



Adsorption of some organic phenolic compounds using activated carbon from cypress products

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ABSTRACT

Phenolic organic compounds are extremely highly generated from many resources which disposed to sewerage system without treatments that increase the risk of contaminating water resources. This study is focused on preparing and studying the properties of activated carbon produced from cypress fruit by chemically activation using phosphoric acid (H_3PO_4) as an activating agent. The activated carbon used to adsorb p-nitrophenol (PNP) from the aqueous solution. Result show that the activated carbon produces from cypress fruit gives good percentage yields which reach up to 51.8%. Surface area determined by iodine number showed $524.1m^2/g$. The adsorptive properties of CFAC were investigated in terms of adsorbent dose, PNP concentration, pH, and temperature and contact time in a batch system. Results indicate that the optimum percent of PNP removal 90.9 % when adsorbent dosage 0.3g and PNP concentration 80mg/L and percentage removal of PNP increase when the concentration of PNP decrease which maximum percentage removal reach 93.2% when PNP concentration 20mg/L and 0.1g CFAC. The effect of temperature on adsorption by CFAC has also been investigated in the range of 15-45 °C. The results indicate that the temperature slightly affected effectiveness of CFAC adsorption. The results showed that equilibrium time for PNP adsorption is 150min, but most the adsorption attained within the first ten minute. Results investigate that the produced cypress fruit activated carbon (CFAC) adsorption equilibrium is represented by both Freundlich and Langmuir equilibrium model, but Langmuir model describe very well the adsorption. The main properties of Langmuir equation can be expressed in term of separation factor, R_L . The R_L equal 0.053 that indicate the adsorption favorable.

Keywords: adsorption, kinetics, cypress, p-nitrophenol, carbon, activation.

INTRODUCTION

Industry revolution process especially in agroindustry, petrochemical industry, and oil industry generate phenol and phenolic derivative which appear in wastewater which usually more than the threshold level which considers as a safe level. Management of this wastewater becomes the main challenge in management of wastewater.

Several methods used to remove phenol and other phenolic compounds from waste water [1-4]. Biochemical oxidation is the base of many methods but these methods suffer from limitation as; low efficiency in the removal of low concentration. Using activated carbon (AC) consider effective method used to remove the contaminant especially phenols and phenolic compounds because it has large surface area, micro-porous properties, capacity of adsorption, high purity and easy to available [1]. Many studies investigate the adsorption of phenol and other phenolic compounds on AC which prepared from agricultural by-product, and low-rank coals [5].

The AC can be derived from many agricultural raw materials for example rice husk, olive waste, date stone and other agricultural residuals. Using agricultural by-product and unused agricultural product have many advantages as; change unwanted agricultural product or agriculture waste to useful product, increasing used of AC to remove contaminants of environment. Producing AC based on two main steps. First, carbonization under nitrogen gas flow with high temperature. Second, activation which may achieve chemically using chemical or physical agents [6].

In this study we investigated the adsorption mechanisms, three simplified kinetic models, i.e., pseudo-first-order, pseudo-second-order, second order and intraparticle diffusion were applied to fit the kinetic data obtained by for the adsorption of PNP into CFAC. The rate constants for the four models were determined and the correlation coefficients were calculated. The kinetic data for adsorbents supports pseudo-second order model for PNP adsorption.

EXPERIMENTAL SECTION

2.1 Materials

Precursor: Cypress fruit were used as the precursor in the preparation of activated carbon. Dried cypress fruit were collected from the cypress trees near Nablus area, crushed using moulinex device, and sieved. Only the fractions of particle sizes comprised between 0.3mm to 1.4mm were selected for the preparation of activated carbon. The activator was Phosphoric acid H_3PO_4 with purity of 98% used as chemical reagent for activation of cypress crushed fruit. The P-nitrophenol of purity higher than 97 % was used as adsorbate in this study. All other chemical used such as hydrochloric acid, sodium thiosulfate, iodine and sodium hydroxide were of analytical grades and purchased from Aldrich, USA.

2.2. Instrumentation

Tubular regulated furnace (Lindberg 9001) with a 0.25 cm thick cylindrical stainless steel tube (4 cm inner diameter and 74 cm length) was used for carbon activation. A UV-visible spectrophotometer was used to determine the PNP concentrations. The UV-visible spectrophotometer made by Shimadzu UV-160A.

2.3 Preparation and characterization of activated carbon

To prepare activated carbon from crushed cypress fruit different chemical reagents were used. Phosphoric acid solution (450 mL, 50% w/w) was used in chemical activation of cypress fruit. This concentration was reported in literature to give the best surface area for carbon activated by phosphoric acid H_3PO_4 [7]. The solution was added to the crushed cypress fruit sample and the mixture was left for 24 h at room temperature.

After these impregnation procedures, the solution was filtered to separate the residual acid. The crushed cypress fruit then washed several times with deionized water, dried in an oven at 110°C, before carbonization at 700°C with constant rate increase of 10°C /min for 1h under inert atmosphere (N_2 gas 99.9%) at a flow rate of (0.5 L min^{-1}). At the end of activation time the carbonized samples were withdrawn from the furnace and allowed to cool to room temperature under the same nitrogen flow. The sample was repeatedly washed with deionize water and then dried at 110°C for 24 h. Finally the sample was stored in covered beaker inside desiccator for further use.

