Removal of Phenolic Compound and COD from Landfill Leach ate by Commercial Activated Carbon

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***Abstract*--A laboratory adsorption study was conducted on real life MSW landfill leachate to evaluate the attenuation capacity of commercial grade powdered activated carbon (PAC) for the removal of phenol and COD containing in leachate sample. Batch studies were performed to examine the effects of various experimental parameters like adsorbent dose, pH, speed of agitation on the removal of phenol and COD by PAC and to establish the different adsorption kinetics and isothermal models. All the above experimental parameters were found significant in the adsorption process and test data showed that PAC adsorption is an effective tool for the removal of phenol and organic load from the MSW landfill leachate.In the present study the optimum dose was found to be 0.4 g/L and the equilibrium time was observed as 3hr. The phenol and COD removal were found to be 97% and 92% respectively at optimum condition.The equilibrium data was fitted to pseudo second-order adsorption kinetic model with rate constant value as 9.347 mg/mg/min and 0.026 mg/mg/min for phenol and COD respectively. The equilibrium data of phenol absorbed onto PAC were analyzed by Langmuir, Freundlich, Tempkin, Dubinin–Radushkevich and Redlich–Peterson adsorption isotherm models. Redlich–Peterson adsorption isotherm model found to be the best fit with the experimental data for removal of phenol and Tempkin isotherm for COD removal.**

***Key words: MSW; leachate; phenol; COD; adsorption; activated carbon; kinetics*.**

I. INTRODUCTION

Potential environmental pollution by hazardous chemical substances as released in landfill leachates due to uncontrolled and unscientific solid waste dumping in peri-urban area of municipal cities is an important ecological issue related to municipal solid waste management practice.Leachate as released from such sites, contains high amount of organic load (COD)and phenolic compounds besides other hazardous and recalcitrant substances Therefore, to reduce the environmental risk, it is desirable to lower the concentrations of such toxic chemical substances in leachates.[1]The characteristics of MSW landfill leachate changes with time and from site to site depending on the type of wastes disposed, rainfall, age of the landfill and design of the landfill etc [2].Phenol have a high toxic and great risk to human health and theenvironment, which can cause negative health effects onbrain, digestive system, eyes, heart, kidney, liver, lungs,skin as well as causing genetic damage. Thus, the removal of phenol from liquid phase of leachate is of great importance, and has been received particular concerns in the recent past [3].Combined Physico-chemical process based on chemical coagulation-flocculation and carbon adsorption has been reported to be an efficient treatment method for MSW landfill leachate [4].However, the conventional chemical sedimentation methods were difficult to remove these phenolic compounds effectively in polluted source water[3].Therefore secondary treatment is mandatory in case of MSW landfill leachate for the removal of phenolic as well as organic load. Out of various technologies studied by various researchers such as chemical oxidation, catalytic oxidation, biodegradation, membrane separation etc.adsorption process proved to be one of the low cost and reasonably the rational option and for which the process is extensively used for the removal of phenol. Several earlier investigators advocated for adsorption method for attenuating different types if toxic substances by different materials using as adsorbent since the process is considered to be highly efficient, reasonably cost effective too [5], [6], [7], [8], [9], [10], [11], [12]. Activated carbon is most widely used adsorbent as it exhibits excellent adsorption abilities for the removal of organic compounds from MSW landfill leachate in addition to its application for [8],[13]. The performances of adsorption of COD and phenol have been investigated by few researchers on various adsorbents [14] However, very few literatures are available on adsorptive removal of phenol and COD from real life MSW landfill leachate.

The objective of the present study was to investigate the adsorptive performance of commercial grade activated carbon for removal of phenol and COD for treatment of the real life MSW landfill leachate. Various influencing parameters affecting uptake rates on phenol, and COD absorption were also studied. Furthermore, the equilibrium and kinetic studies were performed to describe the adsorption isotherm models.

