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Separation of bisphenol A and phenol from water by polymer adsorbents: Equilibrium and kinetics studies

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ABSTRACT

The adsorption behaviours of some polymer adsorbents (Dowex Optipore L493, Diaion SP825 and Diaion SP 207) were evaluated for the removal of bisphenol A (BPA) and phenol from aqueous solutions. Dowex L493 exhibited higher adsorption affinity to BPA showing an adsorption capacity of 1.35 mmol/g, while having less adsorption affinity to phenol with a capacity of 0.84 mmol/g. The adsorption equilibrium data for BPA and phenol removal tested by Dowex Optipore L493, Diaion SP825, and Diaion SP207 were mainly fitted well to Freundlich isotherm model. According to the kinetic test results, both removal rates of BPA and phenol were obtained faster for Dowex L493 than Diaion SP825 and Diaion SP207. Results of kinetic study indicated that the adsorption kinetics were explained better by the pseudo-second-order kinetic model for BPA and phenol removal using all type adsorbents. Rate controlling step has been accepted as particle diffusion for almost all adsorbents. The capability of Dowex Optipore L493 to remove both BPA and phenol from aqueous solutions was found to be the highest among the adsorbents tested.

1. Introduction

Recently, endocrine disrupting compounds (EDCs) detected in drinking water has emerged as a serious problem. EDCs are released into the environment by humans, animals, and industry, mainly through wastewater treatment plants. Particularly, the growing population and development in the industry have resulted in increased production and discharge of these compounds into the environment. EDCs are organic chemicals with one or more phenolic group(s), including natural estrogens such as estrone, 17β-strodial, and estriol, artificially synthesized estrogens such as ethynylestrodial as well as other industrial compounds such as bisphenol A (BPA), nonylphenol, and a number of other phenolic compounds. Due to their structures and physicochemical properties, these molecules can disrupt endocrine systems and contribute adverse effects on aquatic life and human health [1–4]. They can block or mimic the activity of natural hormones, thus interfering with the reproductive systems of wildlife and humans, reduces fertility and increase the possibility of breast, ovarian, and testicular cancers [1,5,6].

BPA, one of the phenolic EDCs, is widely used as a monomer for the manufacture of polycarbonates and epoxy resin in the plastic industry. Due to the frequent use of polycarbonate plastics, epoxy resins and polyvinylchloride in industry and households, BPA has been observed in rivers, seas and soils [3,4,7]. As expected, BPA enters water during manufacturing process, leaches from plastic products and is detected in different water sources [8,9]. The primary sources of BPA released to environmental water are expected to be the discharge of municipal effluent and industrial wastewater [6]. Similarly, phenol and its derivatives are organic compounds those widely used by many industries such as pharmaceuticals, pesticides, paint, plastics, leather, steel mills, coke oven plants, petroleum and petrochemical, nylon, and polycarbonates [10–12]. Phenolic compounds are very harmful to human being and animals even at low concentrations, and is considered one of the major pollutants in wastewater. The permissible concentration of phenolic contents in potable waters is 1 $\mu g\,L^{-1}$ according to the recommendation of World Health Organization (WHO) [12,13]. Because of their ubiquitous nature in the environment and its negative effects to human health and organisms, it is important to reduce the potential risks regarding adverse health effects [6,8,9,11,14].

Accordingly, there is a significant need for developing efficient, simple, and cost-effective technologies for the rapid removal of EDCs from water and wastewaters. However, BPA and phenol cannot be completely removed by conventional treatments such as activated sludge processes [12,15,16], clarification, disinfection, and sand filtration[9] to meet the quality criteria requested by the legislation and an additional treatment stage must be imple-

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mented to provide the final polishing of effluents prior to discharge. Some techniques, including adsorption, membrane filtration, electrochemical technique, and advanced oxidation processes have been proposed in the literature for removal of these compounds [3,4,8,12,13]. Among them, adsorption has been found to be superior in terms of cost, ease of design and operation, and the lack of the possibility of producing secondary harmful substances. However, the use of an efficient adsorbent is crucial in providing the required treatment efficiency for wastewater [2,4,7]. Activated carbon is widely used adsorbent for removal of organic pollutants from wastewater. The porous nature of this adsorbent material and its high internal surface area are among the required properties for efficient adsorption. However, due to its relatively high cost, difficulty in regeneration and restricted applications, there has been an increasing attempt for efficient, easier for regeneration and dispose, and cheaper adsorbents to remove a variety of toxic organic compounds [7,8,12,14].