2.4. Effect of contact time (kinetic study)

The effect of contact time on adsorption was studied. PNP solution (50 mL, 80 mg/L) was added to adsorbent sample (0.1 g) at pH 4. The mixture was shaken at 25°C. Aliquots (0.5 mL each) of the clear solution were withdrawn out at different time intervals until equilibrium was reached after 150 minute.

2.5 Effect of adsorbent dosage

Different amounts (0.02-0.3 g) of prepared activated carbons prepared from cypress fruit (CFAC) were placed into conical flasks, then solutions (50 mL, 80 mg/L PNP) were added to each flask and the pH was 4. The mixtures were then shaken for 150 minute at 25°C.

2.6 Effect of temperature

The effect of temperature on adsorption was studied. PNP solutions (50 mL, 80 mg/L each) were added to adsorbent samples (0.10g) at pH 4. The mixtures were shaken for 150 minute at different temperatures in the range 15-45°C.

2.7 Effect of PNP concentration

In each adsorption experiment, PNP solutions of a given concentration ranging from 0 to 100 mg/L PNP was added to 0.10g adsorbent, with initial pH 4. The flasks were shaken at 25°C for 150 minute to reach equilibrium. The initial and final concentrations of PNP were measured.

The amount of adsorption at equilibrium, q_e (mg/g), was calculated by Equations 1-2:

$$q_e = \frac{(C_o - C_e) V}{W} \quad (1)$$

$$PR (\%) = \frac{C_o - C_e}{C_o} \times 100 \quad (2)$$

Where PR percentage removal (%), C_o and C_e are the initial and equilibrium concentrations of PNP solution (mg/L), respectively, V is the volume of solution (L), and W is the weight of activated carbon used (g).

2.8. Adsorption equilibrium model

Adsorption equilibrium models are a good way to explain the adsorption system. Its provide information about the adsorbate and adsorbent surface properties. There are several equilibrium isotherm models applied including, Freundlich, Langmuir, Temkin...etc. These models developed to fit adsorption system with various conditions [8]. However, the most important and used in the field of adsorption are Freundlich and Langmuir.

2.8.1 Langmuir equation

To determine the maximum PNP adsorption capacity of prepared activated carbon, the experimental adsorption data obtained were fitted to the Langmuir isotherm model. It assumes a mono layer adsorption onto a uniform adsorbent surface with energetically identical sorption sites. Langmuir isotherm equation is given by the following equation (Equation. 3):

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \quad (3)$$

Where q_e is the amount of PNP adsorbed per unit mass of activated carbon (mg/g), q_m is the maximum amount of PNP adsorbed per unit mass of activated carbon (mg/g), C_e is the equilibrium concentration of the PNP (mg/L), and b is the Langmuir constant (L/mg). The Langmuir isotherm model which based on the assumption of a homogeneous adsorbent surface with identical adsorption sites has been successfully used by many researchers to correlate the experimental adsorption data of phenolic compounds on activated carbons [9].

2.8.2 Freundlich equation

It describes equilibrium on heterogeneous surfaces and hence does not assume mono layer capacity [10]. The well-known logarithmic form of the Freundlich isotherm is given by Equation 4.

$$\log q_e = \log KF + \frac{1}{n} \log C_e \quad (4)$$

Where C_e is the equilibrium concentration of the adsorbate (mg/L), q_e is the amount of adsorbate per unit mass of adsorbent (mg/g), KF and n are Freundlich constants. n giving an indication of how favorable the adsorption process is. $K_F ((\text{mg/g}) (\text{L/mg})^{1/n})$ is related with adsorption capacity of the adsorbent. The slope ($1/n$) ranging between 0 and 1 is a measure of surface heterogeneity, becoming more heterogeneous as it's value gets closer to zero [11]. A value for (n) below one indicates a normal Langmuir isotherm, while (n) above one is indicative of efficient adsorption [12].

2.9 Adsorption kinetics

Adsorption kinetics study is important, since they give information about the adsorption system behaves and the rate at which specific constituent is removed using certain adsorbent. In addition, they provide information about whether the adsorption process is chemical or physical and which specifically is the rate limiting step. There is several models describe the adsorption process. In this research we will use adsorption reaction models, which classified into three models.

2.9.1 Pseudo-first order kinetic models

A pseudo-first order kinetic model is considered the earliest models develop to the kinetic process of adsorption. The final integrated form equation of this model as follows (Equation 5):

$$\log(qe - qt) = \log qe - \frac{k_1}{2.303} t \quad (5)$$

Where q_e and q_t (mg/g) are the adsorption capacity at equilibrium and at time t (min), respectively. k_1 (min^{-1}) is the pseudo-first order rate constant [13].

