

# INTRAPARTICLE DIFFUSIVITIES IN LIQUID-PHASE ADSORPTION WITH NONLINEAR ISOTHERMS

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Diagrams showing concentration change in an agitated tank adsorber are presented to estimate intraparticle diffusivities under nonlinear isotherms, where either pore or surface diffusion is assumed to be controlling. The adsorption kinetics of DBS and phenol from dilute aqueous solutions onto two kinds of activated carbon are measured, and the intraparticle diffusivities are estimated with diagrams. An approximation method for the case where surface diffusion is controlling, which is a modification of the Dryden and Kay method, is also proposed. This method gives good agreement with the diagrams obtained numerically.

## Introduction

Recently liquid-phase adsorption processes using activated carbon have been successfully applied to advanced waste-water treatment. For rational design of fixed-bed adsorbers, which are mostly employed in waste-water treatment, the resistances of both external mass transfer and intraparticle diffusion have to be evaluated. The mass transfer resistance at the external surface of the adsorbent may be easily calculated by means of correlations of the interphase mass transfer coefficients in fixed-beds<sup>2)</sup>. Although the mechanism of intraparticle mass transfer in the liquid-phase adsorption has not been clarified, pore diffusion and surface diffusion are considered to be possible mechanisms<sup>4-6)</sup>.

Methods have been presented for determining intraparticle diffusivities under linear adsorption isotherms<sup>1,3)</sup>, and recently Suzuki and Kawazoe<sup>11)</sup> have proposed a method for estimating the pore diffusivities for a rectangular isotherm system by analyzing the concentration change in an agitated tank. However, the isotherm which is usually encountered in waste water adsorption processes may be represented by a nonlinear equation such as the Freundlich or the Langmuir equation.

Dryden and Kay<sup>9)</sup> have proposed a method which approximates a nonlinear isotherm by a linear equation for the case where surface diffusion is controlling. Smith *et al.*<sup>10)</sup> modified this method by introducing adjustable parameters which are experimentally determined, but no clear meanings were given for these adjustable parameters.

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The purpose of this paper is to present a method for estimating intraparticle diffusivities for nonlinear isotherm systems where either pore or surface diffusion is assumed to be controlling. The Freundlich and the Langmuir isotherms were employed as the nonlinear isotherm. (A similar method was presented by Suzuki and Kawazoe<sup>12)</sup> for the Freundlich isotherm at the same time that a part of this paper was presented.) Furthermore, for the case where surface diffusion is controlling, a modification of the Dryden and Kay method is developed.

To examine the validity of the two methods presented in this work, the kinetics of the adsorption of DBS and phenol from dilute aqueous solutions onto two kinds of activated carbon in an agitated tank were studied, and the intraparticle diffusivities were estimated.

## Basic Equations

The driving force for pore diffusion is the concentration gradient of the adsorbate in the pore space, whereas that for surface diffusion may be represented in terms of the gradient of the amount of adsorbate on the pore walls. Thus the flux for each diffusion mechanism can be written as follows:

$$\text{Pore diffusion: } N_p = -D_p(\partial c/\partial r) \quad (1)$$

$$\text{Surface diffusion } N_s = -D_s\rho(\partial q/\partial r) \quad (2)$$

where  $\rho$  is the particle density, and  $D_p$  and  $D_s$  are the pore diffusivity and surface diffusivity, respectively.

The following assumptions are made to develop the basic equations for both surface and pore diffusion controlling:

- (1) Adsorbent particles are spherical, of radius  $R$ .
- (2) The pore structure inside the particle is uniform.
- (3) The diffusivity is constant.

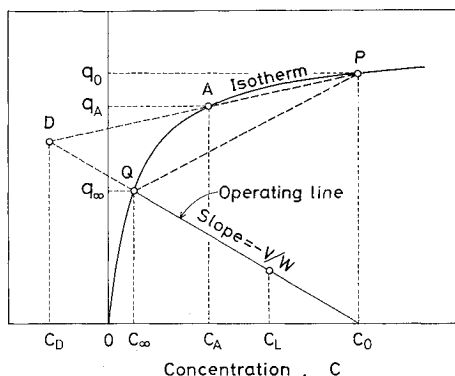


Fig. 1 Isotherm and operating line

- (4) The mass transfer resistance at the particle surface is negligible.
- (5) The rate of adsorption is much faster than the rate of diffusion, and hence local equilibrium is maintained between the adsorbate in the pore space and that on the pore walls.

