

Analysis of the Constant Molar Flow Semi-Batch Adsorber Loaded with Inert Core Adsorbents

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Abstract—An analytical solution for a constant molar flow semi-batch adsorber loaded with inert core adsorbents is obtained for the linear isotherm coupled with intraparticle diffusion and external film diffusion. The solution obtained in this study consists of two parts. One is the asymptote, to which the bulk concentration approaches as time is sufficiently large. The other is the relaxation term, which dictates how the mass in the bulk phase flows into the adsorbent within the adsorption vessel. For a given value of the inert core radius, the slope of the asymptote is a function of the equilibrium parameter only and the intercept is a function of the kinetic parameter as well as the equilibrium parameter. The intercept of the asymptote consists of two parts. One is the contribution due to the external film resistance; the other is the contribution due to the intraparticle diffusion resistance. Hence, we can obtain the information of the equilibrium constant, the film mass transfer coefficient, and the effective diffusivity from the asymptote of the solution presented in this study.

Key words: Constant Molar Flow, Inert Core Adsorbent, Semi-Batch Adsorber, Porous Media, Effective Diffusivity

INTRODUCTION

The diffusion in porous media is important in the sense that in most adsorption processes diffusion is the rate-controlling step owing to the fact that the intrinsic adsorption rate is usually much faster than the diffusion rate. The measurement of the effective diffusivity in porous media can be obtained by using one of several experimental techniques, such as the gas chromatography [Schneider and Smith, 1968], the diffusion cell [Dogu and Smith, 1976], the gravimetric method using microbalance [Gray and Do, 1991], the zero length column [Eic and Ruthven, 1988], the differential adsorption bed [Do et al., 1991], batch adsorber [Garg and Ruthven, 1972], the fixed bed breakthrough [Rosen, 1952; Glueckauf, 1955; Kim et al., 2001; Yang et al., 2003] and the constant molar flow (CMF) semi-batch adsorber [Do, 1995]. The measured effective diffusivity is then used to predict the performance of the industrial process, such as fixed-bed chromatography and various types of adsorbers and catalytic reactors. The advantages and disadvantages or limitations of various methods to measure the effective diffusivity are given in the reference [Park et al., 1996].

Among the experimental techniques for the measurement of the effective diffusivity, the CMF semi-batch adsorber was proposed most recently [Do, 1995]. This method has been successfully applied to measure equilibrium and dynamic parameters in porous adsorbent particles [Prasetyo and Do, 1998, 1999; Do et al., 2000]. Park and Do [1996] presented an exact analytical solution for the solute concentration in the CMF semi-batch adsorber for adsorbents having a bidispersed pore structure. Park [2002] presented an exact analytical solution for the CMF semi-batch adsorber with external film diffusion.

In the CMF semi-batch adsorber, a constant flow of adsorbate is introduced in a pre-evacuated adsorption vessel. The pressure re-

sponse of the adsorption vessel is then monitored as a function of time. After a short period of time, the response reaches a linear asymptote, of which the slope and the intercept can be utilized to determine the equilibrium parameter and the kinetic parameter, respectively.

The determination of model parameters from the experiment and then the prediction of performance of a particular process using the measured parameters are closely related to the mathematical modeling of the corresponding physical system. The model equations concerned with diffusion and adsorption in porous media are usually expressed as a system of partial differential equations. Hence, it is not always a simple matter to solve the model equations for a diffusion model in porous media. While numerical analysis is the last resort to the solution of the diffusion model especially for a nonlinear system, an analytical solution is most desirable because it shows explicit parameter dependences. In design and simulation the system behavior as parameters change is quite critical [Rice and Do, 1995].

Inert core adsorbents were recently developed to improve the separation performance of proteins in an expanded-bed adsorption process [Chanda and Rempel, 1997, 1999; Li et al., 2003a, b, 2004]. These inert core adsorbents have increased density by the incorporation of heavier inert core and are reported to be suitable for stable expansion at high flow rates in expanded-bed. The exact analytical solution for inert core adsorbents has been presented in the reference for the batch adsorber [Chanda and Rempel, 1997, 1999; Li et al., 2003a] and for the fixed bed breakthrough [Li et al., 2003b, 2004].

