

Comparison of vapor adsorption characteristics of acetone and toluene based on polarity in activated carbon fixed-bed reactor

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Abstract—Adsorption characteristics according to polarity of acetone and toluene vapors on coconut based activated carbon were investigated by using a fixed bed reactor. Single vapor and binary vapor adsorption of acetone and toluene were conducted. In the single vapor system, the equilibrium adsorption capacity of toluene vapor on activated carbon was five times higher than that of acetone vapor because of polarity difference between adsorbent and adsorbate. The breakthrough curve of acetone vapor in the binary vapor was quite different from that of single acetone vapor. Acetone adsorbed in the binary vapor was substituted with toluene due to the affinity difference during adsorption process and its outlet concentration increased to 1.6 times than inlet concentration. The temperature changes in activated carbon bed during adsorption of acetone vapor and toluene vapor occurred in the time ranges of 10-30 min. The temperature change for acetone vapor adsorption was about 3 °C; however, that for toluene vapor adsorption was increased to 33 °C maximally.

Key words: Activated Carbon, Adsorption, Acetone Vapor, Toluene Vapor, Breakthrough Curve

INTRODUCTION

Enormous quantities of various organic solvents have been annually produced from a variety of industrial sources. The emission of these organic solvents in the atmosphere has caused harmful effects on human health and on the environment. Since they are directly related to both human health and environment, there is much public concern about widespread environmental pollution. The volatile organic compounds (VOCs) among organic solvents are critical toxic substances. These substances may have harmful effects on human health and could even cause cancer if people are exposed for a long time.

Many methods to treat these organic solvents have been developed. They are classified in two broad groups: oxidation method and recovery method. Oxidation method is simply to destruct the organic solvents into harmless materials such as CO₂ and H₂O, but it is impossible to reuse the VOC, and this method is expensive and energy exhausting. In this context, the recovery method is able to reuse the used organic solvents and has many advantages including organic solvent recovery, energy savings, and environmentally safe aspect. The recovery methods to recover the used organic solvents in the air are condensation, adsorption by adsorbents, absorption by absorbing agents, ion-exchange, and membrane filtration [Kang et al., 2000]. Among these methods, the adsorption method is considered as the most promising technology in view point of organic solvent recovery and energy savings. Also, the adsorption method is regarded as effective technology because it is possible to separate adsorbate selectively according to pore structure of adsorbent. In adsorption, the adsorption efficiency is governed by characteristics of adsorbate and adsorbent. Therefore, it is very important to choose the appropriate adsorbent.

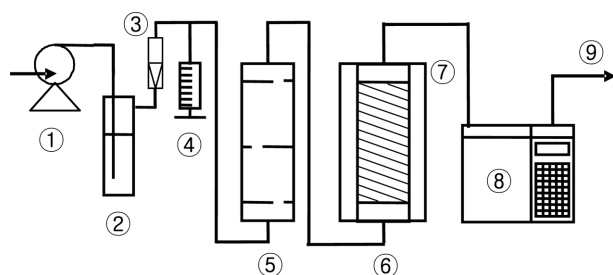
Activated carbon is one of the most effective adsorbent materials and is widely used in adsorption process. The large surface area, well developed microporous structure, and rapid adsorption capability of activated carbon provide very effective performances for removing pollutants from the effluent air stream. Many researchers have studied the elimination and the recovery of VOCs using various activated carbons [Hwang et al., 1998; Lee et al., 1999; Kim et al., 2002]. Popescu et al. [2002] studied the behavior of adsorption and desorption of toluene, butyl acetate, and butanol on activated carbons, and Chiang et al. [2001] published the experimental results of toluene according to pore structure of activated carbon and adsorption temperature. Harikrishnan et al. [1998] investigated adsorption characteristics of ethyl benzene according to pressure and temperature, and Kang et al. [2000] published the experimental results using activated carbon in the benzene recovery process. Kim et al. [2001b] studied the adsorption characteristics of benzene in an activated carbon fixed bed reactor, and Kim et al. [2001a] studied the adsorption of three major solvent vapors such as n-hexane, toluene, and MEK on two pelletized commercial activated carbons. Yun et al. [1997] studied the dynamic behavior of non-ideal gas mixtures in an isothermal fixed-bed adsorber. But these studies were mainly concerned with adsorption of nonpolar organic compounds, and they were hardly concerned with adsorption of polar organic compounds. Although Huang et al. [2003] and Lee et al. [2004] studied for acetone and polar organic compounds, they merely estimated the equilibrium adsorption capacity and adsorption ability according to temperature. But VOCs generated from plants and factories are composed of polar and nonpolar organic compounds mixed with each other. So adsorption characteristics between polar and nonpolar organic compounds were thought to be different.

