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REMOVAL OF POTASSIUM PERMANGANATE FROM AQUEOUS SOLUTION BY ADSORPTION ONTO ACTIVATED CARBON PREPARED FROM ANIMAL BONE AND CORN COB

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ABSTRACT
This study investigates the physicochemical properties of animal bone and corncob derived activated carbon on the removal of potassium permanganate ($KMNO_4$). The effects of condition variables such as initial dye concentration, adsorbent dose, pH and contact time were studied. The result shows that corncob derived activated carbon have good potential of $KMNo_4$ removal than animal bone derived activated carbon. The corncob revealed higher adsorption capacity of 1.25, intensity of 2.80 and surface area of 420m^2/g. the removal efficiency increases as adsorbent dose increases. This makes it an interesting option for dye removal from aqueous solution of dye.

Keywords: Adsorption; adsorbate; adsorbent; isotherm.

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1 INTRODUCTION

Potassium permanganate ($KMNO_4$) is widely used commercially for the purification of many different products. Its uses in potable water treatment to oxidize iron, manganese, and sulfides, and for the destruction of taste producing compounds, is well documented with over 500 municipalities reported on the list of users. Its characteristic pink or purple color is rapidly reduced by hydrogen sulfide, to the yellow or brown color of manganese dioxide.

The effectiveness of adsorption of dye removal from wastewater has made it an ideal alternative to other expensive treatment methods. The effluents from many chemical manufacturing processes contain large quantities of dye which are easily visible when it enters a waterway. The discharge of dyes in the environment is worrying for both toxicological and esthetical reasons. Industries such as confectioneries, leather, paper, plastics, pharmaceuticals etc, are some of the sources for dye effluents. The discharge of organic color containing effluent causes a huge damage to environment. Hence their removal has recently become the subject of interest. The removal of characteristics pink or purple color (reduced to yellow or brown by the action of hydrogen sulphide) from waste water is often more important than the removal of the soluble colorless organic substance which usually contribute to the major fraction of the biochemical oxygen demand (BOD).

Many methods have been reported for removing potassium permanganate from wastewater, among which are membrane filtration, coagulation/flocculation, precipitation, flotation, adsorption, ion exchange, ion pair extraction, ultrasonic mineralization, electrolysis, advanced oxidation and chemical reduction. Biological techniques include bacterial and fungal biosorption and biodegradation in aerobic anaerobic or combined anaerobic/aerobic treatment processes (Kan, 2011).

Adsorption is one of the most effective methods for removing potassium permanganate from waste water

and activated carbon is the preferred adsorbent widely employed to treat waste water containing different classes of dyes. Recognizing the economic drawback of commercial activated carbon, activated carbon has been widely used in waste water treatment to remove organic and inorganic pollutant. Many investigators have studied the feasibility of using low cost plant materials (residues) like babul seed (Sujatha, Geetha, Sivakumar and Palanisamy, 2008), barley husk (Robinson, Chandran, Naidu and Nigam, 2002), sunflower stalks (Gang, Sun and Xu, 1997), peel of sucumis sativa fruit, orange peel and lemon peel (Agboinghale and Ugbesia, 2005) as carbonaceous precursors for the removal of dyes from water and waste water. Previously the work on removal of textile dye from aqueous solution by adsorption technique using natural waste was investigated in the laboratory (Azhar, Liew, Suhardy, Hafiz and Hafim, 2005). The present study was undertaken to evaluate the efficiency of a carbon adsorbent prepared from Animal bone and corncob, an agricultural waste for removal of potassium permanganate dye in aqueous solution. The main aim of the current study has been to visualize the pattern of adsorption of this dye to various situations such as initial dye concentration, adsorbent dose, pH and contact time.

The effects of various operating parameters monitored and experimental conditions were decided. These fundamental data will be useful for further applications in the treatment of practical waste or process effluents.

2 Materials and Method

2.1 Adsorbate

Potassium permanganate (*an electrolytic dye*) was used for the adsorption studies. $KMNo_4$ was manufactured by Gurr, BDH chemicals Ltd, Poole England. A stock solution of the dye with a concentration of 1000mg per liter was prepared. This was done by weighing 1.0g of dye and dissolving it in a 1000ml volumetric flask. The working solution was prepared by diluting the stock solution with deionized water to give the appropriate concentration of the working solutions. The concentration of the residual dye solution was measured using UV-visible spectrophotometer at a wavelength corresponding to the maximum adsorbance for the dye solution. Batch adsorption studies were performed at room temperature ($30^o \pm 1^oC$).

Table 1 enlists the properties of potassium permanganate used.