In this study, we will present an exact analytical solution for the solute concentration in the CMF semi-batch adsorber loaded with inert core adsorbents. In the outer adsorbent shell, the homogeneous particle diffusion model is assumed. The linear adsorption isotherm is assumed and the external film diffusion is included.

MATHEMATICAL MODEL

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1. Mass Balance

Consider an adsorption vessel in which inert core adsorbents are loaded. The inert core adsorbent particle consists of the outer shell layer of radius R and the inert core of radius R_c . The vessel is initially evacuated. At time $t=0$, a pure gas is introduced into the vessel with constant molar flow rate \dot{N} . The simplifying assumptions of the CMF model [Do, 1995; Park and Do, 1996; Park, 2002] for inert core adsorbents [Chanda and Rempel, 1997, 1999; Li et al., 2003a, b, 2004] are: (1) ideal mixing in the vessel; (2) local linear adsorption equilibrium at the particle surface; (3) homogeneous particle diffusion within the outer shell layer of particle; (4) non-permeable, inert core; (5) negligible swelling or shrinking during sorption. The mathematical model based on the above assumptions is as follows:

Intraparticle Mass Balance in Outer Adsorbent Shell:

$$\frac{\partial C_\mu}{\partial t} = D_e \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial C_\mu}{\partial r} \right) \quad (R_c \leq r \leq R) \quad (1a)$$

$$\text{at } t=0 \quad C_\mu = 0 \quad (1b)$$

$$\text{at } r=R_c \quad \frac{\partial C_\mu}{\partial r} = 0 \quad (1c)$$

$$\text{at } r=R \quad \left[D_e \frac{\partial C_\mu}{\partial r} \right]_{r=R} = k_f (C_b - C_s) \quad (1d)$$

where C_μ is the concentration in the adsorbent, C_b is the concentration in the adsorption vessel outside the adsorbents, C_s is the concentration in the gas phase at adsorbent surface, D_e is the effective diffusivity, k_f is the external mass transfer coefficient, r is the coordinate variable of particle and t is the time variable. Note that the symmetric boundary condition at the particle centre in Park [2002] (i.e., at $r=0$, $\partial C_\mu / \partial r = 0$) is replaced by Eq. (1c) to account for the impermeable inert core.

Mass Balance around the Adsorption Vessel:

$$V \frac{dC_b}{dt} + V_{\mu 0} \left(1 - \frac{R_c^3}{R^3} \right) \frac{d\langle C_\mu \rangle}{dt} = \dot{N} X(t); \quad \langle C_\mu \rangle = \frac{3}{R^3 - R_c^3} \int_{R_c}^R r^2 C_\mu dr \quad (2a)$$

$$X(t) = U(t) \quad (2b)$$

$$\text{at } t=0 \quad C_b = 0 \quad (2c)$$

where V and $V_{\mu 0}$ are the volumes of the continuous phase and the dispersion phase within the vessel, respectively. The non-dimensional unit forcing function $X(t)$ is introduced in Eq. (2a). The constant molar flow rate \dot{N} is the intensity of the forcing function. Note that $X(t)$ is simply the unit step function for the CMF semi-batch adsorber of this study, as shown in Eq. (2b).

Linear Adsorption Equilibrium at the Adsorbent Surface:

$$C_s|_{r=R} = K C_s \quad (3)$$

where K is the Henry's constant.

2. Non-Dimensionalization

With the dimensionless variables and parameters defined in Table 1, the model equations given in the previous section are rewritten

Table 1. Definition of dimensionless variables and parameters

Define C_0 reference concentration

$$\begin{aligned} y_b &= \frac{C_b}{C_0}; & y_s &= \frac{C_s}{C_0}; & x &= \frac{r}{R}; & x_c &= \frac{R_c}{R}; & y_\mu &= \frac{C_\mu}{K C_0} \\ \tau &= \frac{D_e t}{R^2}; & \xi &= \frac{k_f R}{K D_e}; & \beta_0 &= \frac{V_{\mu 0}}{V} K; & \beta &= \beta_0 (1 - x_c^3); & \Omega &= \frac{R^2 / D_e}{V C_0 / \dot{N}} \end{aligned}$$

in dimensionless forms as follows:

