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Batch Comparative Adsorption Studies of Fluoride Using Activated Carbon Derived from Magnifera Indica (Mango) Seeds and Peltophorum Pterocarpum (Copper Pod) Seeds

Keerthi. B. Gurani
Asst Professor, Department of Civil Engineering
KCT Engineering College, Gulbarga.
kvgurani@gmail.com
Shashikant. R. Mise
Professor, Department of Civil Engineering
PDA College of Engineering, Gulbarga.
srmise45@yahoo.com

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Abstract: Activated Carbon (AC) is frequently used for purification due to its ability to remove pollutants either from air or water. Its porous structure allowed it to capture pollutants and is widely recommended for numerous applications in water and wastewater treatment. However, AC applications fields are restricted due to high costs. In this project, AC was prepared from locally available Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum(Copper Pod) seeds. The removal of Fluoride from the synthetic sample by adsorption on physically activated carbon prepared from Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum(Copper Pod) seeds have been carried out at room temperature 32 ± 1oC. Batch adsorption processes were conducted for the removal of Fluoride on physically activated carbon for optimum time, optimum dosage, optimum pH sorption kinetics sorption equilibrium. The optimum contact time, adsorbent dose, and pH for carbon was 35 min, 100 mg and 8.00 and 40min, 160mg and 8.50 respectively, with a fluoride removal efficiency of 78.96%, 96%, 97% and 45%, 58%, 96% respectively have been observed. Adsorption kinetics and sorption isotherm models were studied by altering different process parameters like adsorption time and dosage respectively. Langmuir, Freundlich, and Temkin isotherms are studied through this experimental data and found to be well followed.

Keywords: comparative studies batch adsorption fluoride

I. INTRODUCTION

Water is life, but we are still unable to provide everyone with safe drinking water that is both accessible and inexpensive. Water is an essential component of life for all living things. However, only a small percentage of the population has access to potable drinking water anymore. Others drink polluted water to varying degrees. The topic of providing clean drinking water is causing a lot of anxiety around the world, especially in emerging and poor countries. Because India is a developing country with a large population living in villages with limited infrastructure, a high rate of illiteracy, and a lack of awareness of sanitation and hygiene, the concept of safe drinking water takes on increased significance. However, the majority of the rural population relies on contaminated ground water for drinking, which is contaminated with a variety of salts and minerals.

One of them is an overabundance of fluoride in the water, which has a negative impact on people's health. Fluoride is a hazardous substance that is more toxic than lead but less toxic than arsenic, and it is an accumulative toxin. Continuous use of polluted water causes dental and skeletal fluorosis, as well as a variety of other health problems such as gastrointestinal issues. Because "Fluorosis" is considered an incurable condition, the only way to solve this problem is to prevent it. As a result, investigations on the de-fluoridation of water utilizing a range of adsorbents have become increasingly important in recent years. De-fluoridation can be accomplished using a variety of ways, including reverse osmosis, electrodialysis, and ion exchange. NEERI's Nalgonda de-fluoridation process has recently gained popularity, however it has its own set of limitations. De-fluoridation activity is also tested with a variety of adsorbents. De-fluoridation capacity of activated alumina is said to be high.

II. OBJECTIVIES

1. The preparation of physical activated carbons.
2. To determine the characteristics of the prepared carbons.
3. To determine the fluoride removal by adsorbent as a function of contact time, adsorbent dosage and pH.
4. To study sorption kinetics and sorption isotherm models.