II. MATERIALS AND METHODS

A. *Materials*

MSW Landfill leachate

Leachate samples were collected from the Dhapa landfill, Kolkata, West Bengal, located in the eastern side of Kolkata metropolitan city of India at longitude and latitude 22.82°N 88.20°E respectively, in the state of West Bengal. The site is neither having any bottom liner nor any proper leachate collection and treatment system. Therefore, all the leachate generated finds its paths into the surrounding low level ditches and natural drainage outlets. Some of the leachate samples were collected from the base of solid waste heaps where the leachate was drained out by gravity. The climate in Kolkata is more or less tropical and 70 -75 rainy days per year. The average temperature is 26.6^{0}C. Average annual precipitation in Kolkata is 1600 mm, of which over 95 percent falls in the monsoon months of June through September. The Dhapa Disposal Site is managed by the Kolkata Municipal Corporation.Leachate samples were collected in a 5L capacity plastic bottle and kept in an ice container. The samples were collected from some specific points of leachate emergence point and immediately brought in the laboratory and preserved in deep freezer (at- 4ºC). Onsite dissolved oxygen (D.O.) fixation was done. A sum total of 12 leachate samples for different seasons were collected for analytical purpose. Major parameters were estimated in the laboratory with three replicates. The tests were carried out according to the "Standard Methods" (APHA)[15].

Test Adsorbent

Commercial grade activated carbon (powdered form) made by Merck Limited, Worli, Mumbai-400018 was used for carrying out the present experiment.

B. *Analytical methods*

The concentration of phenol was determined by using 5530-D treatment of various types of industrial wastewater. Direct Photometric Method of standard method APHA (1998) with wave length set at 500 nm using UV-visible spectrophotometer. COD was determined by using Closed Refluxing method in Hach apparatus. 5220. C: [15]

C. *Adsorption experiments*

Adsorption Isotherms

Batch adsorption experiments were performed in 250 ml plastic bottles in which 100 ml of the coagulant treated stabilized leachate bringing out from jar test apparatus was poured with the appropriate amount of powdered activated carbon in the range of 0.1–0.5 g/L and then shaken for a period of 3 hrs at a speed of 200 rpm. The sample then settled for 1 hrs and the aliquots were analyzed with respect to its COD content and residual phenol content after centrifugation and filtration through Whatman 42 paper.The uptake capacity of phenol and COD by adsorbent was calculated as per equation (1) given below:-

$$q_e = \frac{(C_0 - C_e)V}{M} \qquad (1)$$

Where, q_e is the amount of adsorbed phenol or COD at equilibrium (mg/mg); $C_0$and C_e are initial and equilibrium concentrations of phenol or COD (mg/L) respectively; V is the solution volume (L); and M is the mass of the adsorbent (mg).

Adsorption kinetics

All the kinetics studies were conducted in a series of 250 ml plastic bottles containing leachate sample of 100ml with 400mg/L adsorbent doses and placed on the mechanical shaker at room temperature at a speed of 200 rpm without pH adjustment. At different time intervals such as 20, 40, 60, 90, 120, 150, 180 min,samples were withdrawn for analysis ofresidual phenol and COD. The removal by PAC at a specific time was defined as follows Eq. (2):

$$q_t = \frac{(C_0 - C_t)V}{M} \qquad (2)$$

Where, q_t is the amount of adsorbed phenol at time t (mg/mg) ; V is the solution volume (L); C_0 and C_t are the initial and remaining phenol or COD concentrations in solution before reaction and at time t (mg/L), respectively; and M is the mass of the adsorbent (mg).

III. RESULTS AND DISCUSSION

A. *Adsorption of phenol and COD by PAC*

- Effect of Dose

The effect of adsorbent doses of PAC on removal of phenol and COD was also investigated on leachate sample. The concentration of phenol and COD were 10.02 mg/L and 5520mg/L respectively maintaining at initial pH of 8.4 at room temperature.The amount of dose of PAC added into the sample was ranging in between 0.1 g/L to0.5g/L. Fig. 1 shows an increase in percentage removal of phenol and COD with the increase in dose of adsorbent up to a certain limit and then it remains almost constant. This result was also compatible with the result of Ola Abdelwahab (2007).[11].Increase in the adsorption with increasing dose of adsorbent is expected due to the increase in more available surface area and the availability of more adsorption sites [16].

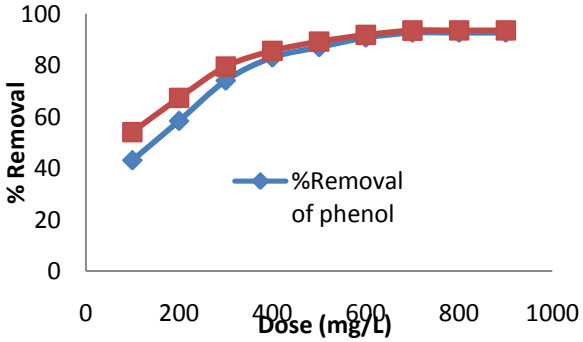

Fig 1. Effect of different doses of PAC on Removal of phenol and COD

- pH Effect

The experiments were performed in various initial pHs(2.0–9.0)with the optimum dose of 0.4 g/L as obtained from earlier plot and contact time equal to the equilibrium time of 3 hrs. Theinfluence of pH on the adsorption behavior of PAC for phenol and COD is presented in Fig. 2. This shows that the removal efficiency increased marginally when the pH increased from 2.0 to 8.0, and descended with the increase of pH higher than 8.0.The optimum pH was found to be pH 8.0 for both phenol and COD. The maximum removal efficiency was 92% and 93% respectively corresponding to above pH.

