 h. The adsorption profile of the test charcoal samples was evaluated using acetic acid adsorption method and Freundlich isotherm model to determine adsorption constants, (K) and (n). It was observed that *M. indica* (K – 0.076), showed the highest adsorption capacity while at the same time was statistically significant, followed by *A. Indica* (K - 0.026) whereas corn cobs (K – 0.013) and standard (K – 0.013) ranked equally with the lowest specific adsorption property. Activated charcoal with excellent performance as well as cost-effective for use in pharmaceutical medicine can be sourced from local plants and farm wastes.

Keywords: Locally sourced, cost-effectiveness, excellent, activated charcoal, alternative, pharmaceutical medicine.

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INTRODUCTION

As delineated, adsorption is a process that occurs when a gas or liquid solute accumulates on the surface of a solid or a liquid (adsorbent), forming a molecular or atomic film (the adsorbate) (Kopecky et al., 1996). This involves the inter-phase accumulation of concentration of substances at a surface or at the inter-phase (Tien, 1994).

Generally, adsorption is categorized into physical process that is multilayer and exhibits dipole- dipole dispersions, hydrogen bond and London van der Waals interaction. The chemical adsorption on the other hand is monolayer and shows formation of strong chemical bonds between adsorbate molecules and specific surface locations also referred to as chemically active sites. The equilibrium relationship of sorbent and adsorbate at constant temperature is then the adsorption isotherm with constant which can be very informative. Three isotherm models have been identified namely: Langmuir, Freundlich and Brunaun Emmeth – Teller (BET). However, the Freurdlich equation seems to work better with adsorption of solutes than gases. Abdul and Aberuagba (2005) observed that a high Freurdlich (K) value is consistent with better adsorption capacity.
In Freurdlich equation:

$$X/m = K.C^{1/n}$$

It then implies that:

Log X/m = 1/n log C + log K

Where the constants K and 1/n represent the Freundlicch empirical parameters for the specific sample as intercept and slope respectively, mass of the adsorbent (activated charcoal) – m in grams; amount of the adsorbate (acetic acid)- X in mol/dm^3 representing the difference between the equilibrium concentration of adsorbate after reaction with the adsorbent in solution – C_1, and the initial concentration of the adsorbate - C_o
At dynamic equilibrium:

$$K_{ad} = \frac{[AS]}{[A][S]}$$

Where A is adsorbate and S is the sorbent or adsorbent. The adsorption constant, being dynamic equilibrium constant however obeys Van't Hoffs equation:

$$\frac{\partial \ln K}{\partial 1/T} = \frac{\Delta H}{R}$$

Where: T is thermodynamic temperature, ΔH is change in enthalpy or energy content of the system, R is Universal gas constant and K is surface coverage constant.
   Charcoal can be prepared from vegetable matters some of which are farm wastes (Yagi, 2011; Oliver, 1999). Examples include corn cobs, groundnut shells, sand dust, rice-husks, cellulose-residue, wood, lignite, petroleum, pits, coke to mention but these few.
   The adsorption power of any given activated charcoal is affected not only by the manufacturing techniques, but also by the total available surface area, incorporating external and internal surfaces, affinity of solute to the adsorbent, contact time up to dynamic equilibrium between the amount of adsorbate adsorbed and the amount of adsorbate in solution as well as the pore size and distribution. Further, adsorption tends to increase as pH and temperature decrease (Helmenstine, 2012), whereas large organic molecules are adsorbed better than smaller ones (Pan and Xing, 2008).
   The wide variety of applications of charcoal is in purification processes (Austin et al., 1984) including decolorization, decontamination, respirators for protection against toxic gases, as desiccants and or medicinal agents in detoxification (Sisca et al., 2009), as well as in flatulence. According to Hall, "It is the effective single agent available as an emergency decontaminant in the gastro intestinal tract (Hall and Strother, 1989). Activated charcoal is used for drug over dose or poisoning. At a dose of 50 to 100 g at once and repeated every 2 to 4 h at a dose equal to 12 g per hour. For children, lower dose (10 to 25 g) is administered (Anon, 1999). This successful indication in children is consistent with observation affirmed by Holt (2010), as well as its antibacterial and antifungal effect (Greenyarn, 2005).
   There is an evidence that one teaspoonful of activated charcoal has a surface area of more than 10,000 square feet. It can adsorb; poisons, impurities and contaminants, thousands of times its own weight in gases, heavy metals, poisons and other chemicals thus making them ineffective or harmless (James and Mark, 2008).
   In this research, acetic acid was the choice adsorbate since it is an organic pollutant with a specific surface area close to that of nitrogen which is most often used in adsorption experiments (Robert, 1991).
   A plot of log (x/m) against log C produced an isotherm which was quite instrumental in the evaluation of test activated charcoal samples as well as the standard.
The equation of the reaction:

