Adsorption Process of Iron, Cadmium, Copper, Lead from Aqueous Solution using Palm Bunch Adsorbent

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Abstract

The potential of an agro-based industrial waste, dry empty oil palm bunch (DEOPB), obtained from a local oil palm processing mill was explored to treat effluent from a petrochemical plant. The adsorbent was subjected to pyrolysis and chemically activated using a 0.1mol hydrochloric acid (HCl). The adsorption behaviors of iron, copper and cadmium were found to obey Pseudo Second Order kinetic model, Brunauer, Emmet and Teller (BET) and the Langmuir isotherm models were used to fit the iron, copper and cadmium adsorption isotherm onto the DEOPB activated carbon with their equilibrium capacities agreeing with experimental data except for the lead component that showed no effect. The heavy metals contaminants investigated using this technique was found to have been removed from the wastewater stream in the following percentage efficiency: iron; 57%, copper; 99.6% and cadmium; 90% but was ineffective in the removal of the lead component as its concentration remained unchanged throughout the experiment. The locally produced activated carbon was subjected to laboratory analysis and its properties (Density; $0.369g/m^3$, Particle Size; 0.18mm, Ash Content; 5.73%ww, Porosity; 90%, Surface Area; $691.85 m^2/g$) were found to favorably compare with commercial activated carbon.

Keywords: heavy metal contaminants, effluent, multi-layer adsorption, bio-adsorbent.

Significance of the work

- Application of a cheaper and active adsorbent from waste biomass material
- Efficient removal of copper, cadmium and iron ions from aqueous solution with minimum duration adsorption rate.
- Converting waste material to useful adsorbent

Introduction

The growth of industrialization has resulted in the release of several heavy metal ions causing major environmental concern. Heavy metal ions are mostly generated as effluent waste from various industrial activities such as electroplating and metal surfaces treatment, Printed Circuit Board (PCB) manufacturing, wood processing industries, petroleum refining etc. All these industrial activities produce different heavy metal ions that can be categorized as hazardous waste requiring extensive waste treatment in large quantities of waste waters, residues and sludge [1] and [2]. Adsorption amongst other methods/techniques like co-precipitation, biosorption, electro coagulation, ultra-filtration, employed for the treatment and removal of heavy metal ion contaminants from industrial effluent streams have been found to be less expensive and a better alternative in handling these hazardous compounds. The conventional methods of heavy metals removal from industrial waste water include many processes such as chemical precipitation, flotation, adsorption, ion exchange and electrochemical desorption.

The adsorption capacity of a compound increases with: increasing molecular weight, a higher number of functional groups such as double bonds or halogen compounds, increasing the polarization potential of the molecules [3]. It has also been established that the adsorption force is the sum of all the interactions between all the atoms. The short range and additive nature of these forces results in activated carbon having the strongest physical adsorption forces of any known materials. To be adsorbed, an adsorbate molecule must find its way to the adsorbent particle by convection, diffuse through the fluid film surrounding the particle, travel by diffusion along the length of a pore until it finds a vacant adsorption site, and then adsorbs onto the solid surface. These mass transfer steps are driven by a departure from equilibrium [4]

The kinetic models have been used to investigate the mechanism of sorption and the potential rate controlling steps, which is an important tool used in selecting optimum operating conditions for full scale batch processes of adsorption. The notable models that have been used are the Pseudo-First order, Psuedo-second order, intra-particle and Banghan. The removal of Cu (II) ion from aqueous solutions by biosorption onto agricultural waste sugar beet pulp by Aksu and Isoglu [5]; phenol removal from aqueous systems by tender leaf refuse by Abdelwahad, [6]; adsorption of phenol and dye from aqueous solution using chemically modified date peat activated carbon by Ahmedna [7]; chromium (VI) removal from aqueous solution and industrial waste water by modified date palm trunk by Sunil et al, [8]; amongst other researchers have all applied this various models in their studies.

Other researches have gone into this area of study with aim of developing the best techniques to remove

heavy metal ion contaminants from effluents include Fenglian and Qi [9]. Dhiraj et al [10] having studied and investigated the conventional techniques for removal of toxic metal contaminants generated from ever growing industrial activities, found that these techniques were not economical as it was too expensive and will further generate huge quantity of toxic chemical sludge [11]; compared the various techniques in modern day heavy metal contaminated effluent waste water treatment at the same water pH and temperature. At the end of their investigation, they were able to state the limitation of the various techniques and came up with the conclusion that the amount of contaminant removal from stream by adsorption was higher making it (adsorption) the most viable of all. Sandhya et al, [12], reviewed the technical feasibilities of various low cost adsorbent for removing heavy metal contaminated waste water which included expensive materials like zeolites, chitesan chemical activated carbon and locally made activated carbon. The results obtained were compared with that of chemical activated carbon showed that these low cost materials has high adsorption capacities and remove more heavy metal ion impurities.