The basic equations are represented in the following dimensionless forms:

**Case (a)** Surface diffusion controlling

$$\frac{\partial z}{\partial \theta} = \frac{\partial^2 z}{\partial x^2} + \frac{2}{x} \frac{\partial z}{\partial x} \quad (3)$$

Boundary and initial conditions are:

$$z=0 \quad \text{for } \theta=0, 0 \leq x < 1 \quad (4)$$

$$z=1 \quad \text{at } \theta=0, x=1 \quad (5)$$

$$\partial z / \partial x = 0 \quad \text{for } x=0, \theta > 0 \quad (6)$$

$$\frac{\partial z}{\partial \theta} = -3 \frac{W}{V} \left( \frac{dq}{dc} \right) \frac{\partial z}{\partial x} \quad \text{for } x=1, \theta > 0 \quad (7)$$

**Case (b)** Pore diffusion controlling

$$\frac{C_0}{q_0} \left( \frac{dq}{dc} \right) \frac{\partial y}{\partial \tau} = \frac{\partial^2 y}{\partial x^2} + \frac{2}{x} \frac{\partial y}{\partial x} \quad (8)$$

Boundary and initial conditions are:

$$y=0 \quad \text{for } \tau=0, 0 \leq x < 1 \quad (9)$$

$$y=1 \quad \text{at } \tau=0, x=1 \quad (10)$$

$$\partial y / \partial x = 0 \quad \text{for } x=0, \tau > 0 \quad (11)$$

$$\frac{\partial y}{\partial \tau} = -3 \frac{W}{V} \left( \frac{q_0}{C_0} \right) \frac{\partial y}{\partial x} \quad \text{for } x=1, \tau > 0 \quad (12)$$

where  $z=q/q_0$ ,  $y=c/C_0$ ,  $x=r/R$ ,  $\theta=(D_p/R^2)t$ ,  $\tau=(C_0/\rho q_0)(D_p/R^2)t$ . These are all dimensionless groups.

The great difference between case (a) and case (b) is that the nonlinear term  $(dq/dc)$ , which represents the slope of an equilibrium curve, is involved in the boundary condition (7) for case (a), whereas it is involved in the basic equation itself for case (b). As a result, the mathematical treatment of case (a) is less difficult than that of case (b).

The equilibrium curves for the Freundlich and the Langmuir isotherms are represented by the following dimensionless equations:

$$\text{Freundlich: } z=y^\beta \quad (13)$$

$$\text{Langmuir: } z=y/[K+(1-K)y] \quad (14a)$$

where  $K$  is called the separation factor and is defined as:

$$K=1/(1+kC_0) \quad (14b)$$

The term  $(dq/dc)$  in the basic equations can be calculated from Eq. (13) or (14). Then Eqs. (3) to (12) may be solved numerically when  $C_\infty/C_0$ , the ratio of the equilibrium concentration to the initial concentration, and  $\beta$  or  $K$  are given. When surface diffusion is controlling, an approximate method is presented which is a modification of the Dryden and Kay method<sup>9)</sup>.

### Numerical Solution

Equations (3) to (12) were solved numerically on a computer using the so-called Saul'yev average method<sup>7)</sup>. This is a stable numerical technique for the integration of parabolic partial differential equations and has the features of the implicit and explicit methods.

The solutions  $z(x,\theta)$  (or  $z(x,\tau)$ ) were integrated over the sphere volume by Simpson's rule to obtain the average amount adsorbed  $q(\theta)$  (or  $q(\tau)$ ), and the fractional approach to equilibrium,  $E=q(\theta)/q_\infty$ , was calculated for each time level.

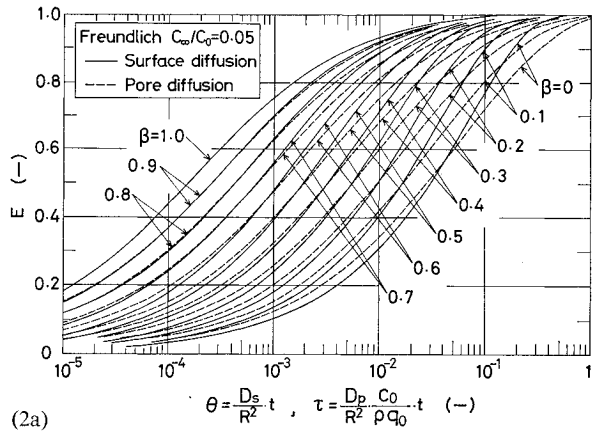
As the term  $(dq/dc)$  becomes infinite at  $c=0$  for the Freundlich isotherm, Eq. (8) cannot be solved under B.C. (9). To overcome this difficulty the isotherm was approximated over the range  $0 \sim y_1$ , where  $y_1$  was chosen close to zero, by the following Langmuir isotherm having a finite slope at  $y=0$ :