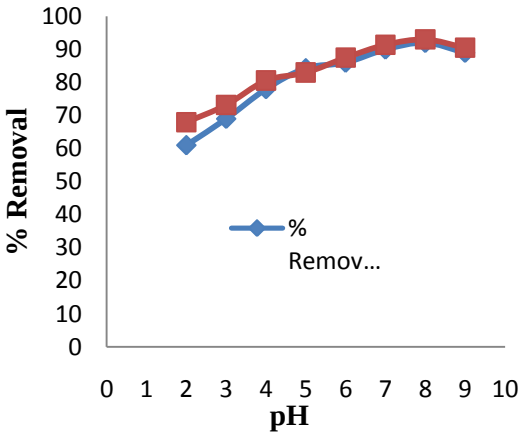

Fig 2. Effect of pH on removal phenol and COD

- Effect of agitation rate

The percentage removal of phenol and COD was found to be increased progressively and maximum (91 and 94%) was attained at 300 rpm agitation rate and it reduces gradually as the agitation rate decreases. The effect of agitation speed on the removal of phenol and COD has been plotted in Fig. 3. From the above figure it is found that the removal of sorbate (COD and phenol) depended on the speed of shaking device. However, the saturation of adsorption is entirely independent of agitation rate as also reported by V P Vinod and T S Anirudhan (2002) [17].

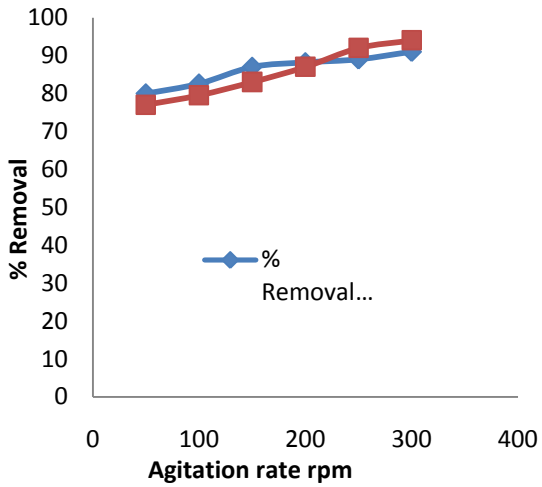

Fig 3. Effect of agitation rate on removal of phenol and aCOD

B. Adsorption kinetics

The kinetic investigation was conducted at optimum dose of 0.4 g/L of PAC, pH of the sample was 8.4 and agitation rate was 200rpm at room temperature on the leachate sample containing phenol concentration of 10 mg/L and COD concentration 5520 mg/L.The amount of phenol and COD adsorbed at various time intervals under the above mentioned conditions is shown in Fig. 4.It can be seen that the amount adsorbed was initially 80% and 77% for phenol and COD respectively in the first 10 minutes, with progressively increase in contact time it gets increased and reached almost constant values of 97.5% and 92% at the equilibrium time of 3 hrs. This may be attributed to the formation of a monolayer of phenol and organic molecules on the outer surface of the adsorbent. Later, a slower rate of removal was controlled by pore diffusion onto the inner surface of the adsorbent particles through the monolayer film due to continuous agitation [18].

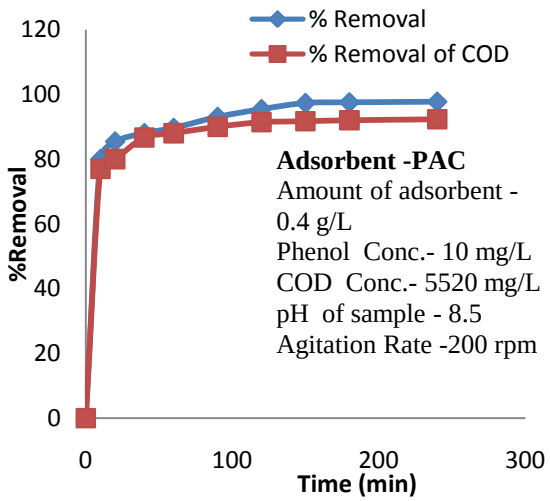

Fig 4. Time-Concentration kinetic study for phenol and COD removal

Sorption kinetic model

The batch kinetic results were also tested in four kinetic reaction models viz. first order reaction model based on the