Apart from adsorption method using activated carbon adsorbent for removal of heavy metal ion contaminants from waste water/aqueous solutions, other methods were also investigated. Nural et, al [13], studied the removal of heavy metals ions from mixed solutions via polymer enhanced ultra-filtration using standard as water soluble bio polymer. It was found at the end of the study that only zinc, zn(ii) and chromium Cr (III) and (IV) gave higher rejection (ie removed by the starch based polymer recovery than lead, (Pb) which was among the contaminants in the stream, thereby making this techniques deficient as compared to adsorption.

Barakat, [14] was of contrary opinion as to the work done by Nural et al, in his article, where recent development and technical applicability of various treatment of heavy metals contaminants removal from industrial effluent waste water such adsorption on new adsorbents, electro dialysis photocatalysis, and membrane filtration was reviewed. His work was able to establish that amongst of the mentioned techniques, only adsorption and membrane filtration handled these contaminants optimally.

Activated carbon with surface modification was studied and found to be most suitable for the removal of heavy metals from waste water when compared to conventional and expensive methods [15]. Other studies were also able to show that surface modification is of great advantage to adsorption process as it decreases the adsorption equilibrium time [16]. Adsorption has advantages over other methods for remediation from waste water because its design is simple. It is sludge-free and can be of low capital intensive except that its manufacturing cost is quite high [17].

Adsorption process reduces energy consumption, increases yield, selectivity, non-generation of toxic sludge etc. It is for these reasons that researchers have gone into these areas with a view to looking at the ways of producing low cost adsorbent. A variety of materials such as Bamboo trees,[18], [19],[20] Periwinkle shell, Tree barks, coconut shells, poultry dung's etc. have all been used as basic raw materials in the local production of activated carbon but have all been found to have their limitations/setbacks as the use of these sources either results in deforestation, or are useful raw materials for road construction and land filling or even useful in agriculture as organic manure, hence the choice of the dry empty oil palm bunch (DEOPB) source raw material in the local production of activated carbon as the DEOPB is not only a seemingly worthless agricultural waste material constituting a menace in our local communities, but also helps to convert solid waste to wealth which in turns create a healthier environment. It is worth noting that the dry empty oil palm bunch (DEOPB) is a by-product from local palm oil production which are usually left littering the local mill area or sometimes burnt openly, releasing volumes and volumes of poisonous carbon soots into our atmosphere polluting it.

Materials and Methods

Preparation of the bio-sorbent

The air dried empty oil palm bunch cut into bits was pyrolyzed at controlled temperature of about 600 to 800°C in the laboratory of the department of Chemical/Petrochemical Engineering Rivers State University of Science and Technology Nkpolu Port Harcourt, for chemical activation and then modified with 0.5mol magnesium oxide (MgO).

Determination of Surface Area

Surface area is an important property of activated carbon as it determines to a great extent the adsorptive level of the activated carbon. Hence, for the activated carbon to have a good commercial value and be applicable for industrial use, it needs to have an incredibly large surface area per unit volume and a network of submicroscopic pores where adsorption actually takes place. After producing the adsorbent locally, the surface area was determined experimentally as briefly described below:

A quantity of 10grams of the already produced activated carbon was measured using an Analytical Weighing Balance, Denver Germany, model AE223 & 03/2015 and soaked in a 50ml of methylene blue solution for exactly two hours (2hrs). At the end of the two hours, the mixture was filtered using a 'Whiteman's filter paper' and the extract collected to determine the volume (weight of adsorbed N₂) from which the surface area



was calculated gravimetrically as carried out by Song et al [21].

Surface Area,
$$SA(m^2/g) = \frac{X_m \times N \times A}{M}$$

Where,

 $Xm = Adsorbed \ adsorbate \ voloume = \frac{2.225}{22.4} = 9.933 \times 10^{-2}$

N = (Avogadro's number) = 6.02×10^{23}

A = (Contact surface Area of N₂ Molecule) = 16.2×10^{-20} M = (Molecular weight of Nitrogen) = 14.0016 Then,

Surface Area,
$$SA(m^2/g) = \frac{Xm \times N \times A}{M}$$

9.933×10⁻²× 6.02×10²³×

$$= \frac{9.933 \times 10^{-2} \times 6.02 \times 10^{23} \times 16.2 \times ^{-20}}{14.0016}$$
$$= \frac{9687.0589}{14.0016}$$

Therefore,

Surface Area,
$$SA(m^2/g) = 691.85 \frac{m^2}{g}$$

Surface area of adsorbent from activated carbon could be determined from condemned tyre as carried out by Alaa [22].