Intraparticle Mass Balance in Outer Adsorbent Shell:

$$\frac{\partial y_\mu}{\partial \tau} = \left(\frac{\partial^2 y_\mu}{\partial x^2} + \frac{2}{x} \frac{\partial y_\mu}{\partial x} \right) \quad (x_c \leq x \leq 1) \quad (4a)$$

$$\text{at } \tau=0 \quad y_\mu = 0 \quad (4b)$$

$$\text{at } x=x_c \quad \frac{\partial y_\mu}{\partial x} = 0 \quad (4c)$$

$$\text{at } x=1 \quad \left[\frac{\partial y_\mu}{\partial x} \right]_{x=1} = \xi (y_b - y_s) \quad (4d)$$

Mass Balance around the Adsorption Vessel:

$$\frac{dy_b}{d\tau} + \beta_0 (1 - x_c^3) \frac{d\langle y_\mu \rangle}{d\tau} = \Omega X(t); \quad \langle y_\mu \rangle = \frac{3}{1 - x_c^3} \int_{x_c}^1 x^2 y_\mu dx \quad (5a)$$

$$X(\tau) = U(\tau) \quad (5b)$$

$$\text{at } t=0 \quad y_b = 0 \quad (5c)$$

Linear Adsorption Equilibrium at the Adsorbent Surface:

$$y_s|_{x=1} = y_s \quad (6)$$

ANALYTICAL SOLUTION

1. Solution in Laplace Domain

The general solution of (4a) in the Laplace domain is:

$$\bar{y}_\mu = c_1 \frac{\cosh(qx)}{x} + c_2 \frac{\sinh(qx)}{x}; \quad q = \sqrt{s} \quad (7a)$$

where c_1 and c_2 are arbitrary constants to be determined by application of the boundary conditions:

$$c_1 = \frac{\bar{y}_b \left[\cosh(x_c q) - \frac{\sinh(x_c q)}{x_c q} \right]}{\left(1 - \frac{1}{\xi} + \frac{1}{\xi x_c} \right) \cosh[q(1 - x_c)] + \left(1 - \frac{1}{\xi} + \frac{q^2 x_c}{\xi} \right) \frac{\sinh[q(1 - x_c)]}{x_c q}} \quad (7b)$$

$$c_2 = c_1 \frac{\cosh(x_c q) - x_c q \sinh(x_c q)}{x_c q \cosh(x_c q) - \sinh(x_c q)} \quad (7c)$$

Taking the Laplace transform of the second equation in Eq. (5a), substitution of Eqs. (7) and integration give the vessel transfer function $F(s)$, which relates the bulk concentration (i.e., \bar{y}_b) to the mean of y_μ (i.e., $\langle \bar{y}_\mu \rangle$):

$$F(s) = \frac{3}{(1 - x_c^3) q^2} \begin{bmatrix} (1 - x_c) q \cosh[q(1 - x_c)] \\ - (1 - x_c q^2) \sinh[q(1 - x_c)] \\ q \left[x_c + \frac{(1 - x_c)}{\xi} \right] \cosh[q(1 - x_c)] \\ + \left[1 - \frac{(1 - x_c q^2)}{\xi} \right] \sinh[q(1 - x_c)] \end{bmatrix} \quad (8)$$

Taking the Laplace transform of the first equation in Eqs. (5), then substitution of Eq. (8) gives the overall transfer function $G(s)$, which relates the forcing function (i.e., $\Omega \bar{X}$) to the response (i.e., \bar{y}_b):

$$G(s) = \frac{1}{s[1 + \beta_0 (1 - x_c^3) F(s)]}$$

That is, since $\bar{X}(s) = 1/s$. The solution for the bulk concentration

in the Laplace domain is expressed as:

$$\frac{\bar{y}_b}{\Omega} = \frac{1}{s^2 [1 + \beta_0(1 - x_c^3)F(s)]} \quad (9b)$$