2.9.2 Pseudo-second order kinetic models

This model employed to describe the adsorption of metal ion and polar functional group such as, ketones, aldehydes, dyes, herbicides, and phenolics compounds from aqueous solution assuming that Langmuir equation applies.

The linearized integral form of the model is (Equation 6):

$$\frac{t}{qt} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (6)$$

Where q_e and q_t (mg/g) are the adsorption capacity at equilibrium and at time t (min), respectively, k_2 is the rate constant ($\text{g}/(\text{mg}\cdot\text{min})$) [14].

2.9.3 Second order kinetic models

The typical form of second order rate equation can be applied to describe the adsorption process based on adsorbate uptake rate. The integrated forms of (equation 7) as follow [15].

$$\frac{1}{C_t} = k_2^* t + \frac{1}{C_0} \quad (7)$$

Where C_0 and C_t (mg/l) is the concentration of solute at $t=0$ and at time t (min), respectively, and k_2^* ($\text{L}/(\text{mg}\cdot\text{min})$) is the rate constant.

2.9.4 Intra-particle diffusion model

In a liquid-solid system, the theory proposed by Weber and Morris to link the fractional uptake of solute on particles varies proportionally with $t^{1/2}$. The Weber and Morris equation (Eq.8) is:

$$qt = k_b t^{1/2} + A \quad (8)$$

q_t (mg/g), k_b ($\text{mg}/\text{g}\cdot\text{min}^{1/2}$) is the rate constant of intra-particle diffusion and (A) gives an idea about the thickness of the boundary layer. The value of k_b will be calculated from the slope of plotting $qt \text{ v } t^{1/2}$ [16].

RESULTS AND DISCUSSION

3.1 Characterization of cypress fruit activated carbon

The properties of CFAC prepared were determined and summarized in Table 1. Ash content of CFAC equal 66 % and this value relatively high compared with commercial activated carbons which reach 17.1% [17]. Surface areas were measured using the iodine number method. The results show that the iodine number equal 494.9mg/g., this result resemble the result that reported an iodine number of Haimour and Emeish (2006) that finds the iodine number of activated carbon prepared by chemical activation of date stones using phosphoric acid 495 mg/g [18].

Table 1: Characteristics of CFAC sample.

Characteristic	Activated carbon
Yield of activation (%)	51.8
bulk density (g/ml)	0.472
Electrical conductivity($\mu\text{s}/\text{m}$)	991
surface area (m^2/g)	524.1
ash content (%)	66
Volatile matter (%)	44
moisture content (%)	7.4
pH	4
iodine number (mg/g)	494.9

3.2 Effect of adsorbent dosage

The effect of CFAC dosage on PNP removal was studied, using 0.02 to 0.3g CFAC at an adsorption time of 150 min. to reach equilibrium. The results are plotted as in Fig. 1.

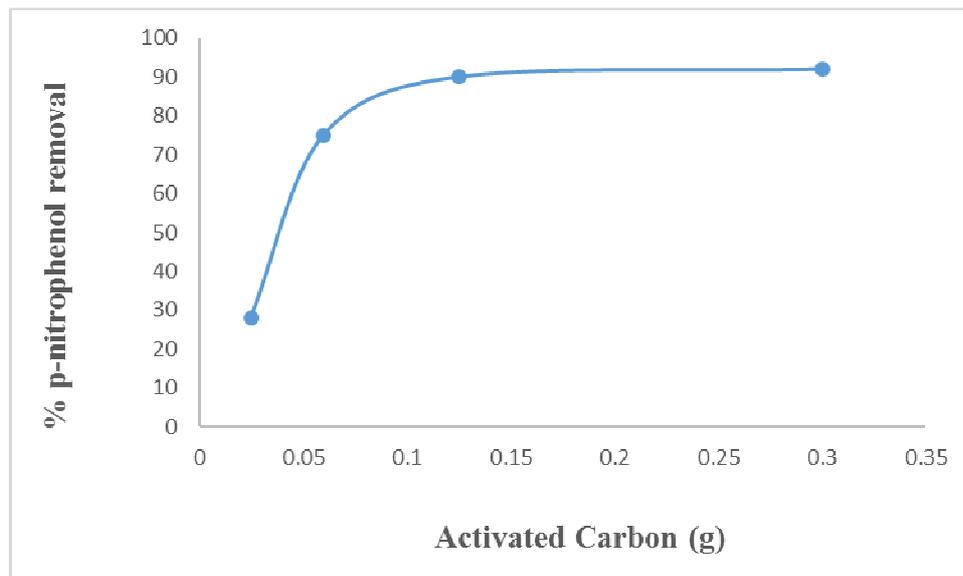


Figure 1: Effect of adsorbent dosage on % of PNP removal by CFAC at (initial conc: 80mg/L, initial pH: 4, temperature: 25 °C and contact time:150 min)

The percent of PNP removal increased by increasing adsorbent dosage. Adsorption increases up to 90.9 % with adsorbent dosage of (0.3 g/50 mL), because increasing adsorbent dosage at fixed PNP concentration provided more available adsorption sites and thus increased the extent of PNP removal.

