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Adsorption of bisphenol A (BPA) from aqueous solutions by carbon nanotubes: kinetic and equilibrium studies

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ABSTRACT

Contamination of water resources by bisphenol A (BPA) is considered as a critical environmental problem. In this study, the feasibility of BPA adsorption from aqueous solutions by single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) was investigated in a batch system. In order to define the kinetic and isotherm of the reaction, specific experiments were performed at BPA concentrations of 5–50 and 2–50 mg/L, respectively. The BPA uptake by both adsorbents was found to be rapid; also, uptake reached to equilibrium in 60 min for all of the cases. The kinetic data of both adsorbents were also described by the saturation model ($R^2 > 0.99$) as well as the pseudo-second-order rate equation ($R^2 > 0.99$). Based on the saturation rate model, the maximum reaction rates (k_m) of BPA adsorption by the SWCNTs and MWCNTs were in the ranges of 0.023–0.089 mg/L min and 0.013–0.060 mg/L min, respectively. In the pH range of 3–11, the optimized condition of pH for BPA adsorption by both the adsorbents was 9.0. The isotherm data of the SWCNTs and MWCNTs were found to be in the best fitness with the Freundlich–Langmuir and Langmuir isotherm models ($R^2 > 0.99$), respectively. According to the Langmuir model, the maximum adsorption capacities of BPA by the SWCNTs and MWCNTs were, respectively, 71 and 111 mg/g. Therefore, it can be concluded that the MWCNTs were more efficient adsorbent for BPA than the SWCNTs.

Keywords: Bisphenol A; Carbon nanotubes; Adsorption; Kinetic; Isotherm

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1. Introduction

Endocrine disruption compounds (EDCs) are exogenous substances that disturb the endocrine system by mimicking, blocking or disrupting the function of hormones. Bisphenol A (BPA), also called 4,4′-(propane-2,2-diyl)diphenol, with the chemical formula $(CH_3)_2C(C_6H_4OH)_2$ is a phenolic EDC which can be synthesized by condensation of acetone and phenol.

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In fact, BPA is being used in manufacturing the polycarbonate plastic, epoxy resins, linings inside drinking water pipes, etc. However, BPA can cause cardiovascular disease, type II diabetes, liver damage, pancreatic β-cell function disruption, thyroid hormone disruption, normal cell function disruption, brain damage, sexual abnormalities, and cancer. There are various ways in which this compound could end up in the environment, including the effluent of municipal wastewater treatment plants, migration from BPA-based products, effluent of BPA production facilities, effluent of industrial wastewater treatment plants, landfill leachate, and natural degradation of polycarbonate plastics [1–8].

There are several methods by which BPA can be removed from water and wastewater, including adsorption, advanced oxidation processes, membrane processes, and ozonation. On the other hand, it has been reported that some of these methods produce harmful by-products. Adsorption process is an effective and fast method for removal of organic pollutants; also, it is noteworthy that this method does not result in the production of the harmful by-products [2,9–11].

Carbon nanotubes (CNTs) were discovered by Sumio Iijima in 1991. Because of their exclusive properties, wide ranges of researches have been performed on their usability in various industrial applications. The properties of CNTs are discussed in depth in the following references [12–14]. The CNTs can uptake gases, metal ions, pesticides, drugs, phthalate esters, and phenolic compounds. Previously conducted studies indicated that the CNTs could be more efficient than the activated carbon for the removal of heavy metals, phenolic compounds, and other organic chemicals, due mostly to their larger surface area and shorter equilibrium times [10,15–17]. The studies of Liao [15], Martino [13], Zhang [18], and Shirmardi [19] showed that CNTs effectively remove resorcinol, 4-chloro-2-methylphenoxyacetic acid, aromatic compounds, and Acid Red 18 (Azo-dye).

The present work was aimed to investigate the performance of the CNTs in the adsorption of BPA from aqueous environments. In addition to study the adsorption kinetic and isotherm, the effects of different types of CNTs and pHs on the adsorption process were examined.