In recent years, there has been an increasing interest in utilizing low cost and efficient adsorbents such as carbon nanotubes, silica, montmorillonite, organophilic clays, surfactant-modified zeolites, hypercrosslinked polymeric adsorbents and others to adsorb organic molecules [10–12,14,15]. Among them, synthetic polymeric adsorbents have been considered as efficient adsorbents in wastewater treatment due to its good mechanical strength, high surface area, feasible regeneration for repeated use, as well as the potential recovery of some valuable organic compounds from wastewater. Also, their basic physicochemical properties can be adjusted by varying the polymerization conditions so that, by introducing some special functional groups into the chemical structure of the polymer it is possible to improve its adsorption capacity towards specific organic compound present in the wastewater [4,12,13]. Several studies have reported the adsorption efficiency of BPA onto different kinds of adsorbents such as organoclays synthesized from montmorillonite [3], organic–inorganic hybrid mesoporous material [7], carbon nanotubes/Fe_3O_4 nanocomposites [15], polymer grafted on porous polyethylene vinyl acetate disk [17], acetylaniline modified hyper-crosslinked polymeric adsorbent [4], carbon nanomaterials [1], fibric peat [8], hypercrosslinked polymeric adsorbents MN-200, NDA-150, MN-150, MN-100 [18], mesoporous carbon CMK-3 [9], polyethersulfone–organophilic montmorillonite hybrid particles [19], water-soluble and water-insoluble b-cyclodextrin polymers [20], surfactant-modified zeolite [2] and the adsorbents tested for phenol removal such as N-butylimidazolium functionalized strongly basic anion exchange resin [13], N-methylacetamide-modified hypercrosslinked resin [12], organomontmorillonite [21], unmodified carbon nanotubes [22], organobentonite [23], carbon nanospheres [10], surface modified fly ash based zeolite [14], and poly (styrene-co-divinylbenzene) functionalized materials [24].

The objectives of the present investigation are (1) to study the potential performance of some polymeric adsorbents (Dowex Optipore L493, Diaion SP825, and Diaion SP207) for the uptake of BPA and phenol from aqueous solutions; (2) application of Langmuir and Freundlich isotherm models to get an idea about the adsorption mechanism, and (3) to adapt various kinetic models to analyze the kinetic data.

2. Experimental

2.1. Materials

The polymer adsorbents Diaion SP825 and Diaion SP207 were kindly sent by Mitsubishi Co. (Japan) and Dowex Optipore L493 was provided by Dow Chem. (USA). The properties of the polymer adsorbents are given in Table 1.

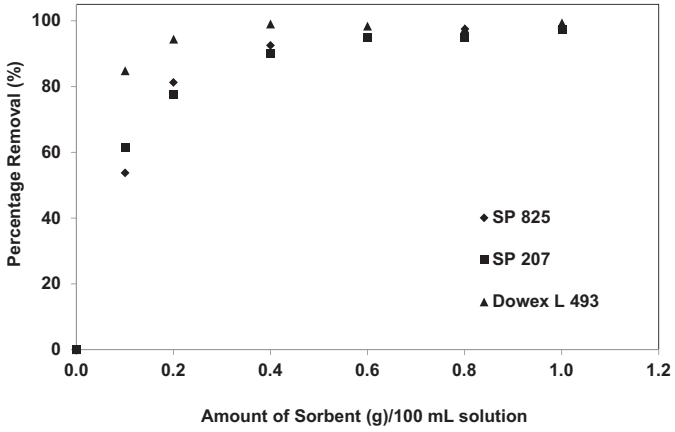

Fig. 1. Determination of optimum adsorbent amount for BPA removal from aqueous solution.

2.2. Determination of optimum adsorbent concentration for BPA and phenol removal

A 100 mL of 0.4 mM of BPA and 0.4 mM of phenol solutions were separately spiked on the polymer adsorbents Dowex Optipore L493, Diaion SP825, and Diaion SP207 at a particle size range of 0.50-0.71 mm using different adsorbent amounts (0.1-1.3 g) with continuous shaking in a shaker at 25 °C for 24 h.