$CH_3COOH + NaOH \rightarrow CH_3COONa + H_2O$
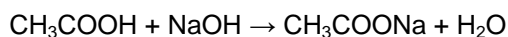
The aim of this study therefore is to explore a more cost-effective, locally sourced and easily accessible activated charcoal alternative with better adsorbent property than the ones currently available commercially. Also, by use of farm wastes such as corn cobs as raw material, public health could be enhanced through environmental mop up of those wastes. In this research, the value for (K) that is known to have a direct correlation to the specific adsorption capacity, was also applied in the statistical analysis of the various samples against the standard sample, using ANOVA at $P \leq 0.05$.

MATERIALS AND METHODS

The activated charcoal samples were prepared by using corn (maize) cobs and the wooden parts of *Mangiferaa indica* (mango plant) and *Azandirachta indica* (neem plant), collected from the University of Port Harcourt, community in Rivers State, Nigeria. Each of the samples was chopped into chips, air dried in the laboratory ambient temperature for 4 weeks and then carbonized using muffler furnace (Model SXL, China), at 600 to 700°C for 30 min (Fiyyaz et al., 2000; Ademiluyi et al., 2009). Each of the carbonized sample residues was further, pulverized, sieved (Mesh size no. 250) and activated in the oven (New Life DHG, England) for 2 h and same subjected to adsorption process.
   Blank titrations were carried out using 1.0N NaOH solution (AR CAS 1310–73–2) as titrant against the various acetic acid (99.8% Sigma Aldrich) concentrations namely: 1.0, 0.8, 0.6, 0.4 and 0.2 N, respectively. Average of triplicates titres was taken. Then, 2.0 g of commercially available activated charcoal (standard) was placed in each of 5 of 250 ml Erlenmeyer flasks and 50 ml of each of the acetic acid concentrations added. The flasks were stoppard and agitated periodically for 30 min and allowed to stand for 1 h to attain dynamic equilibrium (Dina et al., 2012). First, 5 ml of the filtrate was carefully discarded and thereafter 10 ml of same filtrates titrated against 1.0 N NaOH. This procedure at room temperature of 25°C was repeated for the three plant samples of activated charcoal respectively to determine the equilibrium concentrations (C) of acetic acid.
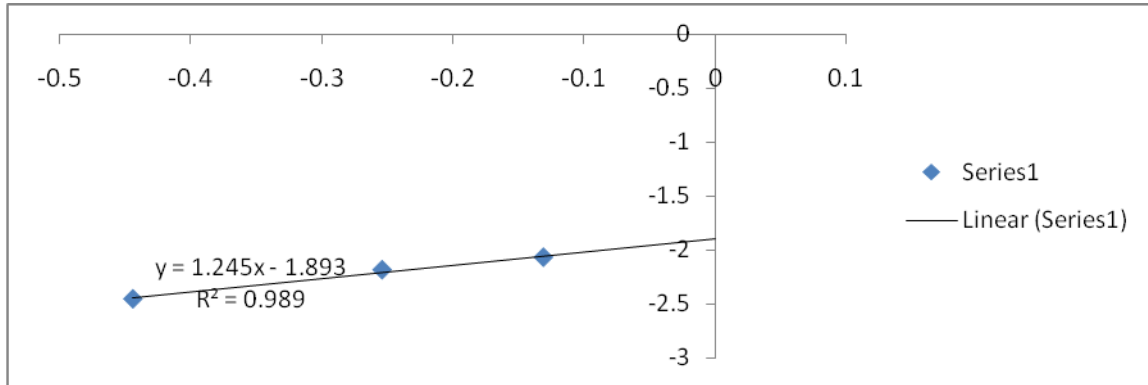   With the respective titre values so obtained, an isotherm of log

