

ADSORPTION OF BINARY AND TERNARY ORGANIC SOLVENT VAPOR IN AIR BY A FIXED BED OF GRANULAR ACTIVATED CARBON

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To obtain design criteria of fixed-bed adsorbers for solvent recovery, experimental breakthrough curves were measured for single-, binary- and ternary-solvent vapor-activated carbon systems and for the same concentration ranges and the same flow rate as those used in industry. Equilibrium relationships for each system were determined from experimental results. It became clear that the binary and ternary adsorption equilibria could be expressed by the Markham-Benton equation. Analyses of the experimental breakthrough curves by the extended MTZ method and by a rather rigorous numerical calculation showed that the intraparticle diffusion of each component was governed by surface diffusion. Furthermore, break times for binary and ternary systems were well predicted by use of the extended MTZ method, with the same accuracy as when the numerical calculation was adopted.

Introduction

Organic solvents have long been used in many industrial processes such as rubber production and magnetic tape production, and the solvents used have been recovered from effluent air mostly by activated carbon adsorption. Since the solvents used are generally a mixture of alcohols, ketones and aromatics, it is necessary to make clear how these solvents are adsorbed on and desorbed from the carbon. Since adsorption equilibria are nonlinear in these cases, it is considered that the most accurate results are given by numerical methods. However, the numerical methods necessitate tedious computational work even for single-component adsorption.¹¹⁾ For binary systems, several analytical methods have been proposed to calculate fixed-bed breakthrough curves, e.g., by Cooney *et al.*²⁾ and by Miura and Hashimoto.⁹⁾ Also, Takeuchi *et al.* reported the extended MTZ method as a simple method of predicting break times in fixed-bed adsorption for water-soluble multicomponent systems.^{12,14,15)} In this paper, as an extension of the method to gas systems, breakthrough curves are shown for the methyl-ethyl ketone (hereafter designated MEK)-toluene vapor system, the toluene-cyclohexanone (hereafter designated as anone) vapor system and the MEK-toluene-anone vapor system, respectively, on a kind of commercial granular activated carbon.

First, the adsorption equilibrium data obtained for each system were compared with those predicted from

single-component adsorption. The mechanism of intraparticle diffusion for binary and ternary systems was examined and break times obtained from the experimental breakthrough curves were compared with those predicted by use of the extended MTZ method and by a rigorous numerical calculation, respectively.

1. Experimental

1.1. Adsorbent

A granular activated carbon, Kurare-coal 3GS-N produced by Kurare Chem. Co. Ltd., was chosen as adsorbent and its 16- to 20-mesh fraction was used after crushing. The mean particle diameter of the sample was determined by screening to be 9.165×10^{-4} m. The particles were pretreated in a nitrogen stream at 413 K for more than 5 hours to remove moisture.

Physical properties of the particles were measured according to conventional methods. The surface area determined from a nitrogen adsorption method was 9.7×10^5 m²/kg, and the particle density was determined to be 730 kg/m³ from a simple calculation using the values of true density, determined to be 2020 kg/m³ by a *n*-butanol displacement method and from the values of pore volume measured by a mercury porosimeter.

1.2. Experimental procedure

A schematic diagram of the experimental apparatus is shown in Fig. 1. Air from a blower was fed to a silica gel column to dry the air. A part of the pretreated air was fed to evaporators to load objective solvents and was mixed with the pretreated air stream. The

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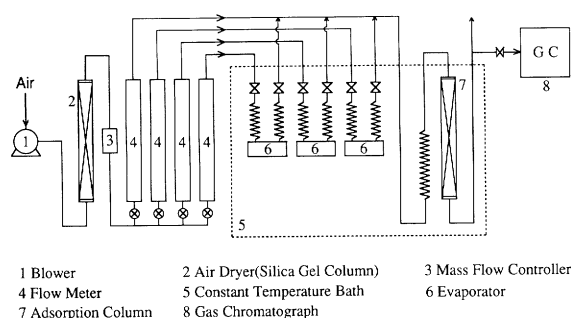


Fig. 1. Schematic diagram of experimental apparatus

solvent-laden air thus obtained, with constant concentrations, constant flow rate and constant temperature, was fed to a fixed bed of granular activated carbon. Then the concentrations history at the exit of the carbon bed was measured by a gas chromatograph (Shimadzu GC-9A) equipped with a flame ionization detector (FID).

