Chemical Engineering Journal 160 (2010) 683–690

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Experimental determination of intraparticle diffusivity and fluid film mass transfer coefficient using batch contactors

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ARTICLE INFO

Article history:
Received 19 October 2009
Received in revised form 2 April 2010
Accepted 6 April 2010

Keywords:
Adsorption
Intraparticle diffusivity
Fluid film mass transfer coefficient
Concentration decay curve
Numerical calculation

ABSTRACT

The mass transfer properties of adsorbates are generally critical factors for design of a fixed bed adsorber. For single-component systems, the completely mixed batch reactor (CMBR) method is a convenient and frequently used experimental technique for determining mass transfer properties. However, the effect of fluid film mass transfer resistance is neglected in the conventional analytical technique of CMBR method when estimating the intraparticle diffusivity (D_S). This effect should be considered to determine the diffusivities more accurately. In this study, a new analytical technique was proposed for determination of D_S and fluid film mass transfer coefficient (k_F) from one concentration decay curve and it was applied in practical experiments. This technique will be useful to estimate diffusivities when the fluid film mass transfer resistance cannot be regarded as being negligible. Under the experimental conditions of this study, the fluid film mass transfer resistance could be neglected at high stirring speeds (i.e., >200 rpm). Therefore, values of D_S can be estimated by the conventional technique from kinetic experiments performed at relatively high stirring speeds. On the other hand, values of k_F obtained by the proposed technique showed a similar tendency to values estimated from Hixson's empirical equation.

1. Introduction

Adsorption by activated carbon has been used in advanced water and wastewater treatment [1–4]. The fixed bed adsorption process is considered the most efficient method for removing organic pollutants such as phenols. The equilibrium and mass transfer properties of the adsorbates are significant factors when designing a fixed bed reactor. Techniques for determining the equilibrium parameters of single- and multi-component systems have been extensively discussed [5,6]. Many techniques have been proposed for determining mass transfer properties, including fixed bed breakthrough curves [7–9], the shallow bed technique [10–12] and the completely mixed batch reactor (CMBR) method [12–14].

Conventionally, the intraparticle diffusivity (D_S) is determined from breakthrough curves. However, values of D_S determined from breakthrough curves include both approximation and estimation errors, because the linear-driving-force approximation and an empirical equation are used in the analytical procedure. To obtain more accurate values of diffusivities, a procedure needs to be developed that does not use approximations and empirical equations.

For single-component systems, a convenient experimental technique is the CMBR method and its analytical procedure is also straightforward. In the conventional analytical technique of CMBR method, the fluid film mass transfer resistance is neglected when estimating D_S. This assumption is valid only for intraparticle diffusion control systems. Then, kinetic experiments have to be performed on the systems (i.e., high stirring speed). However, when relatively small adsorbent particles are employed, the fluid film mass transfer contributes to the adsorption rate. Consequently, it is difficult to use it to accurately estimate the value of D_S for intraparticle diffusion and fluid film mass transfer control systems. Techniques have been proposed for determining the fluid film mass transfer coefficient (k_F) for the CMBR method [13,15]. In the studies, the value of k_F was determined from the initial differential value of the concentration decay curve (i.e., the gradient of the decay curve at time zero). The value of k_F obtained by this technique might be strongly affected by initial experimental errors. Thus, to obtain more accurate values of diffusivities, they should be determined from the whole range of the experimental decay curve (EDC).

Sonetaka et al. proposed an analytical technique for determining mass transfer properties at internal and external adsorbent particles that involves performing rigorous numerical calculations for the shallow bed technique [16,17]. In the present paper, this technique is applied to the CMBR method to estimate diffusivities. In this new determination technique, the values of D_S and k_F are obtained by curve fitting the EDC and the theoretical decay curve (TDC) obtained by rigorous numerical calculations over their full ranges. More accurate diffusivities can be obtained than those

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doi:10.1016/j.cej.2010.04.012

Nomenclature

a_P	surface area based on solid particle [cm^2/cm^3-particle]
c_S	fluid concentration at $r = r_P$ [mg/L]
c_t	fluid concentration at $t = t$ (c_0 at $t = 0$) [mg/L]
D_S	effective surface diffusivity (diffusivity based on solid concentration difference) [cm^2/s]
k_F	fluid film mass transfer coefficient [cm/s]
m	weight of adsorbent [g]
q_0	amount adsorbed in equilibrium with fluid concentration c_0 [mg/g-adsorbate]
q_m	amount adsorbed at $r = r$(q_S at $r = r_P$) [mg/g-adsorbate]
q_t	average amount adsorbed in solid particles [mg/g-adsorbate]
r	internal radial length (length from the solid center) [cm]
r_P	particle radius [cm]
t	time [s]
V	volume of vessel [cm^3]
ρ_S	apparent adsorbent density [g/cm^3]

obtained by conventional techniques, since experimental errors have little effect on the value of k_F in the new technique.

Moreover, the effect of the fluid film mass transfer resistance on various contactors was investigated using the new technique. In the CMBR method, k_F is a function of the stirring speed and the particle size [13,15,18]. Suzuki et al. studied the effect of the mass transfer properties on their systems and suggested that the CMBR method can satisfy the agitation condition for which the fluid film mass transfer resistance can be regarded as being negligible [19,20]. However, specific experimental conditions for which the fluid film mass transfer resistance can be neglected have not been determined. This present study focuses on the dependence of k_F on the systems used. Several CMBR systems have been proposed and three typical systems were employed in kinetic experiments [12,13,15].