$$z = \frac{y_1^{\beta-1} y}{\beta + (1-\beta)(y/y_1)} \quad (15)$$

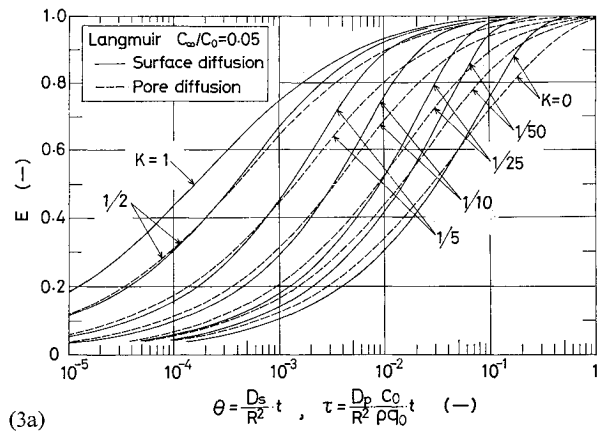
The above equation was determined so that both the absolute value and the derivative of the Langmuir equation at  $y_1$  coincide with those of the Freundlich isotherm. If values of  $y_1$  less than  $10^{-10}$  were chosen, no significant errors were found in the calculation of  $E(\tau)$ , and  $y_1=10^{-10}$  was employed for the computation.

The results for the Freundlich isotherm are shown graphically in Figs. 2a~2d; where the ordinate is  $E$ , the abscissa is  $\theta$  or  $\tau$ . Here the values of  $C_\infty/C_0$  are taken as 0.05, 0.1, 0.2 and 0.5. Similar plots for the Langmuir equation are given in Figs. 3a~3d. In all Figures, the solid lines show surface diffusion controlling, and the dotted lines represent pore diffusion controlling. The discrete parameter for the Freundlich equation is  $\beta$ , and that for the Langmuir equation is  $K$ .

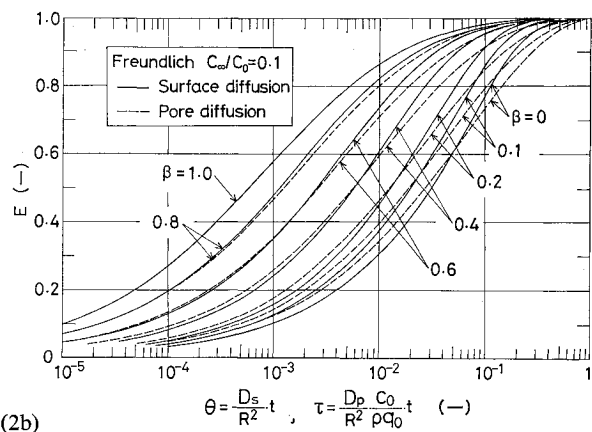
The case where  $\beta$  or  $K$  equals 1.0 corresponds to a linear isotherm. In this case the following analytical solution derived for an unsteady heat transfer problem<sup>8)</sup> is usable, because the basic equation for this



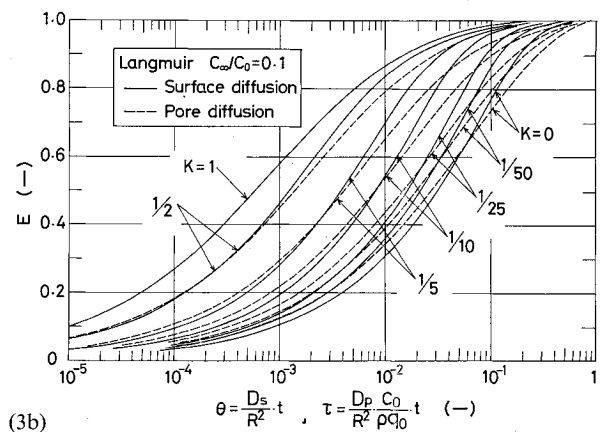
(2a)



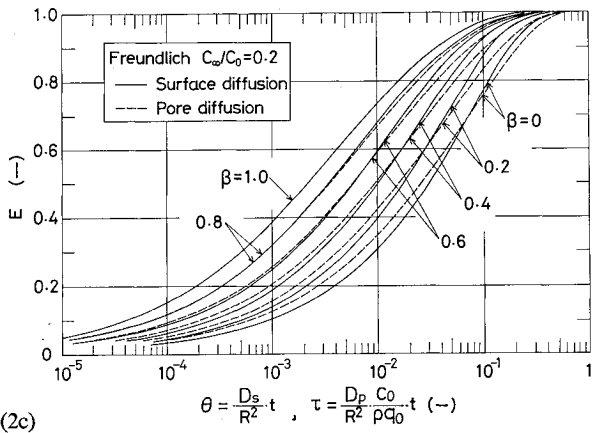
(3a)