The granular activated carbon was packed in a glass column of 0.016 m i.d. to a height of from 0.04 to 0.3 m. The respective solvents used as adsorbates were MEK, toluene and anone of specific grade. The flow rate of the feed air was set to a certain value by a mass flow controller. The temperature was maintained usually at 308.2 K by a constant-temperature bath. Experiments were performed under the following conditions; linear flow rate of from 0.343 to 0.427 m/s, influent concentration of vapors from 500 to 3000 ppm (vol.) and bed density of about 450 kg/m³.

A small amount of effluent air was taken into a micro-syringe at suitable intervals and was analyzed by a gas chromatograph (Shimadzu GC-9A). When the effluent concentration of most-adsorbable component of a binary or ternary system arrived at the influent concentration, measurement of effluent air was stopped.

The amount adsorbed of solvent vapors in equilibrium with various initial concentrations was determined from graphical integration of the experimental breakthrough curves and from the weight increase of the carbon bed. Experimental data were used for subsequent analysis when the relative error of the values of total amount adsorbed obtained from the above two methods was less than 5%.

2. Results and Discussion

2.1. Adsorption equilibria

Adsorption isotherms for each single-solvent vapor are shown in Fig. 2. It became clear that the adsorbability of the three adsorbates to carbon was in the order of anone, toluene and MEK. The adsorption equilibrium data for single-component systems could be expressed by the Langmuir equation described by Eq. (1) and the two constants, K and

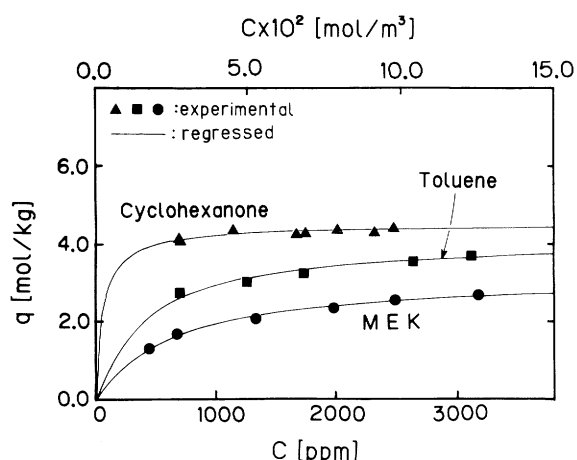


Fig. 2. Adsorption isotherms for single-component solvent vapor-activated carbon systems (308.2 K)

Table 1. Langmuir constants

	$q_{inf.}$ [mol/kg]	Single K [m ³ /mol]	Binary K_i [m ³ /mol]	Ternary K_i [m ³ /mol]
MEK	3.233	36.06	18.38*	25.28
Toluene	4.150	58.16	101.0*	144.2**
Anone	4.466	405.1	463.0**	404.1

* MEK-Toluene system ** Toluene-Anone system

$q_{inf.}$, were determined by regression of the experimental data, as listed in Table 1.

$$q = q_{inf.} K C / (1 + K C) \quad (1)$$

Theoretically, the values of $q_{inf.}$ expressed in terms of moles per unit weight of adsorbent, mol/kg, should be the same irrespective of adsorbates when the molecular sizes of the adsorbates are the same. Because of the small difference in their molecular sizes, however, the values of $q_{inf.}$ were a little different from one another, as can be seen from the table. Consequently, it was assumed that $q_{inf.}$ for each component was the same for each adsorbate irrespective of the presence of other adsorbates, and the langmuir type or Markham-Benton equation as shown in Eq. (2) was applied to binary and ternary systems.

$$q_i = q_{inf.} K_i C_i / (1 + \sum K_j C_j) \quad (2)$$

where values of the constant K_i were selected to be a set of K_i giving minimum error between the values of experimental amount adsorbed and those calculated,¹³⁾ as listed in Table 1, because multicomponent adsorption included the process which the less-adsorbable component is replaced by the more-adsorbable one, and the ratios of K_i determined for binary and ternary systems were different from those obtained for single-component systems. The most

