REGULAR PAPER

Removal of Endocrine Disruptors by Selective Sorption Method using Activated Carbons[†]Boo Ok YOON,^{a)} Shin KOYANAGI,^{a)} Takao ASANO,^{a)}
Mariko HARA,^{a)} and Akon HIGUCHI^{*a)}

(Received September 20, 2002; Accepted October 28, 2002)

We undertook a fundamental investigation of the removal of endocrine disruptors from aqueous solution by a sorption method using activated carbons, and compared the removal ability of endocrine disruptors by activated carbons to that by polydimethylsiloxane (PDMS) membranes. The activated carbons exhibited high removal ability of 1,2-dibromo-3-chloropropane (DBCP), 100 fold increased adsorption over that with PDMS membranes from aqueous solution of DBCP. The removal ratio of the endocrine disruptors, *e.g.*, 1,2-dibromo-3-chloropropane, dibenzo-*p*-dioxin, and hexachlorocyclohexane, increased linearly in an aqueous solution with increasing the logarithm of P_{ow} of the endocrine disruptors. Almost complete removal of the endocrine disruptors (*i.e.*, >85%) was observed above $\log P_{ow}=3.5$ in 25 mL of an aqueous solution using 0.05 g activated carbon. One aim of the present study is to use preferential sorption of chemical contaminants onto activated carbons for removal of the chemical contaminants from aqueous solutions including drinking water, beverages, milk, and human milk. When the removal of endocrine disruptors contained in milk or human milk is targeted, water soluble vitamins in the milk, such as vitamin B₂, vitamin B₁₂, and niacin are expected to be co-currently adsorbed on the activated carbons. Therefore, the present sorption method using activated carbon was applied to the removal of endocrine disruptors, such as di-*n*-octylphthalate, from the mineral water and 89% of di-*n*-octylphthalate was removed.

1. Introduction

Endocrine disruptors, such as polychlorinated biphenyls (PCBs), dioxins, and certain phthalates exhibit estrogenic activity.^{1), 2)} They adversely affect the development and reproduction of humans and animals³⁾⁻⁵⁾ and thus their presence in the environment is of major concern. In this work, we examined the feasibility of removing endocrine disruptors from aqueous solutions including drinking water, beverages, milk, and human milk by sorption with activated carbons.

Activated carbons are adsorbents which are widely used in many processes for gas and water purification.⁶⁾ Although activated carbons have been known for a long time for their abilities to adsorb organic compounds, there have only been a few reports of the adsorption characteristics of endocrine disruptors by activated carbons.^{7), 8)} Cheong *et al.* investigated the adsorption of pesticides such as Fenobucarb (BPMC), Simazine, and Chlorothalonil in aqueous solution onto granular activated carbons and activated carbon fiber.⁷⁾ Nakano reported adsorption characteristics of pesticides (*i.e.*, herbicides, insecticides, and bactericide) onto activated carbons, $\log P_{ow}$ values of which ranged from 1.65 to 5.01⁸⁾ where $\log P_{ow}$ is a partition coefficient of a substance between water and *n*-octanol, and

exhibits an index of the hydrophobicity of substance.

Human milk often contains trace amounts of chemical contaminants known to be endocrine disruptors, *e.g.*, DDT, hexachlorocyclohexane (HCH), polychlorinated biphenyls (PCBs), and dibenzo-*p*-dioxin.⁹⁾⁻¹²⁾ The concentrations of PCBs and dioxins in human milk in some countries are so high that the breast-fed infant's intake exceeds the TDI (tolerable daily intake).⁹⁾⁻¹³⁾ Human milk has unique nutritional and immunological properties and is the most natural and important food source for infants. There are many advantages to breastfeeding infants based on medical studies, *e.g.*, protection against infectious diseases¹⁾⁻³⁾ and allergies,⁴⁾ and enhanced development and intelligence.³⁾ However, no method for removal of endocrine disruptors from human milk has yet been reported.