The new procedure proposed in this study will be useful for estimating D_S and k_F when the effect of the fluid film mass transfer resistance cannot be regarded as being negligible. Under the experimental conditions of this study, the intraparticle diffusivity (D_S) can be estimated using conventional technique from kinetic experiments performed at relatively high stirring speeds (>200 rpm) on any system because the fluid film mass transfer resistance can be neglected at the condition. Moreover, values of k_F obtained by use of the proposed technique showed a similar tendency to values estimated from Hixson's empirical equation [21].

2. Theoretical considerations

In general, the adsorption rate is related to both the fluid film mass transport rate and the intraparticle diffusion rate, because the final adsorbing step onto the adsorbent surface is very rapid [22]. Intraparticle diffusion includes parallel pore and surface diffusion [22,23]. However, in the case of adsorption by granular activated carbon, the intraparticle diffusion is governed by surface diffusion [24].

As mentioned above, k_F is neglected when estimating D_S in the conventional analytical techniques of CMBR method. However, both adsorption kinetic parameters, D_S and k_F, are necessary for intraparticle diffusion and fluid film mass transfer control systems. In this study, the kinetic equation at fluid film was included in the fundamental equations to determine the diffusivities more accurately. The fundamental equations for the CMBR method are as follows:

Intraparticle diffusion

$$\left(\frac{\partial q_m}{\partial t}\right) = \left(\frac{D_S}{r^2}\right)\frac{\partial}{\partial r}\left(r^2\frac{\partial q_m}{\partial r}\right) \tag{1}$$

$$q_m = q_s, \qquad \text{at } r = r_P \, (q_s = f(c_s))$$

Fluid-to-solid film transport

$$\rho_S\left(\frac{\partial q_t}{\partial t}\right) = k_F a_P (c_t - c_S) \tag{2}$$

Interface transport

$$\left(\frac{\partial q_t}{\partial t}\right) = -D_S a_P \left(\frac{\partial q_m}{\partial r}\right)_{r=r_P} \tag{3}$$

Mass balance within vessel

$$\left(\frac{\partial q_t}{\partial t}\right) = -\left(\frac{V}{m}\right)\left(\frac{\partial c_t}{\partial t}\right) \tag{4}$$

Average amount adsorbed

$$q_t = \frac{\int_0^{r_P} 4\pi\, q_m\, r^2\, dr}{(4/3)\pi r_P^3} \tag{5}$$

Equilibrium relationship

$$q_S = k c_S^{1/n} \tag{6}$$

These equations are solved to obtain the theoretical decay curve (TDC). In this study, dimensionless variables (Eqs. (7)–(9)) were introduced to reduce the amount of rigorous numerical calculation. These dimensionless variables minimize the number of parameters. For example, the Biot number (Bi) was used to replace D_S and k_F (see Eq. (8)). Bi represents the ratio of the transport rate across the liquid layer to the intraparticle mass transfer rate [8,17].

$$T = \left(\frac{D_S}{r_P^2}\right) t \tag{7}$$

$$Bi = \frac{k_F r_P}{D_S \beta \rho_S} \tag{8}$$

$$R = \frac{r}{r_P}, \quad \beta = \left(\frac{q_e - q_0}{c_0 - c_e}\right), \quad Q_m = \left(\frac{q_m - q_0}{q_e - q_0}\right), \quad Q_t = \left(\frac{q_t - q_0}{q_e - q_0}\right),$$
$$C_t = \left(\frac{c_t - c_e}{c_0 - c_e}\right), \quad C_S = \left(\frac{c_S - c_e}{c_0 - c_e}\right) \tag{9}$$

The following dimensionless fundamental equations (Eqs. (10)–(14)) were used to calculate the TDC for determining D_S. They were derived from fundamental equations for the CMBR method with dimensionless variables.

Intraparticle diffusion

$$\left(\frac{\partial Q_m}{\partial T}\right) = \left(\frac{1}{R^2}\right)\frac{\partial}{\partial R}\left(R^2\frac{\partial Q_m}{\partial R}\right) \tag{10}$$

Fluid-to-solid film transport

$$-\left(\frac{\partial Q_m}{\partial R}\right)_{R=1} = Bi(C_t - C_S) \tag{11}$$

Mass balance within vessel

$$\left(\frac{\partial Q_t}{\partial T}\right) = -\left(\frac{V}{m\beta}\right)\left(\frac{\partial C_t}{\partial T}\right) \tag{12}$$

Table 1
Properties of GAC.

Property	Value
Surface area	$1520\,m^2/g$
Pore volume	$0.64\,cm^2/g$
Pore diameter	2.4 nm
Average particle radius	0.0603 cm[a]
Apparent particle density	389 g/L[b]

[a] The value calculated from size of sieves opening (#12/16).
[b] The value determined by experiment using sieved GAC.