III. LITERATURE REVIEW

Materials such as thermally activated Neem and Kikar leaves (Sunil Kumar1,et al.,2008), graphite (M Karthikeyan & K P Elango*,2008), Gulmohar fruit shell (Pallavi Vijyakumar & S.R.Mise,2008-09), Rice Husk (Waheed S. Deshmukh,et al.,2009), Cynodon Dactylon (G. Alagumuth*,et al,2010), Phyllanthus Emblica (Veeraputhiran V. and Alagumuthu G.*2011), by Multani Matti and red soil (Gandhi N.[1], et al, 2012), Acacia Auriculiformis scrap wood char (Monal Dutta, et al,2012), possotia(vitex negundo) leaf by (pranjal saikia et al,2017), Fired clay pots by (G.P.Kof et al, 2017), As adsorbents, (Das Kumar et al 2011) utilised commercial and natural adsorption, while (Salwa A Ahmed et al, 2015) used mango seeds. Activated carbon made from a variety of basic materials is effective at removing fluoride from drinking water. There have been no reports of de-fluoridation using Phoenix Dactylifera (Date Palm) seeds. As a result, the purpose of this research is to see if carbon made from Phoenix Dactylifera (Date Palm) seeds and Magnifera Indica (Mango) seeds can be utilised as an adsorbent to remove fluorides from drinking water.

IV. MATERIALS AND METHODS

4.1 Materials:

The material used for preparing carbons was fruit of Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum(Copper Pod) seeds. Physical characteristics of carbon like moisture content, decolourising power, pH, surface area, bulk density and specific gravity has been carried out and shown in Table 4.1 below.

4.2 Characteristics of activated carbon:

It is necessary to understand some of the characteristics of Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum (Copper Pod) seeds carbon before using it as an adsorbent. These characteristics include moisture content, ash content, decolorizing power, surface area, pH, specific gravity, bulk density, and the prepared carbons. Table 4.1 displays the results.

Table.4.1 Characteristics of Prepared Activated Carbons

S.No	Characteristics	Physical Activated Carbon	
		Mango Seeds	Copper Pod Seeds
1	Moisture content (%)	6.76	5.00
2	Ash content (%)	15.20	16.63
3	Decolorizing power (mg/g)	45.0	116.7
4	Surface area (m^2/g)	580.64	596.90
5	pH	7.2	9.01
6	Specific gravity	1.218	1.5
7	Bulk Density (g/cm^3)	0.262	0.240

The analysis was carried out according to the 20th edition of "Standard Methods."

4.3 Methods:

4.3.1 Preparation of Physical Activated carbons:

The seeds of Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum(Copper Pod) seeds were cleaned, and broken into pieces, then washed in distilled water for about 8 to 9 times. The powder was then oven dried at 105±5°C for 24 hours. The oven dried powder was filled in small container in three layers, by compacting each layer without any air space to avoid the loss in weight of the powder, otherwise, the substance would be directly burned, leaving only the ash behind. The small container was then placed into a bigger container, such that sand surrounded the small container completely, the lid of the bigger container was tightly fitted. Then the setup was kept in muffle furnace and heated at steady rate to attain the temperature of 800°C.

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Thirty minutes after attaining the 800°C temperature the furnace was allowed to cool for about 10 hours and then the container is taken out. The activated Carbon thus obtained was sieved to 300 Micron in size, then packed in polythene bags and stored in desiccator.

4.4 Determination of optimum contact time:

The contact time has a major impact on adsorption. To investigate the effect of contact time, 100mL of 5mg/L fluoride solution was mixed with 100mg of activated carbon, and the mixture was agitated at various contact times ranging from 5 to 30 minutes (10mins, 20mins, 30mins up to 120mins). Then, using a UV-visible spectrophotometer, the filtrate was tested for fluoride concentration.

4.5 Determination of optimum dosage of adsorbent:

To establish the best dosage of activated carbon from Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum (Copper Pod) seeds, it was put to a conical flask holding a known concentration of fluoride solution (5 mg/L in 100mL) at several dosages ranging from (20mg, 40mg, 60mg up to 180mg). The filtrate was tested for residual fluoride content using a spectrophotometer after the solution in the conical flasks was stirred for optimum contact time.

4.6 Determination of optimum pH on Fluoride:

The pH at which adsorption occurs has a significant impact on the amount of adsorption that occurs. Equilibrium adsorption studies at various beginning pH values, ranging from 2.0 to 9.0, were used to investigate the effect of pH on fluoride adsorption. 0.1N H_2SO_4 or 0.1N NaOH were used to alter the pH of the solution. The activated carbon from Magnifera Indica (Mango) seeds and Peltophorum Pterocarpum(Copper Pod) seeds was blended and agitated for the duration of the optimum contact period, and the filtrate was tested for residual fluoride content. The pH at which maximum fluoride elimination occurs is referred to as the optimal pH.