3.3 Effect of pH

The variation of PNP adsorption onto CFAC was investigated in the pH range 2-12. The effect of CFAC dosage on PNP removal was studied, using 0.1g AC at an adsorption time of 150 min. to reach equilibrium. The results are presented in Fig. 2.

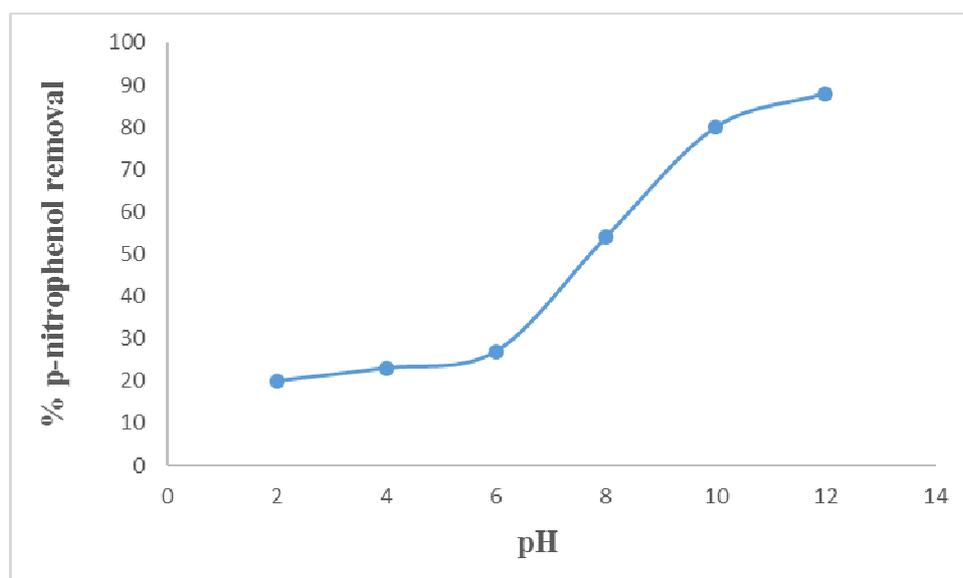
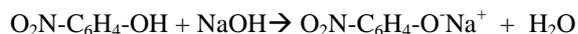


Figure 2: Effect of pH on to the UV-vis absorbance at (initial conc: 80 mg/L, temperature: 25 °C, contact time: 150 min and solid/liquid ratio 0.1 g/50 mL)

Figure 2 shows that the small amount of absorbance increase at pH 2-6. This range of pH which the other factors in this research was studied. When we add sodium hydroxide solution to control pH of PNP solution we note high degree of yellow color this due to high degree of conjugation in their conjugate base will give colored solution, so

absorbance increase strongly. This change of absorbent can due to ionization of PNP by a base to produce sodium p-nitrophenolate which increases both the wave length and the intensities of the absorption bands as shown in the following equation.



3.4 Effect of Temperature

The effect of temperature on PNP adsorption onto CFAC was investigated in the range 15-45 °C. The results are shown in Fig. 3.

Figure 3 revealed that PNP adsorption increased with increase temperature. The highest percentage adsorption performance at 45 °C which reach to 83.58 %.

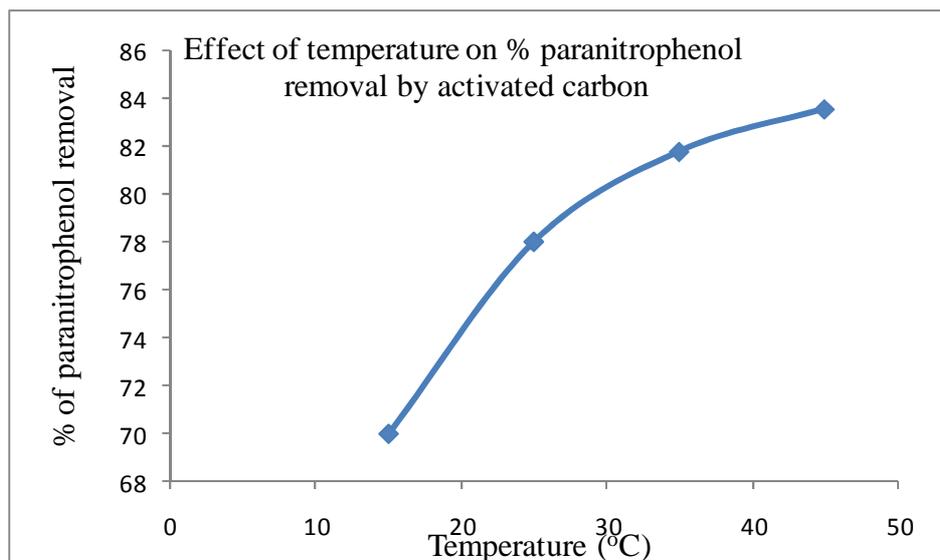


Figure 3: Effect of temperature on % PNP removal by CFAC at (initial conc: 80mg/L, initial pH: 4, contact time: 150 min and solid/liquid ratio 0.10 g/50 mL)