BET Theory

This BET theory for multi-layer adsorption is the expansion of Langmuir equation, which is for the single layer adsorption is shown in equation 1 [23]

$$\frac{P}{V(P^{\circ}-P)} = \frac{1}{CV_m} + \frac{C-1}{CV_m} \cdot \frac{P}{P^{\circ}}$$

 $C = e^{(q_t - q_L)/RT}$ (C: constant)+

Vm: Monolayer adsorption amount+

V: Adsorption amount at the equilibrium pressure P

(1)

This theory well fits to type II and IV isotherms as shown in figure 1 at the relative pressure between 0.05 and 0.35. The monolayer adsorption amount (Vm) and C parameter are calculated from the slope and intercept of BET-plot by using the least square fitting. C parameter represents the surface interaction energy, so it must take a positive value.



Fig. 1: The specific surface area is calculated from this Vm and cross sectional area (σ).

(2)

$$S_{BET} = \frac{Vm}{22414} \cdot 6.02 \times 10^{23} \cdot \sigma \cdot 10^{-18}$$

Classification of isotherms into five types are shown in figure 2. Based on these profiles and the results obtained the isotherms are ascertained in this work.



Fig. 2: Classification of isotherms into five types of Brunaeur, Deming, Deming and Teller (24)

Pseudo Second Order kinetic model

The differential equation for a pseudo – second – order model that describes the kinetics of sorption as applied by Ho and Mckay [25] [26] is given as shown in equation (3):

$$\frac{t}{q_e} - \frac{1}{k_2 q_e^2} = \frac{t}{q_t}$$
(3)

where k_2 , q_e , and q_t are the concentrations of metal ions (mg/L) adsorbed on the sorbent at equilibrium and at time, t respectively; k_2 is the second order rate constant [1/(time.conc.)].

The plot of t/q_t against t gives a linear relationship from which the constants q_e , k_2 are determined.

The Langmuir isotherm

The kinetics of processes in which one substance reacts and the products do not inhibit the process, we have the Langmuir equation as shown in equation (4) [27].

$$\theta = \frac{Kp}{1 + Kp} \tag{4}$$

where K is the adsorption equilibrium constant or the adsorption coefficient, p is the pressure of the gas, θ is the adsorption centers.

However, the thermodynamic studies of the potential of Agave lechuguilla biomass for chromium Cr (III) removal from aqueous solutions, [28] have shown that apparent equilibrium constant, (K) of the bio-sorption is defined as shown in equation (5):

$$K_c = \frac{C_{adeq}}{C_{eq}} \tag{3}$$

where C_{adeq} and C_{eq} are the concentrations of the chromium on the adsorbent and residual chromium concentration at equilibrium respectively.

Source of Industrial Effluent Waste Water used

Waste water sample was collected from effluent of one of the petrochemical plants in Rivers State, Nigeria and characterized. The results obtained are shown in table 1. The analysis of the sample with respect to the four contaminants after treatment are also shown in table 1.

Results and Discussions

TABLE 1: EFFECT OF TIME ON ADSORPTION CAPACITY OF Fe, Cu, Pb, Cd WITH 10 CM BED AND 10G WEIGHT OF ADSORBENT

S/N	Time	Iron conc	Copper conc	Lead conc	Cadmium	Length of	Weight of
	(min)	(ppm)	(ppm)	(ppm)	conc (ppm)	Activated	Activated
						carbon bed	Carbon used
						(cm)	(g)
1	0	0.1400	0.0438	0.0038	0.0020	10	10
2	10	0.1100	0.0021	0.0038	0.0020	10	10
3	20	0.0800	0.0021	0.0038	0.0018	10	10
4	30	0.0700	0.0017	0.0038	0.0013	10	10
5	40	0.0600	0.0017	0.0038	0.0009	10	10
6	50	0,0600	0.0017	0.0038	0.0006	10	10
7	60	0.0600	0.0017	0.0038	0.0002	10	10
8	70	0.0600	0.0017	0.0038	0.0002	10	10

Table 1 shows that the adsorbent could not remove lead contaminant from the slurry. A study carried out by Sasireka, et al [29] showed that *cassia fistula* seed carbon (CFSC) was capable to remove lead from aqueous solutions. It was noted that high carbon content in the activated carbon formed from CFSC played a major role in the adsorption process.

Further analysis of the adsorption capacity of the adsorbent on iron, copper and cadmium with their specific gravities 7.87, 8.96 and 8.65 [30] respectively yielded the results obtained in table 2 and table 3.