V. RESULTS AND DISCUSSION

Fluoride removal effectiveness is measured in terms of.

- 5.1 Effect of contact time.
- 5.2 Effect of dosage.
- 5.3 Effect of pH.
- 5.4 Sorption Kinetics
- 5.5 Sorption Equilibrium

5.1. Effect of Contact Time:

The adsorption process is mostly influenced by contact time. Figure 5.1 depicts the influence of contact time on the elimination of fluoride from a synthetic sample. It has been discovered that the extent of Fluoride adsorption grows with time and reaches equilibrium at a specific time. As a result, Table-5.1 lists the optimum contact time for all produced carbons.

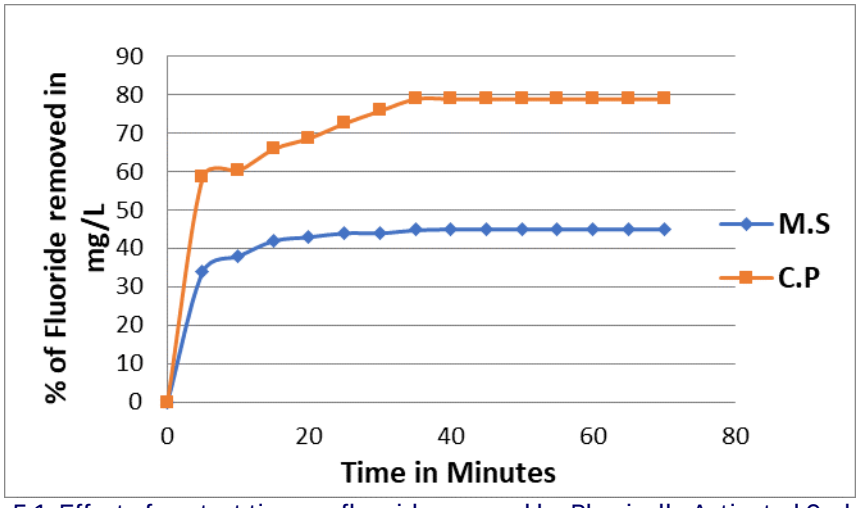

Fig.5.1. Effect of contact time on fluoride removal by Physically Activated Carbon

5.2 Effect of Adsorbent Dosage:

The effect of adsorbent dosage is investigated, and a graph of Fluoride removal percentage vs dosage is presented in Figure 5.2. As the carbon dosage increases, the amount of residual fluoride reduces dramatically and reaches equilibrium, as seen in the graph. The dosage that achieves maximum removal is referred to as the optimum dosage. As a result, Table-5.1 lists the recommended dosages for all types of produced carbon.

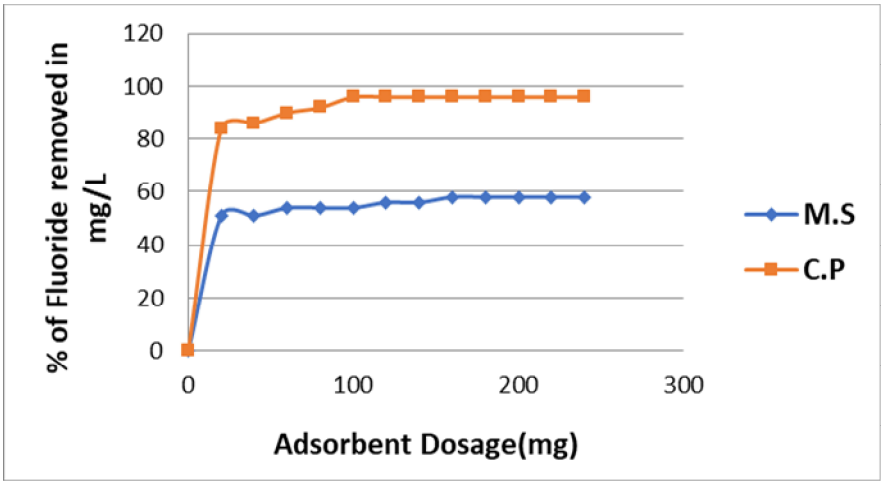

Fig.5.2. Effect of contact dosage on fluoride removal by Physically Activated Carbon

5.3. Effect of pH on Fluoride Removal:

The pH at which adsorption occurs has a significant impact on the amount of adsorption that occurs. The extent of adsorption removal effiencies of Fluoride by produced activated carbon was impacted by the pH of the solution at different pH values, as shown in Fig 5.3 and Table-5.1.

