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# Removal of bisphenol A from aqueous solution by hydrophobic sorption of hemimicelles

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**ABSTRACT:** In this work, the hydrophobic sorption of hemimicelles was proposed as an innovative method for removing bisphenol A from aqueous solution with esterified carboxyl cotton as sorbent and cetyl trimethyl ammonium bromide as cationic surfactant. In order to optimize the sorption process, the effect of sorbent dose, initial pH, surfactant dose, sorbate concentration, contact time and temperature was investigated in batch system. The maximum value of bisphenol A removal appeared in the pH range 4-10. The bisphenol A removal ratio came up to the maximum value beyond 12 time of surfactant/bisphenol A. The isothermal data of bisphenol A sorption conformed well to the Langmuir model and the maximum sorption capacity ( $Q_m$ ) of esterified carboxyl cotton for bisphenol A was 87.72 mg/g. The bisphenol A removal equilibrium was reached within about 4 h and the removal process could be described by the pseudo-second-order kinetic model. The thermodynamic study indicated that the bisphenol A sorption process was spontaneous and exothermic.

Keywords: Cetyl trimethyl ammonium bromide; Endocrine disrupting chemical; Esterified carboxyl cotton; Sorption

# **INTRODUCTION**

Endocrine disrupting chemicals (EDCs) are naturally occurring compounds or man-made chemicals that act like hormones in the endocrine system and disrupt the physiologic function of endogenous hormones. Bisphenol A (BPA), known as one of EDCs since 1936 (Dodds and Lawson, 1936), has aroused the public concerns. BPA is widely used as the monomeric material for the production of polycarbonate and epoxyphenolic resins and as the stabilizer or antioxidant for many types of plastics (Staples et al., 1998), thus it is inevitably released into the aquatic environment through various routes. The potential adverse effects of BPA on human health and reproductive biology include breast and prostate cancer, sperm count reduction, abnormal penile/urethra development in males, early sexual maturation in females, neurobehavioral problems, prevalence of obesity, type 2 diabetes and immunodeficiency.

The removal of BPA from wastewater is important in the protection of the ecological environment and human health. A number of methods such as electrochemical process (Tanaka *et al.*, 1999), sonochemical degradation (Inoue *et al.*, 2008), ozonation (Irmak *et al.*, 2005), chemical oxidation (Yoshida *et al.*, 2001; Ioan *et al.*, 2007; Li *et al.*, 2008), enzymatic oxidation (Fukuda *et al.*, 2001; Li and Nicell, 2008), photooxidation (Zhou *et al.*, 2004), solvent extraction (Fan *et al.*, 2008), membrane filtration (Dong *et al.*, 2008), sorption (Asada *et al.*, 2004), phytoremediation (Nakajima *et al.*, 2004; Noureddin *et al.*, 2004) and biodegradation (Kang and Kondo, 2002; Chai *et al.*, 2003; Lee *et al.*, 2005) have been employed to eliminate BPA from wastewater.

The sorption technique has extensively been used in industrial wastewater treatment and removing heavy metal from aqueous solution using low cost adsorbents (Adel-Ghani *et al.*, 2007; Malakootian *et al.*, 2008). If a sorbent is inexpensive and ready for use, the sorption process will be a promising technique. Recently, a few materials had been used as sorbents for removal of BPA from aqueous solution, which included chitosanbearing  $\beta$ -cyclodextrin (Aoki *et al.*, 2004), polysulfone bead (Zhao *et al.*, 2004), mineral (Shareef *et al.*, 2006), carbonaceous material (Nakanishi *et al.*, 2002), calix[4]crown derivative (Tsue *et al.*, 2005), hydrophobic zeolite (Tsai *et al.*, 2006), Fe(III)/Cr(III) hydroxide (Namasivayam and Sumithra, 2007), carbon

