Isotherm and kinetics studies for the adsorption of bisphenol A from aqueous solution by activated carbon of *Musa acuminata*

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Isotherm and kinetics studies for the adsorption of bisphenol A from aqueous solution by activated carbon of *Musa acuminata*

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Abstract. Bisphenol A (BPA) has been widely used in plastic industries. The chemical waste from industry is sometimes dumped into rivers and lakes then these surface waters can become polluted. Therefore, this study aims to investigate the adsorption process of BPA by banana fronds (*Musa acuminata*), an agricultural waste material. Batch experiment was performed by varying different operational parameters including contact time, concentration of BPA, concentration of adsorbents, agitation, and pH. The results confirmed 63.83 % as the highest removal efficiency at 3-hour contact time, 9 mg/L of adsorbent concentration, 40 mg/l of BPA initial concentration, pH of 5, and agitation speed at 150 rpm. In explaining the sorption potential of adsorbents, Langmuir isotherm was better fit with the experimental isotherm data (R^2 = 0.9876) compared to other employed isotherm models. In addition, the pseudo-first-order outperformed (R^2 = 0.8493) in the kinetic behaviors compared to other employed kinetic models. The analysis by the Field Emission Scanning Electron Microscopy (FESEM) exhibited the specific area of the adsorbent identified as homogeneous surface. Fourier Transform Infrared Spectroscopy (FTIR) analysis confirmed the surface functional groups of the adsorbent before and after BPA removal. The present study indicated that the activated carbon is an alternative of low-cost product as an adsorbent in BPA removal.

1. Introduction

Environment pollution is usually an important issue that can uptake of hazardous compounds in food chains, and also damage the living organism. Many improvement has been produced on the latest decade to remove commercial air pollution, major cases of chemical substance launch still happens [1]. Bisphenol A (BPA) is an organic chemical compound which is commonly used in chemical

industries such as polycarbonate plastics (PC), epoxy resins, food packaging, polyester, flame retardants, thermal paper and brake fluid. BPA is a white solid compound that has low vapor pressure, low volatility, and moderate water solubility at room temperature with a mild phenolic odor . It is one of persistent organic pollutants (POPs) in the environmental because of its toxicity, mutagens, and carcinogenic behaviors [2]. The molecular formula of BPA is $C_{15}H_{16}O_2$ and the molecular weight for BPA is 228.29 g/ mol.

The presence of BPA in the production of PC, will provide clear thermoplastic, highly versatile, light weight, durable, high tensile strength, high melting point, high elasticity and shatter resistant that become a choice of plastic industries in generating high quality of plastic productions [3]. Meanwhile, BPA in the production of epoxy resins is commonly used as a coating for consumer and industrial applications [4]. BPA has been classified as a potential endocrine disruptor compounds (EDCs) that can be possible to cause malfunction in normal bodily function of human and animals because once it absorbed into the body, it can cause or mimic normal hormones and body functions [2,5]. Higher exposure of BPA towards human may cause chronic health such as cardiovascular disease, diabetes, infertility, breast cancer, and genital tract abnormalities [6,7]. The major problem of BPA in the environment is its capability to change the characteristic of surface water even at low concentration. In addition, long lasting presence of BPA can affect life chain of aquatic life. It may decrease aquatic population fertility [8]. In addition, it will also give some impact to the farm production if the water supply is polluted by BPA.

Effluents industrial plastics discharged into the surface water are expected to be the primary sources of BPA. Landfill leachate and sewage sludge are also considered as point sources of BPA to the surface water [8]. Exposure in humans to this pollutant is mainly through food, water supply, and drinking water contributes by the bottle itself [9]. In recent, method of treatment such as adsorption, ozonation and advanced oxidation processes has been used to control the BPA pollutant [10]. Most of researcher used adsorption method by activated carbon to minimize the BPA pollutant in surface water since it is the most efficient methods [11]. Adsorption is the process by which molecules with particular characteristics of polarity and size are attracted and held to the adsorbents surface by mass transfer operation. Chemical and physical methods are commonly used for the adsorption process. Chemical process of adsorption is through an ion exchange while physical process is a process of dissolving the contaminants in aqueous solution, adhere and immobilized onto the surface of adsorbents. By comparing with other methods of treatment, adsorption is a method that commonly been used due to require less investment in terms of initial development cost compare to the other methods. Besides that, it does not need precipitation and sedimentation steps that require large area of operation and simple in design. By using this method, it also can reduce the concentration of hydrophobic substances which are difficult to remove by other methods and free from or less generation of toxic substances.