The present work focuses on the investigation of the adsorption characteristics according to polarity of adsorbate. The breakthrough experiments were conducted as a function of inlet concentration and linear velocity of adsorbates, acetone and toluene using acti-

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Table 1. Characteristics of activated carbon

Properties	Value
Material	coconut
Particle size (mesh)	8-12
Iodine number (mg/g)	1,100
BET specific surface area (m ² /g)	1,145
Total pore volume (cm ³ /g)	0.41
Average pore diameter (Å)	19.0

**Fig. 1. Schematic diagram of experimental apparatus.**

- | | |
|------------------|------------------|
| ① Air pump | ⑥ Adsorption bed |
| ② Drying bottle | ⑦ Jacket |
| ③ Flow meter | ⑧ GC |
| ④ Syringe pump | ⑨ Vent |
| ⑤ Mixing chamber | |

vated carbon fixed bed reactor. The temperature changes by heat of adsorption in activated carbon bed were investigated. Competitive adsorption in the mixed vapor of acetone and toluene was also investigated.

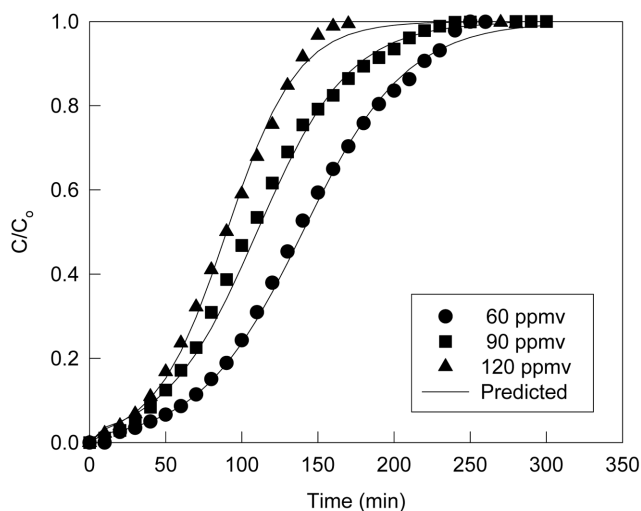
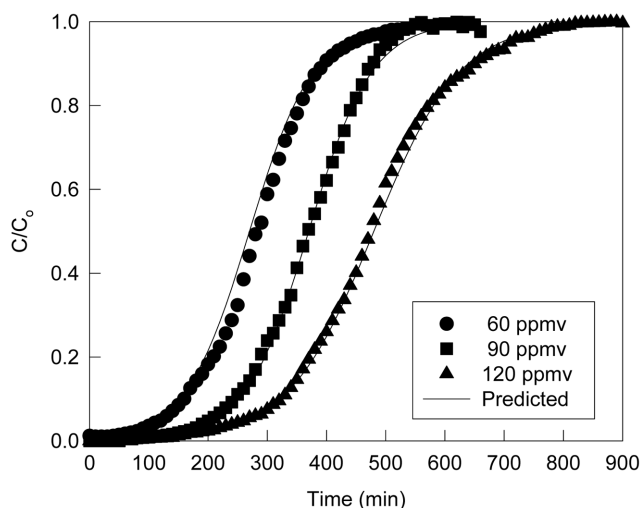
EXPERIMENTAL

Coconut based activated carbon was prepared in the particle size of 8-12 mesh, and was used as adsorbent as in the previous paper [Lim et al., 2005]. Characteristics of the activated carbon are listed in Table 1. BET specific surface area was 1,145 m²/g, total pore volume was 0.41 cm³/g, average pore diameter was 19 Å, and Iodine number was 1,100 mg/g. Toluene and acetone of 99.9% grade, obtained from Samchun Chemical Co., Ltd., Korea were used as adsorbate without purification in this experiment. Adsorption experiments were conducted by using the apparatus shown in Fig. 1. The experimental apparatus consisted of vapor generation part and fixed bed for adsorption.