solution concentration, pseudo-first order equation on the basis of adsorbent uptake capacity, second order reaction model based on the solution concentration and pseudo-second order reaction model based on the solid-phase sorption [19].The experiment was carried out to evaluate the best fit sorption reaction order and mechanism. Based on experimental data kinetic constants as obtained from different sorption kinetic models and isotherm analysis for both phenol and COD removal by PAC adsorbent are presented in Table 1 and Table 2. respectively.

First order reaction model

The linear form of the first order rate equation is expressed as cited in Benefield & Randall (1980).[20]

$$lnC_t = lnC_o - K_1 C_t \qquad (3)$$

Where, C_t is residual concentration of solute at time t,$C_o$is initial concentration of adsorbate, and K_1 is first order reaction rate constant.Figure5.shows the plot between log CtVs t .The best fit line was drawn and R^2 was found as 0.93 for phenol and 0.758 for COD. From the R^2 values it can be revealed thatthe adsorption followed reasonably the first order model in phenol removal but it was poor in case of COD removal

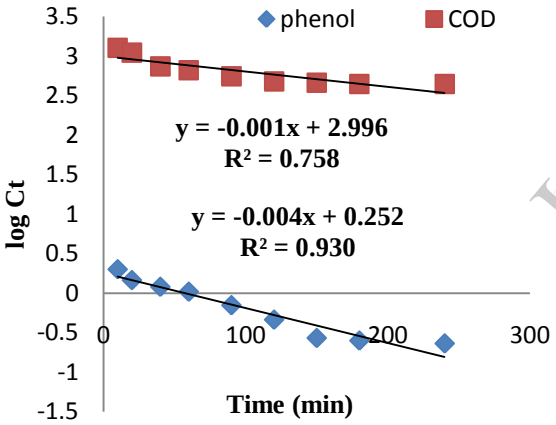

Fig 5. First order reaction model for removal of phenol and COD by PAC

Pseudo-first order reaction model

Linear form of equation as per Pseudo-first order reaction model which was described in Lagergren (1998) [21] was formulated based on the solid capacity for the sorption analysis.

$$\ln(q_e - q_t) = lnq_e - K_{S1}t \qquad (4)$$

Where,

$q_e$is amount of solute adsorbed per unit weight of adsorbent at equilibrium,

q is amount of solute adsorbed per unit weight of adsorbent at time t,

$K_{S1}$is pseudo-firstorder reaction rate constant.

After plotting the graph of straight line between ln(qe-qt) Vs. t as shown in Fig 6 it can be stated that R^2 value for COD removal is 0.991 which is better fitted than first order reaction model. But R^2 value of

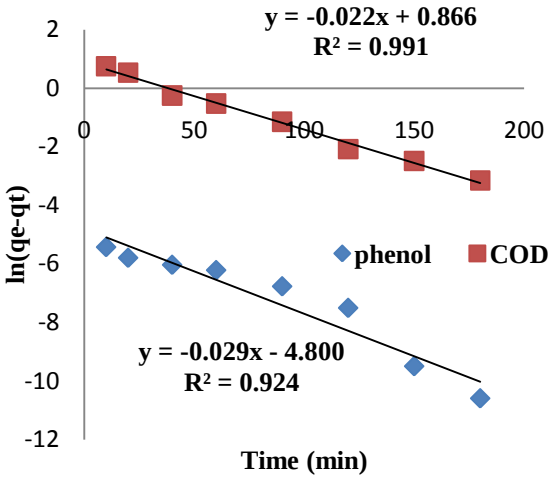

Fig. 6.Pseudo-first order reaction model for removal of phenol and COD by PAC

phenol removal is 0.9243 which is very nearer to the value in first order reaction model.

Second order reaction model

Thelinear form of second order reaction model as was proposed by Ho and McKay (1999) [22]is given hereunder

$$\frac{1}{Ct} - \frac{1}{C0} = K_2\, t \qquad (5)$$

Where,

K_2 is second order reaction rate constant.

A linear plot of 1/Ct against t gives the reaction rate of Second order as shown in Fig 7, which demonstrates the R^2 value for phenol removal as 0.936 and for COD removal as 0.8358

Pseudo-second order reaction model

As per [22], the Pseudo-second order reaction order is expressed as below

$$\frac{1}{(qe - qt)} = \frac{1}{qe} + Kt \qquad (6)$$

.