Table 2. Comparison of equilibrium concentrations obtained from experimental breakthrough curves with those estimated by the extended MTZ method

Run No.	C_{01}	$C_{02} \times 10^2$ [mol/m ³]	C_{03}	$C_{e1}^* \times 10^2$ [mol/m ³]	C_{e1}^{**}	Error [%]	$C_{e2}^* \times 10^2$ [mol/m ³]	C_{e2}^{**}	Error [%]
1	7.3	5.3		11.06	12.58	9.0			
2	7.0	6.2		11.24	11.67	3.8			
3	5.9	2.6		7.71	8.35	8.3			
4	8.8	4.1		11.75	12.34	5.0			
5	9.5	9.8		16.53	17.09	3.4			
6	1.0	3.9		14.03	14.52	3.5			
7	15.4	5.4		19.80	19.78	0.1			
8	7.3	4.9		11.09	10.76	3.0			
9	11.0	6.1		16.01	15.74	1.7			
10	8.5	3.4		11.07	11.23	1.5			
11	6.1	5.3	6.5	15.03	13.86	7.8	10.09	10.47	3.8
12	4.6	3.7	5.1	10.28	10.17	1.1	7.28	7.69	5.6
13	4.0	4.0	9.7	12.26	12.54	2.3	11.07	12.06	8.9
14	9.0	5.1	9.3	17.76	18.77	5.7	10.92	12.58	15.2
15	12.7	10.9	6.4	25.95	25.23	2.8	15.35	15.98	4.1

* From experimental breakthrough curves

** By the extended MTZ method

suitable K_i values obtained for the system are listed in Table 1.

2.2. Equilibrium concentrations

For a ternary adsorption, the distribution of the amount adsorbed and the concentration of each component in a fixed bed can be shown schematically as seen in Fig. 3.¹⁵⁾ Here, three components are designated as Components 1, 2 and 3 in the order of their appearance at the exit of the bed. Details of the calculation of the lengths or concentrations of each zone are presented elsewhere.^{10,12,14,*)} To estimate the equilibrium concentrations in Zone III for a ternary system by use of the extended MTZ method, the procedure is a little complicated. However, since the values of equilibrium concentration of Component 1 in Zone III (C'_{e1}) were always almost the same as that of the initial concentration (C_{01}), the equilibrium concentration of Component 2 in Zone III (C_{e2}) was estimated assuming the amount adsorbed of the first, i.e. the least-adsorbable, component (q'_{e1}) was the same as that in Zone I (q_{01}). In short, the distribution of the first component in Zone II and that in Zone III were assumed to be shown by the chained-line in Fig. 3. The results of equilibrium concentrations estimated for binary and ternary systems by the extended MTZ method are listed in Table 2. The concentrations C_{e1}

* Equations obtained by solving Eqs. (7-68) and (7-69) in literature reference No. 10 are incorrect and should be written as follows.

$$C_{1e} = \frac{-p + \sqrt{p^2 + 4g_1(1-d)}}{2K_1}, \quad p = 1 - g_1 - d(1 + g_2),$$

$$g_1 = K_1 C_{10}, \quad g_2 = K_2 C_{20} \quad \text{and} \quad d = \frac{K_1 q_{1\infty}}{K_2 q_{2\infty}}$$

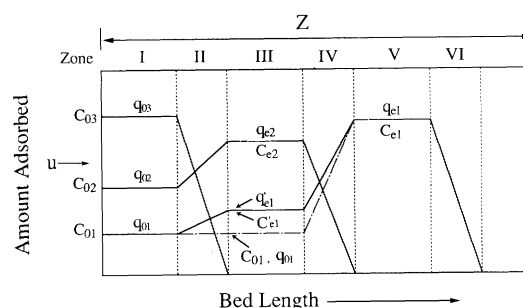


Fig. 3. Schematic diagram of distribution of amount adsorbed in a fixed bed for the ternary system

estimated for the two binary systems agreed well with experimental results. For the ternary system, however, the values of equilibrium concentrations C_{e2} estimated for Component 2 (toluene) were a little larger than the experimental values. This disagreement must arise from the assumption of $C'_{e1} \approx C_{01}$ in Zone III. As the difference between the two values was less than about 10% in terms of relative error, it was thought that a serious error in further calculation of break times might not occur.