2.3. Kinetic tests

Batch kinetic tests were performed using 750 mL of 0.4 mM BPA and 0.4 mM phenol solutions by spiking with 7.5 g of polymer adsorbents Dowex Optipore L493, Diaion SP825, and Diaion SP207 at a particle size range of 0.50-0.71 mm at 25 °C and at a stirring rate of 250 rpm in a three-necked-flask. The concentrations of BPA and phenol in the samples taken in the specified time intervals were monitored to evaluate the kinetic behavior of polymer adsorbents.

2.4. Analytical methods

The concentrations of BPA and phenol were determined using a Shimadzu UV-1800 model spectrophotometer at the wavelengths of 276 and 269 nm, respectively.

3. Results and discussion

3.1. Determination of optimum adsorbent concentration for BPA and phenol removal

The performance of Dowex Optipore L493 was found to be higher than those of Diaion SP825 and Diaion SP 207 for BPA removal from 0.4 mM BPA solution. Fig. 1 shows that the removal efficiency of Diaion SP825 ve Diaion SP207 for BPA removal was obtained as 95% with 0.6 g adsorbent/100 mL-solution whereas 0.4 g adsorbent/100 mL-solution was found to be the optimum amount for Dowex Optipore L493 with 99% of removal efficiency. Besides, the optimum adsorbent concentration for phenol removal from 0.4 mM phenol solution was found to be 1.1 g adsorbent/100 mL-solution with 71% of phenol removal for Diaion SP825 and Diaion SP207 (Fig. 2). Similar to efficiency of BPA removal, a higher removal efficieny was obtained as 95% using 1.0 g adsorbent/100 mL-solution for Dowex Optipore L493. Consequently, the capability of Dowex Optipore L493 to remove both BPA and phenol from aqueous solution is the highest among the adsorbents tested due to its lower average pore diameter and higher surface area in comparison with the other adsorbents. So that, the opportunity of

Table 1
Properties of polymer adsorbents.

Polymer Adsorbent	Dowex L 493	Diaion SP 825	Diaion SP207
Matrix	Styrene-DVB, macroporous	Styrene-divinylbenzene	Brominated styrene-divinylbenzene
Surface Area	>1100 m^2/g	~1000 m^2/g	650 m^2/g
Average Pore Diameter	46 Å	90–125 Å	105 Å
Particle Size Distribution	20–50 mesh	20–60 mesh	20–60 mesh

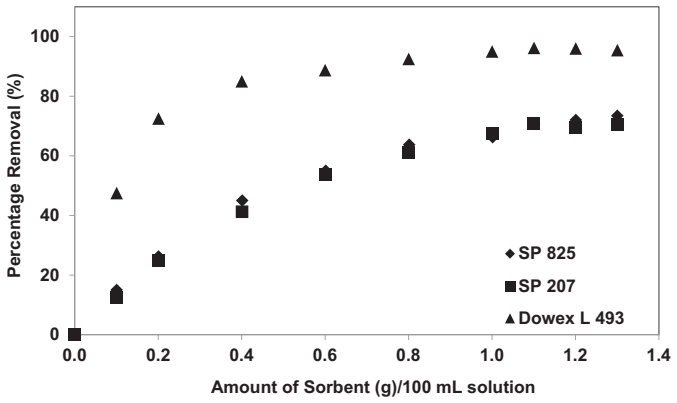

Fig. 2. Determination of optimum adsorbent amount for phenol removal from aqueous solution.

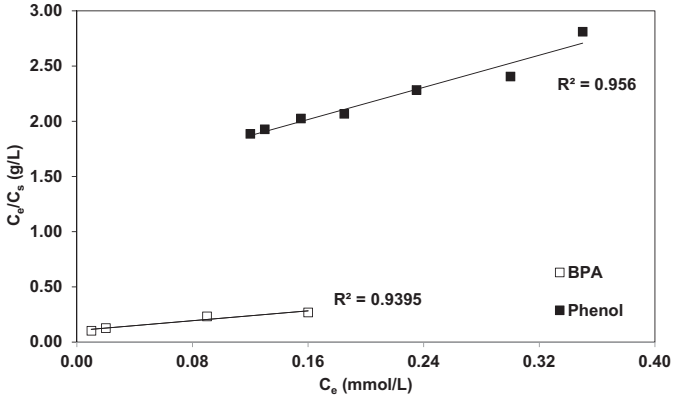

Fig. 3. Linear plots of Langmuir isotherms of BPA and phenol sorption on Diaion SP 207.