Average amount adsorbed

$$Q_t = 3\int_0^1 Q_m R^2 dR \tag{13}$$

Equilibrium relationship

$$Q_S = C_S^{1/n} \tag{14}$$

These equations are solved numerically to obtain the TDC for determining D_S (D_SDC). Simpson's rule was used to calculate Eq. (13) and finite difference equations were employed to evaluate these equations with the equilibrium relationship. The D_SDC was obtained with the help of a decay curve simulator (DC simulator), which is a computer program written in Borland C++ Builder and it was used for the numerical calculations in this study. The D_SDC was converted into the TDC for determining k_F (k_FDC) using Eq. (15). Thus, T was transformed into T'. The new determination procedure is described in Section 4.2.

$$T' = \left(\frac{k_F}{\beta\rho_S r_P}\right)t = Bi\left(\frac{D_S}{r_P^2}\right)t = BiT \tag{15}$$

3. Experimental

3.1. Materials

The adsorbent employed in this study was granular activated carbon (GAC; Toyo Calgon Ltd., Tokyo, Japan). The GAC was sieved to select particles with diameters in the range 1.41–1.00 mm. The sieved GAC was washed with distilled water to remove fine carbon dust and dried at 383 K for 24 h. After drying, the GAC was stored in a desiccator. The physical properties of the GAC are listed in Table 1.

The adsorbate used in this study was p-nitrophenol (PNP; Wako Chemical Co., Tokyo, Japan). PNP was used without further purification.

3.2. Isotherm experiments

The equilibrium isotherm was determined by a conventional batch bottle technique. The equilibrium concentration of the PNP solution was measured by UV spectrophotometry (Shimadzu, UV1700, Japan). The amount adsorbed was determined using the following relationship:

$$q = (c_0 - c_e)\frac{V}{m} \tag{16}$$

where c_0 and c_e are initial and equilibrium concentrations of PNP, respectively. V is the solution volume and m is the mass of adsorbent. Thus-obtained isotherm was used to check the accuracy of the kinetic experiment results and calculate theoretical decay curves. The isotherm is plotted in Fig. 1 together with the operational line in the kinetic experiment.

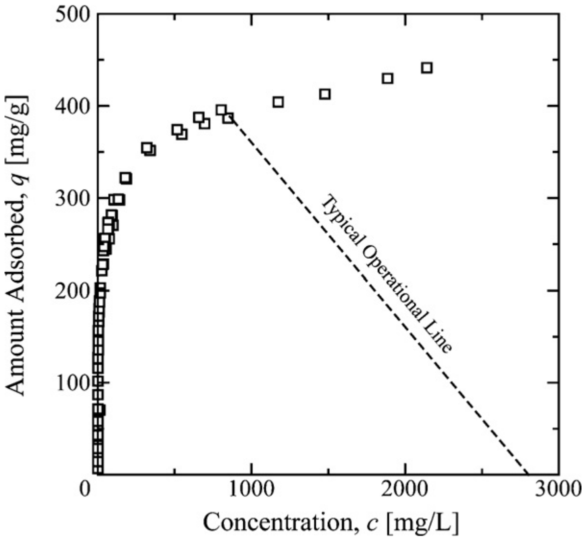

Fig. 1. Isotherm and operational line.

3.3. Kinetic experiments (CMBR method)

Three CMBR systems were employed in this study: a stirred tank reactor (STR), a paddle basket reactor (PBR) and a baffle basket reactor (BBR). Schematic illustrations of these systems are shown in Fig. 2.

For the STR, about 5 g of GAC particles were immersed in the PNP solution, whereas the GAC particles were packed in baskets for the PBR and the BBR. Each basket was filled with an equal amount of GAC particles. A 1 L solution of known concentration (about 2.8 g/L) was prepared and added to a 2 L glass vessel, and agitation was started immediately. This was taken as time zero for the kinetic experiment. The vessel was immersed in a constant-temperature bath (298 K). For all three reactors, experiments were performed at stirring speeds of approximately 0, 50, 80, 100, 150, 200 and 300 rpm. The stirring speed was measured by a tachometer (HT-4200, Ono Sokki Co., Japan). The change in concentration was determined by measuring the UV absorbance of a periodically sampled 1 mL solution. The volume of the solution was assumed to remain constant throughout the experiment.

4. Results and discussion

Kinetic experiments were performed with the three CMBR systems. Fig. 3 shows the concentration decay curves obtained from the kinetic experiments for PNP. To compare the effectiveness of the proposed determination procedure, the values of D_S were determined from the results using the conventional analytical procedure and the proposed analytical procedure. The results analyzed by each procedure are given in subsequent sections. The process used to obtain the values of D_S and k_F in the proposed determination procedure is also described.