2. Materials and methods

2.1. Materials

The BPA (purity 97%) and CNTs (purity 95%) were provided from Merck and Iranian Research Institute of Petroleum Industry, respectively. In this study, two types of CNTs, namely, single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) were applied. Outer diameters of the SWCNTs and MWCNTs were 1–2 and 10–30 nm, respectively; also, their inner diameter were, respectively, 0.8–1.1 and 3.8 nm, and their lengths were 10 mm. The pH of solution, if required, was adjusted by the addition of 0.1–1.0 M HCl and/or NaOH.

2.2. Adsorption experiments

All of the experiments were conducted in a batch system. Also, 50 mL of aqueous solution was added into each vessel. A BPA stock solution with a concentration of 100 mg/L BPA was prepared in order to make different concentrations of BPA. Then, 0.5 g/L of CNTs was added into each vessel. It is noteworthy that each vessel was mechanically shacked at 250 rpm. The CNTs were separated from the solutions by centrifuge (at 5,000 rpm for 15 min) when the desired contact time was met. After that, the solution was passed through membrane filter (0.22 μm). Then, the concentrations of BPA were measured using a UV–vis spectrophotometer (Lambda 25; PerkinElmer Inc.) at the wavelength of 504 nm according to Standard Method [20]. It should be noted that the experiments were conducted at room temperature (20 ± 1℃).

In addition, the kinetic experiments were conducted with the concentrations of 5–50 mg/L BPA and the contact times of 5–120 min. Also, isotherm experiments were done with the concentrations of2–50 mg/L BPA and the contact time of 60 min. The effect of pH on the removal of BPA was studied at 5, 20, and 50 mg/L concentrations of BPA, and 60 min contact time. Eq. (1) shows the BPA adsorption capacity by the CNTs:

$$q_t = \frac{(C_0 - C_t)V}{m} \tag{1}$$

where q_t (mg/g) is the adsorption capacity at any contact time, C_0 (mg/L) is the initial concentration, C_t (mg/L) is the concentration at any contact time, V (L) is the solution volume, and m (g) is the mass of the CNTs [10,21].

2.3. Calculations

2.3.1. Kinetic models

In this study, the kinetic data were analyzed using three models: the pseudo-first-order, pseudo-second-order, and saturation rate equations. Lagergren suggested pseudo-first-order kinetic model (Eq. (2)) which expresses the adsorption rate of dissolved materials from aqueous environments [21,22]:

$$\ln\frac{(q_e - q_t)}{q_e} = -k_1 t \tag{2}$$

where q_e (mg/g) is the equilibrium adsorption capacity, k_1 (1/min) is the pseudo-first-order rate constant, and t (min) is the contact time.

The pseudo-second-order and saturation kinetic models are presented respectively in Eqs. (3) and (4) [21,23]:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \tag{3}$$

$$\frac{1}{t}\ln\frac{C_0}{C_t} = \frac{k_m}{K} + \frac{1}{K}\left(\frac{C_0 - C_t}{t}\right) \tag{4}$$

where k_2 (g/mg min) is pseudo-second-order rate constant, k_m (mg/L min) is the maximum saturation reaction rate, and K (mg/L) is the saturation constant [21,23].

2.3.2. Isotherm models

In this study, the isotherm data were analyzed using four models: the Langmuir, Freundlich, Freundlich–Langmuir, and Temkin isotherm models. The Langmuir isotherm model is based on the assumptions that the adsorption is occurred in a single layer on adsorbent, energy of adsorption is constant, and there is no transmigration of adsorbate in the plane of the adsorbent surface. The Langmuir isotherm model is given by Eq. (5) [24]:

$$q_e = \frac{Q_0 b C_e}{1 + b C_e} \tag{5}$$

where Q_0 (mg/g) is maximum monolayer coverage capacity, b (L/mg) is the Langmuir isotherm constant, and C_e (mg/L) is equilibrium concentration of adsorbate [24].

Another important parameter related to the Langmuir model is a dimensionless separation factor, R_L, which is given by Eq. (6) [24]:

$$R_L = \frac{1}{1 + b C_0} \tag{6}$$

R_L value indicates the adsorption nature which can be unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$), or irreversible ($R_L = 0$) [24].

The isotherm data of heterogeneous adsorption can be interpreted by the empirical Freundlich isotherm model which is presented below as Eq. (7) [24,25]:

$$q_e = K_f C_e^{1/n} \tag{7}$$

where K_f and n are the Freundlich model constants; K_f (mg/g (L/mg)$^{1/n}$) indicates adsorption capacity and n is a function of adsorption intensity [24].