Where,

K is proportionality rate constant. The above equation is the integrated rate law for a pseudo-second order reaction, which after rearrangement is expressed in the following form

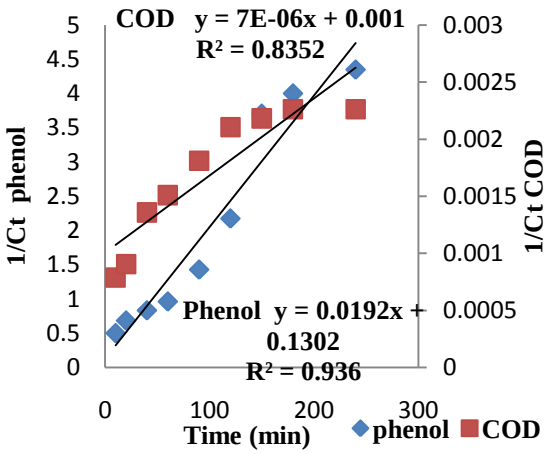

Fig. 7 Second order reaction model for removal of phenol and cod by PAC

$$\frac{1}{qt} = \frac{1}{h} + \frac{1}{qe}t \qquad (7)$$

Where, h = $K_{S2}qe^2$ (K_{S2} is pseudo-second order reaction rate constant)

Pseudo-second order kinetics model is the linear graph of t versus t/qt as shown in Fig 8.The plot clearly presents that Pseudo-second order kinetic model is the best fit kinetic model for the adsorption study for the removal of phenol and COD by PAC from MSW landfill leachate. The R^2 values for the plot are 1 and 0.999 respectively

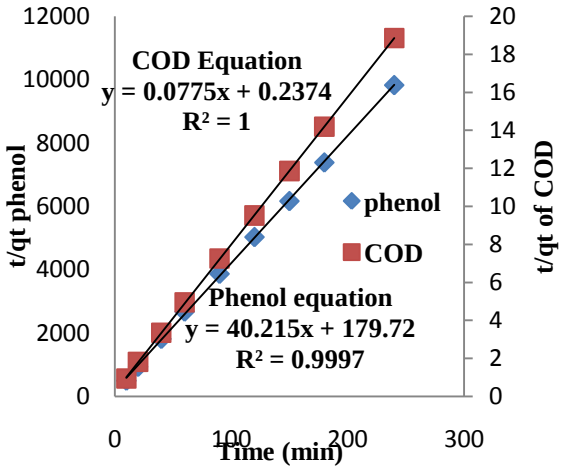

Fig 8 Pseudo-second order model for removal of phenol and COD by PAC

C. Adsorption isotherms

In this study, Langmuir isotherm, Freundlich isotherm, Temkin isotherm,Dubinin–Radushkevich isotherm and Redlich–Peterson isotherm on phenol and COD adsorption on PAC were investigated.

- Langmuir isotherm

The Langmuir isotherm equation is expressed as:

$$\frac{\mathbf{1}}{\boldsymbol{qe}} = \frac{\mathbf{1}}{\boldsymbol{K}} + \frac{\mathbf{1}}{(\boldsymbol{K} \times \boldsymbol{C0})} \times \frac{\mathbf{1}}{\boldsymbol{Ce}} \qquad (8)$$

Where,
qe (mg/mg) is the amount of adsorbate adsorbed per unit mass of adsorbent;
K is Langmuir constant;
Ce is the equilibrium concentration of adsorbate;
C_0 is original concentration of adsorbate.
Linear expression is obtained by plotting the graph of $\frac{1}{Ce}$ verses $\frac{1}{qe}$ as shown in Fig 9. and Fig 10.

Table 1. First order kinetic model, Pseudo- first order kinetic model, Second order kinetic model and Pseudo- second order kinetic model linear equations and R^2 values for phenol and COD adsorption by PAC.