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nanomaterial (Pan et al., 2008), molecularly imprinted polymer (Lin et al., 2008), and polyethersulfoneorganophilic montmorillonite (Cao et al., 2009). In this work, a new hydrophobic sorption mechanism of hemimicelles for removing BPA from aqueous solution was proposed. It was based on that at certain concentration level called critical hemimicellar concentration (CHC), the ionic surfactant was sorbed on oppositely charged sorbent through coulombic attraction and formed hemimicelles and then BPA was sorbed on sorbent through hydrophobic interaction between the hydrophobic moieties of surfactant on hemimicelles and aromatic alkyl groups of BPA. The proposed sorption mechanism had been applied to remove BPA from aqueous solution with esterified carboxyl cotton (ECC) as sorbent and cetyl trimethyl ammonium bromide (CTAB) as cationic surfactant. Compared with other hydrophobic sorbents, not only BPA but also other cationic pollutants can be removed from complicated wastewater by this method with ECC as sorbent. ECC was a stable sorbent and could be used at least 20 times (Gong et al., 2007).

This study was carried out in the Key Laboratory of Biotic Environment and Ecological Safety of Anhui Province, Anhui Normal University, Wuhu, China during February, 2008 to January, 2009.

# **MATERIALS AND METHODS**

### Reagents and instrumentations

All chemicals were of analytical reagent grade unless stated otherwise. Doubly distilled deionized water was used throughout this study.

The standard BPA (Sinopharm Chemical Reagent Shanghai Co., Ltd., China) stock solution (200 mg/L) was prepared by dissolving 0.100 g of solid BPA in 5 mL of ethanol, then diluting in 500 mL volumetric flask with distilled water and stored under refrigeration. The working solutions were obtained by diluting the stock solution to different initial concentrations for removal experiments. The stock solution (2 g/L) of CTAB (Sigma-Aldrich, St. Louis, USA) was prepared by dissolving the required amount of CTAB in distilled water.

#### Preparation of ECC

The ECC was prepared according to the method described in reference (Gong et al., 2007).

#### BPA removal experiments

Except for the experiments of sorption thermo-

dynamics conducted at different temperature, other removal experiments were carried out in a rotary shaker at 150 rpm and ambient temperature using 250 mL shaking flasks containing 100 mL of BPA working solutions with known final concentration and desired initial pH values. The CTAB at different doses were previously introduced to BPA solutions and the initial pH values of BPA solutions were previously adjusted with diluted HNO<sub>2</sub> or NaOH using a pH meter. 0.1 g of ECC was respectively added into each flask and then the flasks were sealed up to prevent volume change of the solutions during the experiment. After shaking the flasks for predetermined time intervals, the samples were separated by sedimentation/centrifugation and the residual concentration of BPA in the supernatant solution was estimated by HPLC under the following conditions: column, TSKgel ODS-80Ts 4.6 mm i.d. × 250 mm (Tosoh Corp., Japan); eluent, methanol/distilled water (7:3, v/v); flow rate, 1 mL/min; temperature, 40 °C and eluent detection at 270 nm. The amount of BPA sorbed on ECC was calculated by the mass balance equation.

The 1 g/L of ECC dosage, chosen by preliminary experiments, was used for the further experiments.

The effect of initial pH was studied over the pH range from 3 to 11 with other experimental parameters fixed as follows: BPA concentration (20 mg/L), ECC dose (1 g/L), CTAB/BPA ratio (12 w/w) and contact time (4 h).

The influence of CTAB dose was investigated over the CTAB/BPA ratio range from 2 to 16 (w/w) with other experimental parameters fixed as follows: BPA concentration (20 mg/L), ECC dose (1 g/L), contact time (4 h) and initial pH (natural).

The sorption isotherm was studied over the BPA concentration range from 10 to 80 mg/L with other experimental parameters fixed as follows: ECC dose (1 g/L), CTAB/BPA ratio (12 w/w), contact time (4 h), and initial pH (natural).