2. Solution in Time Domain

By taking the inverse transform of Eq. (9b) using the method of residues, the solution in the time domain can be obtained as:

$$\frac{y_b}{\Omega} = \left(\frac{y_b}{\Omega} \right)_1 + \left(\frac{y_b}{\Omega} \right)_2 \quad (10a)$$

$$\left(\frac{y_b}{\Omega} \right)_1 = \delta_0 \tau + \beta_0(1 - x_c^3) \delta_0^2 (\delta_f + \delta_a) (1 + \lambda_n^2 x_c) \sin[\lambda_n(1 - x_c)] \quad (10b)$$

$$\left(\frac{y_b}{\Omega} \right)_2 = 6\beta_0 \sum_{n=1}^{\infty} \frac{-\lambda_n(1 - x_c) \cos[\lambda_n(1 - x_c)]}{\lambda_n^4 \left\{ A_n \sin[\lambda_n(1 - x_c)] + B_n \cos[\lambda_n(1 - x_c)] \right\}} \exp(-\lambda_n^2 \tau) \quad (10c)$$

$$\delta_0 = \frac{1}{1 + \beta_0(1 - x_c^3)} \quad (10d)$$

$$\delta_f = \frac{1 - x_c^3}{3\xi} \quad (10e)$$

$$\delta_a = \frac{1}{15} \left(1 - \frac{4(1 - x_c^3)x_c^3}{1 - x_c^3} + \frac{5(1 - x_c)x_c^5}{1 - x_c^3} \right) \quad (10f)$$

$$A_n = 2 \left(1 - \frac{1}{\xi} \right) + \left(3\beta_0 - \frac{\lambda_n^2}{\xi} \right) - \left(1 + \frac{2}{\xi} \right) \lambda_n^2 x_c + \left(\lambda_n^2 + 3\beta_0 - \frac{\lambda_n^2}{\xi} \right) x_c^2 \quad (10g)$$

$$B_n = \lambda_n \left[\left(1 + \frac{2}{\xi} \right) + \left\{ 3\beta_0 - \frac{\lambda_n^2}{\xi} + 2 \left(1 - \frac{1}{\xi} \right) \right\} x_c - \left(3\beta_0 - \frac{\lambda_n^2}{\xi} \right) x_c^2 \right] \quad (10h)$$

where eigenvalues λ_n for $n=1, 2, \dots, \infty$ are given by the positive roots of the transcendental equation, $1 + \beta_0(1 - x_c^3)F(s = -\lambda_n^2) = 0$. That is, λ_n is given by the positive roots of the following transcendental equation:

$$\tan[\lambda_n(1 - x_c)] = \lambda_n^2 x_c + \left(\frac{\lambda_n^2}{\xi} - 3\beta_0 \right) (1 - x_c) \quad (11)$$

RESULTS AND DISCUSSION

1. Significance of Solution

We have obtained the exact analytical solution for the CMF semi-batch adsorber having inert core adsorbents in the previous section. The bulk concentration in the adsorption vessel increases with time as a result of the introduction of adsorbate into the vessel, and as time is sufficiently large it asymptotes to a straight line, which corresponds to $(y_b/\Omega)_1$ given in Eq. (10b). The slope of the asymptote δ_0 is a function of the equilibrium parameter only and the intercept is a function of the kinetic parameters as well as the equilibrium parameter. The intercept of the asymptote consists of two parts: One is the contribution due to the external film resistance, which corresponds to δ_f given by Eq. (10e), and the other is the contribution due to the intraparticle diffusion resistance, which corresponds to δ_a given by Eq. (10f). When $\xi \rightarrow \infty$, δ_f becomes zero and the model behavior is limited by the intraparticle diffusion step.

The adsorbate introduced into the vessel flows then into adsorbents within vessel; hence the rate of increase in the bulk concentration is relaxed as a result of the mass flow into adsorbents. The mass flow into adsorbents is dictated by $(y_b/\Omega)_2$ given by Eq. (10c).

When the radius of the inert core approaches zero, the inert core adsorbent becomes a conventional adsorbent. The author reported the analytical solution for the CMF semi-batch adsorber having a conventional adsorbent [Park, 2002]. It should be noted that Eq. (10) reduces to the solution for the conventional adsorbent at $x_c \rightarrow 0$. When $x_c \rightarrow 1$, the inert core adsorbent becomes the impermeable particle. In this case, the model corresponds simply to the physically filling vessel, of which the response to the step input is dictated by the following equation: $(y_b/\Omega)_1 = \delta_0 \tau = \tau$.