TABLE 2: ANALYSIS OF RESULTS IN TERMS OF VOLUME AND PRESSURE

THEE 2. THATE TO BOT RESCENDED IN TERMS OF TO ECOMETINE TRESSORE						
(P/P^{o})	0.1	0.2	0.3	0.4	0.5	0.6
$V(m^3 liquid N_2/kg solid x 10^6)$	66.7	75.2	83.9	93.4	108.4	130.0
$((P/P^{o})/V) \ge 10^{-6}$	1500	2660	3576	4283	4613	4615
$(P/P^{o})/[V(1-P/P^{o})]$	1666	3333	5109	7138	9296	11538

TABLE 3: ADSORPTION CAPACITY OF VARIOUS COMPONENTS WITH RESPECT TO RESIDENCE TIME ON THE ADSORBENT

Duration (min)	$Fe(m^{3}/kg)$	$Cu (m^3/kg)$	$Cd (m^3/kg)$
10	4225	2676	-
20	2112	2676	578000
30	1811	2651	165200
40	1583	2651	105100
50	-	-	82580
60	-	-	64230



Fig. 3: Adsorption isotherm showing BET and Langmuir

Comparison of the profiles in figure 3 and those in figure 2 one can obtain the isotherm classification.



Fig. 4: Concentration profiles of adsorption rate of metal contaminants on adsorbent Fig. 4 depicts the profile of the dynamic concentration of the iron (Fe), copper (Cu) and cadmium (Cd) with their various initial concentrations at time zero. It shows clearly that as the adsorption treatment with the DEOPB based activated carbon proceeds, the concentration of the copper component in the wastewater stream happens to reduce sharply in the first ten minutes unlike the other components present in the stream. The iron contaminant got removed from the effluent and maintained equilibrium at 40 minutes, while cadmium maintained equilibrium from 60 minutes. This simply explains the effectiveness of the DEOPB based activated carbon.

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Figure 5 shows the adsorption rate profiles of iron and copper ions contaminants on the locally produced adsorbent. It shows that for iron, the rate of adsorption was significant within the first 30 minutes of application. However, for copper the rate of adsorption could be seen to be constant between 10 to 40 minutes. Figure 6 shows significant increase of adsorption rate for cadmium ion contaminants.



Fig. 6: Adsorption rate profile of Cd ions on DEOPB based activated carbon



Fig. 7 : Plot for the determination of adsorption constant for Fe concentration



Fig. 8 : Plot for the determination of adsorption constant for Cu concentration



Fig. 9: Plot for the determination of adsorption constant for Cd concentration

Figure 7 was used to obtain the adsorption rate constant for Iron (Fe). From the model, the adsorption rate constant for Fe was calculated as 12.23g.min⁻¹ mg⁻¹. In like manner, figure 8 was used to obtain the adsorption rate constant for copper (Cu). From the model, the adsorption rate constant for Cu was calculated as 2264g.min⁻¹ mg⁻¹; and figure 9 could be used to obtain the adsorption rate constant for cadmium (Cd). From the model, the adsorption rate constant for cadmium (Cd). From the model, the adsorption rate constant for cadmium isotherm, the following results for iron, copper and cadmium were obtained:



Fig. 10: Plot for the determination of Langmuir constant for Fe concentration



Fig. 11: Plot for the determination of Langmuir constant for Cu concentration



Fig.12: Plot for the determination of Langmuir constant for Cd concentration

Figures 10, 11 and 12 show the Langmuir constants for iron, copper and cadmium respectively as depicted in equations (4) and (5). The distinct profiles of the three plots for the various contaminants metal components show that:

Molecules are adsorbed on definite sites on the surface of the adsorbent and that each site can accommodate only one molecule. The principle that the adsorption energy is the same at all sites and that the area of each site is a fixed quantity determined solely by the geometry of the surface. The adsorbed molecules cannot migrate across the surface but only on the adsorbed sites [31].

Conclusion

The characterization of the produced activated carbon obtained from DEOPB shows that the product can be compared with other AC obtained from other sources favorably.

Its applicability in the treatment of heavy metal ion contaminated industrial effluent wastewater as seen in the percentage removals of iron (57%), copper (99.6%) and cadmium ((90%) except for lead (0%) shows that it can be widely applied in the treatment of industrial effluent before being discharged into water bodies, replacing the expensive commercial activated carbon with similar characteristics properties.

The heavy metal ions contaminants investigated using this technique was found to have been removed from the wastewater stream in the following percentage removals; iron; 57%, copper; 99.6% and cadmium; 90% but was ineffective in the removal of the lead component as its concentration remained unchanged throughout the experiment. The locally produced activated carbon was subjected to laboratory analysis and its properties (Density; 0.369g/m³, Particle Size; 0.18mm, Ash Content; 5.73%ww, Porosity; 90%, Surface Area; 691.85 m²/g) were found to favorably compare with commercial activated carbon.

The treatment of effluent wastewater shows that the second order kinetic constants for iron, copper and cadmium contaminants are 12.23g.min⁻¹ mg⁻¹, 2264g.min⁻¹ mg⁻¹ and 49.6g.min⁻¹ mg⁻¹ respectively.

The authors recommend that further work on the adsorption of lead on DEOPB based activated carbon should be carried out with varying temperature and pH.

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