Table 1: Characteristics of potassium permanganate (Dye).

Molecular formula	(C_4H_4) $KMNO_4$ (S)
Molecular weight	201.242
CAS. Number	55589 – 62 – 3
Maximum Absorbance λ wave) water solubility	270g/L at 25^oC
Appearance	White crystalline power
Density	$1.81g/cm^3$
Melting point	225^oC
Maximum absorbance λ wave	

At room temperature appears as a solid, purple or pink powder that yields yellow or brown solution in water. It is 200 times sweeter than sucrose. It has a slightly bitter after taste, especially at high concentrations (Pavani et al, 2008)

Potassium permanganate oxidizes iron and manganese. It also oxidizes odor and taste – causing compounds. The use as disinfectant has been proven very effective against certain viruses (Ezeugo, 2012) Potassium permanganate, however, has a lot of disadvantages. It has a tendency to give water a pink color. It is toxic and irritating to skin and mucous membranes. No byproducts are generated when preparing the feed solution, however this dark purple/black crystalline solid can cause serious eye injury, is a skin and inhalation irritant, and can be fatal if swallowed. Over – dosing is dangerous and may cause health problems such as chemical jaundice and drop in blood pressure.

Removal of the excess permanganate can be monitored qualitatively by observing the disappearance of the pink color characteristic of permanganate. In plants that do not utilize flocculation and sedimentation processes permanganate dosing should be closely monitored (Montgomery, 1985).

In industrial treatment processes with permanganate, the AB and CC derived activated powdered carbon will be added downstream of permanganate because it may consume permanganate, rendering it unavailable for the oxidation of target organics. (Montgomery, 1985).

2.2 Adsorbents

The corn cobs CC and Animal bone AB used were collected from local area of Eke Uke market in Idemili North Local Government Area, Anambra State, Nigeria in clean plastic bags. These waste

materials were thoroughly washed with distilled water to remove the dirt and impurities. Size reduction of the bone was achieved by manual crushing using cutlass and hammer. It was later sundried for four days. The dried bone was carbonized for 4hrs to remove volatile compounds such as hydrocarbons at a temperature of 700^{o} – 1100^{o}C using muffler furnace. The carbonization process was followed by steam activation at a temperature of 700^{o}C using muffler furnace to increase its surface area. 1000g of the sample was measured, placed in a bucket. This was followed by treatment with activating agent, tetraoxosulphate IV acid of 50% concentration until uniform mixture was obtained. The mixture was allowed to digest for 24hrs in order to increase the surface area of the adsorbent.

After which excess acid was washed off from the sample with sodium hydroxide and distilled water. The resulting black product was kept in an air–oven maintained at 105^{o}C for 12hrs followed by washing with $NaHCo_3$ and water until free from excess acid and dried at $120^{o}C \pm 5^{o}C$ to get AB activated carbon. Product obtained was screened through a mesh sieve with a particle size range of 180–200µm and the physical properties are analyzed by usual standard method.

2.3 Activated Carbon from Corn Cobs

Corn cobs collected from the same source as animal bone was sundried. After which, the size reduction was achieved by crushing first with *morta* daughter. This was followed by milling to smaller grain size. It is then screened through a mesh sieve with a particle sizes range of 180–300µm. The carbon was prepared by chemical activation. 1000g of the dried sample was measured and treated with concentrated sulphuric acid in a weight ratio of 1:1 for 24hrs to increase the surface areas of the sample. The sample was allowed to digest for 48hrs. After which the resulting black product was oven dried for 12hrs at 105 ± 5^{o}C followed by washing of excess acid with sodium hydroxide and distilled water until free from excess acid and dried at 105 ± 5^{o}C to get CC derived activated carbon. The sundried activated carbon product obtained was ground well as fine powder and the physical properties analyzed by usual standard methodologies.

2.4 Batch Adsorption Studies

Adsorption experiments were performed at room temperature (40 ± 1^{0}C); 1g of adsorbent was mixed with known initial dye concentration (100 – $800mgL^{-1}$) $KMNO_4$ dye solution with 1g adsorbent dose variation study. For pH variation, 10ml of 100mg L^{-1} solution was agitated with 1g of the sample for 4hrs at 300rpm. The aliquot was spectrophotometrically analyzed for residual $KMNO_4$ concentration using UV-visible spectrophotometer (spectrum, model 752s) at a wavelength corresponding to the maximum absorbance for the dye solution (λmax = 630nm).

3.0 Results and Discussion

3.1 Characteristics of the Adsorbent.

The data for proximate analysis, physical properties of the prepared activated carbon were determined by standard procedures. Surface area was determined by BET method. The physicochemical properties are listed in table 2.