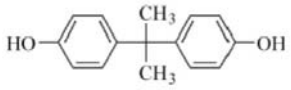
Since commercial activated carbon will contribute cost issues, agricultural waste activated carbon have been used to replace the commercial activated carbon and investigated its feasibility during adsorption processes. Previous researchers have investigated the use of agricultural waste for adsorption process such as coconut husk, oil palm empty fruit bunch, coconut shell, and durian shell [2,12,13]. In this study, banana fronds activated carbon was used as adsorbent to determine the adsorption rate and capacity of BPA. Malaysia has covering about 26,000 ha for banana plantation. The residues of banana plant generate an abundance waste that cause environmental problems. As the problem arise, banana fronds was selected to be an activated carbon agricultural waste since it is discarded all over the world as useless material. Effect of contact time, adsorbents dosage, initial concentration, pH, and agitation, isotherm and kinetic studies were also investigated. FESEM and FTIR characterizations were carried to study chemical and physical characteristics of the adsorbents.

2. Materials and method

2.1. Materials

Banana fronds (*Musa accuminata*) was used as an adsorbent in this experiment. BPA was supplied by Aldrich, USA. Zinc chloride ($ZnCl_2$) used to activate the pore volume of adsorbents was purchased from Qrec™. Table 1 shows the characteristics of BPA. The other chemical reagents such as sodium hydroxide (NaOH), and hydrochloric acid (HCl) that used for pH effect towards BPA were purchased from Qrec™.

Table 1. Characteristics of BPA molecules

Parameter	Properties
Chemical Structure	 BPA
Synonym	2,2 - bis (4 - hydroxyphenyl) propane 4,4' - (1 - methylethylidene) bisphenol) 4,4' - Isopropylidenediphenol
Appearance	White flakes
Physical State	Solid
Solubility	Soluble in acetone, ethanol, ether and benzene. Slightly soluble in carbon tetrachloride.
Dye Content	~50%
Melting Point	150 - 155°C
Boiling Point	220ºC (533 Pa)
Specific gravity	1.195
Vapor pressure	$5.3 \text{-} 10^{-6}$ Pa (25ºC)
Molecular Weight	228.29 $gmol^{-1}$
Molecular Formula	$C_{15}H_{16}O_2$
Maximum wavelength	277nm

2.2. Activated carbon production and batch studies

Banana fronds were collected and washed several time to remove all dirt by using tap water. The banana fronds then were dried in an oven at 110 °C for 24 h to remove moisture. Then 20% $ZnCI_2$ solution was prepared to impregnate the adsorbent prior to the carbonation stage. The adsorbent was carbonized by using a horizontal furnace at 300°C for 1 h.

A stock solution was prepared by dissolving 50mg of BPA granules with 500mL of distilled water in volumetric flask. The solution was heated and stirred by using magnetic bar until it is completely dissolved. The stock solution was performed by the series of 20-100 mg/L of BPA. Batch experiments were carried out by duplicating the sample to perform adsorption tests. 50mL of various concentration of BPA solution were performed in 100mL conical flask. Each flask containing 50 mL series of BPA standard solutions were added with various weight of banana fronds activated carbon within 7 h to study the influence of mass of adsorbents loading onto capacity of BPA removal.

Then, the BPA solutions were shaken by using mechanical shaker at ranging 140-180 rpm to evaluate the effect of agitation towards BPA removal. To ensure equal mixing of solution, the speed of mechanical shaker should be constant throughout the experiment. Each experiment was performed within the contact time ranging from 0-4 h to evaluate the adsorption kinetics. Initial BPA concentration also was prepared variously within 20 - 100 mg/L. the effect of the initial pH on the adsorption process was studied by adjusting the pH range of 2-11. At the predetermine time, the samples were withdrawn from the shaker and filtered by using Advantec™ filter paper. The removal of BPA and the adsorption capacity were calculated by the following equation:

$$\text{BPA removal } (\%) = \frac{C_o - C_e}{C_o} x100 \quad (1)$$

$$\text{Adsorption capacity} = \frac{C_o - C_e}{X} xV \quad (2)$$

where C_o and C_e are representing the initial and equilibrium concentrations (mg/L), V is the volume of solution (L), and X the weight of adsorbent (gm) [14].