Kinetic model	Phenol Removal		COD Removal	
	Equation	R^2	Equation	R^2
First order	Y=0.0044X+0.25	0.937	Y=-0.001 X + 2.996	0.758
Pseudo-First order	Y=-0.029X- 4.8	0.924	Y=- 0.022 X - 0.866	0.991
Second order	Y=0.0192X+ 0.13	0.936	Y=7 *10-6 X + 0.001	0.835
Pseudo second order	Y=40.21X+ 179.7	0.999	Y= 0.077 X + 0.237	1

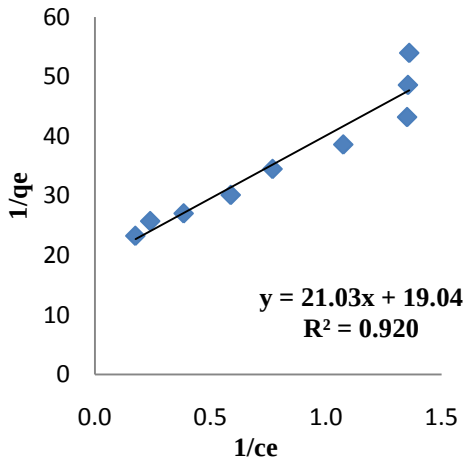

Fig. 9 Langmuir Isotherm for removal of phenol by PAC

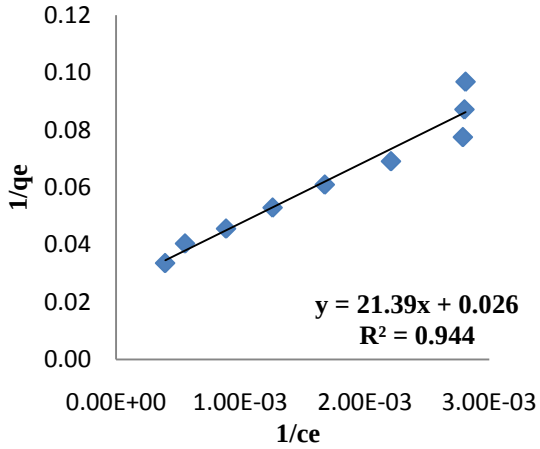

Fig. 10 Langmuir Isotherm for removal of COD by PAC

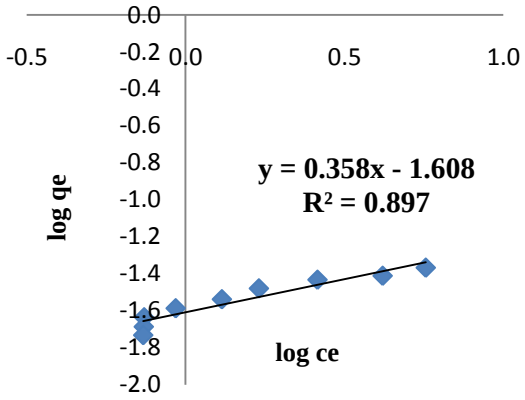

Fig. 11 Freeundlich Isotherm for removal of phenol by PAC

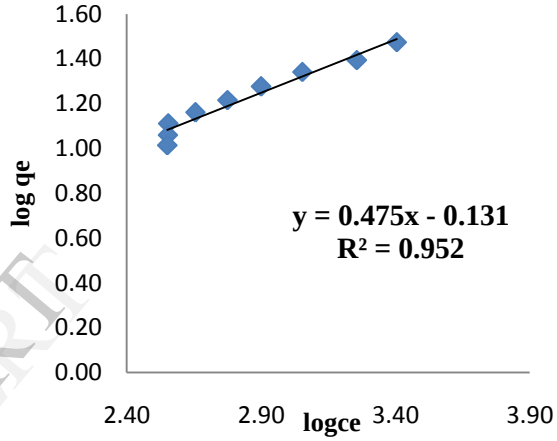

Fig. 12 Freeundlich Isotherm for removal of COD by PAC

- Freundlich isotherm

The Freundlich equation is expressed as follows

$$\log qe = \log kf + \frac{1}{n} \times \log Ce \qquad (9)$$

Where,
n is the Freundlich constants. The value of n is indicative of adsorption intensity.
kf is the distribution coefficient and indicative of the relative capacity of the adsorbent. The
value of n is indicative of adsorption intensity.
The plot is obtained by plotting the data of logCe versus log qe as shown in Fig 12 and Fig 13.

- Temkin Isotherm

The Temkin Isotherm can be expressed as

$$\boldsymbol{qe = \frac{R_T}{b_T} \times A_T + \left(\frac{R_T}{b_T}\right) lnCe} \qquad \boldsymbol{(10)}$$

Where,
$\frac{R_T}{b_T}$ is slope of the line,
R is universal gas constant (8.314 J/mol K)
T is temperature in Kelvin (K),
A_T is the equilibrium binding constant.
The graph is obtained by plotting the data of lnCe verses qe as shown in Fig 14.and Fig. 15.