Table 2: Characteristic of activated carbon of Animal bone and Corncobs.

S/ No	Properties	Animal bone activated carbon	Corncobs activated carbon
1.	pH	5.9 ± 0.02	6.0 ± 0.02
2.	Moisture content (%)	5 ± 0.00	5.9 ± 0.01
3.	Ash content (%)	22 ± 0.25	24 ± 0.22
4.	Porosity	0.823 ± 0.005	0.810 ± 0.003
5.	Apparent density	0.449 ± 0.058	0.248 ± 0.056
6.	Solubility in water (%)	0.36 ± 0.01	0.44 ± 0.01
7.	Volatile matter	38.32 ± 0.15	37.33 ± 0.15
8.	Surface area (m^2g^{-1})	$420m^2/g \pm 1.00$	$500m^2g^{-1} \pm 1.00$

3.2 Effect of Dye Concentration

To study the effect of different initial concentrations of ($KMNO_4$) potassium permanganate on adsorption behavior, six concentrations (100, 200, 300, 400, 500 and 600mg/L) were used and the amount adsorbed were calculated and given in tables 3 and 4. The amount of dye adsorbed (mgg^{-1}) increased with increase in time and the equilibrium for dye removal attainment was achieved after 240 mins. The initial dye concentration provides the necessary driving force to overcome the persistence to the mass transfer of $KMNO_4$ between the aqueous and solid phases. The maximum adsorption obtained by AB and CC are

93.01% and 91.1% respectively (Somasekhara, 2006).

Table 3: Effect of different initial dye concentration on dye removal using AB (Adsorbent dosage: 1g/10ml, initial pH: 5.0, Size: 180–300µm, agitation speed: 300rpm, Time: 240 mins)

Initial dye concentration (mg/L)	Percentage of potassium permanganate removal with time (mins)					
	40	**80**	**120**	**160**	**200**	**240**
100	23.15	38.56	50.16	66.22	80.74	84.30
200	24.34	40.67	52.73	68.26	82.86	86.26
300	22.40	37.50	58.74	72.24	84.90	87.20
400	25.53	45.92	58.24	71.24	85.92	87.25
500	28.64	50.82	57.98	76.64	87.84	90.78
600	38.49	54.86	61.50	75.52	89.91	91.01

Table 4: Effect of different initial dye concentration on dye removal using CC (Adsorbent dosage: 1g/10ml, initial pH: 5, Size: 180 – 300µm, agitation speed: 300rpm, Time: 24mins).

Initial dye concentration (mg/L)	Percentage of potassium permanganate removal with time (mins)					
	40	**80**	**120**	**160**	**200**	**240**
100	34.16	48.45	63.18	73.43	87.41	88.10
200	24.50	42.90	58.20	68.54	88.15	89.05
300	20.18	36.88	50.28	67.98	88.86	90.16
400	33.40	45.98	64.40	78.60	89.45	91.08
500	29.50	50.56	59.22	79.40	89.60	91.02
600	34.15	50.33	65.01	79.80	90.80	91.10

3.3 Effect of Adsorbent dose Variation

The effect of adsorbent dose on removal of $KMNO_4$ was studied by varying the dose of adsorbent (0.002, 0.004, 0.006, 0.008, 0.01 and 0.012gL^{-1}) in the test solution while keeping the initial dye concentration 100mg/L (Temperature $40\pm1^{o}C$) at pH: 5. Experiments were carried out at different contact time for 240 mins.

Table: 5 and 6 show that percentage of adsorption increased with increasing adsorbent dose. The increase in the percentage removal of dye with the increase in adsorbent dosage is due to the availability of large surface area with more active functional groups. The maximum adsorption of $KMNO_4$ by AB and CC are 91.01% and 93.9% respectively.

Table 5: Effect of different adsorbent dosage AB and dye removal (Initial concentration: 100mg/L, contact time: 240mins, pH of solution: 5, Agitation speed: 300rpm, Temperature: 40°C, Size: 180–300µm).

Adsorbent dose (g/100ml)	Percentage of dye removal with time (min)					
	40	**80**	**120**	**160**	**200**	**240**
0.2	14.62	32.28	35.64	54.10	70.80	71.76
0.4	18.64	38.26	65.68	79.53	87.28	88.28
0.6	39.10	54.16	66.87	79.93	88.06	88.20

0.8	35.33	54.18	70.02	80.10	89.04	90.24
1.0	23.31	44.40	69.04	81.02	90.68	90.98
1.2	22.31	45.01	68.08	82.01	91.01	91.01

Table 6: Effect of different adsorbent dosage (Corn Cobs) or dye removal (Initial concentration: 100mg/L, Initial pH of solution: 5.0, contact time: 240mins, Agitation speed: 300rpm, Temperature: 40°C, Size: 180 – 300µm).