The Freundlich–Langmuir isotherm model was developed to improve the fitness found by the Freundlich or Langmuir model. Eq. (8) presents the Freundlich–Langmuir model [26]:

$$q_e = \frac{b Q_0 C_e^{1/n}}{1 + b C_e^{1/n}} \tag{8}$$

where Q_0, b, and n are the Freundlich–Langmuir model parameters [26].

The Temkin isotherm model considers linear reduction of adsorption heat of all the molecules in a layer with coverage. By using Temkin model (Eq. (9)), determining the heat of adsorption is possible [24]:

$$q_e = \frac{RT}{b_T}\ln(A_T C_e) \tag{9}$$

where R (8.314 J/mol K) is the universal gas constant, T (K) is the absolute temperature, b_T (J/mol) is the Temkin constant related to heat of sorption, and A_T (L/g) is the Temkin isotherm equilibrium binding constant [24].

3. Results and discussion

3.1. Adsorption kinetic

There are several adsorption kinetic models upon which adsorption mechanisms can be modeled [18]. In this study, the pseudo-first-order, pseudo-second-order, and saturation kinetic models were applied. In all of the examined initial concentrations of BPA, the BPA uptake by the CNTs reached to equilibrium at contact time of 60 min. As it is shown in Figs. 1 and 2 and Table 1, the kinetic data of BPA adsorption by both the CNTs were found to be in the best fitness with the saturation model. The data also showed good correlation with the pseudo-second-order rate equation ($R^2 > 0.99$). According to the saturation rate model (Table 1), the maximum reaction rates (k_m) of BPA adsorption by the SWCNTs and MWCNTs were determined to be in the ranges of 0.023–0.089 mg/L min and 0.013–0.060 mg/L min, respectively.

Liu et al. [2] indicated that the kinetic of the BPA adsorption by activated carbon is well fitted with the pseudo-second-order kinetic model. Yoon et al. [21] observed that the kinetic of the BPA adsorption by mesoporous carbon CMK-3 obeys the pseudo-second-order kinetic model. The pseudo-second-order model also describes the kinetic of aromatic compounds removal by CNTs and activated carbons [18]. Guo et al. [27] reported