Therefore, a goal of our study is to use preferential sorption of chemical contaminants onto activated carbons for the removal of the chemical contaminants from aqueous solutions including drinking water, beverages, milk, and human milk. Because endocrine disruptors, such as polychlorinated dioxins, DDT, PCBs, and agricultural chemicals, have hydrophobic characteristics similar to hydrophobic estrogen, these chemical contaminants are expected to be absorbed from lipid micelles and proteins in the milk onto activated carbons or hydrophobic membranes such as polydimethylsiloxane membranes, after these adsorbents come into contact with the aqueous solution or milk containing the chemical contaminants. In this study,

^{a)} Department of Applied Chemistry, Seikei University (Mushashino 180-8633, Japan)

* Corresponding author. Tel: +81-422-37-3748;
Fax: +81-422-37-3871; e-mail: higuchi@ch.seikei.ac.jp

[†] Congratulations on winning the 2002 Nobel Prize

Congratulation, Prof. Fenn. I remember I visited Prof. Fenn's laboratory at Yale university, when Dr Nohmi was proceeding his research at Prof. Fenn's laboratory. I am happy to hear Prof. Fenn received Nobel Prize in 2002.

we fundamentally investigated the removal of endocrine disruptors from aqueous solution by a sorption method using activated carbons, and compared the removal of endocrine disruptors by activated carbons to that by polydimethylsiloxane membranes.

2. Experimental

2.1 Materials

Spherical activated carbon (Active Carbon Beads-M, GL Science, Inc.) with a surface area of 800–1200 m²/g and a particle diameter of 0.25–0.59 mm was used as received in this study. For comparison of its sorption ability with other adsorbent, flat-sheet polydimethylsiloxane (PDMS) membranes (SR50, Tigers Polymer, Co., Ltd.) with a thickness of 500 μ m (3.6×10^{-3} m²/g) were also used. Prior to use, the PDMS membranes were rinsed ten times with 2-propanol for 24 hours and then dried subsequently under vacuum at 25°C for 12 hours. Activated carbons used for analysis of mineral water were also rinsed three times with methanol for 24 hours and then dried under vacuum at 120°C for 10 hours, on purpose to remove impurity from the activated carbons.

Model endocrine disruptors were purchased from Wako Pure Chemicals Industries, Ltd., and are listed together with their physical properties in Table 1. Mineral water (Minami alpes no tennensui) was purchased from Suntory, Ltd.

Casein (Wako Pure Chemicals Industries, Ltd.) and γ -globulin (bovine; from Cohn Fraction II, III, Sigma Chemical, Co.) were used as the model proteins. Vitamin B₂ (riboflavin), vitamin B₁₂, and niacin (nicotinic acid) were purchased from Wako Pure Chemicals Industries, Ltd.

Other chemicals were of reagent grade and were used without further purification. Ultrapure water was used throughout the experiments.

2.2 Sorption experiments

25 mL of 1 ppm or 10 ppm aqueous solution of the endocrine disruptors, listed in Table 1, were introduced into a jar with a lid and agitated by using a shaker (Uni Ace shaker NCS-1300, Tokyo Rika Kikai Co., Ltd.) at 25°C. After the activated carbons or PDMS membranes were inserted into aqueous solutions containing endo-

crine disruptors, the concentration of endocrine disruptors, C_s (ppm), was analyzed by a gas chromatograph (GC-14B, Shimadzu Co.) equipped with a flame ionization detector (FID) or by a gas chromatograph (GC)-mass spectrometer (MS) (GCMS-QP5050A, Shimadzu Co.). The removal ratio (R) of endocrine disruptors determined by the sorption method was obtained as

$$R(\%) = (C_b - C_s) / (C_b) \times 100 \quad (1)$$

where C_b (ppm) is the initial concentration of endocrine disruptors solution.