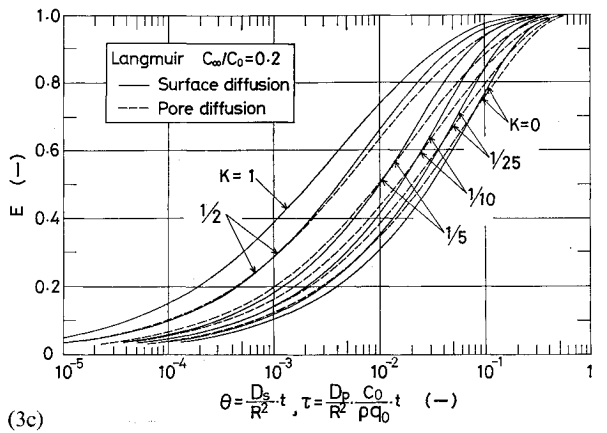
(2b)



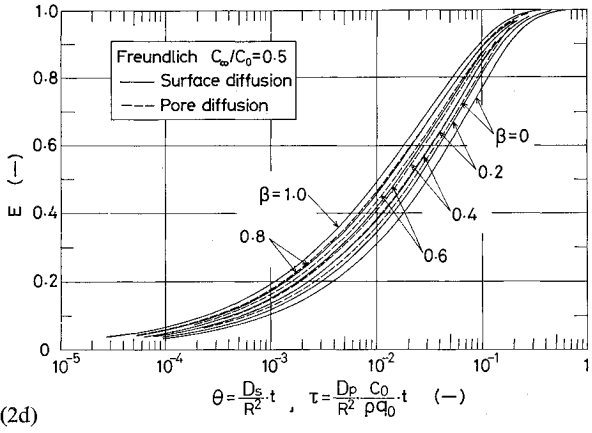
(3b)



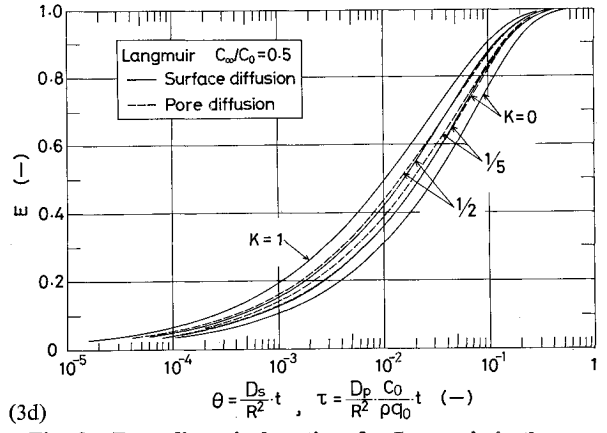
(2c)



(3c)



(2d)



(3d)

Fig. 2 *E* vs. dimensionless time for Freundlich isotherms

Fig. 3 *E* vs. dimensionless time for Langmuir isotherms

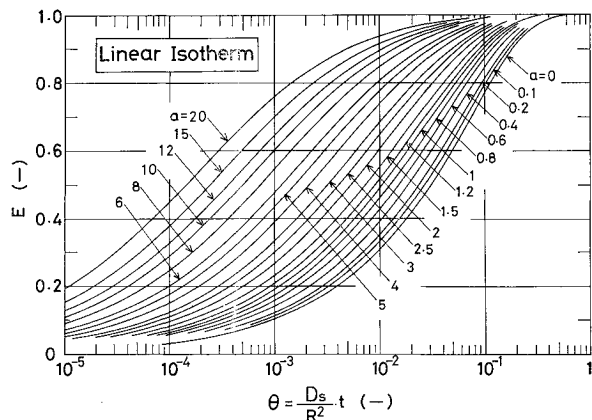


Fig. 4  $E$  vs. dimensionless time for linear isotherms

diffusion problem is analogous to that for the heat transfer problem.

$$E = 1 - (2/3a) \sum_{n=1}^{\infty} \frac{\exp(-s_n^2 \theta)}{1 + s_n^2 / [9a(1+a)]} \quad (16)$$

where  $s_n$  is the  $n$ -th positive root of the equation

$$s \cot s = 1 + s/3a \quad (17)$$

and  $a$  is given as

$$a = \frac{1 - C_{\infty}/C_0}{C_{\infty}/C_0} \quad (18)$$

When  $a=0$ , Eq. (16) becomes a well-known equation:

$$E = 1 - \left( \frac{6}{\pi^2} \right) \sum_{n=1}^{\infty} \frac{\exp(-n^2 \pi^2 \theta)}{n^2} \quad (19)$$

The curves of  $\beta=1$  in Figs. 2a~2d and those of  $K=1$  in Figs. 3a~3d were computed by means of Eq. (16). Figure 4 also indicates  $E$ - $\theta$  curves for the linear isotherms computed using Eqs. (16) and (19) varying the parameter  $a$  from 0 to 20.