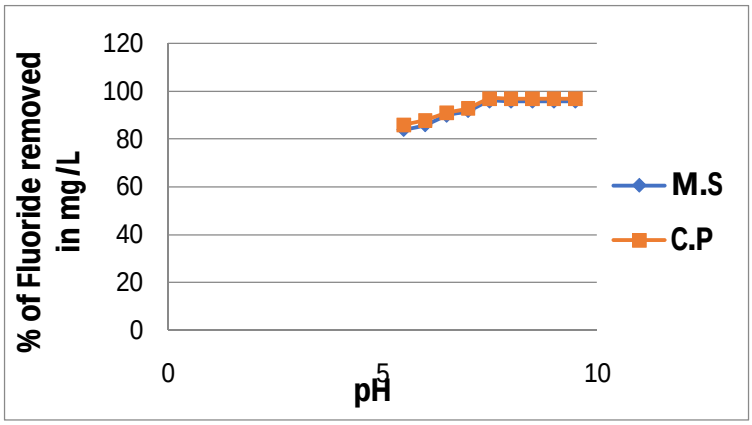

Fig.5.3. Effect of pH on fluoride removal by Physically Activated Carbon

Table.5.1, Optimum Time, Dosage and pH for Prepared Carbons

S.No	Type of carbon (Phyically Activated Carbons)	Optimum Time (min)	Optimum Dosage (mg)	Optimum pH
1	Mango seeds	40	160	8.50
2	Copper Pod seeds	35	100	8.00

5.4 Sorption Kinetics:

At room temperature, the kinetics of fluoride removal were studied at varied time intervals of adsorption. Table 3 shows the model values of the reaction rate constant for various carbons, with the resulting plots given in Figure 4. Table 4.2 shows the calculated and graphical values of 'K' for prepared carbon. The first order reaction was tested using batch kinetic data for fluoride adsorption. The first order reaction's rate equation is provided as level spiel.

L_n Ca / Co = K*T

i.e., 2.303 $\log_{10}$ a / (a – x) = K * t

i.e., $\log_{10}$ a / (a–x) = (k / 2.303) t

i.e., k = 2.303 / t x $\log_{10}$ a / (a –x)

Where a = initial concentration of the fluoride

x = the amount of fluoride that has been absorbed at any given time 't'.

a –x = residual amount of fluoride

K = rate constant

Equation (5) is of the type Y = mx

Where,

Y = $\log_{10}$ a / (a –x) and

x = t,

m = k / 2.303

Substituting the values in equation (5) we get

y = mx

As a result, a straight line graph with slope = k / 2.303 is produced. From the values of graphical and calculated "K" values, it is observed that adsorption of fluoride follows first order rate equations. Rate constant for each carbon comes to be constant, also it is observed that "K" values increases with the increase of Impregnation ratio and graphical values are slightly lower than the calculated values for physical activated carbons as shown as in Fig 5.4 and Table-5.2.