2.3. Intraparticle diffusion

To predict break times by use of the extended MTZ method, adsorption zones for each component must be estimated using values of the intraparticle mass transfer coefficient. Regarding binary and ternary systems, the intraparticle mass transfer coefficient can be obtained by a simple method described elsewhere.^{14,15)} The procedure is as follows. When each apparent adsorption isotherm^{12,14)} which can be obtained by use of the extended MTZ method introducing the Markham-Benton equation is ex-

pressed by the Langmuir equation, and two constants, K and q_{inf} , corrected to fit the data, are determined, the number of transfer units can be calculated by use of these constants. The values of intraparticle mass transfer coefficients for each component were determined by use of $r-\zeta$ method by Kawazoe.⁸⁾

The values thus obtained for the intraparticle mass transfer coefficients of the components and for single, binary and ternary systems are shown in Figs. 4 to 6 against the slope of the operating line. The broken lines in the figures denote average values of the coefficient for the solvents in the respective binary systems. Regarding the binary systems, toluene was the more-adsorbable component in the MEK-toluene system but was the less-adsorbable component in the toluene-anone system. However, the values of the intraparticle mass transfer coefficient for toluene were always almost the same irrespective of the presence of another component. Furthermore, it became clear that the values of intraparticle mass transfer coefficient for the ternary system were not so different from those for single and binary adsorption, though the values for toluene in the ternary system seemed to be a little larger than those for the binary systems, probably because the estimated equilibrium concentrations of toluene for the ternary system were a little larger than those obtained from the experiments and, as a result, smaller No_s gave larger $k_s a_v$ for the same condition of Zone III.

Next, comparison of the above results with those obtained from curve-fitting of experimental breakthrough curves to those from numerical calculation was performed.^{1,5)} The effect of axial dispersion was considered to be negligible, because a particle Reynolds number (Re_p) of about 20 was used throughout the study. Therefore, the following equation can be given.

Material balance for an infinitely small bed:

$$u \left(\frac{\partial \bar{C}}{\partial Z} \right) + \gamma \left(\frac{\partial \bar{q}}{\partial t} \right) + \varepsilon \left(\frac{\partial \bar{C}}{\partial t} \right) = 0 \quad (3)$$

Rate equations are described as follows.

For fluid film transfer:

$$\gamma \left(\frac{\partial \bar{q}}{\partial t} \right) = k_f a_v (\bar{C} - C_s) \quad (4)$$

At the solid-fluid interface:

$$\gamma \left(\frac{\partial \bar{q}}{\partial t} \right) = -D_s \rho_s a_v \left(\frac{\partial q}{\partial r} \right)_{r=b} \quad (5)$$

For intraparticle diffusion, surface diffusion is assumed to be predominant:

$$\left(\frac{\partial q}{\partial t} \right) = D_s \left(\frac{\partial^2 q}{\partial r^2} + \frac{2}{r} \frac{\partial q}{\partial r} \right) \quad (6)$$