Adsorption experiments were conducted at 20 °C, and the fixed bed prepared by Pyrex tube of 16 mm inside diameter was used. Air from an air pump (Young Nam Yasunaga Co. Ltd., LP-40A) was first fed to the bottle packed with silica-gel in order to minimize the effect of moisture during adsorption. Acetone vapor and toluene vapor were prepared by vaporizing acetone and toluene flowed out from syringe pump (Cole Parmer International, U.S.A., p-74901-10). Vaporized acetone and toluene were mixed thoroughly with dried air passing through the mixing chamber to set up a certain concentration, and those vapors flowed into the bottom side of the reactor and flowed out from the upside. The upside of the reactor was connected to the injection port of the gas chromatograph (GC, Donam DS-6200). The analysis conditions of GC are as in Table 2.

Table 2. Analysis conditions for acetone vapor and toluene vapor by gas chromatography

GC	DONAM DS 6200
Detector	FID (Flame Ionization Detector)
Oven temperature	130 °C
Injector temperature	250 °C
Detector temperature	200 °C
Flow rate (N ₂ : H ₂ : Air, ml/min)	30 : 30 : 300

**Fig. 2. Effect of inlet concentration of acetone vapor on breakthrough curve (linear velocity: 0.42 m/s, activated carbon: 2 g).****Fig. 3. Effect of inlet concentration of toluene vapor on breakthrough curve (linear velocity: 0.42 m/s, activated carbon: 2 g).**

RESULTS AND DISCUSSION

1. Effect of Inlet Concentration of Adsorbate on Breakthrough Curve

To investigate breakthrough characteristics according to inlet concentration of acetone and toluene vapors, an adsorption experiment

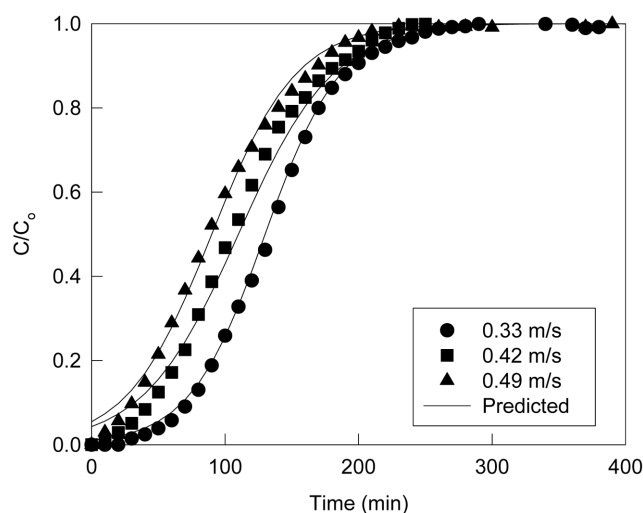
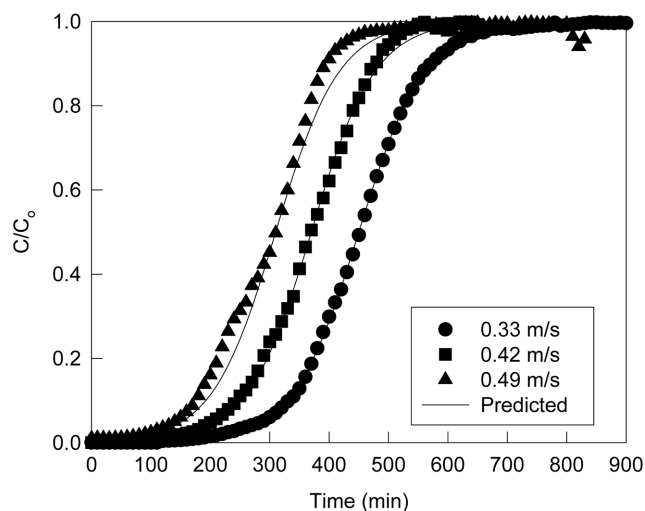
Table 3. Effect of inlet concentration of adsorbate on breakthrough time and adsorption capacity