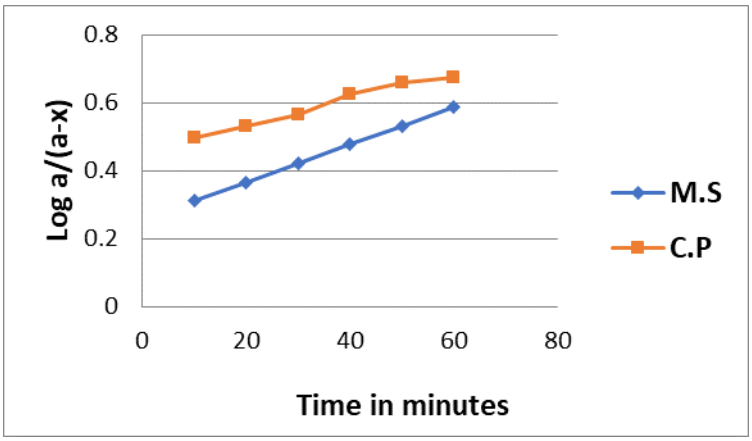

Fig.5.4 Plot of Reaction Rate Constant for Physically Activated Carbon

5.4.1 Pore Diffusion:

Further, the rate of pore diffusion can be determined by,

$C / C_0 = K_P \times t^{½}$

Where C = Concentration of sorbate at any time "t" (hr) in mg/L

C_0 = Initial concentration of sorbate in mg/L.

t = time taken for sorption

K_P = rate of pore diffusion

The plot of C / C_0 Vs $t^{1/2}$ for fluoride is shown which is straight line graph. The model values of C/Co and $t^{1/2}$ are shown. The calculated and graphical values of K_P for prepared carbon are shown. It is observed that graphical values of K_P values are greater than theoretical values as shown as in Fig 5.5 and Table-5.2. It demonstrates that pore diffusion reduces with time, resulting in an increase in adsorption. Data on pore diffusion confirms the rise in adsorption with time.

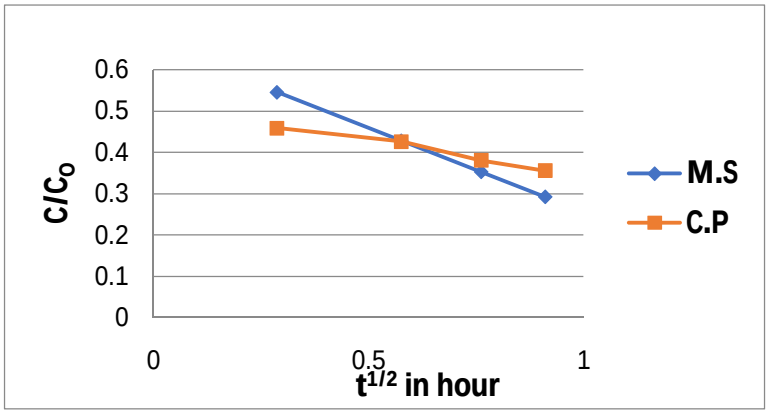

Fig.5.5 Webber and Morris Plot for Physically Activated Carbon

TABLE.5.2: SORPTION KINETICS

S.No	Type of Carbon (Physically AC)	K Values from calculation	K values from graph	Kp values from calculation	Kp values from graph
1	Mango seeds	0.323	0.367	0.301	0.406
2	Copperpod seeds	0.287	0.036	0.558	0.436

5.5 Sorption Isotherm Models:

Sorption equilibrium isotherms can be used to calculate the amount of sorbate necessary to sorb a specified amount of sorbate from solution. Freundlich, Langmuir, and Temkin isotherms are the most commonly used equations for presenting adsorption data. For all three isotherms, the sorption equilibrium data is filled.

5.5.1 Freundlich Isotherm

Taking log on both sides is the linear version of the Freundlich isotherm.

log10 = log10 K + 1 / n log10
At equilibrium,

X/M = Amount of sorbate adsorbed per unit weight of adsorbent.
X is the amount of adsorbate extracted in milli grams per liter (mg/L).
M =is the adsorbent's mass or weight.
C = Adsorbate equilibrium concentration in solution
K =is a sorption capacity-related empirical constant.
n = A sorption intensity-related empirical constant.

The equation is of the type y = c + m x. Where, c = $\log_{10}$ K
Y = log10 X / M, X = log10 C, and m = 1/n are the equations.

The plot of $\log_{10}$ X/M versus $\log_{10}$ C gives a straight line with slope 1/n and intercept = $\log_{10}$ K, knowing slope and intercept "1/n" and "K" are calculated. Experimental data are often plotted in this manner as a convenient way of determining whether removal of materials from solution is accomplished by adsorption, as a means of evaluating the constants "K" and "n" as shown as in Fig 5.6 and Table-5.3.