In the three limiting cases mentioned above (i.e., $\xi \rightarrow \infty$, $x_c \rightarrow 0$, and $x_c \rightarrow 1$), all the quantities in Eqs. (10a)-(10h), which affected by ξ and x_c , have physically sound limiting values. For example, $\delta_a = 1/15$ and $\delta_f = 1/3\xi$ at $x_c \rightarrow 0$ [Park, 2002]; $\delta_a = \delta_f = 0$ at $x_c \rightarrow 1$; and $\delta_0 = 0$ at $\xi \rightarrow \infty$.

2. Solution for Conventional Batch Adsorber

With the impulse forcing, that is, $X(\tau) = \delta(\tau)$, the mass balance equation around the adsorption vessel becomes

$$\frac{dy_b}{d\tau} + \beta_0(1 - x_c^3) \frac{d\langle y_u \rangle}{d\tau} = \Omega \delta(\tau) \quad (12a)$$

$$\text{at } \tau=0 \quad y_b=0 \quad (12b)$$

For a given time basis, the above equation means simply that the amount of accumulation in both bulk and adsorbed phases is just balanced with the instantaneous input Ω . Since this input Ω is introduced into the empty vessel instantaneously at $\tau=0$, it is simply that the adsorption vessel is the conventional batch adsorber containing a finite amount of adsorbent and a finite amount of adsorbate. Note that the mass balance around the batch adsorption vessel is usually expressed as [Li et al., 2003a]:

$$\frac{dy_b}{d\tau} + \beta_0(1 - x_c^3) \frac{d\langle y_u \rangle}{d\tau} = 0 \quad (13a)$$

$$\text{at } \tau=0 \quad y_b/\Omega=1 \quad (13b)$$

We can obtain the same analytical solution from the two models, given in Eqs. (12) and (13), respectively:

Solution in Laplace Domain

$$\left(\frac{\bar{y}_b}{\Omega} \right)_{batch} = \frac{1}{s [1 + \beta_0(1 - x_c^3)F(s)]} \quad (14a)$$

Solution in Time Domain

$$\left(\frac{y_b}{\Omega} \right)_{batch} = \delta_0 - 6\beta_0 \sum_{n=1}^{\infty} \frac{-\lambda_n(1 - x_c) \cos[\lambda_n(1 - x_c)]}{\lambda_n^2 \left\{ A_n \sin[\lambda_n(1 - x_c)] + B_n \cos[\lambda_n(1 - x_c)] \right\}} \exp(-\lambda_n^2 \tau) \quad (14b)$$

It should be noted that Li et al. [2003a] reported the same solution for the conventional batch adsorber.

The third method to obtain the solution for the batch adsorber is through the differentiation of the solution for the CMF semi-batch adsorber. We know that in the Laplace domain the impulse response can be obtained directly from the step response via the multiplication of s , and that in the time domain the impulse response can be obtained directly from the step response via differentiation with respect to time. Hence, it is reasonable that Eq. (14a) is just the multiplication of s of Eq. (9b) and Eq. (14b) is just the differential of Eq. (10a).

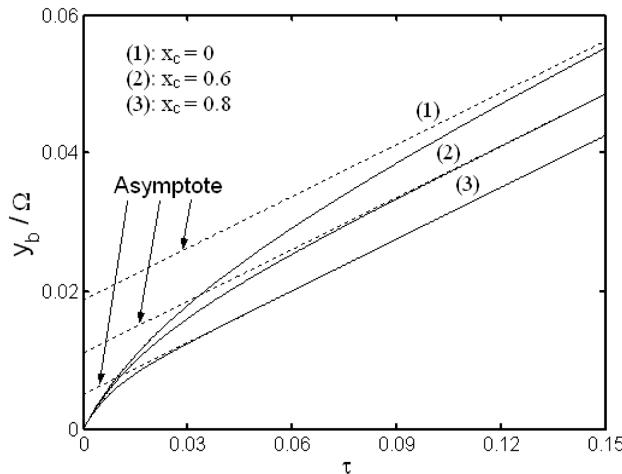


Fig. 1. Effect of inert core size x_c on the bulk concentration for the constant volume of outer adsorbent shell ($\beta=3$, $\xi=10$).