The isotherm has been widely used to characterize the adsorption capacity of organic pollutants. In this study, two types of isotherm have been discussed to study the performance of adsorption capacity between BPA molecules and banana fronds activated carbon. Isothermal study on the adsorption had been discuss through Langmuir, and Freundlich models. The Freundlich model is applicable to heterogeneous sorption while the Langmuir equation is design to homogeneous sorption [14].

$$\text{Langmuir equation , } \frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \quad (3)$$

$$\text{Freundlich equation, } Inq_e = InK_F + (\frac{1}{n})InC_e \quad (4)$$

where C_e (mg/L) is the concentration of BPA at equilibrium; q_m (mg/g) is the maximum monomolecular adsorption capacity; q_e is the amount of BPA absorbed at time; K_L denotes the rate constant of Langmuir related to the affinity between adsorbate and adsorbent; K_F and n are the rate constant of Freundlich that indicates the affinity of the adsorbent towards the biomass.

The kinetics study was analyzed by the influence of contact time on BPA removal using the following model:

$$\text{Pseudo-first-order, In } (q_e\text{-}q_t) = \text{In } q_e\text{-}k_1 t \quad (5)$$

$$\text{Pseudo-second-order , } \frac{t}{q_t} = \frac{1}{k_2 q_e^{\,2}} + \frac{t}{q_e} \quad (6)$$

$$\text{Intraparticle diffusion , } qt = k_{diff}\, t^{\frac{1}{2}} + C \quad (7)$$

where *qe* (mg/g) is amount of BPA biosorbed on the adsorbent at equilibrium time; *qt* (mg/g) is amount adsorbed at time, *t* (min); k_1 is the pseudo-first-order biosorption rate constant; k_2 is the pseudo-second-order biosorption rate constant; k_{diff} (mg/g $\text{min}^{1/2}$) denotes intraparticle diffusion rate constant and C is the intercept that indicates the boundary layer thickness.

2.3. Characterization

FESEM analysis was carried out to investigate its surface texture before and after adsorption. FTIR analysis was applied on the raw and banana fronds activated carbon to determine the functional groups that involved in the adsorption process. The porosity of the adsorbents and the chemical reactivity of functional group at the adsorbent surface influence the adsorption capacity of the adsorbent [2,15]. The concentration of the pollutant was measured by using UV-Vis spectrophotometer obtained from Macherey Nagel at wavelength 276 nm.

3. Results and discussion

3.1. Characterization of adsorbents

Analysis on the surface morphology of the banana fronds was performed by FESEM analysis by observing at a magnification of x1000. FESEM images of raw and treated banana frond is shown in Figure 1. There are no visible pore found at surface structure of banana fronds that able to trap BPA molecules. It support the data gained that raw banana fronds were not effective to absorb the BPA by itself. The image shows that the pore and hilly structure were developed at structure of banana fronds after carbonation process. But the structure formed still unable to absorb the BPA due to small pores size and the hilly structures. However, the size of pore and the quantity pore was increased than that of

before soaked with zinc chloride. Zinc chloride play an important role in improving the pore size of banana fronds and encourage the adsorbents to absorb BPA. Based on the data obtained above, the adsorbent were effective to absorb the BPA after soaked with zinc chloride. This result is similar with previous study that the porosity of adsorbent was significantly developed by pyrolysis and physical activation with high temperature [2].