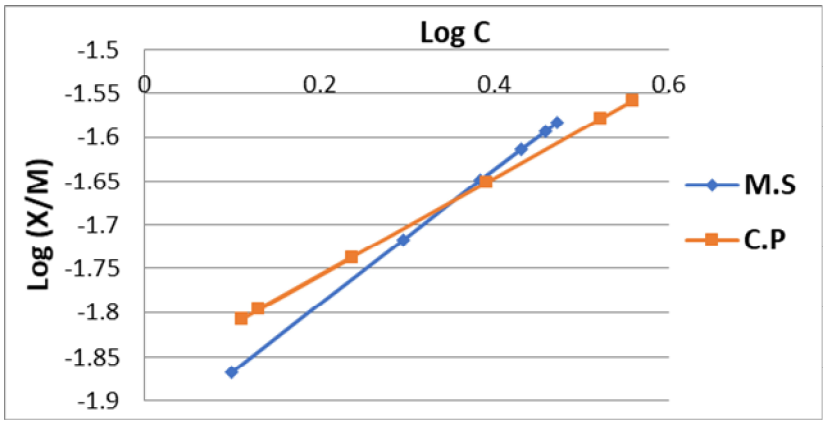

Fig.5.6 Plot of Freundlich Isotherm for Physically Activated Carbon

5.5.2 Langmuir's Isotherm

The Langmuir isotherm is used to describe the single layer adsorption and the linear form of the Langmuir's isotherm is, X/M = abc / (a + bc)

C / (X/M) = 1/ab + c / a
Where X/M denotes the amount of sorbate adsorbed per unit of adsorbent weight.
C= After adsorption, equilibrium or saturated concentration of adsorbent in solution.
'a' and 'b' are emperical constants.
The equation (4.3) is of the type y = c+mx, where c = 1/ab and m=1/a, x = c the plot of C/(X/M) Versus C produces a straight-line graph. The value of constants 1/b and 1/ab are calculated by intercept and slope.
1 / ab = intercept; slope = 1/a

Intercept = 1/b X slope; b = slope / intercept.
Calculated model data are shown in table-4.4. the plot of linearized Langmuir isotherm (C / X/M versus C) for different adsorbents are shown as in Fig 5.7 and Table-5.3. The slope and intercept of the aforementioned graphs are used to evaluate 'a' and 'b.' Least square analysis can be used to verify this, and graphical values are shown. On the basis of the findings, the following equations for various carbons are presented. Webber defines the main properties of the Langmuir isotherm in terms of a dimensionless constant distinct factor or equilibrium parameter, R = 1 / [(1 + a) Co]. Where a= is the Langmuir constant and C0 is the initial solute concentration in milligrams per litre. Webber has offered the following parameter identifying the kind of the isotherm from the preceding equation:

Values of R	Type of Isotherm
R > 1	Unfavorable
R = 1	Linear
0 < R < 1	Favorable
R = 0	Irreversible

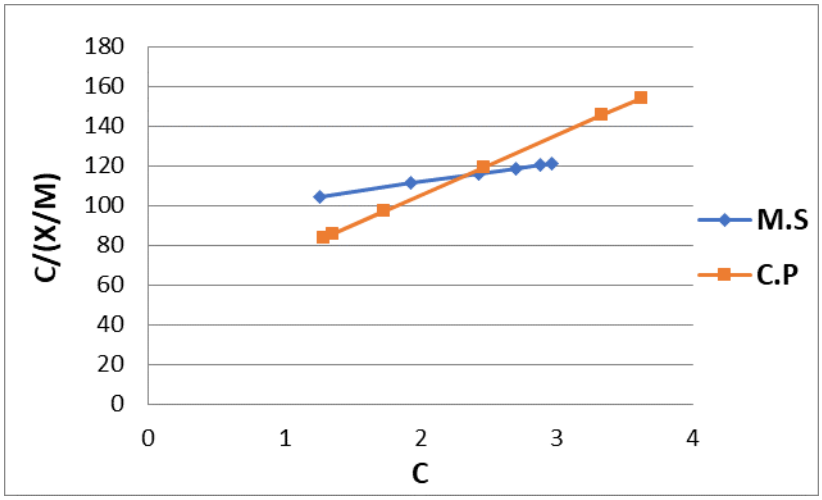

Fig.5.7 Plot of Langmuir Isotherm for Physically Activated Carbon

5.5.3 TEMKIN ISOTHERM:

The simple form of the adsorption isotherm model, which was designed to account for the chemisorption of an adsorbate on to the adsorbent, is X = a + b ln C.