Adsorbate	C_o (ppmv)	t_b (min)	t^* (min)	q (mg/g)
Acetone vapor	60	70	160	54
	90	50	110	64
	120	30	90	74
Toluene vapor	60	330	480	294
	90	250	370	339
	120	170	270	349

was carried out at 20 °C in the following conditions: linear velocity 0.42 m/s, inlet concentration of adsorbate 60, 90, 120 ppmv. The breakthrough curves of acetone vapor according to inlet concentration are compared in Fig. 2. And the breakthrough curves of toluene vapor according to inlet concentration are compared in Fig. 3. As shown in Fig. 2 and Fig. 3, the higher inlet concentration of adsorbate was, the faster breakthrough was, and the slope of breakthrough curve was gradually increased. Also, the variation range of breakthrough curve of acetone vapor was not larger than that of toluene vapor. In Figs. 2 and 3, breakthrough time (t_b) is defined as time when outlet concentration was 10% of inlet concentration, and when outlet concentration was 50% of inlet concentration, it was defined as stoichiometric breakthrough time (t^*). The breakthrough time, stoichiometric breakthrough time, and adsorption capacity (q) according to inlet concentration obtained from Fig. 2 and 3 were listed in Table 3. As inlet concentration of acetone vapor was increased from 60 ppmv to 120 ppmv, the breakthrough time was decreased from 70 min to 30 min. And as inlet concentration of toluene vapor was increased from 60 ppmv to 120 ppmv, the breakthrough time was decreased from 330 min to 170 min. When inlet concentration was 60 ppmv, the equilibrium adsorption capacities of acetone vapor and toluene vapor were 54 mg/g and 294 mg/g, respectively. And when inlet concentration was increased to 120 ppmv, that is twice concentration, the equilibrium adsorption capacities of acetone vapor and toluene vapor were 74 mg/g and 349 mg/g, respectively. Therefore, as inlet concentrations of acetone vapor and toluene vapor were increased, the breakthrough time was decreased, but the equilibrium adsorption capacities of acetone vapor and toluene vapor were increased. At the same concentration, the equilibrium adsorption capacity of toluene vapor was 4.7-5.4 times than that of acetone vapor. This result is explained by the fact that allowable adsorbate molecules are increased as increasing adsorbate concentration; also, the adsorption is fast as much as the increase of diffusion velocity and adsorption velocity into pores of activated carbon, so time to reach equilibrium is reduced. Also, it was thought that the increase of adsorption capacity according to increasing adsorbate concentration was due to the increase of concentration differences, that is, driving force in mass transfer [Hwang et al., 1998; Lee and Yu, 1998].

2. Effect of Linear Velocity of Adsorbate on Breakthrough Curve

To investigate breakthrough characteristics according to linear velocity of adsorbate, an adsorption experiment was carried out at 20 °C in the following conditions: inlet concentration of adsorbate 90 ppmv, linear velocity 0.33-0.49 m/s. The breakthrough curves of acetone vapor according to linear velocity are compared in Fig. 4. And the breakthrough curves of toluene vapor according to linear

**Fig. 4. Effect of linear velocity of acetone vapor on breakthrough curve (Inlet acetone concentration: 90 ppmv, activated carbon: 2 g).****Fig. 5. Effect of linear velocity of toluene vapor on breakthrough curve (Inlet toluene concentration: 90 ppmv, activated carbon: 2 g).**

velocity are compared in Fig. 5. The breakthrough time, stoichiometric breakthrough time, and adsorption capacity according to linear velocity obtained from these breakthrough curves are shown in Table 4. As increasing linear velocity from 0.33 m/s to 0.49 m/s, the breakthrough times of acetone vapor and toluene vapor were reduced from 70 min to 30 min, and from 340 min to 180 min, respectively, but adsorption capacities did not show the difference as 62-65 mg/g and 331-339 mg/g, respectively. The breakthrough time was also decreased according to the increase of linear velocity. This result was similar to breakthrough characteristics according to the changes of inlet concentration. This result can be explained as that the more linear velocity is increased, the more adsorbates per unit time are flowing into the adsorption bed, and the gas film on the surface of activated carbon becomes thin; thus mass transfer coefficient of gas film is increased, and adsorption velocity is fast [Kim and Park, 2003;

Table 4. Effect of linear velocity of adsorbate on breakthrough time and adsorption capacity