On the other hand, the case of  $\beta=0$  or  $K=0$  represents a rectangular isotherm. In this case the solutions given by Suzuki and Kawazoe<sup>11)</sup> and by Eq. (19) are applied directly for pore diffusion controlling and for surface diffusion controlling, respectively.

Figures 2a to 3d are utilized to estimate the intraparticle diffusivity in the following way:

- (1) Obtain the value of  $\beta$  or  $K$  from the isotherm.
- (2) Plot the experimental  $E$  values against  $t$ .
- (3) Read  $\theta$  or  $\tau$  for a number of selected  $E$  values from the corresponding diagram.
- (4) Plot the respective  $\theta$  or  $\tau$  values against the observed time  $t$  for the selected  $E$  values. Either the plot of  $\theta$  vs.  $t$  or the plot of  $\tau$  vs.  $t$  may give a straight line through the origin.  $D_s$  or  $D_p$  is determined from the slope using the following relation:

$$D_s = (\text{slope}) \times R^2 \quad (20)$$

$$D_p = (\text{slope}) \times R^2 \times (\rho q_0 / C_0) \quad (21)$$

## A Modified Dryden and Kay (D-K) Method

As mentioned above, the nonlinear term ( $dq/dc$ ) appears only in B.C. (7) for the case where surface diffusion is controlling. This means the equilibrium relation is necessary only at the outer surface of adsorbent particles.

Dryden and Kay approximated the curved isotherm  $\widehat{PQ}$  by the straight line  $\overline{PQ}$  as shown in Fig. 1. Then the term ( $dq/dc$ ) reduces to a constant value,  $(q_0 - q_{\infty}) / (C_0 - C_{\infty})$ . This approximation enables us to apply Eq. (16). In this case the parameter  $a$  must be interpreted by the following expression<sup>3)</sup>:

$$a = W(q_0 - q_{\infty}) / V(C_0 - C_{\infty}) \quad (22)$$

This approximation may not be successful when the equilibrium point Q is distant from P. To overcome this weakness, we chose a point A between P and Q, and approximated the isotherm curve by  $\widehat{PA}$ . This may improve the original D-K method, at least while the concentration in the vessel changes from  $C_0$  to  $C_A$ . In this case, to obtain the  $E$  values from Fig. 4, the parameter  $a$  must be replaced with  $b$ , defined by the following equation:

$$b = W(q_0 - q_A) / V(C_0 - C_A) \quad (23)$$

The values of  $E$  determined above are represented by  $E_D$ . However, the  $E_D$  values cannot be compared directly with the experimental data, because they are obtained regarding point D as a pseudo-equilibrium point, that is, they give the values of  $(C_0 - C_L) / (C_0 - C_D)$ . Here  $C_L$  is the concentration in the vessel at time  $t$ . Since the true fractional approach to equilibrium is given as  $E = (C_0 - C_L) / (C_0 - C_{\infty})$ ,  $E_D$  must be converted to  $E$  using the relation:

$$E = \frac{1+a}{1+b} E_D \quad (24)$$

Equation (24) is rewritten as

$$E = \frac{q_0 E_A}{q_{\infty} E_A + (q_0 - q_A)} E_D \quad (25)$$

where  $E_A = (C_0 - C_A) / (C_0 - C_{\infty})$ .

To apply the modified D-K method to the experimental data, the value of  $E_A$  must be chosen first so that a part of the isotherm,  $\widehat{PA}$ , in Fig. 1 may be represented approximately by a straight line,  $\overline{PA}$ . Usually the appropriate values of  $E_A$  are found to be 0.5 to 0.8.  $E_A = 1.0$  corresponds to the original D-K method. If  $E_A$  values are chosen, the relation between  $E$  and  $\theta$  can be obtained by use of Fig. 4 and Eqs. (24), (25). Then the  $E$ - $\theta$  curve is compared with the experimental data to determine the surface diffusivity by a procedure similar to that stated in the previous section.

It is apparent from the above discussion that this modified D-K method is applicable even if equilib-

rium relations are not formulated into equations such as the Freundlich or the Langmuir but are given graphically.

### Experimental

To examine the validity of the two methods developed in the previous sections, a series of experimental runs was made. Two kinds of activated carbon, Black Creek base carbon (Carbon A) and CAL (Carbon B), whose physical properties are listed in **Table 1**, were employed as the adsorbents, and DBS (sodium laurylsulfonate) and phenol were used as adsorbate.

Equilibrium isotherms were determined by the usual method: crushed carbon (less than 200 mesh) was added to 300 ml-Erlenmeyer flasks with 200 ml of solution, and were shaken for 2 weeks in a thermostatted bath at 30°C. The final equilibrium concentrations were determined by ultraviolet spectrophotometry (225 nm wave length for DBS, 270 nm for phenol) after the carbon had been filtered off. As is shown in **Fig. 5**, the equilibrium data for the activated carbon B were correlated by the Freundlich equations, whereas those for the carbon A were expressed by the Langmuir equations for each solution.