Figure 1. The Isotherm for pharmaceutical commercial sample (standard) with K = 0.013.
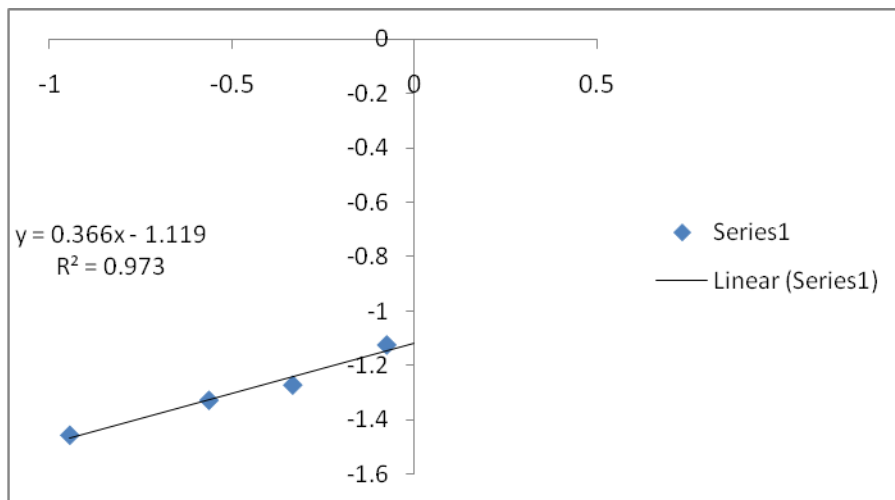
y = 1.245x - 1.893
R² = 0.989

Figure 2. Isotherm for *Mangifera indica* – activated charcoal with K = 0.076.

y = 0.366x - 1.119
R² = 0.973
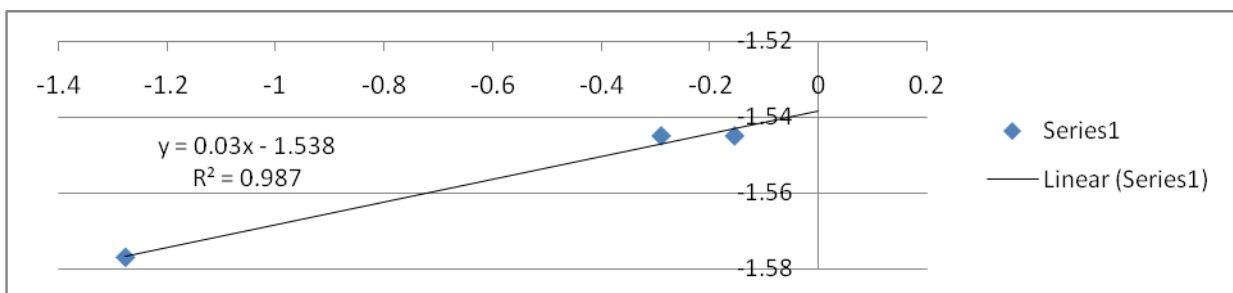
Figure 3. Isotherm for *Azandirachta indica* – Activated charcoal with K = 0.026.

y = 0.03x - 1.538
R² = 0.987

X/m was plotted against the logarithm of the equilibrium concentration C_i (Freundlich isotherm); their respective constants derived and then used in the evaluation of the adsorption properties of the test samples.

RESULTS

The following Freundlich adsorption Isotherm plots shown in Figures 1 to 4 were obtained, for a straight line graph; $y = mx + c$; where m and c are the gradient (slope) and intercepts respectively.

Applying the above equation to the Freundlish plot;