4.1. Conventional analytical procedure for determining D_S (neglects fluid film mass transfer resistance)

In the conventional analytical techniques of CMBR method, the fluid film mass transfer resistance is neglected when estimating the intraparticle diffusivity (D_S). In this section, the conventional determination procedure was used to analyze the experimental results. For the CMBR method, several determination procedures have been proposed for intraparticle diffusion control systems [12,19,20]. In this study, the procedure proposed by Furuya et al. was employed;

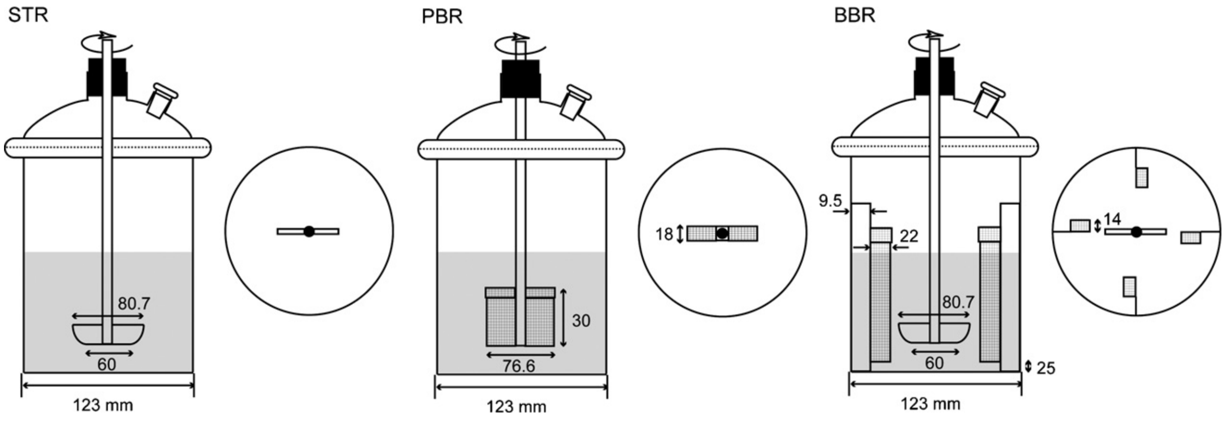

Fig. 2. Schematic illustrations of the CMBR systems used in this study.

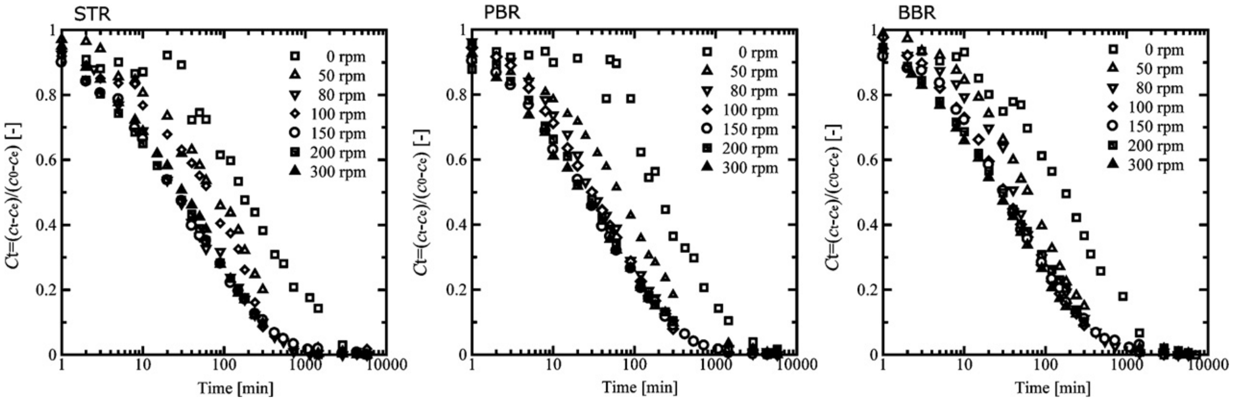

Fig. 3. Experimental decay curves of PNP for the three CMBR systems.

a detailed description of this procedure can be found elsewhere [12].

The values of D_S were determined from the kinetic experiment results. Some EDCs could not be successfully overlaid with the TDC (see Fig. 4). However, we applied the process even when the curves could not be well fitted. As a result, the values of D_S were determined and the dependence of D_S on the stirring speed is shown in Fig. 5.

From Fig. 5, it is clear that the value of D_S is affected by the stirring speed. Fluid film mass transfer resistance is generated by insufficient agitation on all systems at low stirring speeds. As mentioned above, some EDCs could not be successfully overlaid with the TDC. This was observed for EDCs measured at low stirring speeds ($N < 200$ rpm). This means that the conventional determi-

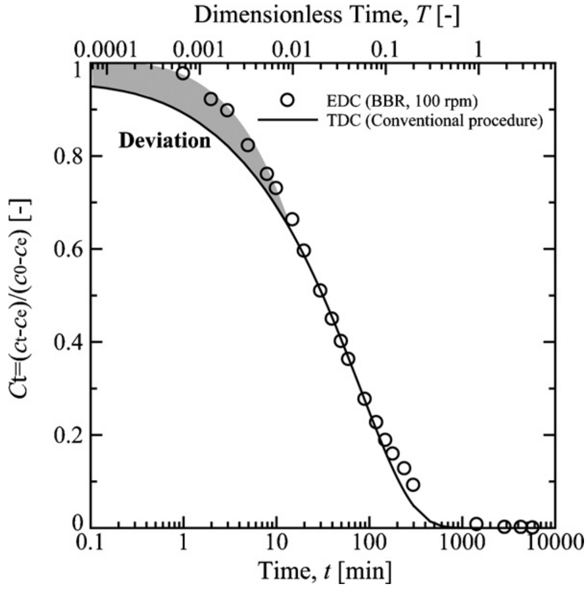

Fig. 4. Curve fitting of experimental decay curve (EDC) with theoretical decay curve (TDC).