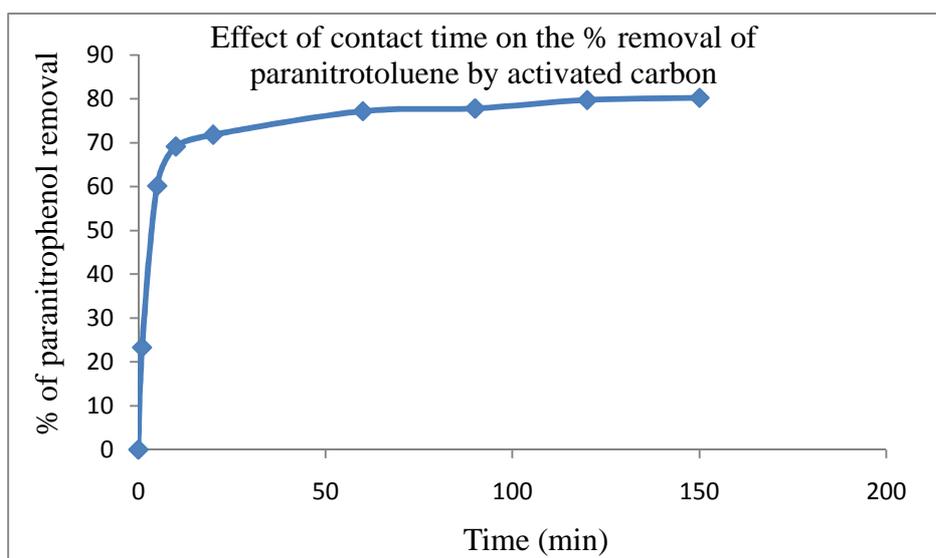


Figure 4: Effect of contact time on the percentage removal of PNP by CFAC at (initial conc: 80 mg/L, initial pH: 4, temperature: 25°C and solid/liquid ratio 0.1 g/50 mL)

3.5 Effect of Contact Time

The effect of contact time on adsorbed amount and removal percentage of PNP is shown in Fig.4. The adsorbed amount of PNP onto CFAC increases with the increase of contact time, as shown in Fig.4 the adsorption reached

equilibrium in about 150 min. for PNP. For PNP adsorption, a rapid increase in adsorbed amount is achieved during the first 10 min. The fast adsorption at the initial stage may be due to the higher driving force making fast transfer of PNP to the surface of CFAC particles and the availability of the uncovered surface area and the remaining active sites on the adsorbent [19-21].

4.7 Adsorption isotherms

In order to design an adsorption system, it is important to study the adsorption isotherms. This is because they give an idea about the adsorption capacity [22]. Adsorption isotherm for PNP onto CFAC at 25°C is given in Figure 5.

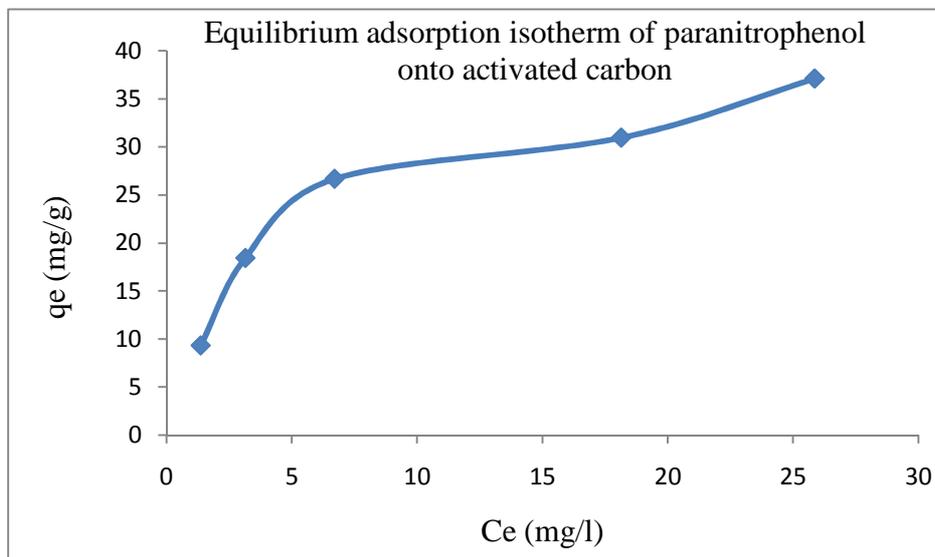


Figure 5: Equilibrium adsorption isotherm of PNP onto CFAC at (temperature: 25 °C, initial pH: 4 and solid/liquid ratio 0.1 g/50 mL)

In this study, Langmuir and Freundlich isotherm models were used to describe the relationship between the amounts of PNP adsorbed and its equilibrium concentration in solution at 25°C. The fitting of adsorption data to Langmuir and Freundlich isotherm equations are shown in Figures 6 -7, respectively.