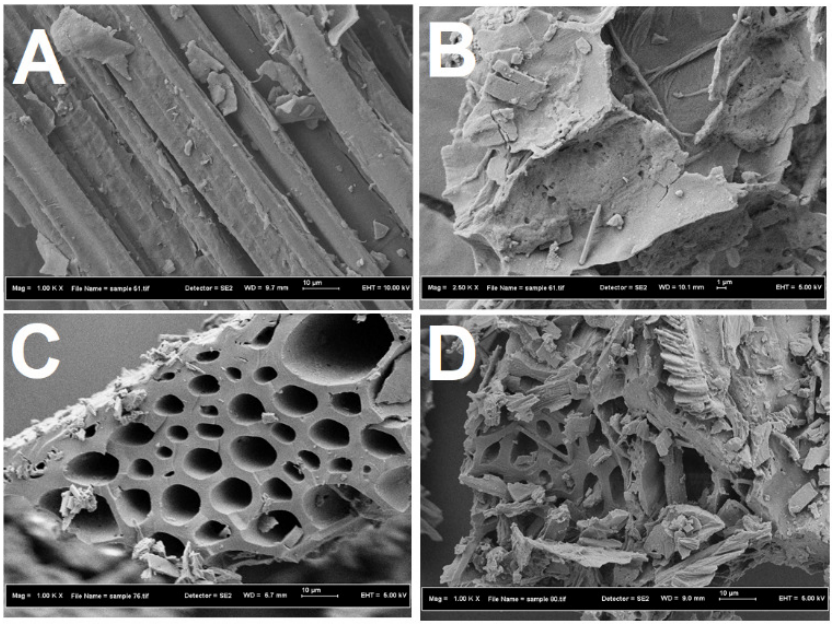


Figure 1. FESEM images of raw banana fronds adsorbent (A), banana fronds activated carbon (B), banana fronds activated carbon soaked with $ZnCI_2$ (C) and banana fronds activated carbon soaked with $ZnCI_2$ after treatment (D)

The results obtained from FTIR spectrum indicated that there are broad and strong bond for raw sample is 3417.91 cm^{-1} was shifted to 3416.07 cm^{-1} after carbon activation and 3431.36 cm^{-1} after carbon activation soaked with $ZnCl_2$. After treatment, the spectrum has changed to 3388.14 cm^{-1} (Figure 2). The bond has assigned to the stretching vibration of the O-H and H bounded with functional groups of alcohols and phenols. The medium absorption peaks for raw samples which is 2923.35 cm^{-1} has been changed to 2929.48 cm^{-1} after carbon activation, 2924.71cm^{-1} after carbon activation soaked with $ZnCI_2$ and getting reduce to 2923.67 cm^{-1} and are assigned as C-H stretch with exist of alkanes functional group (Table 2).

The medium peaks at range 1626.82 cm^{-1} for raw sample was switched to 1623.50 cm^{-1} when the carbon was activated and changed to 1626.40 cm^{-1} after carbon activation soaked with $ZnCI_2$. The frequency of spectrum increased to 1629.75 cm^{-1} due to surge of functional group of 1°amines which is assigned as N-H bend peak. The alkanes functional group was also represented in C-H rock bond at 1382.41 cm^{-1} for raw sample. After carbon activation, the spectrum was shifted to 1386.41 cm^{-1} and 1366.78 cm^{-1} when the activated carbon was soaked with $ZnCI_2$. The absorption peak was reduced to 1364.49 cm^{-1} after treatment process. The peaks at range 1335-1250 cm^{-1} are attributed as C-N stretching vibration of C-N stretch bond which is classified as aromatics amines functional groups.