2.3 GC/GC-MS equipments

Aliquots (5 μ L) of endocrine disruptors solutions except for hexachlorocyclohexane (HCH), biphenyl, and dibenzo-*p*-dioxin were analyzed with a gas chromatograph (GC-14B, Shimadzu Co.) equipped with FID and a Thermon-3000 column (1.6 $\times$ 3.2 mm, Shincarbon A 60/80 Shinwakakou Co., Ltd). The carrier gas was nitrogen and maintained constant at 40 kPa. The temperatures of injector and detector were set at 230°C. The column temperature was programmed as follows: 90°C for 13 min, heating at 2.5°C/min up to 120°C, heating 20°C/min up to 200°C, and keeping at 200°C for 3 min.

On the other hand, aliquots (1 μ L) of dibenzo-*p*-dioxin, biphenyl and HCH solutions were analyzed from splitless injection with GC-MS equipped with a type AOC-20 autosampler and a DB-1 capillary column (0.25 mm inner diameter of 30 m, J&W Scientific Co.). The injection and detection temperatures were set at 250°C. The column temperature was programmed as follows: 60°C for 2 min, heating at 20°C/min up to 250°C, and keeping at 250°C for 10 min unless otherwise specified.

2.4 Adsorption of proteins and water soluble vitamins

Adsorption experiments of nutrients contained in the human milk (*i.e.*, casein, γ -globulin, vitamin B₂, vitamin B₁₂, and niacin) were performed using 0.7 g of the activated carbons and PDMS membranes as the adsorbents. Casein and γ -globulin for the milk proteins, and vitamin B₂, vitamin B₁₂, and niacin for the water soluble vitamins were introduced into 25 mL of ultrapure water in a jar with a lid. The solution was agitated by using a shaker (Uni Ace shaker NCS-1300,

Table 1. Properties of Endocrine Disruptors Used

No.	Endocrine disruptors	MW (Dalton)	Use	Vapor pressure (Torr) ^{a)}	Water solubility (mg/L) ^{a)}	log P_{ow} ^{a)}
1	Didenzo- <i>p</i> -dioxin	184.2		4.125×10^{-4}	1	4.37
2	Biphenyl	154.2	Insecticide	0.0089	7.5	3.16
3	1,2-Dibromo-3-chloropropane (DBCP)	236.3	Insecticide	0.58	1230	2.96
4	2- <i>sec</i> -Butylphenyl-methylcarbamate (BPMC)	207.3	Pesticide	1.425×10^{-4}	420	2.78
5	2,2-Dimethyl-1,3-benzodioxol-4-yl methylcarbamate (Bendiocarb)	223.2	Pesticide	5×10^{-6}	260	1.70
6	<i>n</i> -Butylbenzene	134.2	Liquid crystal	1.064	11.8	4.38
7	Diethylphthalate (DEP)	222.2	Plasticizer	2.1×10^{-3}	1080	2.42
8	Dibutylphthalate (DBP)	278.3	Plasticizer	2.01×10^{-5}	13	4.72
9	1,2,3,4,5,6,-Hexachlorocyclohexane (HCH)	290.8	Insecticide	7.83×10^{-4}	8	3.8

^{a)} refers from the refs. 16–22.

Tokyo Rika Kikai Co., Ltd.) at 25°C. The initial concentration (C_b) of casein and γ -globulin were 1000 ppm, and C_b of vitamin B₂, vitamin B₁₂, and niacin were 10 ppm, 100 ppm and 50 ppm, respectively.

After the activated carbons and PDMS membranes were inserted into aqueous solutions containing proteins or vitamins, the concentrations of proteins or vitamins, C_s (ppm), was analyzed by a UV/VIS spectrophotometer (V-550, JASCO Corporation). The concentration of casein and γ -globulin was measured at 280 nm, and that of vitamin B₂, vitamin B₁₂, and niacin was 267.5 nm, 278.0 nm, and 260.5 nm, respectively.

2.5 Analysis of mineral water

100 mL of commercially available mineral water was extracted with 10 mL of hexane twice. The resulting hexane phase was dehydrated with a suitable amount (about 2.7 g) of anhydrous sodium sulfate. Thereafter, the extract was analyzed by GC-MS. For comparison, ultra pure water was also used instead of the mineral water as a blank test. Further, to estimate the removal ability of di-*n*-octylphthalate (DOP) by the present sorption method, the quantitative detection of DOP was performed from each of the extracts from the mineral water solutions where the same weight (1.6 g) of activated carbons or PDMS membranes were immersed for three hours. All of the injection and detection temperatures were set at 300°C. The column temperature was programmed as follows: 70°C for 1 min, heating at 5°C/min up to 300°C, and keeping at 300°C for 10 min.