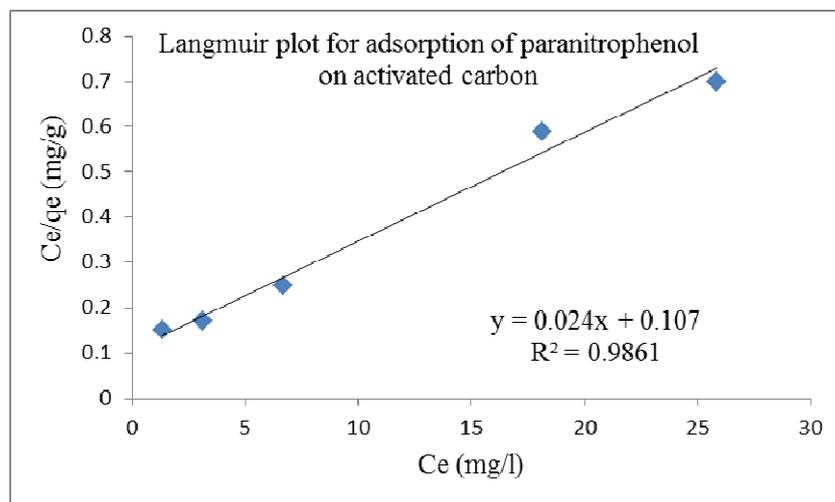


Figure 6: Langmuir plot for PNP adsorption onto CFAC at Temperature; 25 °C, initial pH: 4 and solid/liquid ratio 0.1 g/50 mL)

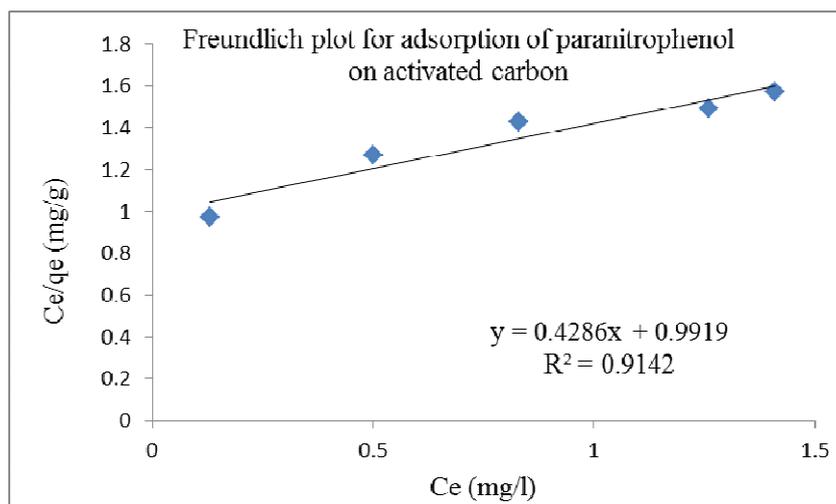


Figure 7: Freundlich plot for PNP adsorption onto CFAC at (temperature: 25 °C, initial pH: 4 and solid/liquid ratio: 0.1 g/50 mL)

It is clear from Table 2 the R^2 values that both Langmuir and Freundlich isotherm describe the experimental data, but Langmuir isotherm model describe data better than Freundlich.

On average, a favorable adsorption tends to have Freundlich constant (n) between 1 and 10. Larger value of n (smaller value of $1/n$) implies stronger interaction between the adsorbent and the adsorbate [23]. It can be seen that (n) value was between 1 and 10 showing favorable adsorption of onto the activated carbon prepared here.

Table 2: Langmuir and Freundlich isotherm model parameters and correlation coefficient of PNP adsorption on to CFAC

Isotherm	Langmuir			Freundlich		
	Parameters			Parameters		
Adsorbate	qm (mg/g)	b (L/mg)	R^2	KF ($(\text{mg/g})(\text{L/mg})^{1/n}$)	n	R^2
PNPh	41.67	0.224	0.9861	9.82	2.33	0.9142

The main properties of Langmuir equation can be expressed in term of separation factor, R_L defined by McKay [24] by equation. 9:

$$R_L = \frac{1}{(1 + bC_0)} \quad (9)$$

Where C_0 is the initial PNP concentration (mg/L) and b Langmuir adsorption constant. The R_L show the adsorption to be un favorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$), irreversible ($R_L = 0$). The value of R_L for CFAC was founded to be 0.053, which show that the adsorption system was favorable.

4.8 Kinetics of PNP Adsorption

In order to investigate the mechanism of PNP adsorption process on CFAC, the pseudo-first-order kinetic model, the pseudo-second-order kinetic model, second order kinetic model and the intra-particle diffusion model were all used to test the experimental data. The results are shown in Figures 8-11.