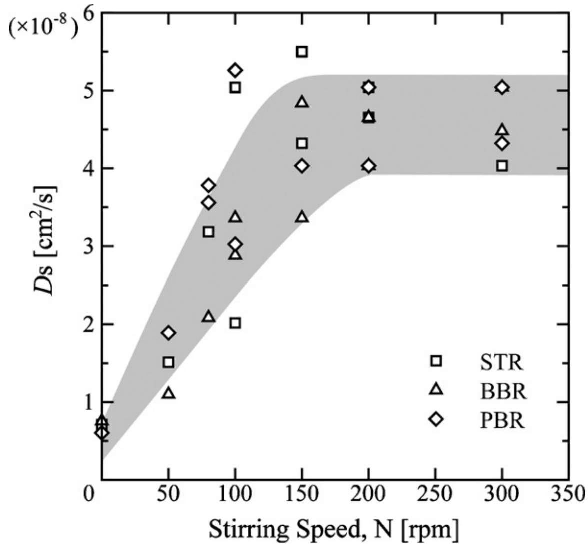

Fig. 5. Dependence of D_S on the stirring speed.

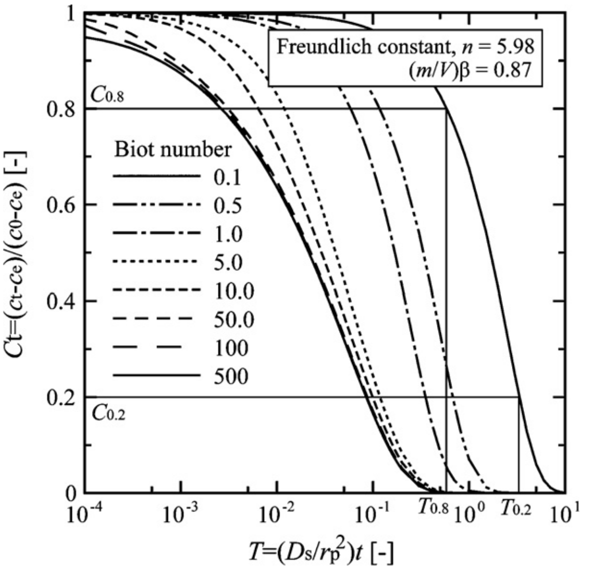

Fig. 6. Theoretical decay curves calculated under experimental conditions.

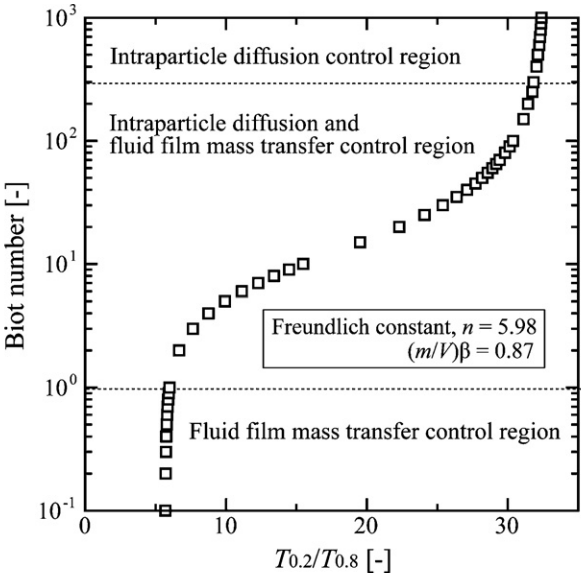

Fig. 7. Plot of Biot number against $T_{0.2}/T_{0.8}$.

nation procedure is not effective for analyzing the experimental results obtained with insufficient agitation, since the fluid film mass transfer resistance cannot be regarded as being negligible. Therefore, a new determination technique is required to analyze the experimental results for intraparticle diffusion and fluid film mass transfer control systems.

4.2. New analytical procedure for determining D_S and k_F

Sonetaka et al. proposed an analytical technique for determining the mass transfer properties at internal and external adsorbent particles using the shallow bed technique [16,17]. This technique was applied to the CMBR method, and hence a new analytical procedure for determining the values was established. This new procedure is described below.

Fig. 6 shows the TDCs obtained by the DC simulator for a wide range of *Bi*. As shown in Fig. 6, the shape of the TDC depends on the value of *Bi*. The larger *Bi* is, the gentler the slope of the TDC is. The ratio $T_{0.2}/T_{0.8}$, which represents the slope of the TDC, is plotted against *Bi* in Fig. 7. $T_{0.8}$ and $T_{0.2}$ are dimensionless concentrations at $C_t = 0.8$ and 0.2, respectively. From Fig. 7, the adsorption rate is controlled by fluid film mass transfer when $Bi < 1$. In the region $1 < Bi < 300$, both the intraparticle diffusion and the fluid film mass transfer are important. In the region $Bi > 300$, the adsorption rate is controlled by the intraparticle diffusion. The value of *Bi* can be obtained from Fig. 7 for a known value of $T_{0.2}/T_{0.8}$. The ratio $T_{0.2}/T_{0.8}$ can be obtained from the EDC.