3. Results and Discussion

3.1 Concentration change of DBCP solutions

To determine the optimal conditions of endocrine disruptor removal from aqueous solution, the time course of concentration of aqueous 1,2-dibromo-3-chloropropane (DBCP) solution adsorbed on activated carbons was investigated using a feed solution of 10 ppm DBCP. The results are presented in Fig. 1. The concentration of DBCP in the solution decreased dramatically by adsorption on the activated carbons over time. The concentration of DBCP in the solution became almost constant after one hour. Hereafter, the period of 3 hours was used as the adsorption equilibrium time on activated carbons.

3.2 Adsorption of DBCP on activated carbons

The adsorption of DBCP on activated carbons was

Fig. 1. Time dependence of concentration of 25 mL of DBCP solution inserted with (closed circle) and without (open circle) 0.05 g of activated carbons at 25°C.

Fig. 2. Dependence of removal ratio of DBCP on the weight of the adsorbents using activated carbons (open circle) or PDMS membranes (open square). The initial concentration (C_b) of DBCP solution was 10 ppm.

compared with adsorption on PDMS membranes under the same conditions as described above. The results are presented in Fig. 2. The activated carbons exhibited high removal ratio, showing a 100 fold increase in endocrine disruptor adsorption over that with PDMS membranes, which are hydrophobic polymeric membranes. This is due to the different sorption mechanisms of activated carbons and PDMS membranes: the high sorption of the activated carbons is attributed to surface adsorption originating from their large surface area, while the sorption in the PDMS membranes is governed by a solution-diffusion mechanism in the membranes.^{14), 15)}

3.3 Removal of several endocrine disruptors

Several model endocrine disruptors listed in Table 1, *i.e.*, agricultural chemicals: 2-*sec*-buthylphenyl methylcarbamate [BPMC] and 2,2-dimethyl-1,3-benzodioxol-4-yl methylcarbamate [bendiocarb]; insecticides: 1,2-dibromo-3-chloropropane [DBCP], 1,2,3,4,5,6-hexachlorocyclohexane [HCH]; liquid crystal: *n*-butyl benzene [*n*-BB]; model PCBs: biphenyl; model 2,3,7,8-tetrachlorodibenzo-*p*-dioxin [TCDD]; dibenzo-*p*-dioxin; plasticizer: diethylphthalate [DEP] and dibutylphthalate [DBP] were selected and the removal ratios of the endocrine disruptors were investigated by dipping the ac-

Fig. 3. Relationship between $\log P_{ow}$ and removal ratio of the endocrine disruptors in aqueous solution by the sorption method. C_b of the endocrine disruptor solution excepted for dibenzo-*p*-dioxin, biphenyl and HCH was 10 ppm. C_b of dibenzo-*p*-dioxin, biphenyl and HCH were 1 ppm. (1, Dibenzo-*p*-dioxin; 2, Biphenyl; 3, DBCP; 4, BPMC; 5, Bendiocarb; 6, *n*-Butylbenzene; 7, DEP; 8, DBP; 9, HCH).