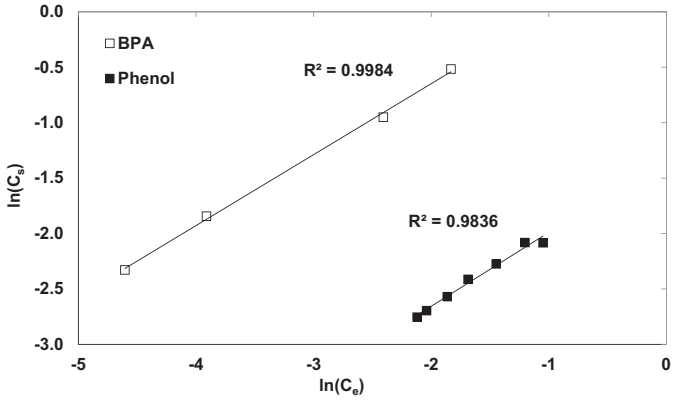

Fig. 4. Linear plots of Freundlich isotherms of BPA and phenol sorption on Diaion SP 207.

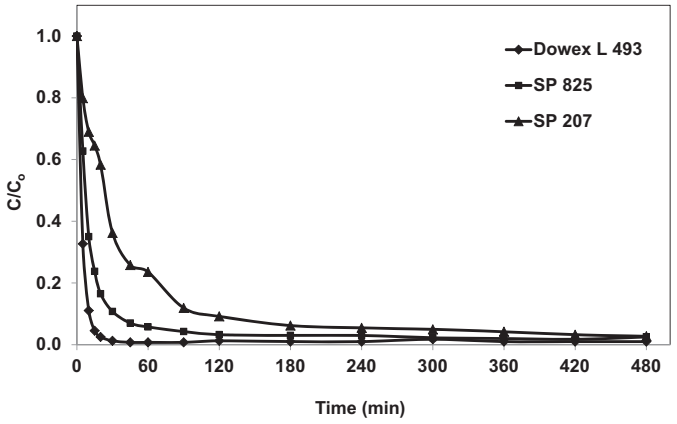

Fig. 5. Comparison of kinetic performances of polymer adsorbents for removal of BPA.

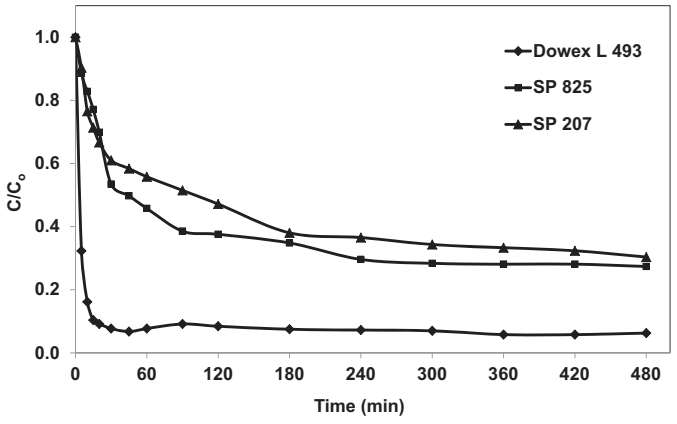

Fig. 6. Comparison of kinetic performances of polymer adsorbents for removal of phenol.

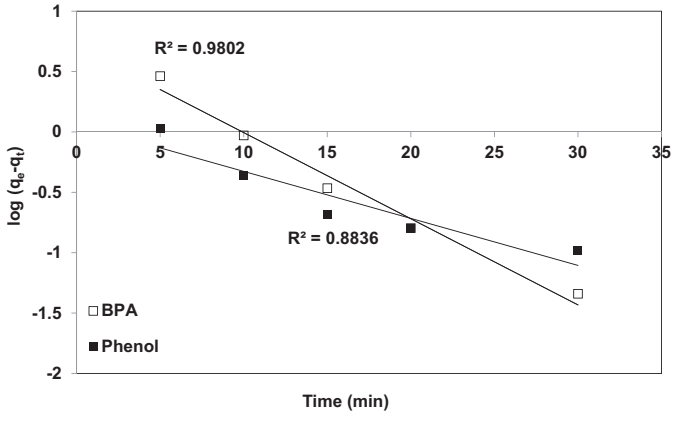

Fig. 7. Pseudo first-order sorption kinetics of BPA and phenol on Dowex L493.

Table 2
Adsorption isotherms parameters for BPA removal.