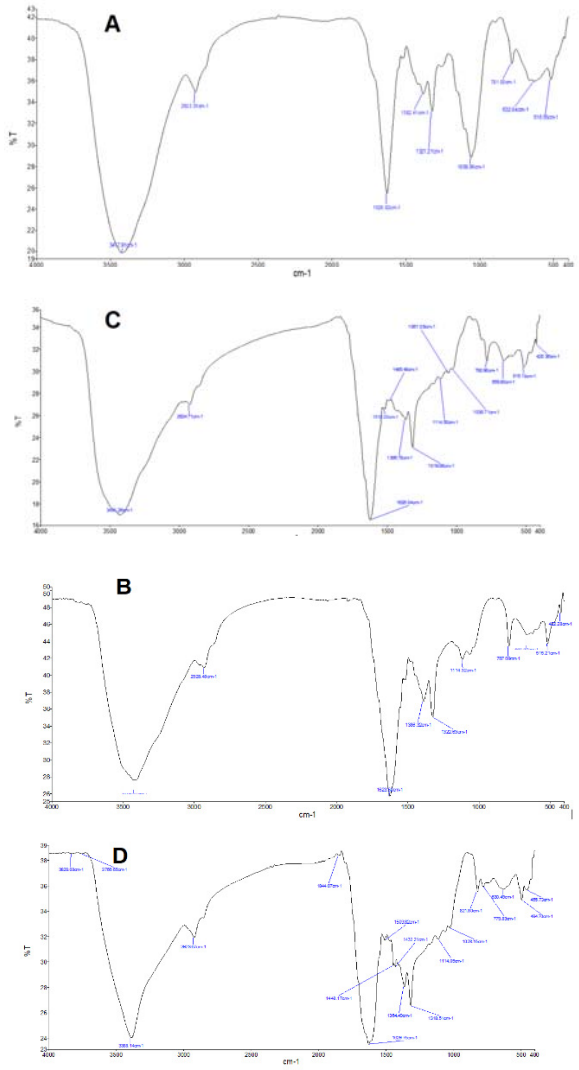


Figure 2. FTIR peaks for raw banana fronds adsorbent (A), banana fronds activated carbon (B), banana fronds activated soaked with $ZnCI_2$ (C), banana fronds activated carbon soaked with $ZnCI_2$ after treatment (D)

Table 2. FTIR spectral characteristics of banana fronds before and after adsorption

Banana fronds frequency (cm-1)				Possible compound
Raw sample	Activated carbon	Soaked with $ZnCl_2$	After treatment	
3417.91	3416.07	3431.26	3388.14	O-H stretch, H bound
2923.35	2928.48	2924.71	2923.67	C-H stretch
1626.82	1623.50	1626.04	1629.75	N-H bend
1382.41	1386.32	1366.78	1364.49	C-H rock
1321.21	1322.63	1319.06	1318.51	C-N stretch
781.00	787.09	780.90	779.83	C-CI stretch
516.55	515.21	515.13	494.70	C-Br stretch

The absorption peaks for raw sample 781.00 cm^{-1} has changed their frequency to 787.09 cm^{-1} when the sample was activated, 780.90 cm^{-1} when the activated carbon was soaked with $ZnCI_2$ and reduce to 779.83 cm^{-1}. The stretching bond was assigned as C-CI and grouping as alkyl halides functional groups. The medium bond located at 516.55 cm^{-1} for raw samples has been shifted to 515.21 cm^{-1} after carbon activation, 515.13 cm^{-1} after carbon activation soaked with $ZnCI_2$ and drop to 494.70 cm^{-1} after treatment. The bonds were assigned as C-Br stretch and the reduction of spectrum frequency due to loss of alkyl halides functional groups. The peak changes of spectrum indicated that there is involvement of functional groups on the surface of adsorbent during sorption process. The potential active sites for absorption of BPA may influenced by the surface functional groups of banana fronds activated carbon that consist of large number of carbonyl groups that represent hydrogen bond and hydroxyl groups that defined as alcohol groups which have high attraction towards pollutants [16].

3.2. Adsorption studies

Contact time is one of the most important fundamental parameter in all transfer phenomena such as adsorption [17]. The effect of contact time onto percentage BPA uptake is shown in Figure 3A. The study was performed with initial concentration of BPA of 9 g/L in order to determine the equilibrium time for BPA uptake. It was observed that, at first stage there was a rapid initial adsorption within 4 h. As a result, the concentration of BPA decreases as time increases but contrary with the percentage removal of BPA increases as time increases until it achieved equilibrium state. The percentage removal and the adsorption capacity were raising parallel as contact time increases. The higher percentage of BPA removal are achieved at 4 hours which is 59.97 % removal with 1.30 mg/g adsorption capacity and it is the optimum contact time for the adsorption of BPA. the adsorbent was saturated and dynamic equilibrium stages since the adsorbents unable to absorb BPA molecules after 4 hours. Previous study revealed that the following stage shows the desorption and adsorption between activated carbon and BPA in a state of equilibrium due to reduction of available external sites. Adsorption equilibrium time is defined as the time needed for BPA concentration to reach a constant value after through adsorption process [18]. At earlier, by mass transfer, the BPA molecules rapidly reach the boundary layer but getting slowly since the available sites increasingly occupied by BPA molecules. As a result, due to formation of repulsive forces between the BPA molecules and the surface layer of banana fronds activated carbon cause difficulty the remaining vacant sites to be occupied in bulk phase [19].