Where C is the adsorbate concentration in solution at equilibrium (mg/L).

X represents the amount of metal absorbed per gram of adsorbent (mg/gm).

a = An adsorption capacity-related empirical constant.

b = An empirical constant related to adsorption intensity.

The energy of adsorption is a linear function of the surface coverage in the Temkin isotherm equation. Only an intermediate range of ion concentrations is valid for the Temkin isotherm. The plot of ln C vs X yields a straight line with slope and intercept; given the intercept and slope, the constants 'a' and 'b' are determined in relation to each other, as shown as in Fig 5.8 and Table-5.3.

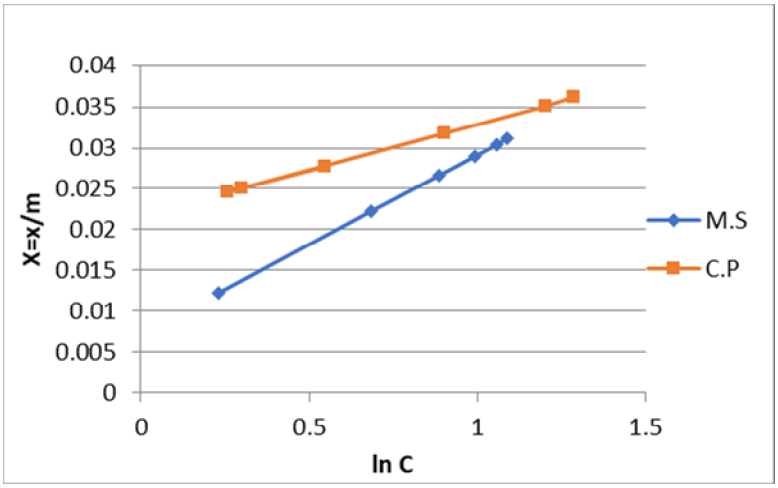

Fig5.8 Plot of Temkin Isotherm for Physically Activated Carbon

TABLE.5.3: SORPTION ISOTHERMS MODEL

S.No	Type of Carbons (Physically Activated)	Freundlich Isotherm		Langmuir Isotherm			Temkin Isotherm	
		Const '1/n'	Const 'k'	Const 'a'	Const 'b'	'R'	Const 'a'	Const 'b'
1	MANGO SEEDS	0.7657	0.0113	0.1023	0.106	0.181	0.0071	0.0219
2	COPPERPOD SEEDS	0.5564	0.0135	0.033	0.672	0.2151	0.0128	0.0110

VI. CONCLUSIONS

Based on the experimental study following conclusions were drawn:

1. The optimum contact time for physical activated carbons of 40 minutes and 35 minutes with removal efficiency of 45.50% and 78.96% respectively.
2. The result of experiment on optimization of dosage of adsorbent reveals that, increase in amount of adsorbent added, increases the removal of fluoride from the solution. Optimum dosage for physical activated carbons 160 mg and 100 mg with removal efficiency of 58% and 96% respectively.

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3. The adsorption of fluoride is mainly pH dependent. The removal efficiency of adsorbent increases with decrease in pH value. It has been observed that maximum adsorption taken place around pH 8.5 and pH 8.0 for physical activated carbons with removal efficiency of 96%, 97% respectively.
4. The rate of adsorption of fluoride obeys first order rate equation. It also follows Webber and Morris equation for pore diffusion.
5. The result of the isotherm models follows Langmuir, Temkin and Freundlich isotherms proves to be an favorable adsorption.

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