Polymer Adsorbent	Langmuir Isotherm			Freundlich Isotherm		
	C_m (mmol/g)	L (L/mmol)	R^2	K_f (mmol/g)	n (−)	R^2
Dowex L493	1.3515	25.0814	0.9470	5.7650	0.6854	0.9915
Diaion SP207	0.8952	10.7105	0.9395	1.8797	0.6399	0.9984
Diaion SP825	0.6833	19.3061	0.9957	1.3986	0.5197	0.9831

Table 3
Adsorption isotherms parameters for phenol removal.

Polymer Adsorbent	Langmuir Isotherm			Freundlich Isotherm		
	C_m (mmol/g)	L (L/mmol)	R^2	K_f (mmol/g)	n (−)	R^2
Dowex L493	0.8376	6.2936	0.9798	1.5994	0.7229	0.9920
Diaion SP825	0.4164	1.6114	0.9401	0.3269	0.7314	0.9911
Diaion SP207	0.2747	2.5390	0.9560	0.2660	0.6662	0.9836

Table 4
Evaluation of kinetic data for BPA removal.

Polymer Adsorbent	Pseudo-First Order Kinetic Model				Pseudo-Second Order Kinetic Model		
	R^2	$q_{e,exp}$ (mg g^{-1})	q_{e1} (mg g^{-1})	k_1 (min^{-1})	R^2	q_{e2} (mg g^{-1})	k_2 (g mg^{-1} min^{-1})
Dowex L493	0.9802	8.97	2.03	0.0714	0.9977	9.81	0.0421
Diaion SP825	0.9626	8.83	2.31	0.0318	0.9846	11.01	0.0095
Diaion SP207	0.9414	8.63	2.57	0.0137	0.9932	10.73	0.0037

$q_{e,exp}$: equilibrium adsorption capacity from experimental results.
q_{e1}: equilibrium adsorption capacity calculated using pseudo-first-order model.
q_{e2}: equilibrium adsorption capacity calculated using pseudo-second-order model.

Table 5
Evaluation of kinetic data for phenol removal.

Polymer Adsorbent	Pseudo-First Order Kinetic Model				Pseudo-Second Order Kinetic Model		
	R^2	$q_{e,exp}$ (mg g^{-1})	q_{e1} (mg g^{-1})	k_1 (min^{-1})	R^2	q_{e2} (mg g^{-1})	k_2 (g mg^{-1} min^{-1})
Dowex L493	0.8836	3.60	1.06	0.0389	0.9990	3.87	0.1334
Diaion SP825	0.9451	2.66	1.49	0.0105	0.9909	3.24	0.0091
Diaion SP207	0.9118	2.50	1.32	0.0047	0.9987	2.04	0.0394

$q_{e,exp}$: equilibrium adsorption capacity from experimental results.
q_{e1}: equilibrium adsorption capacity calculated using pseudo-first-order model.
q_{e2}: equilibrium adsorption capacity calculated using pseudo-second-order model.

Table 6
Diffusional and reaction models [33].

Model	Equation	Rate-determining step
ISV	$-\ln(1-X)=kt$ where $k=D_r p^2/r_0^2$	Film diffusion
ISV	$-\ln(1-X^2)=K_{1i}t$ where $K_{1i}=3DC/r_0\delta C_r$	Particle diffusion
UCM	$X=(3C_{A0}K_{mA}/a_{r0}C_{s0})t$	Liquid film
UCM	$3-3(1-X)^{2/3}-2X=(6D_{eR}C_{A0}/a_{r0}2C_{s0})t$	Reacted layer
UCM	$1-(1-X)^{1/3}=(k_sC_{A0}/a_{r0}C_{s0})t$	Chemical reaction

sorbate molecules to diffuse the available adsorption sites could be improved by using the adsorbents with small pore sizes.

3.2. Adsorption isotherms

Analysis of adsorption isotherms is important for developing a model that can be used for adsorption process design and comparison of the adsorption capacities of different adsorbents under different operational conditions. The most frequently employed adsorption isotherms models, namely, Langmuir and Freundlich, were applied to describe the adsorption of BPA and phenol. The Langmuir adsorption isotherm [25–27] assumes monolayer adsorption such that adsorption occurs on a homogenous adsorbent surface of identical sites that are equally available and energetically equivalent without lateral interaction between the adsorbed molecules. The linearized Langmuir equation is written as

$$\frac{C_e}{q_e} = \frac{1}{q_{max}K_L} + \frac{C_e}{q_{max}}$$

where C_e (mg L^{-1}) is the adsorbate equilibrium concentration, q_e (mg g^{-1}) is the adsorption capacity, q_{max} (mg g^{-1}) is the maximum adsorption capacity, K_L (L/mg) is Langmuir constant. The values of q_{max} and K_L can be calculated from the slope and intercept of the linear plot of C_e/q_e versus C_e, respectively.