In the intraparticle diffusion and fluid film mass transfer control region, the following technique for determining *Bi* can be applied. From Eq. (7), the dimensionless time ratio ($T_{0.2}/T_{0.8}$) can be expressed in Eq. (17). Therefore, the experimental time ratio ($t_{0.2}/t_{0.8}$) is identical to the dimensionless time ratio ($T_{0.2}/T_{0.8}$). Consequently, the EDC can be used to determine the value of *Bi* from Fig. 7.

$$\frac{T_{0.2}}{T_{0.8}} = \frac{(D_S/r_P^2) \times t_{0.2}}{(D_S/r_P^2) \times t_{0.8}} = \frac{t_{0.2}}{t_{0.8}} \tag{17}$$

The adsorption isotherm of PNP on GAC was determined, and Freundlich constants, *k* and *n*, were obtained as 129.01 and 5.98, respectively. With the determined *Bi* and experimental conditions (Freundlich constant, $n = 5.98$, dimensionless ratio of solid to liquid, $(m\beta/V) = 0.87$), the TDC for determining D_S (D_SDC) was calculated by the DC simulator. The D_SDC and the EDC were then overlaid and one of the curves was shifted horizontally until the two curves fit each other, as shown in Fig. 8.

A T/t of 6.67×10^{-4} (min^{-1}) was obtained by curve fitting and it was substituted into Eq. (7) to evaluate the intraparticle diffusivity, D_S (Eq. (18)). The obtained D_S was approximately 4.03×10^{-8} (cm^2 s^{-1}).

$$D_S = \left(\frac{T}{t}\right) r_P^2 = \frac{6.67 \times 10^{-4}}{60} [\mathrm{s}^{-1}] \times (0.0603\,[\mathrm{cm}])^2 = 4.03 \times 10^{-8}\ [\mathrm{cm}^2/\mathrm{s}] \tag{18}$$

To determine k_F, another dimensionless variable (Eq. (15)) was introduced. D_SDC was converted into the plot of C_t versus T' (k_FDC) using Eq. (15). The k_FDC and the EDC also overlapped each other (see Fig. 9). A T'/t ratio of 1.59×10^{-2} min^{-1} was obtained by curve

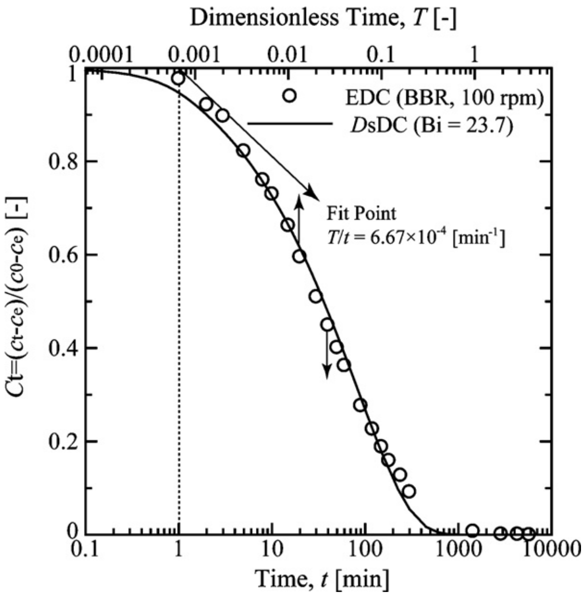

Fig. 8. Curve fitting of experimental decay curve (EDC) with theoretical decay curve for D_S determination (D_SDC).

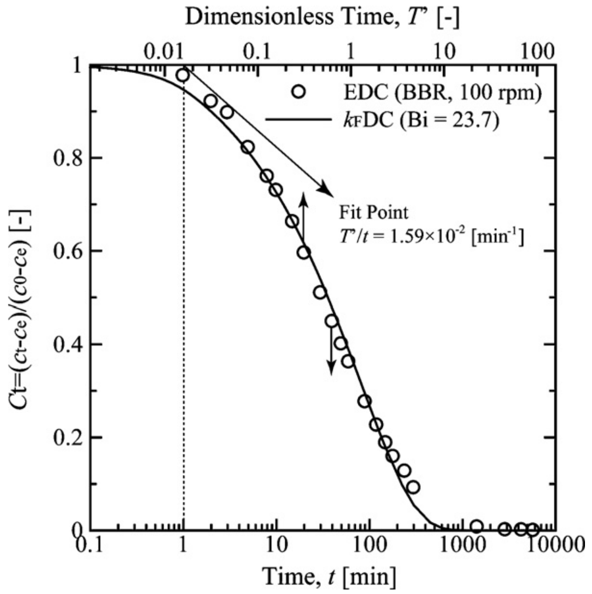

Fig. 9. Curve fitting of experimental decay curve (EDC) with theoretical decay curve for k_F determination (k_FDC).

fitting and it was substituted into Eq. (15) to evaluate the fluid film mass transfer coefficient, k_F (Eq. (19)). The obtained k_F was about 1.08×10^{-3} (cm s^{-1}).

$$k_F = \left(\frac{T'}{t}\right)\beta\rho_S r_P = \frac{1.59\times10^{-2}}{60}\,[\mathrm{s}^{-1}]\times0.174\,[\mathrm{L/g}]\times389\,[\mathrm{g/L}] \times0.0603\,[\mathrm{cm}] = 1.08\times10^{-3}\ [\mathrm{cm/s}] \qquad (19)$$

Finally, Bi was calculated from the evaluated values of D_S and k_F to compare with theoretical Bi determined from Fig. 7 with the experimental time ratio in order to confirm accuracy of obtained D_S and k_F. Both Bi values were consistent with each other.