Adsorbent dose (g/100ml)	Percentage of dye removal with time (min)					
	40	**80**	**120**	**160**	**200**	**240**
0.2	13.34	30.54	37.01	49.63	62.35	63.17
0.4	22.65	46.16	58.81	72.44	86.05	86.25
0.6	44.40	60.43	78.40	78.80	85.55	84.24
0.8	44.60	58.48	76.80	83.21	86.60	87.28
1.0	50.10	60.01	77.01	85.04	87.06	88.08
1.2	52.62	63.45	77.07	86.01	88.09	91.10

3.4 Effect of pH variation

Adsorption is also affected by change in pH of the solution as shown in fig 1. The hydrogen ion concentration (pH) initially affects the degree of ionization of the dye and the surface properties of the adsorbents. Experiment were carried out at 100mg/L, initial dye concentration with 1g/L adsorbent mass at room temperature (39 ± 1°C) for 3hours equilibrium time. It is clear from the fig1. That for each initial concentration value of $KMNO_4$, the percent removal increases as pH rises and after pH 5.0, the percent removal decreases with further increase in pH value. This clearly shows that the optimum pH for the present adsorbate – adsorbent system is 5.0.

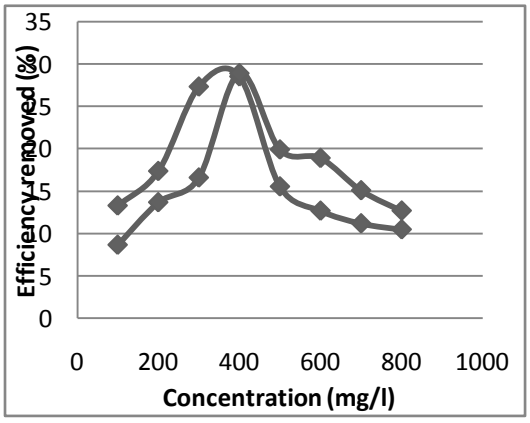

Figure 1: Effect of different initial dye concentration on dye removal using CC and AB derived adsorbent.

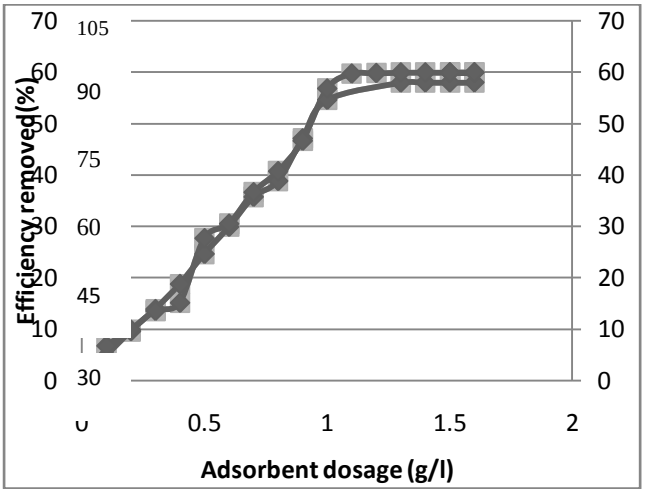

Figure 2: Effect of different adsorbent dosage on $KMnO_4$ removal using CC and AB derived adsorbent.

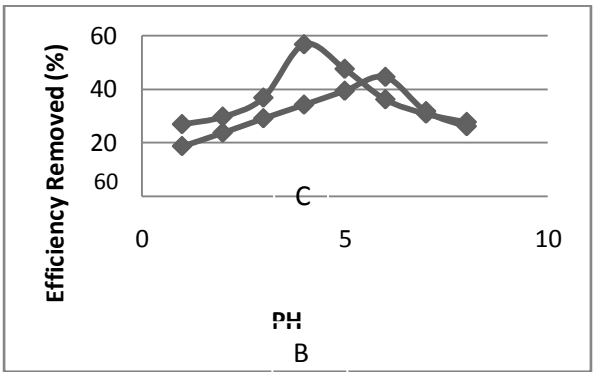

Figure 3: Effect of pH variation on potassium permanganate removal using corn cob and animal bone derived adsorbent.

3.5 Adsorption Isotherm

The distribution of dye between the adsorbent and dye solution when the system is at equilibrium, is important in order to obtain the adsorptive capacity of Animal bone and corn cob derived activated carbons.