The Freundlich isotherm [27–30] fits the experimental data over a wide range of concentrations. This isotherm gives an expression encompassing the surface heterogeneity, the exponential distribution of active sites, and their energies. It describes the equilibrium on heterogeneous surfaces and does not assume mono layer capacity. It can be written in a linear form as:

$$lnq_e = lnK_F + \frac{1}{n}lnC_e \quad (7)$$

where K_F (mg g^{-1}) (L mg^{-1}) and n is Freundlich constant related to adsorption capacity and adsorption intensity of the adsorbent, respectively. The values of K_F and 1/n can be obtained from the

intercept and slope of the linear plot of lnq_e versus lnC_e, respectively.

Adsorption isotherms parameters for BPA and phenol removal were summarized in Tables 2 and 3 along with the linear plots of isotherms for one of the selected polymer adsorbents which is Diaion SP 207 in Figs. 3 and 4. Table 2 shows that the adsorption equilibrium data for Dowex Optipore L493 and Diaion SP207 for adsorption of BPA were better described by Freundlich isotherm model. However, the correlation coefficients of linear plots for both isotherms show that adsorption isotherm obeys Langmuir isotherm using Diaion SP 825 for BPA adsorption. However, adsorption isotherms for phenol removal obey the Freundlich isotherm model for all type of adsorbents. Among the adsorbents, Dowex Optipore L493 has the highest adsorption capacity for both BPA and phenol according to Langmuir and Freundlich isotherm models as shown in Tables 2 and 3.

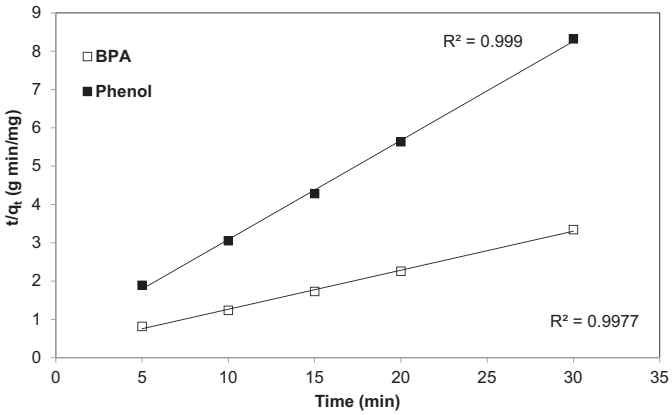

Fig. 8. Pseudo second-order sorption kinetics of BPA and phenol on Dowex L493.

3.3. Kinetic tests for BPA and phenol removal

The kinetic behaviours of the polymer adsorbents Dowex Optipore L493, Diaion SP825 and Diaion SP207 were examined and compared in order to get a comparison of the relative kinetic performance of these adsorbents for BPA and phenol removal. Figs. 5 and 6 show the kinetic data obtained with Dowex Optipore L493, Diaion SP825, and Diaion SP207 for removal of BPA and phenol, respectively. Both BPA and phenol removals were found to be faster with Dowex Optipore L493 than with Diaion SP825 and Diaion SP207. According to the results, equilibrium was approximately reached after 30 min by Dowex Optipore L493 to remove both BPA and phenol from aqueous solution so that it was the shortest period comparing to the equilibrium times obtained by other adsorbents.

In order to examine the rate controlling mechanism of adsorption process, several kinetic models were used to evaluate the experimental data. Firstly, adsorption kinetics were analyzed by pseudo-first-order equation [31,32] given as,

$$\ln(q_e - q_t) = \ln q_e - k_1 t$$

where q_e [mg g^{-1}] and q_t [mg g^{-1}] are the concentrations of BPA or phenol adsorbed on adsorbents at equilibrium and at any time t. k_1 [min^{-1}] is the rate constant. The values of q_e and k_1 can be calculated from the intercept and the slope of the linear plot of $\ln(q_e\text{-}q_t)$ versus t, respectively.