The new procedure for simultaneously determining D_S and k_F for a single-component CMBR system is illustrated in Fig. 10 as a flow diagram. For intraparticle diffusion and fluid film mass transfer control systems, the proposed technique can determine diffusivities more accurately than conventional techniques.

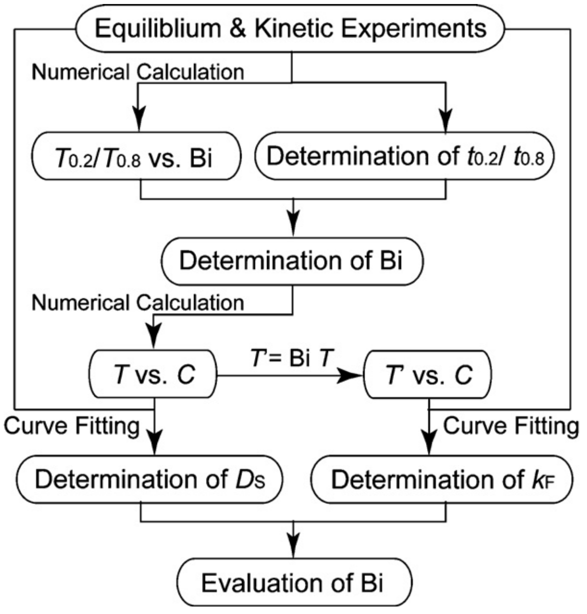

Fig. 10. New procedure for determining D_S and k_F for the CMBR method.

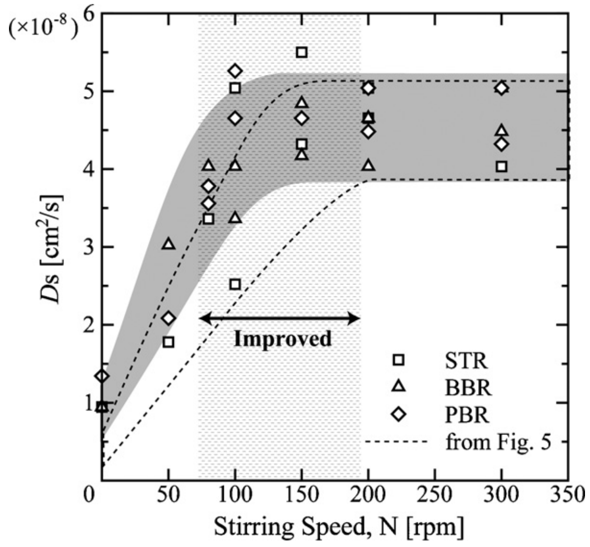

Fig. 11. Dependence of D_S on the stirring speed.

Fig. 3 shows the decay curves obtained from the kinetic experiments for PNP. The values of D_S and k_F were determined from these experimental results by use of the new procedure described above. As mentioned above, the adsorption rate is controlled by intraparticle diffusion in the region $Bi > 300$. It is noted that $Bi > 300$ implies that $T_{0.2}/T_{0.8} > 32$. When $T_{0.2}/T_{0.8} > 32$, the conventional procedure was used to determine the value of D_S. Some values of $t_{0.2}/t_{0.8}$ determined from the EDC exceeded 32. Thus-obtained values of D_S and k_F are listed in Table 2. Since Bi evaluated from obtained D_S and k_F were consistent with Bi determined from the experimental time ratio, the values of D_S and k_F are expected to be correct (see Table 2).

Table 2 shows that, as expected, fluid film mass transfer resistance was observed at low stirring speeds in all three systems. As mentioned above, the conventional determination procedure is not effective for analyzing the experimental results measured with insufficient agitation, since the fluid film mass transfer resistance cannot be negligible. However, since the effect of fluid film mass transfer resistance was considered in the new procedure, curve fitting the EDC with the D_SDC using the new procedure was successful even for EDCs that could not be analyzed by the conventional procedure (compare Figs. 4 and 8).

Fig. 11 shows the dependence of the intraparticle diffusivity (D_S) on the stirring speed. It is obvious from Fig. 11 that the values of D_S, in relatively low stirring speed region ($80 < N < 200$ rpm), obtained by the new procedure were more accurate than those obtained by the conventional procedure. In addition, values of D_S obtained in the stirring speed region more than 80 rpm were almost identical with the value obtained by the shallow bed technique. The value of D_S obtained by the shallow bed technique was approximately 3×10^{-8} (cm^2 s^{-1}) [25]. However, values of D_S obtained in the stirring speed region less than 50 rpm were hardly improved. This is because the fluid film mass transfer might strongly contribute to the adsorption rate. Therefore, more accurate values of diffusivities can be obtained by the new procedure when the adsorption rate is controlled by the intraparticle diffusion and the fluid film mass transfer. Under the experimental conditions of this study, the effect of fluid film mass transfer resistance could be neglected at high stirring speeds (i.e., >200 rpm). Therefore, D_S can be estimated by the conventional procedures from kinetic experiments performed at relatively high stirring speeds.