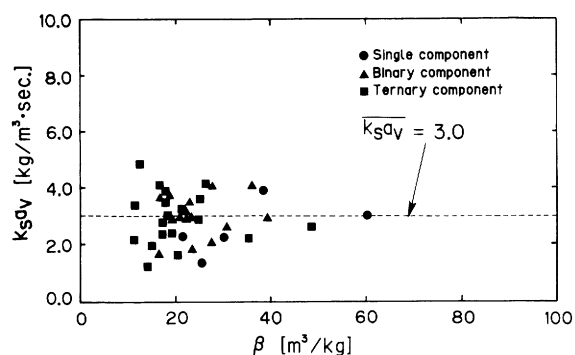


Fig. 4. Dependence of $k_s a_v$ on β for MEK

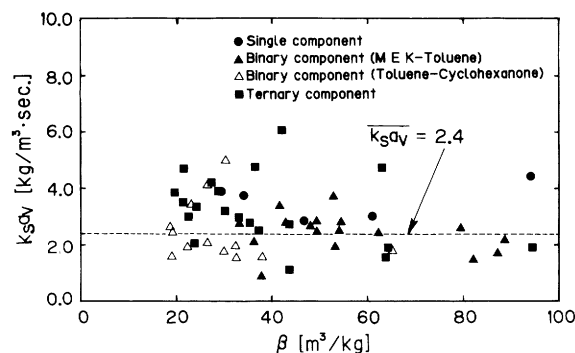


Fig. 5. Dependence of $k_s a_v$ on β for Toluene

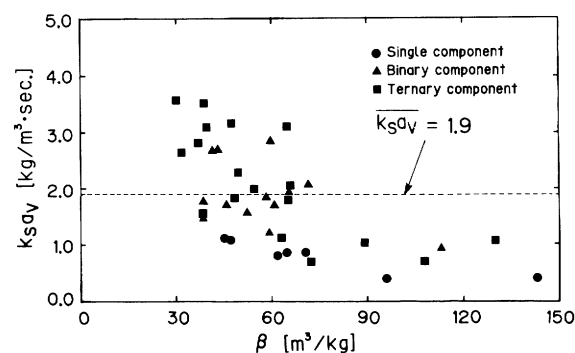


Fig. 6. Dependence of $k_s a_v$ on β for Anone

As the adsorption isotherm is expressed by Eq. (2), the calculation of breakthrough curves can be performed in the same way as described elsewhere.⁵⁾ The above equations are rewritten in dimensionless form and finite-difference equations were derived to replace Eqs. (3) to (6). Integration of Eqs. (3) to (6) was carried out numerically by use of the successive over-relaxation (S.O.R.) method, which is an implicit method for partial differential equations. Comparison of an experimental result with numerical breakthrough curves for the ternary system is shown in Fig. 7. For this system the calculated curves fitted well to the experimental ones. It can be seen that the intraparticle diffusion is determined by surface diffusion⁷⁾ except for anone since the values of intraparticle diffusivities are almost constant irrespective of the adsorption

coefficient, β ($=q_0/C_0$). **Table 3** shows a comparison of the values of intraparticle diffusivity calculated from the extended MTZ method and by use of Glueckauf's equation,⁶⁾ Eq (7), with those obtained from the numerical calculation.

$$k_s a_v = 60 \rho_s (1 - \epsilon) D_s / dp^2 \quad (7)$$

Both sets of values agreed fairly well, except the values for MEK (the first component), which might be obtained with the greatest error due to poor estimation of the isotherm.

2.4. Prediction of break times

By use of the values of optimal $k_s a_v$ for the ternary system determined by the extended MTZ method and the average values of $k_s a_v$ determined using the method for binary systems, break times for binary and ternary systems were calculated as shown in **Table 4**. Equations included in the method are presented elsewhere.¹²⁾ Break times were taken to be the times when the concentration of each component arrived at

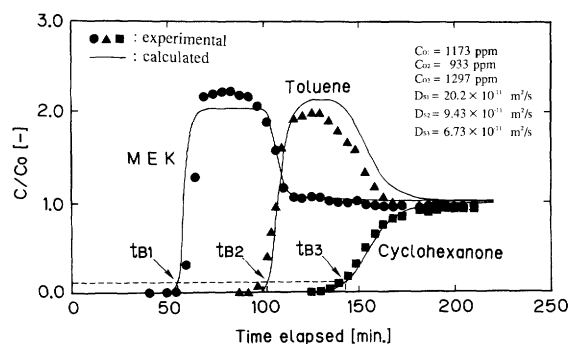


Fig. 7. Comparison between experimental and calculated breakthrough curves for the ternary system

a certain values of the initial concentration C_{0i} , e.g., one-tenth. As the break times can also be predicted from the numerical calculations, these results are also shown in Table 4. The three sets of values agreed well with one another. This agreement seems to be due to the conditions that the values of $k_s a_v$ for ternary system are taken to be the same as those for the binary system and that, as listed in **Table 5**, the lengths of the adsorption zone and of the displacental one are much shorter than the experimental bed lengths, which