Adsorbate	V (m/s)	t_b (min)	t^* (min)	q (mg/g)
Acetone vapor	0.33	70	130	62
	0.42	50	110	64
	0.49	30	90	65
Toluene vapor	0.33	340	450	334
	0.42	250	370	339
	0.49	180	310	331

Table 5. Velocity constants (k') according to inlet concentration (C_o) and linear velocity (V)

Adsorbate	C_o (ppmv)	k' (s^{-1})	V (m/s)	k' (s^{-1})
Acetone vapor	60	0.02943	0.33	0.03544
	90	0.03323	0.42	0.02824
	120	0.04297	0.49	0.03173
Toluene vapor	60	0.0138	0.33	0.0181
	90	0.0187	0.42	0.0187
	120	0.0185	0.49	0.0187

Cho et al., 1999; Jeon et al., 1996].

Yoon and Nelson [1984] proposed an empirical equation describing the breakthrough curve on adsorption as follows:

$$C = C_o \left(\frac{1}{1 + e^{k'(t-t^*)}} \right) \quad (1)$$

Where C is outlet concentration (ppmv), C_o is inlet concentration (ppmv), k' is velocity constant (s^{-1}), t is breakthrough time (min), and t^* is stoichiometric breakthrough time (min).

The experimental data obtained from the adsorption experiment according to concentration (Fig. 2 and 3) and linear velocity (Fig. 4 and 5) was applied to Eq. (1) by using non-linear regression, and the calculated velocity constants (k') are shown in Table 5. The experimental data and breakthrough curve obtained by substituting these velocity constants to Eq. (1) are compared in Fig. 2-5. As shown in Figs. 2-5, the empirical equation of Yoon and Nelson fairly described the breakthrough curves obtained in this experiment.

3. Adsorption Isotherms

Generally, Langmuir and Freundlich equation were used as adsorption isotherm equation. Langmuir equation was expressed as follows:

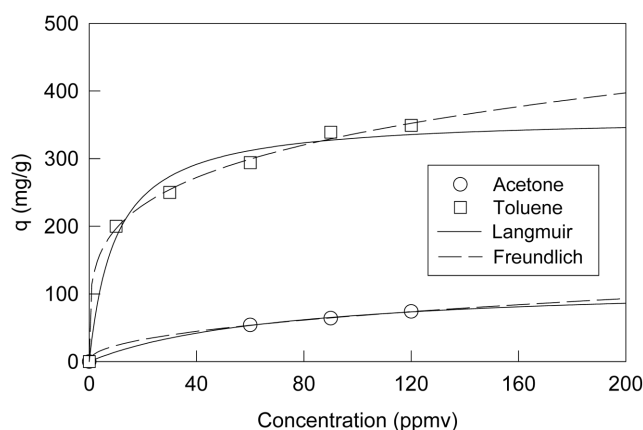
$$q = \frac{q_{max}bC}{1+bC} \quad (2)$$

where q is the equilibrium adsorption capacity (mg/g), q_{max} is the theoretical maximum adsorption capacity (mg/g), C is the equilibrium concentration of adsorbate (ppmv), and b is an experimental constant.

Freundlich equation was expressed as follows:

$$q = KC^{1/n} \quad (3)$$

where q is the equilibrium adsorption capacity (mg/g), C is the equilibrium concentration (ppmv), and K and n are experimental constants.

**Fig. 6. Comparison of adsorption isotherms of acetone and toluene vapors at 20 °C.****Table 6. The constants of Langmuir and Freundlich isotherm**

Adsorbate	Langmuir			Freundlich		
	q_{max} (mg/g)	b	r^2	K	n	r^2
Acetone vapor	117.2	0.01395	0.99	8.279	2.190	0.99
Toluene vapor	362.9	0.1012	0.98	114.3	4.254	0.99

