Single Effective Diffusivities for Dynamic Adsorption in Bidisperse Adsorbents

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To be realistic, diffusion in bidisperse porous structures is often considered in terms of macropore diffusion and micropore diffusion. In this approach the diffusivity and the mass balance are defined separately for each pore system. Ruckenstein et al. (1971) and many investigators (Haynes and Sarma, 1973; Hashimoto and Smith, 1973; Hashimoto et al., 1976; Raghavan and Ruthven, 1985) have used this approach to analyze dynamic diffusion and adsorption in biporous adsorbents, and proposed their respective mathematical models. Nevertheless, it is not uncommon to lump both macropore and micropore diffusion together and model the internal diffusion with a single effective diffusivity as if the pore size distribution is unimodal (Eberly, 1969; Frost, 1974; Dogu and Smith, 1976; Baiker et al., 1982; Wang and Smith, 1983), since the resulting mathematical model is much simpler, and easier to handle than the realistic models. The problem in the latter approach, however, is lack of theoretical basis bridging the diffusion theories for bidisperse and monodisperse pore structures, and thus clear physical meaning cannot be attached to the single effective diffusivity. Because of this, the single diffusivity, once measured, is useful only for the identical system and conditions for which the measurement has been made. It has been demonstrated that the single diffusivity can be related to the macropore diffusivity, the micropore diffusivity, or a combination of both diffusivities (Haynes and Sarma, 1973; Furusawa and Smith, 1973), but the relationship between the single effective diffusivity and the macropore and micropore diffusivities has not been clarified.

The object of this study is thus to present formulas that relate the single effective diffusivity to the macropore and micropore diffusivities for dynamic adsorption in a biporous adsorbent.

Derivation of Formulas

A frequently employed method in estimation of the diffusivities in porous adsorbents is the method of moments. In this method, pulses of adsorbing gas are injected into a fixed bed, and from the moments of the response peaks the parameters are extracted. The method is well developed for monodisperse porous solids (Suzuki and Smith, 1971) as well as for bidisperse porous solids (Haynes and Sarma, 1973; Hashimoto and Smith, 1973). Regardless of the pore structure, whether monodisperse or bidisperse, the internal diffusivities are usually extracted from the second moment of the peaks.

When bidisperse porous material is involved and diffusion in the material is described with macropore and micropore diffusivities, then the contribution of the internal diffusion process to the second moment will be expressed in terms of the two diffusivities. But, on the other hand, if the internal diffusion in the same material is described with an effective diffusivity, then the effective diffusivity alone will have to account for the contribution of the internal diffusion to the second moment. In both cases, the contribution, whether expressed with the two diffusivities or with the single diffusivity, should be the same. And this requirement can yield a relation between the macropore and micropore diffusivities and the single effective diffusivity. We seek such relation from moments for plug flow adsorbers.

Diffusion models with macropore and micropore diffusivities

We consider a tubular adsorber in which bidisperse porous adsorbent pellets are packed. We assume the pellets are spheres of uniform size and are made of microporous spheres. We also consider mass transfer between the flowing phase and the outer surface of the adsorbent pellets. When the internal diffusion is described with the macropore and micropore diffusivities, the dimensionless mass balances for the flowing phase, for the macropore, and for the micropore are

$$\frac{\partial C_2}{\partial t} = -\frac{\partial C_2}{\partial z} - \frac{3(1-\epsilon)}{\epsilon}\frac{\theta D_a}{R_a^2}\frac{\partial C_a}{\partial x}\bigg|_{x=1} \tag{1}$$

$$\epsilon_a\frac{\partial C_a}{\partial t} = \frac{\theta D_a}{R_a^2}\frac{1}{x^2}\frac{\partial}{\partial x}\left(x^2\frac{\partial C_a}{\partial x}\right) - 3(1-\epsilon_a)\frac{\theta D_b}{R_b^2}\frac{\partial C_b}{\partial y}\bigg|_{y=1} \tag{2}$$

$$\epsilon_b\frac{\partial C_b}{\partial t} = \frac{\theta D_b}{R_b^2}\frac{1}{y^2}\left(y^2\frac{\partial C_b}{\partial y}\right) - \frac{\partial Q}{\partial t} \tag{3}$$