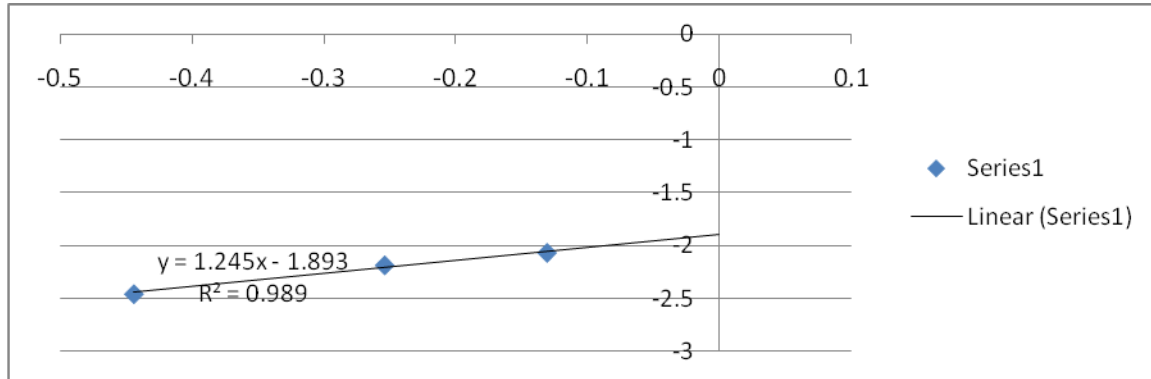$$X/m = K.C^{1/n}$$

Log K and 1/n represent the intercept and slope

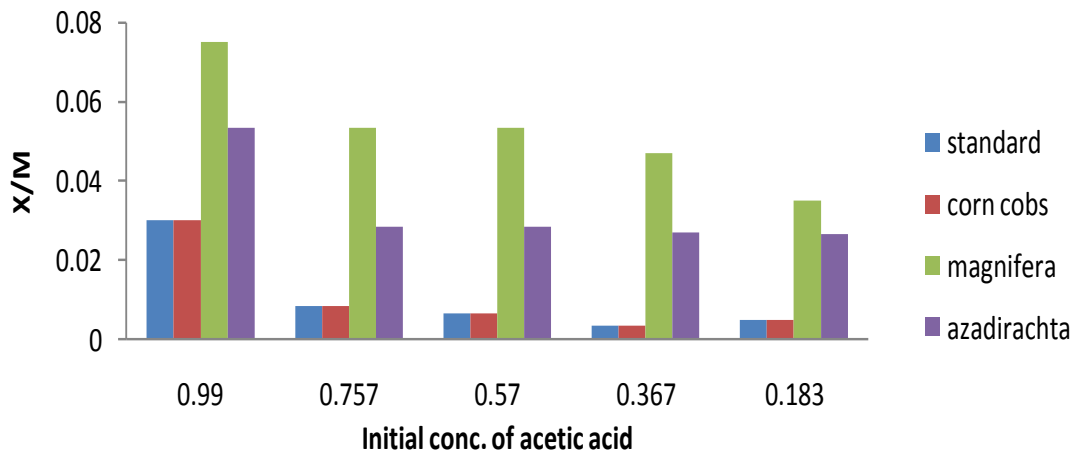**Figure 4.** Isotherm for corn cobs- activated charcoal with K = 0.013.

Figure 5. Bar chart representing the mole of acetic acid adsorbed per gram of adsorbent (activated charcoal samples).

respectively.

In the graphs depicting the adsorption isotherm of the test activated charcoal samples (Figures 1 to 4), the value for K is directly proportional to the adsorption capacity of the samples. Figure 5 showed the respective quantities of adsorbate (acetic acid) adsorbed per gram of adsorbent (activated charcoal) samples and proved consistent with the deductions reached by application of isotherm constants (K).

DISCUSSIONS

From the equations represented above, a plot of log X/m against log C was linear with intercept and slope as log K and 1/n respectively. Evaluation of the constants derivable from the Isotherm of various test samples and standard showed that activated charcoal from different sources exhibited variable specific adsorption capacities.

The preparation technique was the same for all samples hence the likely uniformity of some variable factors such as, pore sizes and distribution, so also, the contact time up to dynamic equilibrium between the adsorbent and the adsorbate as well as pH and temperature of experiment. The likely contributors therefore, to the different specific adsorption properties in this case might include; available internal surface areas that may be intrinsic for a particular sample and then the special affinity of the solute to the adsorbent. This can be expressed in terms of the difference in heat of adsorption (-ΔH) – exothermic changes in enthalpy or energy content of same system and is a measure of energy levels between the mobile and static (ground) states of molecules in the system.

Activated charcoal sample derived from corn cobs showed the best adsorption capacity of all those from farm wastes namely, groundnut shells and rice-husks (Abdul and Aberuagba, 2005), yet same corn cobs from above result ranked equally to the standard sample lower than that of *M. indica* and *A. indica.* On the other hand, adsorption exhibited by the activated charcoal from waste newsprint papers ranked same as that of the commercial standard (Oliver, 1999), thereby implying that the plants

stand out as potential source of local raw material for excellent activated charcoal for use in pharmaceutical medicine.

As the human gastro intestinal tract (GIT), particularly the stomach is an acid-rich environment; it may imply that this *in vitro* research could be very well extrapolated to an *in vivo* system.

Conclusion

In conclusion, as there exist different specific adsorption capacities for different activated charcoal samples, and in consideration of the profound usefulness of medicoal, this study has therefore contributed in the establishment of the fact that activated charcoal alternative much cheaper, easily accessible, locally sourced and with excellent adsorption potentials are practicable.

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