Table 2
Experimental results.

System	Stirring speed [rpm]	$t_{0.2}/t_{0.8}$ [−]	Biot number [−][a]	D_S [cm^2/s]	k_F [cm/s]	Biot number [−][b]
STR	0	16.7	11.2	9.45E−09	1.19E−04	11.2
	50	27.3	41.0	1.78E−08	8.17E−04	41.0
	80	28.6	42.6	3.36E−08	1.62E−03	42.9
	100	31.4	190	2.52E−08	5.22E−03	185
	150	>32	–[c]	4.32E−08	–	–
	200	>32	–[c]	4.65E−08	–	–
	300	>32	–[c]	4.03E−08	–	–
BBR	0	17.6	12.4	9.45E−09	1.31E−04	12.3
	50	14.3	8.6	3.03E−08	2.96E−04	8.7
	80	17.3	12.1	4.03E−08	5.44E−04	12.0
	100	23.3	23.7	4.03E−08	1.08E−03	23.8
	150	25.0	28.5	4.17E−08	1.33E−03	28.4
	200	>32	–[c]	4.03E−08	–	–
	300	>32	–[c]	4.48E−08	–	–
PBR	0	8.6	3.6	1.34E−08	5.44E−05	3.6
	50	28.1	48.0	2.09E−08	1.13E−03	48.3
	80	21.4	19.1	3.78E−08	8.10E−04	19.0
	100	24.6	27.2	4.65E−08	1.42E−03	27.1
	150	30.8	120	4.65E−08	6.18E−03	118
	200	31.8	300	4.48E−08	1.51E−02	300
	300	>32	–[c]	5.04E−08	–	–

[a] *Bi* determined from the experimental time ratio $t_{0.2}/t_{0.8}$.

[b] *Bi* evaluated from D_S and k_F.

[c] Based on the value of $t_{0.2}/t_{0.8}$, the intraparticle diffusion control the adsorption rate. In the case, *Bi* is unnecessary, because the conventional procedure is used for determining D_S.

On the other hand, experimentally obtained values of fluid film mass transfer coefficient (k_F) were plotted against the stirring speed in Fig. 12 along with values estimated from Hixson's empirical equation [21]. It is clear that both values showed a similar tendency. As mentioned above, the fluid film mass transfer coefficient (k_F) is known to be a function of the stirring speed and the particle size in the CMBR method [13,15,18]. Generally, k_F is proportional to N^x, where N is the stirring speed and x is a constant. Experimental k_F values follow the correlation. However, several experimental k_F values were deviated from Hixson's equation. This deviation might be due to experimental conditions. Most previous studies were carried out for dissolution of materials and ion exchange; there have been few reports for adsorption onto activated carbon. In addition, the impeller shape and position and vessel size will affect the results. Detailed factors that affect the value of k_F can be found elsewhere [18,26].

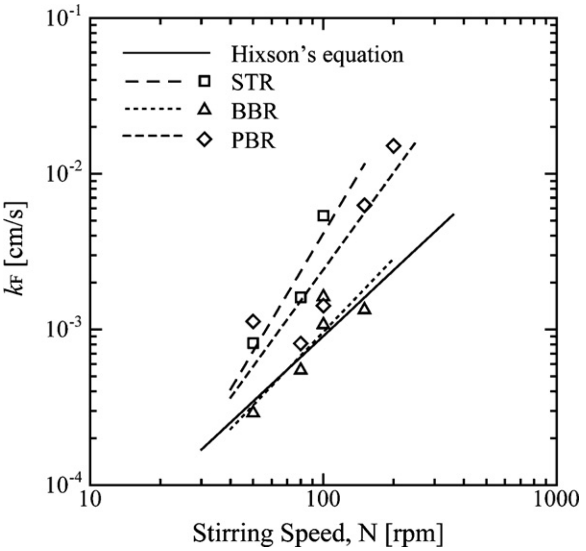

Fig. 12. Dependence of k_F on the stirring speed.

5. Conclusions

A simultaneously determination technique of the intraparticle diffusivity (D_S) and the fluid film mass transfer coefficient (k_F) for a single-component CMBR system was proposed. This technique is described in Section 4.2 and illustrated in Fig. 10. Experimental values of D_S and k_F were obtained from experimental results analyzed by the proposed technique. The values of D_S, in relatively low stirring speed region ($80 < N < 200$ rpm), obtained by the new technique were more accurate than those obtained by the conventional technique. On the other hand, values of k_F obtained by use of the new technique showed similar tendency to ones estimated from Hixson's empirical equation. Therefore, the proposed technique will be useful to estimate D_S and k_F when the fluid film mass transfer resistance cannot be ignored. Moreover, the fluid film mass transfer resistance can be regarded as negligible for stirring speeds greater than 200 rpm under the experimental conditions employed here. Therefore, it should be noted that D_S can be estimated by conventional techniques from kinetic experiments performed at relatively high stirring speeds on any system.

Acknowledgements

The authors would like to gratefully acknowledge partial funding from the Special Research from the Office of Research Advanced and Intellectual Property, Meiji University, Japan. Furthermore, a part of this study was supported by an International Research Award of Meiji University.

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