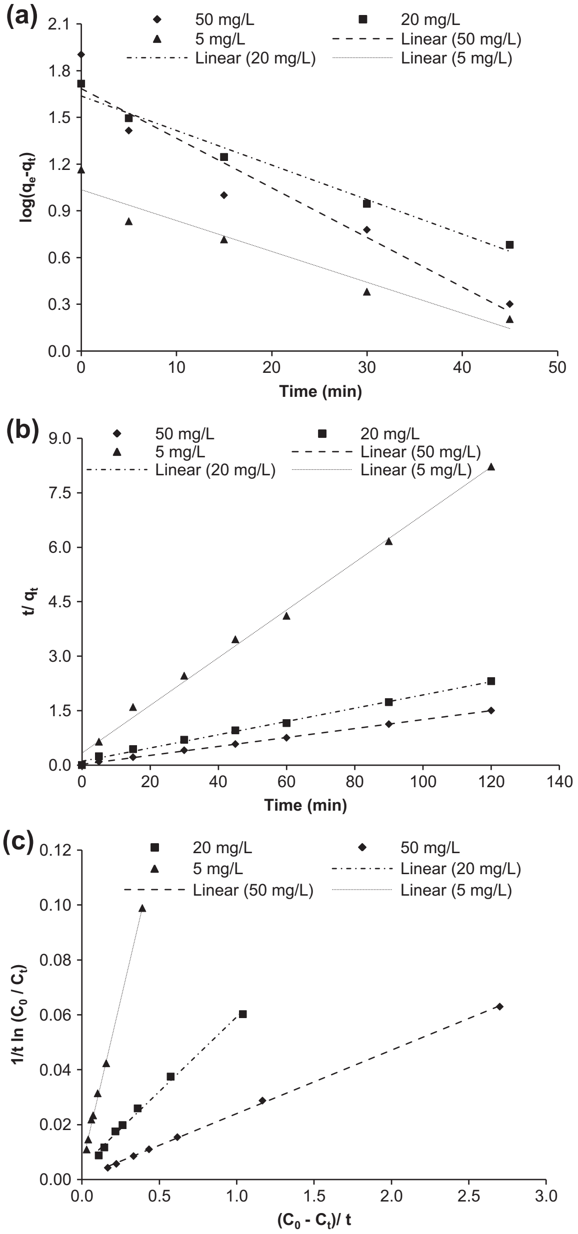

Fig. 1. Kinetic of BPA adsorption onto the SWCNTs: (a) pseudo-first-order, (b) pseudo-second-order, and (c) saturation kinetic models (SWCNTs dosage = 0.5 g/L).

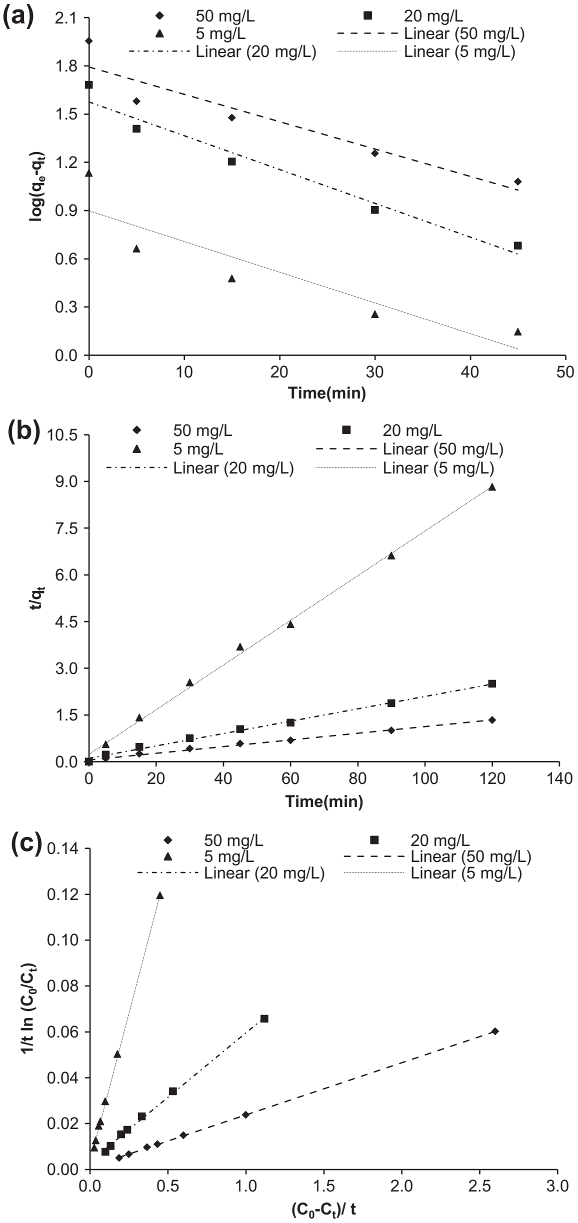

Fig. 2. Kinetic of BPA adsorption onto the MWCNTs: (a) pseudo-first-order, (b) pseudo-second-order, and (c) saturation kinetic models (MWCNTs dosage = 0.5 g/L).

that the kinetic of BPA adsorption using novel molecularly imprinted polymers based on kaolinite/Fe_3O_4 composites obeys the pseudo-second-order model. Pan et al. showed that the kinetic of BPA adsorption by chitosan/γ-Fe_2O_3/fly-ash-cenospheres composites is consistent with the pseudo-second-order rate equation [28]. Considering above-mentioned studies, the pseudo-second-order model was mostly found to be well fitted to describe kinetic of BPA adsorption from aqueous solutions. The result of the present study regarding the kinetic of BPA adsorption is in line with previously conducted researches as well.

3.2. Adsorption isotherm

Isotherm data (statement of q_e as a function of C_e) are basic requirements to design adsorption systems at full scale. In order to make the isotherm data applicable for design purposes, developing an equation to accurately predict the results is necessary [24,25].