tivated carbons into a 1 ppm or 10 ppm aqueous solution of the endocrine disruptors. Figure 3 shows the relationship between the octanol–water partition coefficients ($\log P_{ow}$) and the removal ratio of the model endocrine disruptors in the sorption experiments. The initial concentration (C_b) of endocrine disruptors, except dibenzo-*p*-dioxin, biphenyl, and HCH, was 10 ppm. The initial concentration of dibenzo-*p*-dioxin, biphenyl and HCH was 1 ppm due to their low solubility in water. It was found that the removal ratio of the endocrine disruptors increased linearly in an aqueous solution with increasing the logarithm of P_{ow} up to $\log P_{ow}=3.5$. Almost complete removal of the endocrine disruptors (*i.e.*, >85%) was observed above $\log P_{ow}=3.5$ in an aqueous solution, except for DBP, using 25 mL of the aqueous solution and 0.05 g activated carbons. The relatively low removal ratio of DBP is thought to be due to the DBP molecule having two long alkyl chains and thus being unable to fit into the pores of activated carbons used in this study. This result indicates that the chemicals in an aqueous solution can be removed effectively using activated carbons when the chemicals are significantly hydrophobic, *i.e.*, the chemicals have a high $\log P_{ow}$ value. Because the $\log P_{ow}$ value of the PCBs, TCDD, and DDT are reported to be 7.10, 6.80, and 6.91, respectively,^{16)–22)} it is expected that the most malignant endocrine disruptors such as the PCBs, TCDD, and DDT can be easily removed from aqueous solutions and milk using activated carbons.

3.4 Sorption of proteins and vitamins onto activated carbons

High removal ratio of endocrine disruptors in the aqueous solutions was obtained by using the activated carbons in the present study. When the removal of endocrine disruptors contained in milk or human milk is targeted, one of the disadvantages is expected to be decreased in some nutrients in the human milk due to their co-current sorption into the activated carbons. Important nutrients in human milk are proteins, such as caseins and immunoproteins, which strengthen the immune system of infants. It was reported that human milk contains 1.9% of casein and 0.14% of immunoproteins.²³⁾ Therefore, the removal ratio of casein and γ -globulin into the activated carbons and PDMS membranes was investigated and is shown in Table 2.

The removal ratio of casein and γ -globulin was found to be less than 2.2% using 1000 ppm of casein or γ -globulin solution into 0.70 g of activated carbons or PDMS membranes. This is thought to be due to the protein molecules being too large to be sorbed into the activated carbons and PDMS membranes.

Other important nutrients are water-soluble and water-insoluble vitamins. The removal ratio of vita-

mins from aqueous solution containing 10 ppm of vitamin B₂, 100 ppm of vitamin B₁₂, and 50 ppm of niacin was investigated and results are shown in Fig. 4 and Table 2. The removal ratio of vitamin B₂, vitamin B₁₂, and niacin was observed to be 0%, 0.8%, and 2.7%, respectively, when 0.70 g of the PDMS membranes were immersed in the solutions of the model vitamins. The removal ratio of vitamin B₂, vitamin B₁₂, and niacin, however, was observed to be 98.4%, 42.3%, and 98.8%, respectively, when 0.70 g of the activated carbons was immersed in the vitamin solutions. These results indicate that the usage of activated carbons as an adsorbent may contribute to adsorb co-currently water soluble vitamins in the milk, such as vitamin B₂, vitamin B₁₂, and niacin, although a higher adsorption with activated carbons for the endocrine disruptors from aqueous solution was observed than for sorption using PDMS membranes, as shown in Fig. 2. Therefore, the present sorption method using activated carbon was applied to the removal of endocrine disruptors from the mineral water in the next section.

3.5 Analysis of mineral water by GC-MS

Finally, the selective removal of the endocrine disruptors as contaminants in drinking water was investigated using activated carbons and PDMS membranes. A commercially available mineral water not containing sugars, lipids and proteins was used as the sample drinking water. Figure 5 shows the chromatograms of extracts from a commercial mineral water (a) as determined by GC-MS measurements. Di-*n*-octylphthalate (DOP) was observed at retention time of 37.2 min in the extract from mineral water (a) by measurements in electrical ionization (EI) mode. For comparison, a blank ultra pure water (was not illustrated in Fig. 5) was determined by GC-MS, but DOP was not detect-

Fig. 4. Dependence of removal ratio of vitamin B₂ (circle), vitamin B₁₂ (square), and niacin (triangle) on the weight of PDMS membranes (open symbols) and activated carbons (closed symbols). The initial concentrations of vitamin B₂, vitamin B₁₂, and niacin were 10 ppm, 100 ppm, and 50 ppm, respectively.