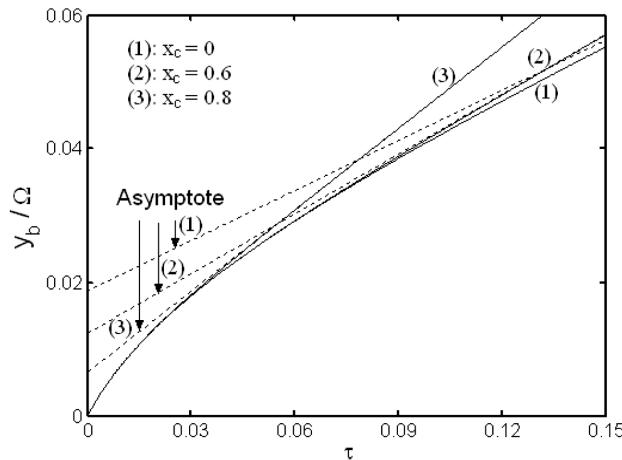


Fig. 2. Effect of inert core size x_c on the bulk concentration for the constant volume of total solid ($\beta_0=3$, $\xi=10$).

3. Simulation of Model

For the constant volume of the outer adsorbent shell, the equilibrium amount of adsorption remains unchanged even when the radius of the inert core changes. This is shown in Fig. 1 for $x_c=0$, 0.6, and 0.8. As we can see in Fig. 1, all the slopes of the asymptotes are same, but the intercepts are different. The intercept is higher for smaller x_c . This is because the diffusion time constant of the outer shell increases with the increase in the radius of the inert core. For the constant volume of the total solids, however, the equilibrium amount of adsorption decreases with increase in the radius of the inert core, which leads the steeper asymptote. This is shown in Fig. 2 for $x_c=0$, 0.6, and 0.8. As we can see in Fig. 2, the intercept is higher for smaller x_c . This is because the slope of the asymptote increases with decreasing x_c , as we can see in Eqs. (10b) and (10d).

Fig. 3 shows the effect of the equilibrium parameter β when the inert core radius x_c is constant. In this case the increase in β means an increase in the amount of adsorbents, which leads to a decrease in both the bulk phase concentration and the slope of the asymptote.

The effect of Biot number is shown in Fig. 4 for the constant values of β and x_c . Since the amount of adsorbents is constant, all the

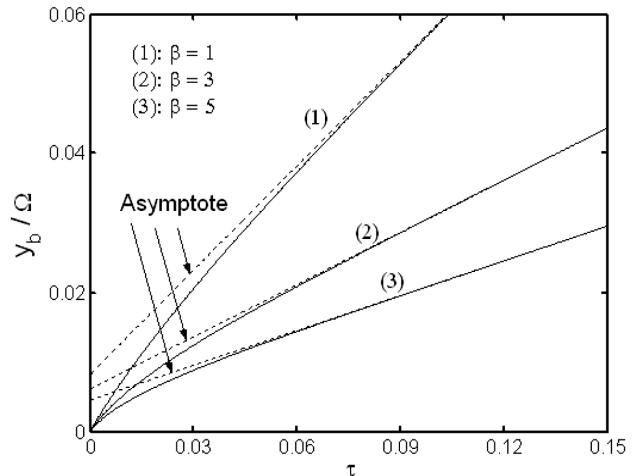


Fig. 3. Effect of the distribution parameter β on the bulk concentration for the constant size of inert core ($x_c=0.6$, $\xi=\infty$).