- Dubinin–Radushkevich isotherm

The Dubinin-Redushkevich isotherm expression is presented as follows

$$ln(q_e) = ln(q_s) - K_{ad} \times \varepsilon^2 \qquad (11)$$

Where,
q_e is amount of adsorbate in the adsorbent at equilibrium(mg/g);
q_s is theoretical isotherm saturation capacity (mg/g);
Kad is Dubinin–Radushkevich isotherm constant (mol2/kJ2) and
ε^2 I s Dubinin–Radushkevich isotherm constant.

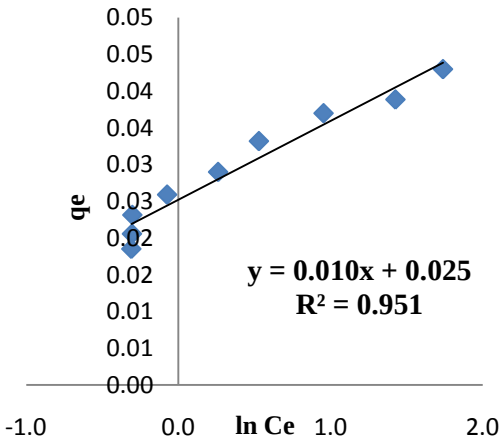

Fig. 13 Temkin Isotherm for removal of phenol by PAC

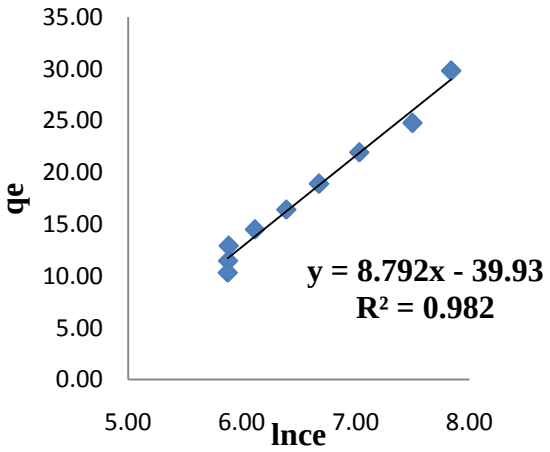

Fig. 14 Temkin Isotherm for removal of COD by PAC

The graph obtained when the adsorption data are plotted as a function of logarithm of amount adsorbed $ln(q_e)$ verses ε^2 as show in Fig. 15 and Fig. 16

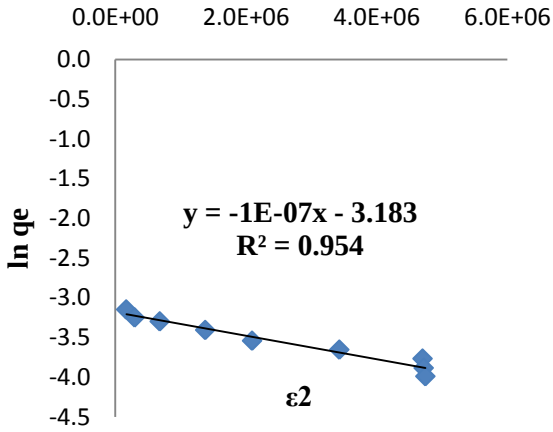

Fig. 15 Dubinin-Redushkevich Isotherm for removal of phenol byPAC

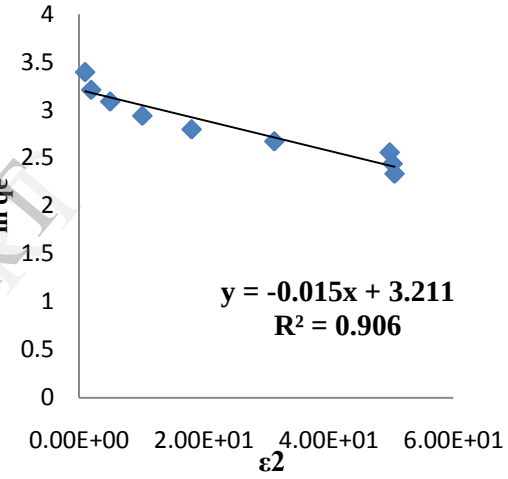

Fig. 16 Dubinin-Redushkevich Isotherm for removal of COD by PAC

- Redlich–Peterson isotherm

The linear equation of Redlich- Peterson isotherm is as follows

$$ln\left[K_R\frac{C_e}{q_e}-1\right]=gln(C_e)+ln(a_R) \qquad (12)$$

Where,
g is Redlich–Peterson isotherm exponent
$K_R=\frac{1}{[1+(K_a\times C_0)]}$ In which k_a is Langmuir isotherm constant; C_0 is initial concentration of adsorbate
a_R Redlich–Peterson isotherm constant (1/mg)
The graph is obtained against the plot of $ln(C_e)$ verses $ln\left[K_R\frac{C_e}{q_e}-1\right]$ as shown in Fig 17