Table 3 summarizes the Q_o and K_L) values for the freundlich isotherm and the correlation coefficients for the two isotherms. The Langmuir isotherms are found to lie between 1 and 10 for beneficial adsorption. This is represented by the following equation.

$$C_e/q_e = 1/Q_o^b + C_e/Q_o \quad (1)$$

Where q_e and C_e are defined as the amount of dye adsorbed (mg/g) and equilibrium liquid phase concentration (mg/L), respectively, K_L is a direct measure of the intensity of the sorption (1mg/L) and Q_o is a constant related to the area occupied by a monolayer of adsorbate, reflecting the maximum adsorption capacity (mgg^{-1}) (Ezeugo, 2012). From the data of C_e/q_e Vs C_e, Q_o and K_L can be determined from M, the slope and intercept. The essential characteristics of Langmuir equation can be expressed in terms of a dimensionless separation factor R_L, which is defined by

McKay *et al* (1985) as:

$$RL = 1/1 + K_L C_o \quad (2)$$

Where co is any adsorbate concentration at which the adsorption is carried out. Favorable adsorption is indicated by $o< R_L<_1^{20}$. The Langmuir isotherm constants were presented in table 7 while linear plots of C_e/q_e against C_e suggest the applicability of the Langmuir isotherms, figures 1, 2, 3.

The value of RL indicates the type of the isotherm to be either favorable ($O<R_L<1$), unfavorable ($R_L>1$), or irreversible (R = 0). The value of R_L was found to be between 0 and 1 for AB and CC suggesting the isotherm to be favorable at the concentration studies. The freundlich equation is also employed for the adsorption of potassium permanganate ($KMNO_4$) on the AB and CC adsorbent. The freundlich isotherm (Freundlich, 1906) is represented as $\log q_e = \log K_f + (1/n) \log C_e$ (3) Where q_e is the amount of potassium permanganate dye adsorbed (mg/g), C_e is the equilibrium concentration of dye in the solution (mg/L) and K_f and n are constants incorporating all factors affecting the adsorption process such as adsorption capacity and intensity respectively. Linear plot of $\log q_e$ versus $\log C_e$ shows that the adsorption of $KMNO_4$ follows also the freundlich isotherm (figure 3 and 5 for AB and CC respectively). The values of K_f and n are given in table 7. As seen from table 7, the increase in negative charges on the adsorbent surface that makes electrostatic forces like Vander Waals between the adsorbent surface and dye ion. The molecular weight, size and radii either limit or increase the possibility of the adsorption of the dye onto adsorbent. However, the values clearly show the dominance in adsorption capacity. The intensity of the adsorption is an indicative of the bond energies between dye and adsorbent and the possibility of slight chemisorptions rather than physisorption. However, the multilayer adsorption of potassium permanganate through the percolation process may be possible. The "n" value for AB and CC activated carbon are evaluated as and was in between 1 and 10 indicating the adsorption is much more favorable for selected adsorbents.

Table 7: Langmuir and freundlich isotherm of activated carbon prepared from AB and CC for $KMNO_4$.

Adsorbent	Langmuir isotherm		Langmuir isotherm		Freunclich isotherm		
	Q_o	K_L	Correlation coefficient (r)	R_L	Intercept (K_f)	Slope (1/n)	Correlation coefficient (r)
Animal Bone (AB)	28.038	0.0500	0.939	0.246	0.486	0.667	0.953
Corncobs (CC)	26.004	0.0320	0.860	0.3461	0.85	0.357	0.778

3.6 Conclusions

The results of the present investigation showed that Animal bone and corn cob derived activated carbon has considerable potential for the removal of potassium permanganate from aqueous solution over a wide range of concentrations. The adsorbed amount of dye increased as the surface area increased with an increasing adsorbent mass. The adsorption capacities and intensities show that the activate carbon from Animal bone is more

efficient for the decolourization of aqueous solution than that of corncob and it is of higher surface area. The surface charge on the adsorbent and the solution pH play a significant role in influencing the capacity of an adsorbent towards dye ions. A decrease in the pH of solution leads to a significant increase in the adsorption capacities of dye onto Animal bone based activated carbon and corn cob derived activated carbon. The adsorbed amounts of dye increased with increase in contact time and reached the equilibrium in 180mins. The equilibrium data have been analyzed using Langmuir and freundlich isotherms. The characterization parameters for each isotherm and related correlation coefficients have been determined from graphs of their linear equations. It was found that the freundlich isotherm appears to fit the isotherm data better than the Langmuir isotherm.

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