The removal of BPA at different dose of banana fronds is shown in Figure 3B. The maximum removal BPA percentage is found to be 59.97% for the adsorbent dosage of 9 g/L respectively. As the adsorbent was added from 6 g/L to 9 g/L, the percentage removal increased from 46.73% to 59.97%. On the contrary with the adsorption capacity was dropped from 1.525 mg/g to 1.305 mg/g when increased the dosage. The adsorption vacant sites for BPA molecules increase when increase the adsorbent dosages that enhanced the adsorption competition between adsorbent and BPA molecules [18]. Theoretically, if the adsorbent dosage increasing, the removal rate was increased due to growing of active adsorption site and surface area of adsorbents. The study on effects of dosages of adsorbent onto BPA is important to determine the capacity of adsorbent to adsorb the given concentration of sorbate since it is related to the availability of the sorbent vacant site for adsorption [20].

The plot of BPA percentage removal versus time with the influenced of various initial concentration of BPA is shown in Figure 3C. The highest removal was 63.83% performed by initial concentration of 40 mg/L within 3 h. the percentage removal was decreased since the adsorbent became saturated after 3 h. The other initial concentrations of BPA show the optimum results at hour four, which is 59.97%, 61.26%, 59.37% and 55.21% for 20 mg/L, 60 mg/L, 80 mg/L and 100 mg/L. As can be seen from the results obtained, the optimum percentage removals were increased as initial concentration decreased because there is less amount of BPA to be absorbed except for 20 mg/L. Theoretically, adsorption occurs mainly on the surface of the adsorbent at lower concentration and the adsorbent had saturated rapidly. Otherwise, more time is needed at higher concentration for adsorption

to occur since there is competition among BPA molecules to be attract with adsorbents pore since there is lack of available active sites (intraparticle diffusion) [2,20].

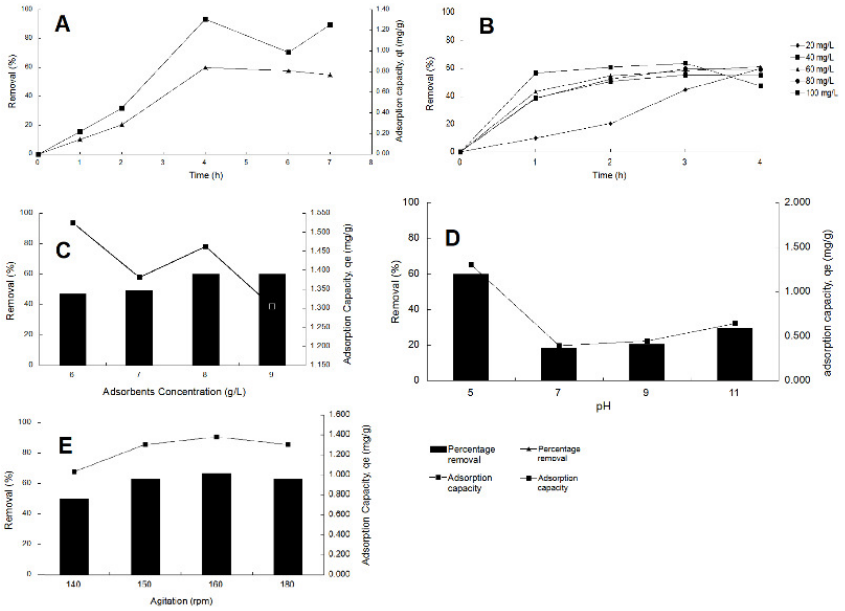


Figure 3. Effect of some parameters on BPA adsorption: contact time (A) dosages of adsorbent (B) initial concentration (C) agitation pH (D).

The effect of agitation on adsorption of BPA with banana fronds activated carbon is shown in Figure 3D. The parameter of 4h contact time, 20 mg/L initial concentration of BPA, and 9 g/L adsorbent concentration were kept constant along the experiments. The graph shows that, the optimum percentage removal of BPA was performed at agitation 160 rpm with 63.37 % removal and 1.379 mg/g adsorption capacity. At agitation 140 rpm, the removal performed was 47.29% with 1.029 mg/g of adsorption capacity, while at agitation 150 rpm and 180 rpm the removal was performed to 59.97% with 1.305 mg/g. This conclude that, at speed 160 rpm, the barrier between the solid-liquid phases was overcome by decrease the boundary layer and increase in the transportation of BPA molecules to the adsorption site [18].