Table 1
Kinetic parameters of pseudo-first-order, pseudo-second-order, and saturation rate equations for BPA adsorption by the CNTs

		Saturation model			Pseudo-second-order model			Pseudo-first-order model		
Type of adsorbent	C_0 (mg/L)	K	k_m	R^2	$q_{e,\ cal}$ (mg/g)	k_2	R^2	$q_{e,\ cal}$ (mg/g)	k_1	R^2
SWCNTs	50	43.2	0.037	0.999	83.33	0.007	0.999	5.20	0.029	0.928
	20	18.4	0.089	0.993	55.55	0.003	0.995	5.38	0.026	0.98
	5	4.2	0.023	0.998	15.38	0.012	0.995	2.75	0.017	0.938
MWCNTs	50	43.9	0.045	0.999	100.03	0.002	0.994	6.05	0.017	0.888
	20	17.8	0.060	0.998	52.63	0.003	0.995	4.83	0.021	0.964
	5	3.8	0.013	0.999	14.08	0.022	0.997	2.45	0.019	0.819

Figs. 3 and 4 illustrates the isotherm analysis of BPA adsorption using the Langmuir, Freundlich, Freundlich–Langmuir, and Temkin isotherm models. The isotherm parameters of these models for BPA adsorption onto the CNTs are listed in Table 2. As indicated in Figs. 3 and 4 and Table 2, the isotherm data of BPA adsorption by the SWCNTs and MWCNTs were found to be in the best fitness with the Freundlich–Langmuir and Langmuir models, respectively ($R^2 > 0.99$). The data also showed relatively acceptable correlation with the other isotherm models ($R^2 > 0.95$). Among the isotherm parameters, the maximum uptake capacity parameter (Q_0) of the Langmuir model is considered to be more important; that is why the parameter is often used as a measure to compare different adsorbents for the same adsorbate. According to the Langmuir equation, the parameter Q_0 of BPA adsorption by the SWCNTs and MWCNTs were obtained to be 71.4 and 111.1 mg/g, respectively. This result indicates that the MWCNTs