All the kinetic studies were conducted at 30°C in a stirred tank, 9.0 cm I.D. and 12.0 cm high, equipped with four baffles (0.9 cm wide), all of which were made of Pyrex glass. The two-bladed impeller, 4.0 cm wide and 1.0 cm high, was rotated at about 500 rpm, where the mass transfer resistance at the particle surface was negligibly small. A small amount of dried carbon particles was placed into the tank with 500 ml of distilled water, and 5 ml of concentrated solution was added. The change of concentration in the tank was detected continuously by circulating a small amount of solution through a micro flow cell set in the spectrophotometer.

The experimental conditions and the diffusivities determined are listed in **Table 2**. **Figures 6a** and **6b** show the comparison between the experimental data and the predicted lines using the two methods.

### Discussion

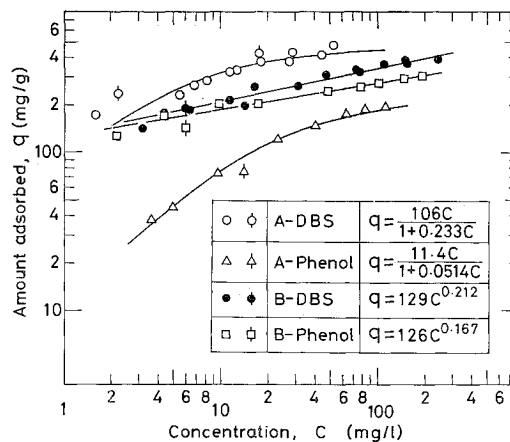
No significant difference was found in the corresponding diagrams between the two limiting mechanisms under the experimental conditions in this work, and it was difficult to determine which mechanism was controlling from the shape of the experimental  $E$  vs.  $t$  curves. Therefore, both  $D_p$  and  $D_s$  were first determined using the diagrams.

The pore diffusivity,  $D_p$ , may be represented by Eq. (26) according to the parallel-pore model.

$$D_p = \varepsilon D_A / \tau_p \quad (26)$$

**Table 1** Physical properties of activated carbons

Carbon	Particle density [g/cm <sup>3</sup> ]	Pore volume [cc/g]	Porosity [—]	Nitrogen surface area [m <sup>2</sup> /g]
A (Black Creek)	0.47	1.93	0.90	1250
B (CAL)	0.82	0.75	0.62	1090



(The keys, ◊, △, ●, □, indicate the data obtained from kinetic experiments.)

**Fig. 5** Experimental plots of adsorption isotherms

where  $D_A$  is the molecular diffusivity in aqueous solution,  $\tau_p$  is the tortuosity factor of pore diffusion, and  $\varepsilon$  is the porosity. The estimated values of  $D_A^{13}$  at 30°C for DBS and phenol are  $4.8 \times 10^{-6}$  and  $1.1 \times 10^{-5}$  cm<sup>2</sup>/sec, respectively. The value of  $\tau_p$  is usually taken as 4<sup>9)</sup>.

The  $D_p$  values computed from Eq. (26) were much less than those determined from the kinetic data for runs A-3 and B-2, where the adsorbate was phenol. This fact suggests that the contribution of pore diffusion is less significant compared with that of surface diffusion, and that surface diffusion may be controlling. Therefore, only the  $D_s$  values are listed for these runs in **Table 2**. On the other hand, for runs A-1, A-2 and B-1 Eq. (26) gave somewhat smaller  $D_p$  values than those obtained using the kinetic data from DBS solution. In this case it was difficult to determine which diffusion mechanism was controlling. Therefore both  $D_p$  and  $D_s$  are listed for these runs in **Table 2**. The predicted lines based on surface and pore diffusion controlling are shown in **Figs. 6a** and **6b**. As was expected, the experimental data for runs A-1, A-2 and B-1 lay between the two limiting cases.

It was noted that the experimental  $E-t$  curve for run B-2 has a plateau near  $E=0.8$ , and that the time required to reach equilibrium was longer than for the other runs. This retardation may be explained by the fact that active carbon B has fine pores into which molecules of phenol can enter while those of DBS cannot.