The adsorption isotherm is to illustrate the equilibrium adsorption capacity at constant temperature according to concentration of adsorbate. Adsorption isotherms of acetone vapor and toluene vapor carried out at 20 °C are compared in Fig. 6. As shown in Fig. 6, equilibrium adsorption capacity was increased with the increase of concentration. The experimental data in Fig. 6 were substituted into Eq. (2) and Eq. (3), and the parameters calculated by non-linear regression method are listed in Table 6. The adsorption isotherms of acetone vapor and toluene vapor well satisfied the Langmuir and Freundlich equation altogether. The correlation coefficients (r^2) between these isotherm equations and experimental data of acetone vapor and toluene vapor were over 0.98 in all. Maximum adsorption capacities (q_{max}) of acetone vapor and toluene vapor calculated by Langmuir equation were 117.2 mg/g and 362.9 mg/g, respectively. Maximum adsorption capacity of toluene vapor was three times as much as adsorption capacity of acetone vapor. This result could be explained by relationship between the surface of activated carbon and these vapors. Acetone molecule has polarity by uncovalent electron dipole of oxygen atom due to C=O bond within its molecule, but toluene molecule has nonpolarity due to C-C bond between benzene and methyl group [William, 2000]. Also, because the activated carbon used in this study has nonpolarity and hydrophobicity, the adsorption ability of toluene vapor with nonpolar nature is higher than that of acetone vapor with polar nature [Jeon and Seo, 2002]. This result indicates that the nonpolar nature of activated carbon displays a high affinity toward nonpolar organic compounds.

4. Temperature Change in Activated Carbon Bed During Adsorption

When organic compounds are adsorbed on activated carbon, the accumulation of heat generated during adsorption progress may set fire to an activated carbon bed. Ko et al. [1997] studied the heat of

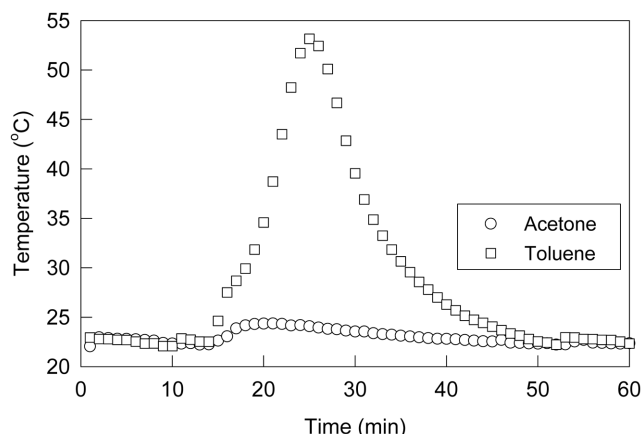


Fig. 7. Temperature changes in activated carbon bed during adsorption of acetone and toluene vapors (Inlet concentration: 120 ppmv, linear velocity: 0.42 m/s, activated carbon: 2 g).

adsorption and ignition of the organic solvent by activated carbon and reported that adsorption heat was increased with the increase of organic solvent concentration and micropores, and with the decrease of linear velocity. They also mentioned that the possibilities of firing within an activated carbon bed were increased with the increase of activated carbon bed length, since the generated heat was easily accumulated in a fixed bed, but the discharge of heat from the bed was difficult.

To measure the heat generated in an activated carbon bed during adsorption of acetone vapor and toluene vapor, a K-type thermocouple was installed in the middle part of fixed bed, and an experiment was carried out at 20 °C in the following conditions: inlet concentration 120 ppmv, linear velocity 0.42 m/s. Temperature changes according to adsorption time are shown in Fig. 7. As adsorption continued, the heat of adsorption was slightly increased until 10 min, but rapid temperature changes showed in the time ranges of 10–30 min. After 30 min, there was no temperature change as a result of cooling by continuously flowing in adsorbate at 20 °C. When acetone vapor was adsorbed on activated carbon, the temperature change was about 3 °C. But when toluene vapor was adsorbed on activated carbon, the temperature change was increased to 33 °C maximally. These results implied that adsorption heat of acetone vapor on activated carbon was low as a result of small adsorption capacity by different polarity between adsorbate and adsorbent. Adsorption heat of toluene vapor on activated carbon was high as a result of large adsorption capacity by similar polarity between adsorbate and adsorbent, and the increment of adsorption capacity was transformed into increment of temperature change [Ko et al., 1997].