Figure 3E shows the graph obtained from the pH data. As the pH increased, the percentage removal of BPA was decreased. Considering at pH 5 was the optimum removal of BPA which achieved 59.97% and 1.305 mg/g adsorption capacity. Basically, pH affected electrostatic attractions between adsorbent and BPA molecules in aqueous solution. Based on the result obtained, the adsorption between adsorbents and BPA molecules was enhanced at acidic medium. The addition of H^+ ion onto BPA was occurred at acidic medium and caused the BPA become positively charged [21,22]. The reaction was more vigorous with present of acids compare to bases because the negative charge of adsorbent was increases as pH decreases [18]. This reason contributed more electrostatic attraction between the adsorbent and BPA molecules performed of more BPA uptake.

3.3. Adsorption isotherms and kinetics

The isotherm and kinetic were widely used to characterize the adsorption capacity of activated carbon derived from natural or synthetic materials. Since natural materials performance are closely comparable to those of synthetic materials [23], this study focuses on the evaluation of a natural material. In this study, two types of isotherm have been discussed to study the performance of adsorption capacity between BPA molecules and banana fronds activated carbon. The results obtained

that, there were strong adsorption interaction between the BPA molecules and the activated carbon. Isothermal study on the adsorption had been discuss through Langmuir, and Freundlich models. The Freundlich equation is applicable to heterogeneous sorption while the Langmuir equation is design to homogeneous sorption [14].

From the Langmuir isotherm, the sorption potential of adsorbents, q_m value can be predicted by plotting q_e/C_e versus C_e as shown in Table 3. The graph of Freundlich isotherm gives the value of K_F and n through graph plotted of loq q_e versus log c_e. The correlation factor (R^2) from a linear curve of Langmuir, Freundlich and Temkin graph plotted can be determined. In explaining the sorption potential of adsorbents, Langmuir isotherm has proved a better fit since the R^2 obtained is 0.9876 and it assumes that the banana fronds activated carbon have homogeneous surface. Besides that, estimation of monomolecular adsorption capacity, q_m that complete monolayer coverage on the banana fronds activated carbon also can be predicted.

Table 3. Isotherm parameters of BPA adsorption

Adsorption isotherm	Adsorption constant	BPA
Langmuir	q_m (mg/g)	33.44
	K_L (mg/L)	0.2856
	R^2	0.9876
Freundlich	n	0.2193
	K_F(mg/g)	51.48
	R^2	0.7266
Temkin	A (L/g)	6.4641
	B (J mol^{-1})	7.1158
	R^2	0.7036

The value of kinetics rate constant of k and q_e can be determined through adsorption kinetics model which are pseudo-first-order, pseudo-second-order and intraparticle diffusion models. Table 4 shows the value of k and q_e obtained from calculated of the graph plotted from three models of adsorption kinetics. From the results obtained in Table 4, amount of sorbed dyes (q_e) for experiment is almost similar as q_e calculated based on pseudo first order models. Furthermore, the R^2 obtained through pseudo first order graph give value of 0.8493 that prove it is better fit compare to pseudo-second-order and intraparticle diffusion models. It suggested that, the adsorption of BPA through banana fronds activated carbon was followed the pseudo first order.

Table 4. Kinetic parameters of BPA adsorption

Adsorption Kinetics	$q_{e,exp}$	$q_{e,cal}$	k_2	R^2
Pseudo-first-order	1.30	1.6114	0.3907	0.8493
Pseudo-second-order	1.30	4.1712	0.0151	0.2735
Interparticle diffusion	1.30	0.1087	0.5104	0.8354

4. Conclusion

Banana fronds activated carbon has the potential to be a great adsorbent in order to remove BPA. Adsorption on BPA by banana fronds activated carbon was faster at parameters 3 h contact of time, 9 g/L of adsorbent concentration, 40 mg/L of initial concentration, 150 rpm of agitation, and pH 5 by removing above 63.83 % concentration of BPA. Langmuir isotherm model and pseudo-first-order

were the best model compared to others by R^2 equal to 0.9876 and 0.8493, respectively. In general, this study has successfully investigated the ability of banana fronds activated carbon to remove BPA.

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