Table 2. Sorption Ability of Proteins and Vitamins in an Aqueous Solution Using PDMS Membranes and Activated Carbons of 0.7 g

Adsorbents used	Removal ratio (%) of proteins		Removal ratio (%) of vitamins		
	Casein $C_b=1000$ ppm	γ -Globulin $C_b=1000$ ppm	Vitamin B ₂ $C_b=10$ ppm	Vitamin B ₁₂ $C_b=100$ ppm	Niacin $C_b=50$ ppm
PDMS membrane	2.1%	0.2%	0%	0.8%	2.7%
Activated carbon	1.9%	1.1%	98.4%	42.3%	98.8%

Fig. 5. Chromatograms of extracts from a commercial mineral water (a), and mineral water solutions, where PDMS membrane (b) or activated carbon (c) were immersed, as determined by the GC-MS method. The arrows show a peak of di-*n*-octylphthalate (DOP) in the chromatograms (a), (b), and (c).

ed in the extract from the ultra pure water. DOP is expected to be used as a plasticizer in polyethylene terephthalate (PET) bottles or their plastic caps. However, the plasticizers such as DEP, DBP, or DOP from the PET bottle and its plastic cap were not detected by IR spectra measured with an ATR-FT/IR Spectrometer (FT/IR-600 Plus, JASCO). This might be due to the concentration of the plasticizers being below the detection limit of the ATR-FT/IR spectrometer (*i.e.*, less than 1 wt%). It is unclear whether the bottle or cap is the origin of DOP released into the mineral water. It is likely that DOP in the mineral water leaked out from some parts of the PET bottle during storage.

To estimate the removal ability for DOP of the present sorption method, quantitative detection of DOP was performed from each of the extracts from original mineral water, and mineral water solutions where adsorbents (*i.e.*, activated carbons or PDMS membranes) were immersed for three hours. This quantitative determination was performed by SIM (Selected Ion Monitoring) mode of GC-MS. The DOP level detected in the original mineral water (*i.e.*, 0.51 ppb) was reduced to 0.05 ppb and 0.33 ppb after the mineral water was treated with the activated carbons and PDMS membranes, respectively. It is found that 89% of DOP can be removed from mineral water using the activated carbons, while 36% of DOP was removed from mineral water using PDMS membrane. It is, therefore, suggested that the endocrine disruptors such as DOP including DEP and DBP can be also removed from drinking water by sorption using the activated carbons or PDMS membranes.

4. Conclusions

Endocrine disruptors, *e.g.*, 1,2-dibromo-3-chloropropane, dibenzo-*p*-dioxin, and hexachlorocyclohexane were removed efficiently by a simple sorption method using activated carbons. It was found that endocrine disruptors in an aqueous solution can be removed effectively with activated carbons when the endocrine disruptors are significantly hydrophobic, *i.e.*, the endocrine disruptors have a high $\log P_{ow}$ value. Because the $\log P_{ow}$ value of PCBs, TCDD, and DDT are reported to be 7.10, 6.80, and 6.91, respectively, it is expected that these most malignant endocrine disruptors can be easily removed from aqueous solutions using the activated carbons.

When compared with the sorption ability of PDMS membranes, it was found that the activated carbons showed higher adsorption of endocrine disruptors from aqueous solution than that of PDMS membranes. However, the usage of activated carbons as an adsorbent is inadequate for the removal of endocrine disruptors from human milk, because water-soluble vitamins, such as vitamin B₂, vitamin B₁₂, and niacin, were co-currently adsorbed on the activated carbons, but not into the PDMS membranes. In contrast, casein and γ -globulin, as the protein components in human milk, did not adsorb on either the activated carbons or into the PDMS membranes. One solution for safe human milk consumption for infants might be to first treat the human milk with the present sorption method using activated carbons and then replenish nutrients that were removed by sorption.

Endocrine disruptors such as di-*n*-octylphthalate were trace detected from mineral water in a polyethyl-

ene terephthalate bottle and were removed from the mineral water by the sorption method using activated carbons and to some extent by PDMS membranes.