5. Breakthrough Curve of Binary Vapor

To investigate breakthrough characteristics of binary vapor of acetone and toluene, an adsorption experiment was carried out at 20 °C in the following conditions: linear velocity 0.42 m/s, inlet concentration of adsorbate 60 ppmv. The breakthrough curves of binary vapor were compared with those of single acetone vapor and single toluene vapor, respectively in Fig. 8. As shown in Fig. 8, the breakthrough curve of toluene vapor in the binary vapor was similar to that of single vapor. However, the breakthrough curve of acetone vapor in the binary vapor was quite different from that of single

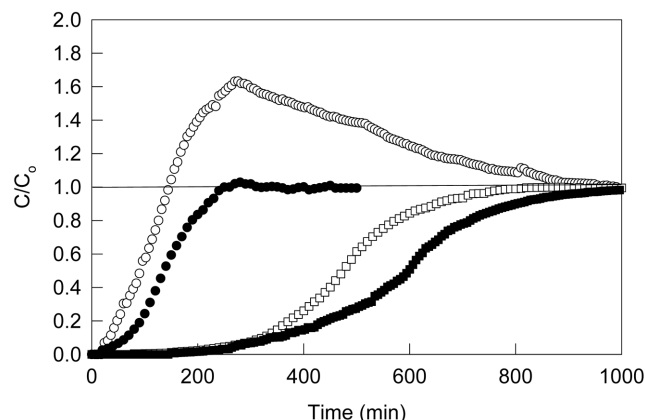


Fig. 8. Comparison of breakthrough curves for single and binary vapor systems of acetone and toluene: (●) acetone vapor, (■) toluene vapor; (○) acetone vapor in binary vapor, (□) toluene vapor in binary vapor (Inlet concentration: 60 ppmv, linear velocity: 0.42 m/s, activated carbon: 2 g).

vapor. The outlet concentration of acetone vapor in the binary vapor reached to the inlet concentration rapidly and increased to 1.6 times than inlet concentration. This result was similar to those of Takeuchi et al. [1995] and Kim et al. [2003]. The breakthrough time of acetone vapor and acetone vapor in the binary vapor was about 36 min and 70 min, respectively. The breakthrough time of acetone vapor in the binary vapor was faster about twice than that of single acetone vapor. But the breakthrough time of toluene vapor in the binary vapor was similar to that of single toluene vapor. This result could be explained by the fact that the affinity difference between activated carbon and adsorbates, acetone and toluene. The affinity of toluene with activated carbon is bigger than that of acetone at competitive adsorption of acetone and toluene. So it was thought that acetone was substituted with toluene due to the affinity difference during adsorption process. Activated carbon is composed of pure carbons with almost C-C bond and has nonpolarity and hydrophobicity. Therefore, it was thought that adsorption ability of toluene on activated carbon is bigger than that of acetone due to polarity difference between adsorbates, acetone and toluene, and activated carbon.

CONCLUSIONS

Adsorption characteristics according to polarity of acetone and toluene vapors on coconut based activated carbon were investigated by using a fixed bed reactor. In the single vapor adsorption experiments of acetone and toluene, the breakthrough time was decreased as increasing inlet concentration and linear velocity of acetone and toluene vapors. Adsorption capacity was increased according to the increment of inlet concentration, while it kept at constant value regardless of the increment of linear velocity. The equilibrium adsorption capacity of toluene vapor on activated carbon was five times higher than that of acetone vapor because of polarity difference between adsorbent and adsorbate. The breakthrough curves obtained from these experimental results were fairly described by the empirical equation proposed by Yoon and Nelson. Adsorption isotherms of acetone vapor and toluene vapor well satisfied the Langmuir and

Freundlich equations altogether. The breakthrough time of acetone vapor in the binary vapor was fast about twice than breakthrough time of single acetone vapor. Also, the outlet concentration of acetone vapor in the binary vapor passing through the fixed bed reactor increased to 1.6 times than inlet concentration. But the breakthrough time of toluene vapor in the binary vapor was similar to that of single toluene vapor.

The temperature changes in the activated carbon bed during adsorption progress of acetone vapor and toluene vapor occurred in the time range of 10-30 min. The temperature changes for toluene vapor adsorption were very high; however, that for acetone vapor was low. Adsorption heat of acetone vapor on activated carbon was low as a result of small adsorption capacity by different polarity between adsorbate and adsorbent, but adsorption heat of toluene vapor on activated carbon was high as a result of large adsorption capacity by similar polarity between adsorbate and adsorbent.

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