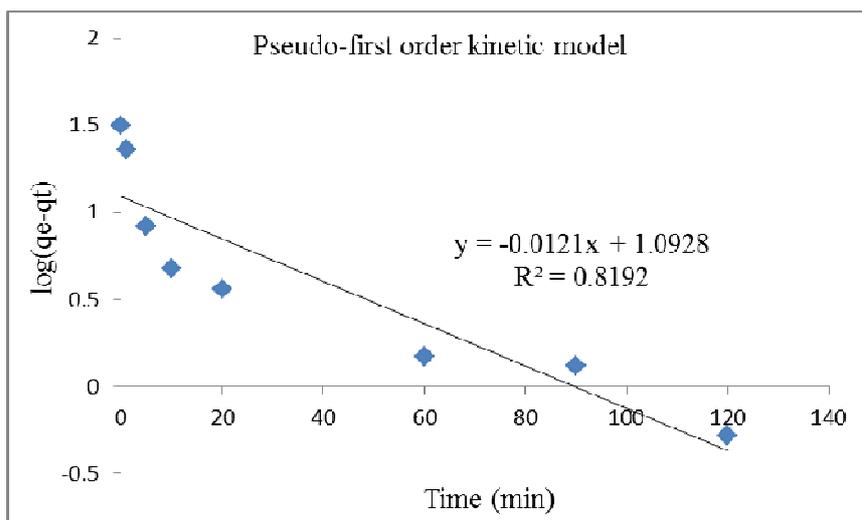


Figure 8: Kinetics of PNP removal according to the pseudo-first-order model by CFAC at (initial conc: 80 mg/L, initial pH: 4, temperature: 25 °C and solid/liquid ratio 0.1 g/50 mL)

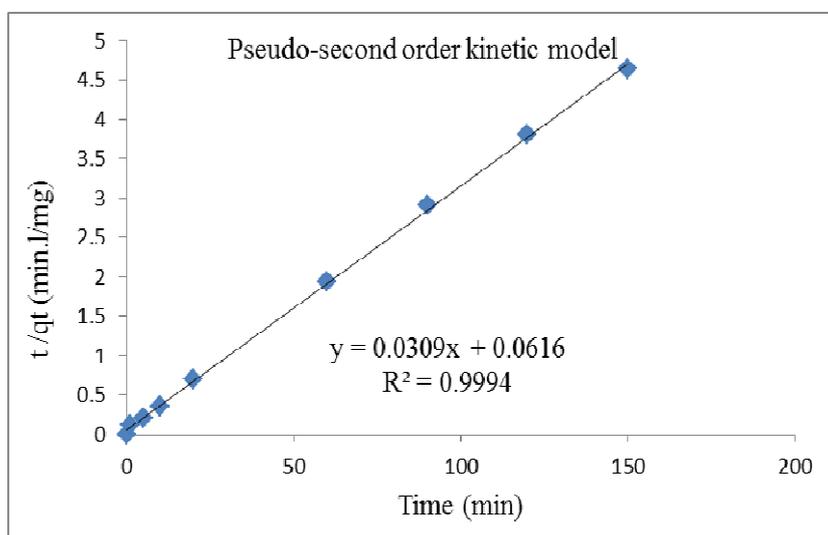


Figure 9: Kinetics of PNP removal according to the pseudo-second-order model by AC at (initial conc: 80 mg/L, initial pH: 4, temperature: 25 °C and solid/liquid ratio 0.1 g/50 mL)

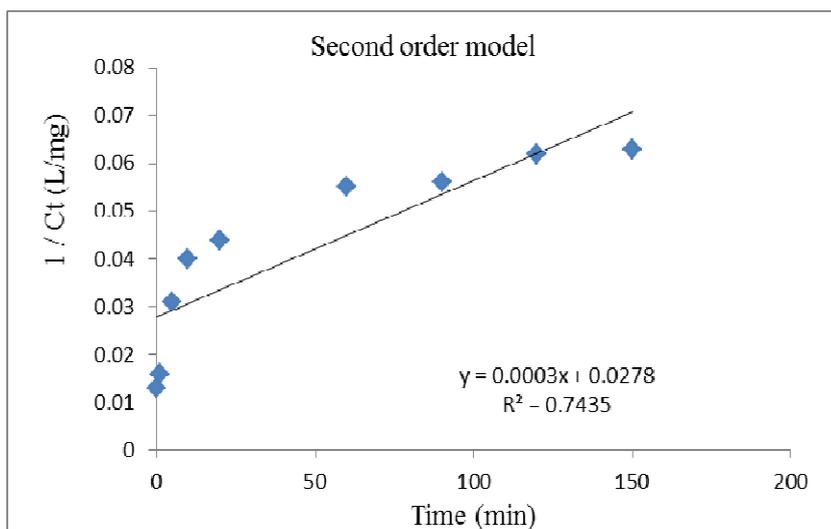


Figure 10: Kinetics of PNP removal according to the second-order model by CFAC at (initial conc: 80 mg/L, initial pH: 4, temperature: 25 °C and solid/liquid ratio 0.1 g/50 mL)

The correlation coefficients and other parameters calculated for the Pseudo-first-order model and pseudo-second-order model are listed in Table 3 and second order model listed in Table 4. From those tables, it is clear that the correlation coefficient values (R^2) of the pseudo-second-order model for adsorbents greater than those obtained for the pseudo-first-order model and second order model. On the other hand, q_e (exp) values for the first-order-rate expression and C_0 for second order model do not agree with the calculated ones obtained from the linear plots. In contrast, q_e (calc) values for the second-order-model are close to q_e (exp) for both adsorbents. Thus, higher correlation coefficients of pseudo-second-order equation and the q_e (calc) values close to the experimental ones indicates that the pseudo-second-order kinetic model might be more suitable to describe the kinetic of adsorption processes of PNP onto adsorbents. This suggests that during the adsorption of PNP, there was chemisorption between the adsorbent surface and adsorbate [24,25].