Table 3. Comparison of intraparticle diffusivity calculated by the procedure included in the extended MTZ method with those obtained by numerical calculation

Run No.	$D_{s1} \times 10^{11}$ [m ² /s] (A)	$D_{s1} \times 10^{11}$ [m ² /s] (B)	$D_{s2} \times 10^{11}$ [m ² /s] (A)	$D_{s2} \times 10^{11}$ [m ² /s] (B)	$D_{s3} \times 10^{11}$ [m ² /s] (A)	$D_{s3} \times 10^{11}$ [m ² /s] (B)
1	9.61	19.5	8.31	6.51	MEK-Toluene system	
2	11.7	20.0	9.48	7.65		
3	8.83	20.3	6.59	6.75		
4	6.04	13.3	8.23	6.62		
5	11.9	16.7	8.05	8.28		
6	6.55	6.58	8.94	9.09	Toluene-Anone system	
7	8.03	8.20	8.94	9.09		
8	6.54	6.59	5.64	6.59		
9	11.6	10.2	5.87	10.2		
10	5.38	6.83	5.04	8.54		
11	7.87	19.6	9.69	8.18	7.47	7.46
12	9.67	20.2	9.29	9.43	3.80	6.73
13	10.7	19.7	10.5	8.20	5.09	6.56
14	7.39	22.8	15.9	9.50	10.8	10.6
15	11.4	20.2	15.8	8.41	11.9	11.8

(A): By the extended MTZ method and by use of Glueckauf's equation

(B): From numerical calculation

Table 4. Comparison of break times obtained from experimental breakthrough curves with those predicted by the extended MTZ method and by numerical calculation

Run No.	t_{B1} [min]				t_{B2} [min]				t_{B3} [min]			
	exp.	calc. 1	calc. 2	calc. 3	exp.	calc. 1	calc. 2	calc. 3	exp.	calc. 1	calc. 2	calc. 3
1	48.0	42.2	36.7	42.3	103.4	99.7	99.6	100.7	MEK-Toluene system			
2	26.3	27.1	22.9	26.4	53.6	58.7	57.9	58.1				
3	63.4	68.3	57.2	69.0	180.8	198.2	200.1	204.7				
4	27.4	29.1	36.4	29.2	78.8	82.6	91.1	86.5				
5	25.1	26.1	24.9	25.4	48.1	51.7	81.7	50.6				
6	40.8	43.7	47.2	44.4	87.6	95.7	96.2	100.7	Toluene-Anone system			
7	27.5	29.1	31.3	29.0	61.2	59.6	61.5	64.6				
8	39.8	43.1	46.7	44.1	63.5	75.4	78.4	80.1				
9	38.4	39.6	40.9	38.3	70.3	66.7	73.4	72.9				
10	55.0	53.2	59.3	55.8	116.7	114.7	116.8	122.1				
11	42.2	40.6	40.7	41.6	68.7	72.1	72.7	71.0	95.6	102.5	103.5	100.4
12	58.1	57.0	55.3	57.2	99.0	101.4	102.7	100.6	139.4	133.3	143.4	141.4
13	87.3	95.0	92.2	94.7	125.6	136.5	136.2	135.1	155.9	164.9	169.6	167.4
14	33.6	29.5	30.9	31.4	63.6	59.4	58.3	57.1	85.2	77.2	77.6	73.9
15	20.5	21.0	21.0	20.6	37.5	41.4	40.6	38.4	71.9	68.6	69.3	60.8

calc. 1: The optimal $k_s a_v$ was used from the extended MTZ method

calc. 2: The optimal $k_s a_v$ was used from numerical calculation

calc. 3: Average values of $k_s a_v$ for binary systems was used from the extended MTZ method

Table 5. Lengths of the adsorption zone and the displacement zone for ternary system