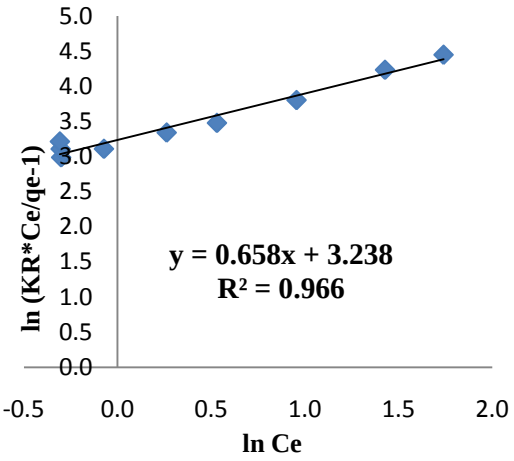

Fig. 17 Redlich–Peterson Isotherm for removal of phenol byPAC

The Redlich- Peterson isotherm model was observed to be not suitable for the removal of COD by PAC because the K_R value is very less as initial concentration of COD is 5520mg/L.

The constants obtained from fitting all the data to different isotherm models as mentioned above are listed in Table 2.The R^2 values of the entire isotherm models were above 0.9 except Freundlich isotherm used for the removal of phenol, though it was close to 0.9. It evince that all the models tested with experimental data well fittedto the adsorption models aswell. The Redlich-Peterson model indicates the best fit results for the phenol adsorption as it gives highest R^2 value amongst remaining isotherm plots. For the COD adsorption experiment Temkin isotherm is the best fit model, its R^2 value is 0.982.The results obtained in previous works are slightly differs from present work. According to Yan Ma et al. (2013)[23], Langmuir isotherm model is perfect fit model for the removal of phenol by adsorption on PAC, Freundlich isotherm works almost equal to Langmuir isotherm. In this study, all the 1/n values were below one, indicating the adsorption follows a normal Langmuir isotherm. The maximum amounts of the phenol adsorption 92% and 97% COD adsorption onto PAC respectively indicating PAC is an effective adsorbent for the removal of both the ingredients present in the MSW landfill leachate.

Table 2. Langmuir, Freundlich, Temkin and Dubinin–Radushkevich and Redlich–Peterson isotherm constants for the adsorption of Phenol and COD on PAC.

Adsorbate	Langmuir Isotherm				Freundlich Isotherm			
	$\frac{1}{K}$	K	Slope m	R^2	$\frac{1}{n}$	n	K_f	R^2
Phenol	19.04	0.053	21.03	0.920	0.358	2.79	0.25	0.897
COD	0.026	38.36	21.39	0.944	0.475	2.11	0.739	0.952

Adsorbate	Temkin Isotherm				Dubinin–Radushkevich isotherm			
	$\frac{RT}{b_T}$	b_T	A_T	R^2	K_{ad}	lnq_s	q_s	R^2
Phenol	0.010	2.53*10^5	2.5	0.951	-10*10^{-7}	-3.18	-0.041	0.954
COD	8.792	288.41	-4.54	0.982	-0.0015	3.211	24.80	0.906

Adsorbate	Redlich–Peterson isotherm			
	g	K_R	a_R	R^2
Phenol	0.658	0.653	25.48	0.966
COD	Not Suitable			

IV. Conclusion

In the present study, the adsorption potential of PAC for the removal of phenol and organic load were tested by conducting batch experiments on real life MSW landfill leachate.Batch kinetic results demonstrated that the equilibrium time for removal of both phenol and COD was 3hrs and percentage removal was 92 and 97% respectively. Different effect studies like dose effect, pH effect and agitation effect were appreciably influences the removal rate in the present investigation. The adsorption behaviors was well described by pseudo second order adsorption kinetic model.Five adsorption isotherms (Langmuir, Freundlich and Tempkin, Dubinin–Radushkevich and Redlich–Peterson isotherm isotherm) were further investigated and it was observed that Redlich-Peterson model indicates the best fit for phenol adsorption whereas,Temkin isotherm is the best fit model for COD removal.

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