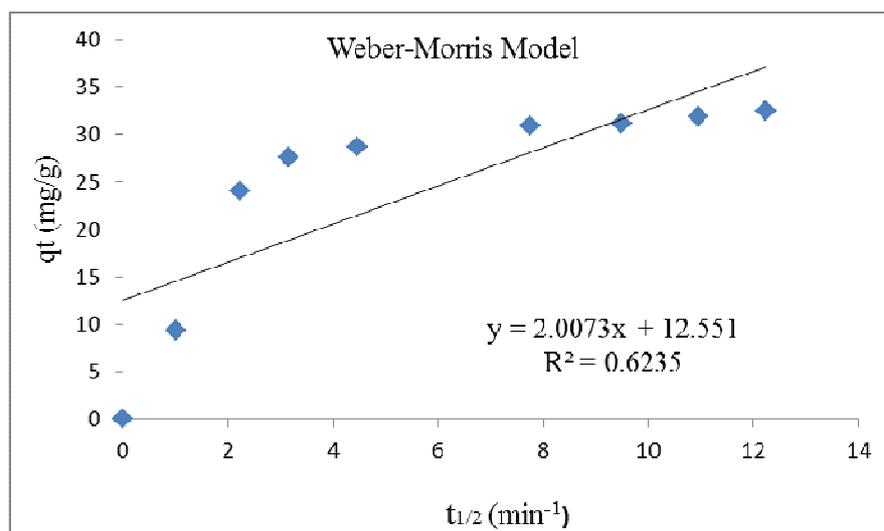


Figure 11: Kinetics of PNP removal according to the intra-particle diffusion model (Weber-Morris model) by CFAC at (initial conc: 80 mg/L, initial pH: 4, temperature: 25 °C and solid/liquid ratio 0.1 g/50 mL)

Table 3: Pseudo-first-order and pseudo-second-order kinetic model parameters for PNP adsorption onto CFAC at 25°C

Adsorbent	q_e (exp) (mg/g)	Pseudo-first-order kinetic model			Pseudo-second-order kinetic model		
		k_1 (min ⁻¹)	q_e (calc) (mg/g)	R_2	k_2 (g/mg min)	q_e (calc) (mg/g)	R_2
CFAC	32.43	0.0279	12.38	0.8192	0.0155	32.36	0.9994

Table 4: Second-order kinetic model parameters for PNP adsorption onto CFAC at 25 °C

Adsorbent	C_0 (exp) (mg/L)	second-order kinetic model		
		K^2 (g/mg.min)	C_0 (calc) (mg/L)	R^2
CFAC	80	0.0003	35.7	0.7435

The Table 5 show that the values of k_2q_e in this work are higher than the value obtained in other work because PNP adsorption more favorable than phenol.

Table 5: Pseudo-second order parameters for phenol and PNP adsorption onto various adsorbents

Activated carbon	Adsorbate	k_2 g/(mg.min)	R_2	k_2q_e (min ⁻¹)	Reference
Pumkin stem	phenol	1.63	0.987	0.0494	(Ektepe, 2011)
Pistachio shell	phenol	0.97	1.000	0.2159	(Tseng <i>et al.</i> , 2010)
CFAC	PNP	0.0155	0.9994	0.502	This work

The values of the intra-particle diffusion constants and the correlation coefficients obtained from the linear plots of qt versus $t^{1/2}$, given in Fig. 11, are summarized in Table 6. Figure 11 implies that the rate of PNP adsorption onto CFAC is limited by mass transfer across the boundary layer.

Table 6: Intra-particle diffusion kinetic model parameters for PNP adsorption onto CFAC 25 °C

Adsorbent	Kb (mg/g min ^{1/2})	R ²	A
CFAC	2.0073	0.6235	12.551

CONCLUSION

1. Activated carbon produced from cypress fruit gives a good percentage which reach up to 51.8% yield.
2. Optimum percent of PNP removal 90.9 % when adsorbent dosage 0.3g at PNP concentration 80mg/L.
3. Percentage removal of PNP increases when the concentration of PNP decreases with maximum percentage removal reaches 93.2% at PNP concentration of 20mg/L and 0.1g CFAC.
4. The results showed that equilibrium time for PNP adsorption is 150min, but most the adsorption attained within the first ten minute.
5. PNP adsorption uptake slightly increases when temperature is increased.
6. The results showed that PNP adsorptions onto CFAC can be described by pseudo-second-order model.
7. Both Frenundlich and Langmuir equilibrium model describe the adsorption of PNP, but Langmuir model describe very well the adsorption.

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