Run No.	$Z \times 10^2$ [m]	$Z_{min} \times 10^2$ [m]	$Z_{II} \times 10^2$ [m]		$Z_{IV} \times 10^2$ [m]		$Z_{VI} \times 10^2$ [m]	
			obs.	calc.	obs.	calc.	obs.	calc.
11	12.60	7.95	2.94	3.41	1.93	2.31	2.97	2.42
12	13.60	7.14	4.10	2.69	1.71	1.91	2.20	2.12
13	24.80	5.50	4.14	3.48	1.78	2.28	2.11	2.28
14	13.00	6.76	2.67	3.65	1.66	2.61	3.99	2.65
15	10.40	18.5	3.21	5.21	1.76	3.15	2.84	3.19

Z_{min} : Calculated by equations^{3,4,12)} introducing the optimal values of $k_s a_v$ for ternary system determined from numerical calculation
 calc.: By use of the r - ζ method introducing the average values of $k_s a_v$ determined for binary systems

are longer than the minimum one, Z_{min} , satisfied to separate adsorption zones from each other and to exhibit breakthrough curves of constant pattern. The equations used to calculate the minimum bed length, Z_{min} , and details of procedure are presented elsewhere.^{3,4,12)} The calculated minimum bed length is also listed in Table 5. Consequently, when the experimental bed length is taken to be long enough, such as calculated Z_{min} , the deviation in the intraparticle mass transfer coefficient does not affect so much the calculation of the break times, and the application of the extended MTZ method is valid.

Conclusions

Regarding solvent recovery, binary and ternary systems of solvent-laden air containing MEK, toluene and cyclohexanone was fed to a fixed bed of granular activated carbon bed of sufficient length to fulfill constant pattern conditions, and equilibria as well as rate data were obtained. The following conclusions were obtained from the results.

1. Adsorption equilibria for binary and ternary systems can be expressed by the Markham–Benton equation, with a small modification of constant values, K_i obtained from single-component systems, to give best fit to the binary and ternary experimental data.

2. The concentrations at equilibrium-attained zones can be estimated well by use of the extended MTZ method with the Markham–Benton equation determined for each system.

3. The values of the intraparticle mass transfer coefficient for toluene were almost the same as those for single component irrespective of the presence of other components. Also, the values of the intraparticle mass transfer coefficient for each component in the ternary system were almost the same as those for the binary systems.

4. Prediction of the break times for binary and ternary systems by the extended MTZ method was found to be sufficiently accurate in comparison to those obtained by numerical calculation.

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Nomenclature

a_v	= Specific surface area in the bed	[m ² /m ³]
b	= Radius of granular activated carbon	[m]
C	= Concentration of solvent vapor in air	[mol/m ³]
C_{oi}	= Influent concentration (constant)	[mol/m ³]
C_{ei}	= Constant concentration at equilibrium-attained zones	[mol/m ³]
C_s	= Concentration at fluid-solid interface	[mol/m ³]
D_{si}	= Intraparticle diffusivity	[m ² /s]
K	= Langmuir constant	[m ³ /mol]
$k_f a_v$	= Fluid film volumetric mass transfer coefficient	[1/s]
$k_s a_v$	= Intraparticle volumetric mass transfer coefficient based on the difference in amount adsorbed	[kg/m ³ ·s]
No_s	= Number of transfer units based on amount adsorbed	[—]
q	= Amount of solvent vapor adsorbed	[mol/kg]
$q_{inf.}$	= Amount adsorbed at monolayer saturation	[mol/kg]
q_{oi}	= Amount adsorbed in equilibrium with C_{oi}	[mol/kg]
q_{ei}	= Amount adsorbed in equilibrium with C_{ei}	[mol/kg]
q'_{e1}	= Amount adsorbed in equilibrium with C'_{e1}	[mol/kg]
r	= Radial distance in spherical particle	[m]
t	= Time elapsed	[s] or [min]
t_{Bi}	= Break time with respect to i -component	[s] or [min]
u	= Linear flow rate of gas	[m/s]
Z	= Bed length	[m]
Z_i	= Length of Zone i	[m]
β	= Adsorption coefficient (slope of operating line)	[m ³ /kg]
γ	= Bed bulk density	[kg/m ³]
ε	= Void fraction of the bed	[—]
ρ_s	= Particle density	[kg/m ³]

<Subscript>

i = Refer to component or zone

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