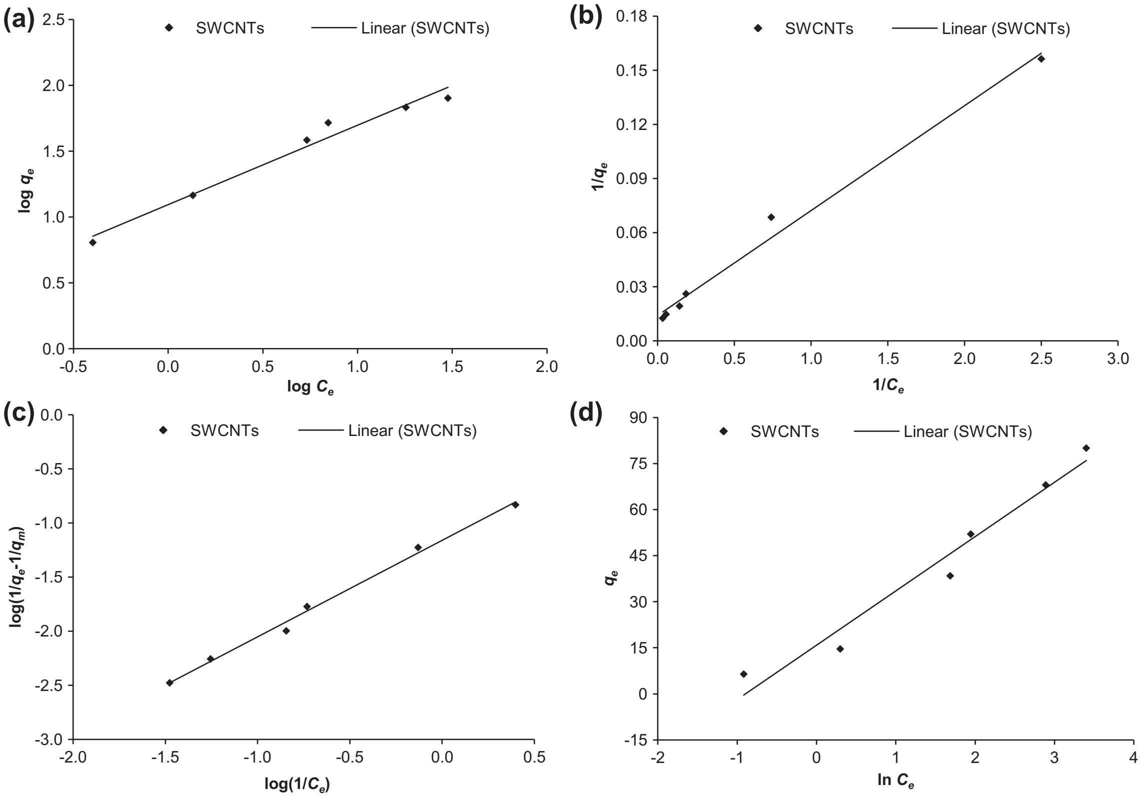

Fig. 3. Isotherms of BPA adsorption by the SWCNTs: (a) Freundlich, (b) Langmuir, (c) Freundlich–Langmuir, and (d) Temkin isotherm models ($t = 60$ min, BPA concentration range = 2–50 mg/L, SWCNTs dosage = 0.5 g/L).

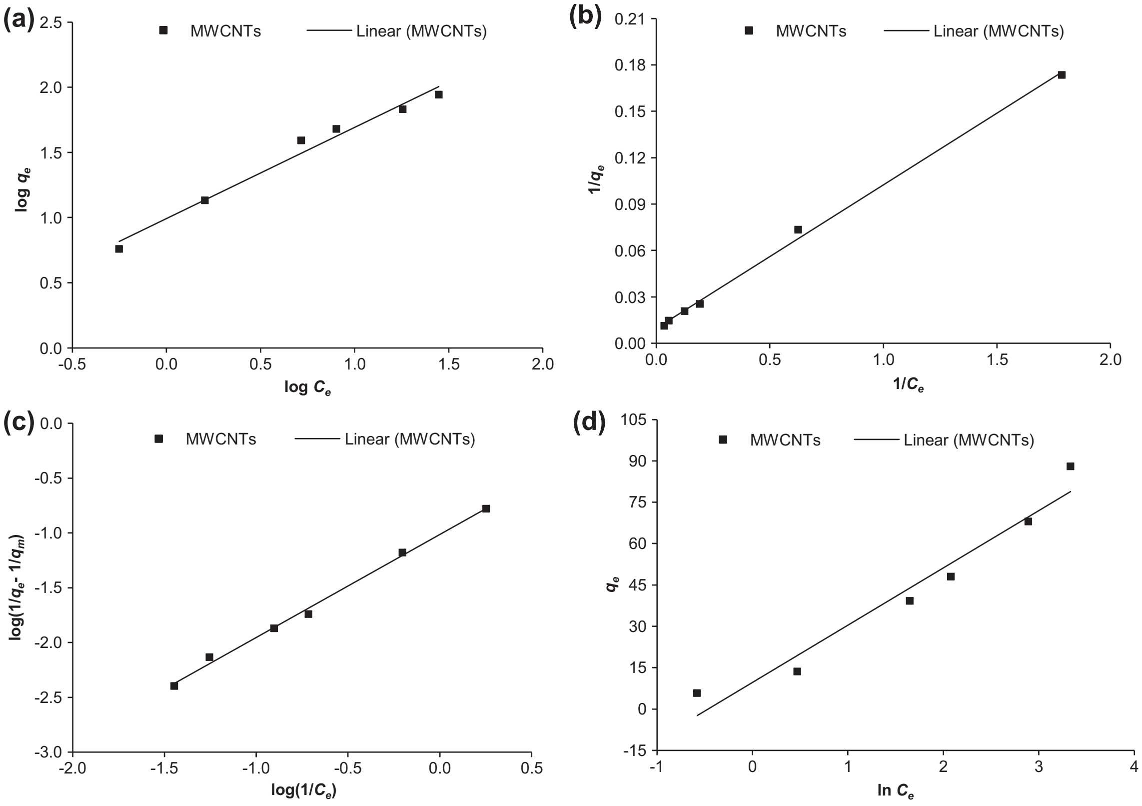

Fig. 4. Isotherms of BPA adsorption by the MWCNTs: (a) Freundlich, (b) Langmuir, (c) Freundlich–Langmuir, and (d) Temkin isotherm models ($t = 60$ min, BPA concentration range = 2–50 mg/L, MWCNTs dosage = 0.5 g/L).