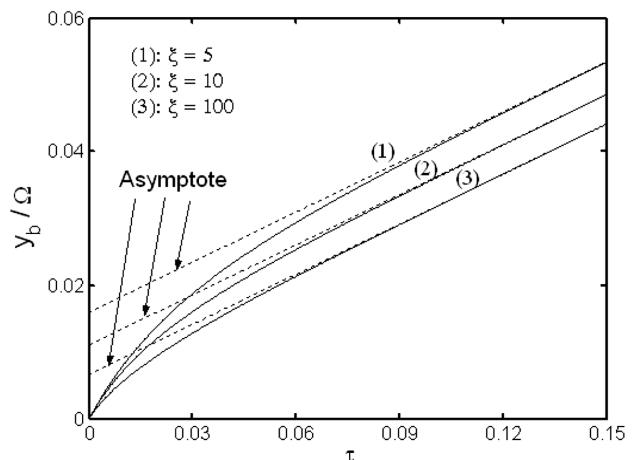


Fig. 4. Effect of the film resistance parameter ξ on the bulk concentration for the constant size of inert core ($x_c=0.6$, $\beta=3$).

slopes of asymptotes shown in Fig. 4 remain unchanged. However, the intercept becomes smaller for higher value of ξ (i.e., for smaller external film diffusion resistance).

CONCLUSIONS

An analytical solution for a constant molar flow semi-batch adsorber loaded with inert core adsorbents is obtained. The inert core adsorbent consists of the outer adsorbent shell and the inert core. As the radius of the inert core increases, the capacity of the outer adsorbent shell decreases, which leads to a decrease in the total amount of adsorption capacity of the adsorbents loaded in the adsorption vessel.

The solution obtained in this study consists of two parts. One is the asymptote, which the bulk concentration approaches to as time is sufficiently large; the other is the relaxation term, which dictates how the mass in the bulk phase flows into the adsorbent within the vessel. For a given value of the inert core radius, the slope of the asymptote is a function of the equilibrium parameter only, and the intercept is a function of the kinetic parameters as well as the equi-

librium parameter. The intercept of the asymptote consists of two parts: One is the contribution due to the external film resistance, the other is the contribution due to the intraparticle diffusion resistance.

In the three limiting cases (i.e., $\xi \rightarrow \infty$, $x_c \rightarrow 0$, and $x_c \rightarrow 1$) considered in this study, all the quantities included in the solution have physically sound limiting values. The solution for the conventional batch adsorber can be readily obtained by the differentiation of the solution for the CMF semi-batch adsorber.

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NOMENCLATURE

C_b	: concentration in the bulk phase [mol/m ³]
C_0	: reference concentration, on which dimensionless concentration is based [mol/m ³]
C_s	: concentration in the bulk phase at the particle surface [mol/m ³]
C_μ	: concentration in the adsorbed phase [mol/m ³]
D_e	: effective diffusivity, defined by Eq. (1a) [m ² /s]
$F(s)$: function of s, defined by Eq. (8)
$G(s)$: function of s, defined by Eq. (9a)
k_f	: external film mass transfer coefficient, defined by Eq. (1d) [m/s]
K	: dimensionless Henry constant of the linear isotherm, defined by Eq. (3)
\dot{N}	: molar flow rate of adsorbate into vessel [mol/sec]
q	: square root of s
r	: radius of adsorbent particle [m]
R	: radius of adsorbent particle [m]
R_c	: radius of inert core [m]
s	: dimensionless Laplace domain variable
t	: time variable [s]
$U(t)$: dimensionless unit step function
$V, V_\mu, V_{\mu 0}$: volume of free space, outer adsorbent shells, and total particles including inert cores within the adsorption vessel [m ³]
x	: dimensionless radial variable within the particle, defined in Table 1
x_c	: dimensionless radius of inert core, defined in Table 1
X	: dimensionless unit forcing function
y_b	: dimensionless concentration in the fluid phase, defined in Table 1
y_s	: dimensionless concentration in the fluid phase at the particle surface, defined in Table 1
y_μ	: dimensionless concentration in the particle phase at the particle surface, defined in Table 1
$\langle y_\mu \rangle$: mean value of y_μ

Greek Letters

β	: dimensionless distribution parameter, defined in Table 1
β_0	: dimensionless distribution parameter at $x_c \rightarrow 0$, defined in Table 1

δ_b , δ_a , δ_f	: dimensionless parameter, defined by Eqs. (10d), (10e), and (10f), respectively
$\delta(t)$: dimensionless Dirac delta function
λ_n	: the n-th eigenvalue, defined in Eq. (11)
τ	: dimensionless time variable, defined in Table 1
ξ	: dimensionless film resistance parameter, defined in Table 1
Ω	: dimensionless inlet molar flow rate parameter, defined in Table 1

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