The sorption kinetics was investigated over the contact time range from 0 to 5 h with other experimental parameters fixed as follows: BPA concentration (20 mg/ L), ECC dose (1 g/L), CTAB/BPA ratio (12 w/w) and initial pH (natural).

The sorption thermodynamics was studied over the temperature range from 10 to 50 °C with other experimental parameters fixed as follows: BPA concentration (20 mg/L), ECC dose (1 g/L), CTAB/BPA

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ratio (12 w/w), contact time (4 h) and initial pH (natural). The experiments were conducted in duplicate and the mean values were used as the experimental data.

# **RESULTS AND DISCUSSION**

# Effect of solution pH

The effect of solution pH on BPA removal was investigated over a range from 3 to 11. As elucidated in Fig. 1, the maximum value of BPA removal appeared in the range of pH 4 to 10. When pH is equal to 4, the formation of hemimicelles on ECC decreased because of protonation of negative charged ECC (Gong et al.,2007), the removal of BPA decreased. Due to the pK<sub>a</sub> value of BPA ranging from 9.6 to 10.2 (Staples et al., 1998), when pH is 10, the hydrophobic interaction between CTAB and BPA decreased because of deprotonation of BPA, together with the electrostatic repulsion between BPA anion and negative charged ECC, the removal of BPA decreased.

When pH is 4 or 10, the decrease of BPA removal could be explained by the following reaction equations:

$$ECC-O^{-} + H^{+} \longrightarrow ECC-OH(pH=4)$$
(1)

$$BPA + OH \rightarrow BPA + H_{2}O(pH=10)$$
(2)

#### Influence of CTAB dose

As the intermediary of BPA sorbed on ECC, the influence of CTAB dose on the removal percentage of BPA was investigated. As shown in Fig. 2, the removal percentage of BPA was enhanced from 72.67 to 95.33 % with increasing the ratio of CTAB to BPA from 2.0 to 12. Beyond 12 times of CTAB/BPA, the maximum ratio of BPA removal was reached and the removal percentage of BPA kept basically unchangeable.

#### Sorption Isotherm

Sorption isotherm of BPA was shown in Fig. 3. The data in Fig. 3 were represented by the following Langmuir and Freundlich models:

$$C_{e}/q_{e} = 1/(aQ_{m}) + C_{e}/Q_{m}$$
 (3)

$$\ln q_e = \ln K + (1/n) \ln C_e \tag{4}$$

Where  $C_{\rho}$  (mg/L) is the concentration of BPA solution at equilibrium,  $q_e$  (mg/g) is the amount of sorbed BPA at equilibrium. In Langmuir equation,  $Q_{m}$  is the maximum sorption capacity and a is Langmuir constant.



Table 1: The isotherm equations and parameters of BPA sorption on ECC

Fig. 2: Influence of CTAB dose on removal of BPA by ECC

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In Freundlich equation, K and n are empirical constants.

The results in Table 1 showed that the Langmuir model represented the sorption isotherm of BPA more reasonably than the Freundlich one. The maximum sorption capacity  $(Q_m)$  of BPA obtained from the Langmuir equation was 87.72 mg/g.

### Sorption kinetics

The BPA sorption kinetics was investigated by examining the influence of contact time on its removal. The kinetic experiment result indicated that the sorption process approached equilibrium within about 4.0 h. The kinetic data of BPA sorption were treated with the following pseudo-first-order and pseudo-second-order models:

$$\ln\left(q_{e} - q_{t}\right) = \ln q_{e} - k_{adl} t \tag{5}$$

$$t/q_{t} = 1/k_{ad2} q_{e}^{2} + t/q_{e}$$
(6)

Where  $q_e$  and  $q_t$  (mg/g) refer respectively to the amount of sorbed BPA at equilibrium and time t (min),  $k_{adl}$  and  $k_{ad2}$  are pseudo-first-order and pseudo-second-order rate constant, respectively.

The correlation coefficients in Table 2 showed that the kinetics of BPA sorption conformed to the pseudosecond-order model better than pseudo-first-order one.