Run	A*-1	A-2	A-3	B**-1	B-2
Adsorbate	DBS	DBS	Phenol	DBS	Phenol
Mean particle radius $R$ [cm]	0.0382	0.0382	0.0538	0.0175	0.0538
Carbon dosage $W$ [g]	0.098	0.102	0.250	0.246	0.166
Initial concentration $C_0$ [mg/l]	100	50	50	100	50
Equilibrium concentration $C_\infty$ [mg/l]	17.3	2.2	14.2	6.0	6.0
Surface diffusivity $D_s$ [cm <sup>2</sup> /sec]	8.9 $\times 10^{-10}$	3.6 $\times 10^{-10}$	2.5 $\times 10^{-8}$	3.2 $\times 10^{-10}$	8.7 $\times 10^{-9}$
Pore diffusivity $D_p$ [cm <sup>2</sup> /sec]	1.8 $\times 10^{-6}$	1.4 $\times 10^{-6}$		9.5 $\times 10^{-7}$	
Surface diffusivity (modified D-K) $D_s$ [cm <sup>2</sup> /sec]	8.9 $\times 10^{-10}$	3.6 $\times 10^{-10}$	2.5 $\times 10^{-8}$		

\*, \*\* A and B represent the active carbon A and B respectively.

\*\*\* The volume of solution is 505 ml for each run.

Table 3 Comparison between numerical and modified D-K method

$\theta$	$E$ (Numerical)	Modified D-K method					
		$E_A$	0.5	0.6	0.7	0.8	1.0
$1 \times 10^{-4}$	0.083		0.082	0.082	0.082	0.082	0.081
$1 \times 10^{-3}$	0.242		0.240	0.239	0.238	0.237	0.232
$5 \times 10^{-3}$	0.473	$E$	0.471	0.467	0.463	0.458	0.440
$1 \times 10^{-2}$	0.605		0.608	0.602	0.594	0.585	0.556
$2 \times 10^{-2}$	0.741		0.761	0.751	0.740	0.725	0.680

As shown in Fig. 6a, the lines predicted by the modified D-K method coincide with those predicted using the diagrams up to  $E$  values of 0.7, and the  $D_s$  values obtained are the same as those determined using the diagrams. The values are listed in the last column of Table 2. Here the value of  $E_A$  was taken as 0.5. To examine which  $E_A$  value should be selected, the  $E$  values were calculated for  $E_A$  ranging from 0.5 to 1.0 for a Freundlich isotherm ( $C_\infty/C_0=0.1$ ,  $\beta=0.4$ ), and were compared with the corresponding numerical solutions. The  $E$  values are listed against  $\theta$  in Table 3. As  $E_A$  approaches unity, which corresponds to the original D-K method, the  $E$  values obtained by the modified D-K method deviate from those obtained numerically. Thus the original D-K method may not be applied except when experimental data are restricted to small values of  $E$ . On the other hand, the modified D-K method gives good approximation to the numerical solutions.

### Conclusion

Diagrams were presented to estimate intraparticle diffusivities for nonlinear isotherms, and these were compared with the adsorption kinetics of DBS and phenol solutions for two kinds of activated carbon. The experimental data showed good agreement with these diagrams.

An approximation method for the case where surface diffusion is controlling, which may be called a modified Dryden and Kay method, was also presented. This method gave good agreement with the numerical solutions.

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### Nomenclature

$a$	$= (W/V)(q_0 - q_\infty)/(C_0 - C_\infty)$	[—]
$b$	$= (W/V)(q_0 - q_A)/(C_0 - C_A)$	[—]
$C_A$	$=$ concentration at point A (see Fig. 1)	[mg/ml], [mg/l]
$C_D$	$=$ pseudo equilibrium concentration	[mg/ml], [mg/l]

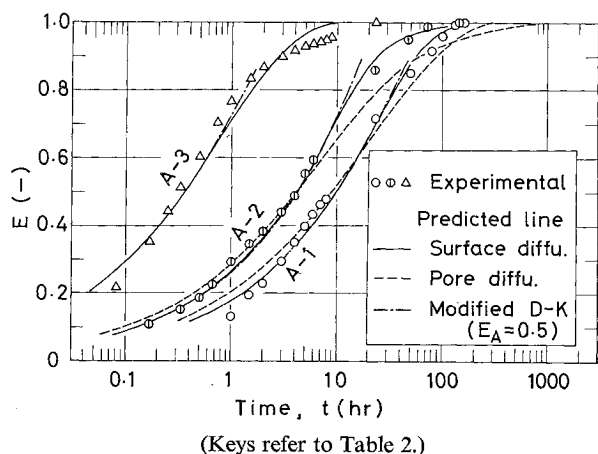


Fig. 6a Comparison between experimental and theoretical  $E-t$  plots for active carbon A

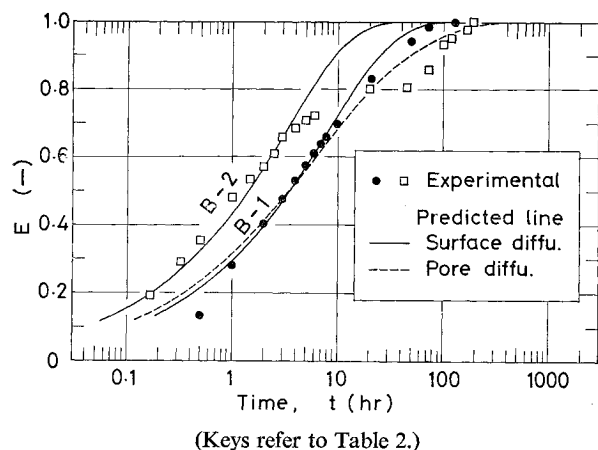


Fig. 6b Comparison between experimental and theoretical  $E-t$  plots for active carbon B