Acknowledgement

This research was partially supported by the Salt Science Foundation.

References

- 1) M. Hahn-Zoric, F. Fulconis, I. Minoli, G. Moro, B. Carlsson, M. Bottiger, N. Raiha, and L. A. Hanson, *Acta Paediatr. Scand.*, **79**, 1137 (1990).
- 2) S. L. Cochi, D. W. Fleming, A. W. Hightower, K. Limpakarnjanarat, R. R. Facklam, J. D. Smith, R. K. Sikes, and C. V. Broome, *J. Pediatr.*, **108**, 887 (1986).
- 3) I. Lopez, M. Quibriac, J. Petitjean, M. Bazin, J. F. Duhamel, and F. Freymuth, *Arch. Fr. Pediatr.*, **46**, 263 (1989).
- 4) R. K. Chandra, S. Puri, and A. Hamed, *Br. Med. J.*, **22**, 228 (1989).
- 5) A. Lucas, R. Morley, T. J. Cole, G. Lister, and C. Leeson-Payne, *Lancet*, **339**, 261 (1992).
- 6) R. C. Bansal, J. B. Donnet, and F. Stoeckli, "Active Carbon," Marcel Dekker, New York (1988).
- 7) Y. E. Cheong, Y. Sudo, J. E. Sohn, and M. Suzuki, *J. Korea Soc. Env. Eng.*, **10**, 1209 (1996).
- 8) S. Nakano, *Kagaku to Kogyo*, **64**(3), 90 (1990).
- 9) A. A. Jensen, "Chemical Contaminants in Human Milk," ed. by A. A. Jensen and S. A. Slorach, CRC Press, Boston, Massachusetts (1991), p. 45-198.
- 10) R. Nakagawa, H. Hirakawa, T. Iida, T. Matsueda, and J. Nagayama, *J. AOAC Int.*, **82**, 716 (1999).
- 11) J. S. LaKind, C. M. Berlin, and D. Q. Naiman, *Environ. Health Perspect.*, **109**, 75 (2001).
- 12) B. C. Gladen, S. C. Monaghan, E. M. Lukyanova, O. P. Hulchiy, Z. A. Shkyryak-Nyzhnyk, J. L. Sericano, and R. E. Little, *Environ. Health Perspect.*, **107**, 459 (1999).
- 13) I. Tsuyumoto, "Dioxin," Natsume, Tokyo (2000).
- 14) C. L. Mantell, "Adsorption," McGraw-Hill Book Co., Inc., New York and London (1951), p. 20.
- 15) J. Crank and G. S. Park, "Diffusion in Polymer," Academic Press, London and New York (1968), Chapter 7.
- 16) B. J. Herbert and J. G. Dorsey, *Anal. Chem.*, **67**, 744 (1995).
- 17) W. Y. Shiu, W. Doucette, F. A. P. C. Gobas, A. Andren, and D. Mackay, *Environ. Sci. Technol.*, **22**, 651 (1988).
- 18) C. Hansch, A. Leo, and D. Hoekman, "Exploring QSAR—Hydrophobic, Electronic, and Steric Constants," American Chemical Society, Washington, D.C. (1995), p. 101.
- 19) W. J. Doucette and A. W. Andren, *Environ. Sci. Technol.*, **21**, 821 (1987).
- 20) D. E. Munnecke and S. D. Vanguhdy, *Annu. Rev. Phytopathol.*, **17**, 405 (1979).
- 21) S. F. Donovan, *J. Chromatogr. A*, **749**, 123 (1996).
- 22) D. A. Hinckley, T. F. Bidleman, and W. T. Foreman, *J. Chem. Eng. Data*, **35**, 232 (1990).
- 23) Y. Tamari, T. Matsuo, E. S. Kim, and T. Mori, "Study on the Breast Feeding," Konan University Sougou Kenkyujyo (1998), Series No. 49.

Keywords: Activated carbon, Sorption, Endocrine disruptors, Membrane, Octanol-water partition coefficient ($\log P_{ow}$)