The pseudo-second-order kinetic model [31,32] is expressed as:

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

where k_2 [g mg^{-1} min^{-1}] is the rate constant. q_e and k_2 are calculated from the slope and intercept of the plot of t/q_t versus t, respectively.

Tables 4 and 5 summarize the correlation coefficients and kinetic parameters for the adsorption of BPA and phenol on different types of adsorbents. The linearity of the plots as presented for Dowex L493 adsorbent in Figs. 7 and 8 shows the empirical applicability of the pseudo-first-order or pseudo-second-order kinetic equations for BPA and phenol adsorption on the adsorbents. According to the kinetic data, BPA and phenol adsorption kinetics obey the pseudo-second-order kinetic model for all adsorbents. The results in Tables 4 and 5 point out that Dowex Optipore L493 has the highest rate constants, k_2, obtained by the pseudo-second-order kinetic model for both BPA and phenol adsorption since the equilibrium during adsorption of BPA or phenol on Dowex Optipore L493 was reached in a shorter time than those on Diaion SP825 and Diaion SP207. Moreover, equilibrium adsorption capacities calculated using pseudo-second-order model agreed well with the adsorption capacities obtained from experimental results for Dowex Optipore L493.

Finally, diffusional and reaction models such as infinite solution volume (ISV) and unreacted core (UCM) models were used to

Table 7
Evaluation of kinetic data for BPA removal according to diffusional and reaction models.

Polymer Adsorbent	R^2				
	ISV		UCM		
	$-\ln(1\text{-}X)$	$-\ln(1\text{-}X^2)$	X	$3\text{-}3(1\text{-}X)^{2/3}\text{-}2X$	$1\text{-}(1\text{-}X)^{1/3}$
Dowex L493	0.9783	0.9839	0.7457	0.9441	0.9275
Diaion SP825	0.9626	0.9821	0.8978	0.9559	0.9185
Diaion SP207	0.9779	0.9894	0.9642	0.9898	0.9642

Table 8
Evaluation of kinetic data for phenol removal according to diffusional and reaction models.

Polymer Adsorbent	R^2				
	ISV		UCM		
	$-\ln(1\text{-}X)$	$-\ln(1\text{-}X^2)$	X	$3\text{-}3(1\text{-}X)^{2/3}\text{-}2X$	$1\text{-}(1\text{-}X)^{1/3}$
Dowex L493	0.9503	0.9559	0.8310	0.9152	0.9166
Diaion SP825	0.9451	0.9567	0.8958	0.9602	0.9307
Diaion SP207	0.9554	0.9907	0.9233	0.9923	0.9454

describe the rate determining steps for BPA and phenol removals. The models for process dynamics includes both the diffusional steps (bulk solution, a film layer at the external surface of the particle, pores) and the exchange reaction on the active sites. Since the resistance in the bulk solution is easily controlled and negligible, three resistances, such as film diffusion, particle diffusion, and chemical reaction, usually determine the overall rate of the ion-exchange process. One approach uses ISV model, whereas the other method uses UCM model to describe the rate determining steps in ion exchange process [33]. These kinetic models developed for spherical particles to specify the rate-determining steps are explained in Table 6. Tables 7 and 8 summarize the linear correlation coefficients obtained from the plots of F(X) function vs. time. The rate determining step for BPA removal was found to be particle diffusion for all type of adsorbents according to both ISV and UCM models. Whereas, it is difficult to distinguish the rate controlling mechanism for phenol removal for particularly Dowex Optipore L493 adsorbent. Since the correlation coefficients for ISV model were found to be almost the same, rate-determining steps could be accepted as both film and particle diffusion. Similarly, particle diffusion and chemical reaction dominate the rate determining step due to the UCM model. On the other hand, particle diffusion determines the rate of the adsorption process for phenol removal using both Diaion SP825 and Diaion SP207.

4. Conclusion

The adsorption behaviours polymer adsorbents such as Dowex Optipore L493, Diaion SP825, and Diaion SP 207 were evaluated for the removal of BPA and phenol from aqueous solutions in terms of equilibrium and kinetc tests. Among the adsorbents tested, Dowex L493 exhibited higher adsorption affinity to BPA with an adsorption capacity of 1.35 mmol/g, while having a capacity of 0.84 mmol/g for phenol. The removal rates of BPA and phenol were found to be faster for Dowex L493 than Diaion SP825 and Diaion SP207.

Acknowledgements

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