The equations apply to biporous adsorbent pellets in which the size of the micropores are much larger than the size of diffusing molecules so that the diffusing phase and the adsorbed phase are discernible in the micropores. Adsorbent pellets such as alumina and silica belong to this class. In zeolite pellets, however, the micropores are so small that the diffusing molecules are never free from the force field of the atoms that make up the pore wall, and the solid diffusion equation, Eq. 11, is usually used in the modeling. It will be dealt with later in this section.

For the adsorption, we assume equilibrium with a linear isotherm. The absorbed phase concentration, Q, based on the microparticle volume is

$$Q = KC_b \tag{4}$$

The initial conditions are

$$C_2(z,0) = 0,\ C_a(x,z,0) = 0,\quad C_b(x,y,z,0) = 0,\ Q(x,y,z,0) = 0 \tag{5}$$

and the boundary conditions are

$$C_2(0,t) = \delta(t),\ C_2(\infty,t) = 0 \tag{6}$$

$$C_2 - C_a|_{x=1} = \frac{D_a}{k_f R_a}\frac{\partial C_a}{\partial x}\bigg|_{x=1},\quad \frac{\partial C_a}{\partial x}\bigg|_{x=0} = 0 \tag{7}$$

$$C_a = C_b|_{y=1},\quad \frac{\partial C_b}{\partial y}\bigg|_{y=0} = 0 \tag{8}$$

$\delta(t)$ in Eq. 6 denotes the unit pulse input of the absorbing gas at the adsorber inlet. The above model equations are formally identical to those proposed by Haynes and Sarma (1973). The moments can be readily obtained by solving the model in the Laplace domain and evaluating derivatives of the solution. The second moment is

$$\begin{aligned} m_2 &= \int_0^\infty t^2 C_2(1,t)\,dt \\ &= m_1^2 + \frac{2}{15}\frac{(1-\epsilon)}{\epsilon}\left[(1-\epsilon_a)(\epsilon_b+K)^2\frac{R_b^2}{\theta D_b} + [\epsilon_a + (1-\epsilon_a)(\epsilon_b+K)]^2\frac{R_a^2}{\theta D_a}\right] \\ &\quad + \frac{(1-\epsilon)}{\epsilon}[\epsilon_a + (1-\epsilon_a)(\epsilon_b+K)]^2\frac{2R_a}{3\theta k_f} \end{aligned} \tag{9}$$

where m_1 is the first moment and given by

$$m_1 = 1 + \frac{(1-\epsilon)}{\epsilon}[\epsilon_a + (1-\epsilon_a)(\epsilon_b+K)] \tag{10}$$

In Eq. 9 the contribution of the internal diffusion to the second moment is expressed with the macropore and micropore diffusion parameters.

For zeolite-type adsorbents the following solid diffusion equation (Shah and Ruthven, 1977) is used for the mass balance in the micropores:

$$\frac{\partial Q}{\partial t} = \frac{\theta D_c}{R_b^2}\frac{1}{y^2}\frac{\partial}{\partial y}\left(y^2\frac{\partial Q}{\partial y}\right) \tag{11}$$

The mass balance for the macropores, Eq. 2, is also modified as

$$\epsilon_a\frac{\partial C_a}{\partial t} = \frac{\theta D_a}{R_a^2}\frac{1}{x^2}\frac{\partial}{\partial x}\left(x^2\frac{\partial C_a}{\partial x}\right) - 3(1-\epsilon_a)\frac{\theta D_c}{R_b^2}\frac{\partial Q}{\partial y}\bigg|_{y=1} \tag{12}$$