$C_L$	= concentration of solution at time $t$	[mg/ml], [mg/l]	$\beta$	= coefficient of Freundlich isotherm	[—]
$C_0$	= initial concentration	[mg/ml], [mg/l]	$\varepsilon$	= porosity	[—]
$C_\infty$	= equilibrium concentration	[mg/ml], [mg/l]	$\theta$	= dimensionless time $= (D_s/R^2)t$	[—]
$c$	= concentration at point $r$ within particle	[mg/ml]	$\rho$	= particle density	[g/cc]
$D_A$	= molecular diffusivity	[cm <sup>2</sup> /sec]	$\tau$	= dimensionless time $= (C_0/\rho q_0)(D_p/R^2)t$	[—]
$D_p$	= pore diffusivity	[cm <sup>2</sup> /sec]	$\tau_p$	= tortuosity factor for pore diffusion	[—]
$D_s$	= surface diffusivity	[cm <sup>2</sup> /sec]	<b>Literature Cited</b>		
$E$	= fractional approach to equilibrium	[—]	1) Boyd, G. E., A. W. Adamson and L. S. Myers: <i>J. Am. Chem. Soc.</i> , <b>69</b> , 2836 (1947).		
$E_A$	= $(C_0 - C_A)/(C_0 - C_\infty)$	[—]	2) Carberry, J. J.: <i>AIChE J.</i> , <b>6</b> , 460 (1960).		
$E_D$	= $(C_0 - C_L)/(C_0 - C_D)$	[—]	3) Dryden, C. E. and W. B. Kay: <i>Ind. Eng. Chem.</i> , <b>46</b> , 2294 (1954).		
$K$	= separation factor	[—]	4) Kawazoe, K.: <i>Shokubai (Catalyst)</i> , <b>15</b> , 59 (1967).		
$k$	= coefficient of Langmuir isotherm	[ml/mg], [l/mg]	5) Knoblauch, K., H. Jüntgen and W. Peters: <i>Chemie Ing. Techn.</i> , <b>41</b> , 798 (1969).		
$N_p$	= flux of pore diffusion	[mg/cm <sup>2</sup> ·sec]	6) Komiyama, H. and J. M. Smith: <i>AIChE J.</i> , <b>20</b> , 728 (1974).		
$N_s$	= flux of surface diffusion	[mg/cm <sup>2</sup> ·sec]	7) Liu, S.: <i>Chem. Eng. Sci.</i> , <b>22</b> , 871 (1967).		
$q$	= amount adsorbed at point $r$	[mg/g]	8) Paterson, S.: <i>Proc. Phys. Soc. (London)</i> , <b>59</b> , 50 (1947).		
$q_A$	= equilibrium amount adsorbed at $C_A$	[mg/g]	9) Satterfield, C. N. and P. J. Cadle: <i>I &amp; E C, Process Des. Develop.</i> , <b>7</b> , 256 (1968).		
$q_0$	= equilibrium amount adsorbed at $C_0$	[mg/g]	10) Smith, S. B., A. X. Hiltgen and A. J. Juhola: <i>Chem. Eng. Progr., Symposium Series</i> , No. 24, <b>55</b> , 25 (1959).		
$q_\infty$	= equilibrium amount adsorbed at $C_\infty$	[mg/g]	11) Suzuki, M. and K. Kawazoe: <i>J. Chem. Eng. Japan</i> , <b>7</b> , 346 (1974).		
$R$	= particle radius	[cm]	12) Suzuki, M. and K. Kawazoe: Preprint of the 8th Autumn Meeting of The Soc. of Chem. Eng. Japan, <b>B-301</b> , Tokyo (1974); <i>Seisan-Kenkyu</i> (Univ. of Tokyo), <b>26</b> , No. 7, No. 8 (1974).		
$r$	= radial position within particle	[cm]	13) Wilke, C. R. and P. Chang: <i>AIChE J.</i> , <b>1</b> , 264 (1955).		
$s_n$	= $n$ -th positive root of Eq. (17)	[—]			
$t$	= time	[sec], [hr]			
$V$	= volume of solution	[ml]			
$W$	= carbon dosage	[g]			
$x$	= $r/R$	[—]			
$y$	= $c/C_0$	[—]			
$y_1$	= value at which Freundlich and Langmuir equations coincide	[—]			
$z$	= $q/q_0$	[—]			