Table 2
Isotherm parameters for BPA adsorption on the CNTs

Isotherm models	Parameters	SWCNTs	MWCNTs
Freundlich	n	1.66	1.43
	K_f	12.42	9.82
	R^2	0.973	0.979
Langmuir	Q_0	71.43	111.11
	b	0.241	0.097
	R^2	0.989	0.997
Freundlich–Langmuir	Q_0	109.04	136.02
	b	0.132	0.075
	n	1.12	1.07
	R^2	0.993	0.995
Temkin	A_T	2.44	1.59
	b_T	140.13	119.34
	R^2	0.961	0.953

were better adsorbent for the BPA than the SWCNTs. Although comparing the results of the previously conducted studies is difficult, due mostly to the difference in their applied experimental conditions, the Q_0 of the CNTs (especially the MWCNTs) for the BPA adsorption exceed from the other adsorbents obtained in the previous studies. Guo et al. [27] reported that the isotherm data of BPA adsorption by polymers based on kaolinite/Fe_3O_4 composites obey the Langmuir model; the parameter Q_0 at 298, 308, and 318 K for magnetic non-imprinted polymers were 112.4, 79.4, and 68.0 mg/g and for magnetic molecularly imprinted polymers were 142.9, 123.5, and 113.6 mg/g, respectively. Pan et al. [28] indicated that the equilibrium data of BPA adsorption on chitosan/fly-ash-cenospheres/γ-Fe_2O_3 (CTS/γ-Fe_2O_3/FACs) are consistent with the Langmuir isotherm model; also, the Q_0 at 298, 308, and 318 K were determined to be 78.0, 90.1, and 103.1 mg/g, respectively. In a study conducted by Cao et al. [29], the parameter Q_0 of the BPA adsorption by polyethersulfone–organophilic montmorillonite hybrid particles was obtained to be in a range of 20.7–32.1 mg/g. Kim et al. [9] observed that the Q_0 of BPA adsorption by phenyl-functionalized mesoporous silica and powdered activated carbon were 351 and 337 mg/g, respectively.

As shown in Table 2, the parameter n of the Freundlich model for the SWCNTs and MWCNTs were 1.66 and 1.43, respectively. It should be mentioned that the values that are more than 1 means the BPA uptake was favorable for both CNTs; also, it shows that the BPA adsorption by the SWCNTs was more intense than the MWCNTs. The parameter b_T for the SWCNTs and MWCNTs were 140.13 and 119.34, respectively. The positive values show that the uptake reaction by both the CNTs was exothermic. The variation of separation factor (R_L) as a function of initial BPA concentration is presented in Fig. 5. As shown in the figure, the R_L values were in range of 0–1 for both the CNTs, indicating that the sorption of BPA on the CNTs was favorable. Also, by increasing C_0, the R_L value diminished to zero that showed sorption of BPA on CNTs is less favorable at high initial BPA concentration.

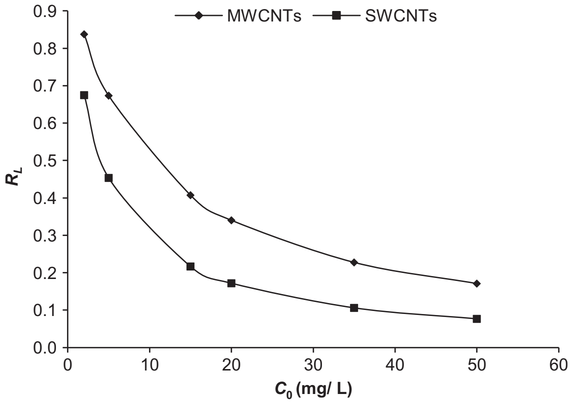

Fig. 5. Variation of separation factor (R_L) as a function of initial BPA concentration.