#### Sorption thermodynamics

The BPA sorption thermodynamics was studied by determining the effect of temperature on its removal. The thermodynamic parameters,  $\Delta H^{\circ}$  and  $\Delta S^{\circ}$ , were obtained from the following Van't Hoff equation:

$$\ln\left(q_{e}/C_{e}\right) = \Delta S^{\circ}/R - \Delta H^{\circ}/RT$$
(7)



Fig. 3: The sorption isotherm of BPA removal by ECC

where  $q_e/C_e$  is the equilibrium constant (mL/g),  $\Delta S^{\circ}$  is standard entropy (J/mol·K),  $\Delta H^{\circ}$  is standard enthalpy (J/mol), *T* is the absolute temperature (K), and *R* is the gas constant (8.314 J/mol/K). The  $\Delta H^{\circ}$  and  $\Delta S^{\circ}$  values could be calculated from the slope  $(-\Delta H^{\circ}/R)$  and intercept  $(\Delta S^{\circ}/R)$  of the linear plot of  $\ln(q_e/C_e)$  versus 1/T.

Based on the experimental data under different temperatures, the Van't Hoff equations of BPA on ECC was given as follows:

$$\ln (q/C_{\star}) = 1559.5/T + 4.6813, R^2 = 0.9964$$
 (8)

The values of  $\Delta G^{\circ}$  was calculated from the following Gibbs-Helmholtz equation:

$$\Delta G^{\circ} = \Delta H^{\circ} - T \Delta S^{\circ} \tag{9}$$

Sorbate		pseudo-firs	t-order model		pseudo-second-order model			
	$\ln(q_e - q_t) = -0.0142t + 1.5643$				$t/q_t = 0.0518t + 0.3902$			
	$K_{ad1}$	$q_e ({ m mg/g})$		$\mathbf{R}^2$	K	$q_e ({ m mg/g})$		$\mathbb{R}^2$
		Calculated	Experimental	ĸ	n aaz	Calculated	Experimental	R
BPA	0.0142	4.779	18.93	0.9574	0.0069	19.31	18.93	0.9999
		Table 3:	The thermodynamic	mic paramet	ters of BPA s	sorption on ECC		

Table 2: The kinetic equations and parameters of BPA sorption on ECC

Sorbate	$\Delta H^2$	$\triangle S^{?}$ (J/mol·K)	$\bigtriangleup G^2$ (kJ/mol)					
	(KJ/MOI)		283.15 K	293.15 K	303.15 K	313.15 K	323.15 K	
BPA	-12.97	38.92	-23.97	-24.38	-24.77	-25.16	-25.55	

Where  $\Delta G^{\circ}$  is the standard free energy change (kJ/mol). The  $\Delta G^{\circ}$  values under different temperatures as well as  $\Delta H^{\circ}$  and  $\Delta S^{\circ}$  values, which were obtained from Eqs. 8 and 9, were presented in Table 3. The negative values of  $\Delta G^{\circ}$  and  $\Delta H^{\circ}$  indicated that the sorption of BPA on ECC was spontaneous and exothermic. Lower temperatures were favorable for the sorption process.

#### CONCLUSION

This work confirmed that the ECC could effectively remove BPA from aqueous solution in the presence of CTAB. By studying the effect of various experimental parameters, the optimal condition of BPA sorption was decided. The maximum value of BPA removal appeared in the range of pH 4 to 10. Beyond 12 times of CTAB/ BPA, the BPA removal ratio came up to the maximum value. The isothermal data of BPA sorbed on ECC followed the Langmuir model better than the Freundlich one. The maximum sorption capacity  $(Q_m)$  of ECC for BPA was 87.72 mg/g. The BPA removal equilibrium was reached within about 4 h and the BPA removal process might be described by the pseudo-second-order kinetic model. The thermodynamic study suggested that the BPA sorption process was spontaneous and exothermic.

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