3.3. Effect of pH

The pH of solution is another important parameter affecting the adsorption process. Fig. 6 shows the effect of pH on equilibrium adsorption capacities of BPA by the CNTs. As shown in the figure, the adsorption capacity of BPA by both the CNTs increased from pH 3 to 9 and then decreased from pH 9 to 11. In fact, it can be said that the optimum condition of pH for BPA adsorption by the CNTs was 9.0. Similar results were observed by Sui et al. [21], wherein the BPA adsorption capacity by mesoporous carbon was approximately constant at pH range of 3–9, but it decreases from pH 9 to 13. Liu et al. [2] indicated that the amount of BPA adsorbed onto activated carbons at pH range of 5.0–9.0 changed slightly; also the lowest adsorption capacity was occurred at pH 11.0. When pH of solution is acidic, the adsorbate and surface of adsorbent become positively charged and the adsorption capacity remains fairly constant. Similarly, in the alkaline pH of solution, especially when the pH of solution is higher than the pKa value of adsorbent (pKa value of BPA is about 9.6), the charge of the adsorbate and surface of adsorbent become negative and the adsorption capacity decreases [10,17].

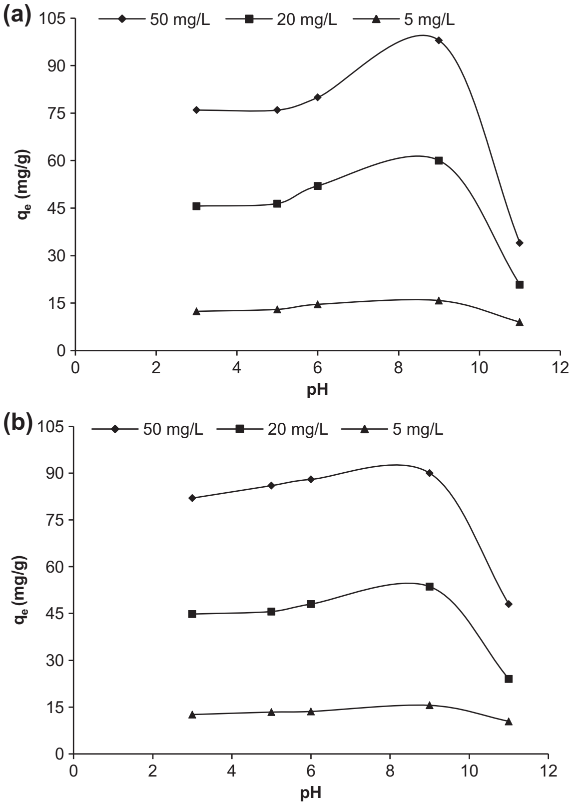

Fig. 6. Effect of pH on BPA adsorption by CNTs: (a) SWCNTs and (b) MWCNTs ($t = 60$ min, CNTs dosage = 0.5 g/L).

4. Conclusions

In this study, the ability of CNTs in the removal of BPA from aqueous environments was evaluated. The results showed that the kinetic data of the SWCNTs and MWCNTs obey the saturation model as well as the pseudo-second-order kinetic model. Isotherm data of the BPA uptake on SWCNTs and MWCNTs were described by the Freundlich–Langmuir and Langmuir isotherm models, respectively. Because the Langmuir parameter Q_0 (maximum adsorption capacity) of the MWCNTs was more than the Q_0 of the SWCNTs, it was concluded that the MWCNTs are better adsorbent for BPA than the SWCNTs. Moreover, the highest adsorption capacity occurred at pH 9 for both the CNTs.

List of symbols

A_T — Temkin isotherm equilibrium binding constant (L/g)
b — Langmuir isotherm constant (L/mg)
b_T — Temkin constant related to heat of sorption (J/mol)
C_0 — initial concentration (mg/L)
C_e — equilibrium concentration (mg/L)
K — saturation constant (mg/L)
k_0 — maximum saturation reaction rate (mg/L min)
k_1 — pseudo-first-order rate constant (1/min)
K_f — Freundlich isotherm constant (mg/g $(\text{L/mg})^{1/n}$)
k_2 — pseudo-second-order rate constant (g/mg min)
m — mass of the CNTs (g)
n — Freundlich model constant
Q_0 — maximum monolayer coverage capacity (mg/g)
q_e — equilibrium adsorption capacity (mg/g)
q_m — Freundlich–Langmuir constant
q_t — adsorption capacity at any contact time (mg/g)
R — universal gas constant (8.314 J/mol K)
R_L — dimensionless separation factor
T — temperature (K)
V — solution volume (L)

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