The boundary conditions for Q are

$$Q|_{y=1} = KC_a,\quad \frac{\partial Q}{\partial y}\bigg|_{y=0} = 0 \tag{13}$$

The mass balance for C_2, and the initial and boundary conditions for C_2 and C_a, are the same as in Eqs. 1 and 5–7. The second moment is

$$\begin{aligned} m_2' &= m_1'^2 + \frac{2}{15}\frac{(1-\epsilon)}{\epsilon}\cdot\left[(1-\epsilon_a)K\frac{R_b^2}{\theta D_c} + [\epsilon_a + (1-\epsilon_a)K]^2\frac{R_a^2}{\theta D_a}\right] \\ &\quad + \frac{(1-\epsilon)}{\epsilon}[\epsilon_a + (1-\epsilon_a)K]^2\frac{2R_a}{3\theta k_f} \end{aligned} \tag{14}$$

where m_1' is the same as m_1 in Eq. 10 with $\epsilon_b = 0$.

Diffusion model with a single effective diffusivity

A much simpler but evidently more unrealistic description of the internal diffusion in the bidisperse porous adsorbent involves modelling the diffusion with an overall diffusivity, D_p. In this approach, macropore concentration, C_a, and micropore concentration, C_b, are not distinguished, but both are lumped together and represented as the overall pore concentration, C_p. The dimensionless mass balances are

$$\frac{\partial C_1}{\partial t} = -\frac{\partial C_1}{\partial z} - \frac{3(1-\epsilon)}{\epsilon}\frac{\theta D_p}{R_a^2}\frac{\partial C_p}{\partial x}\bigg|_{x=1} \tag{15}$$

$$\epsilon_p\frac{\partial C_p}{\partial t} = \frac{\theta D_p}{R_a^2}\frac{1}{x^2}\frac{\partial}{\partial x}\left(x^2\frac{\partial C_p}{\partial x}\right) - \frac{\partial Q'}{\partial t} \tag{16}$$

where ϵ_p is the total internal void fraction which is given by

$$\epsilon_p = \epsilon_a + (1-\epsilon_a)\epsilon_b \tag{17}$$

However, for zeolitic diffusion,

$$\epsilon_p = \epsilon_a \tag{18}$$

because the solid diffusion equation, Eq. 11, regards the microparticles as homogenous solids and thus $\epsilon_b = 0$. The equilibrium relation, Eq. 4, modified for the absorbed phase concentration, Q', based on the adsorbent pellet volume is

$$\begin{aligned} Q' &= (1-\epsilon_a)KC_p \\ &= K'C_p \end{aligned} \tag{19}$$

The initial and boundary conditions for Eqs. 15 and 16 are

$$C_1(z, 0) = 0, C_p(x, z, 0) = 0, Q'(x, z, 0) = 0 \tag{20}$$

$$C_1(0, t) = \delta(t), \quad C_1(\infty, t) = 0,$$

$$C_1 - C_p\big|_{x=1} = \frac{D_p}{k_f R_a} \frac{\partial C_p}{\partial x}\bigg|_{x=1}, \quad \frac{\partial C_p}{\partial x}\bigg|_{x=0} = 0 \tag{21}$$

The second moment is

$$M_2 = m_1^2 + \frac{(1-\epsilon)}{\epsilon}(\epsilon_p + K')^2 \left[\frac{2R_a^2}{15\theta D_p} + \frac{2R_a}{3\theta k_f}\right] \tag{22}$$

Formulas for overall effective diffusivity from moments matching

In order to have the same second moments, it is necessary to match the contribution of the internal diffusion to the second moments of the two-diffusivity models with that of the single-diffusivity model. For alumina or silica adsorbents this leads to

$$\frac{R_a^2}{D_p} = \frac{R_a^2}{D_a} + \frac{(1-\epsilon_a)(K+\epsilon_b)^2}{[\epsilon_a + (1-\epsilon_a)(\epsilon_b + K)]^2} \frac{R_b^2}{D_b} \tag{23}$$

and for zeolite adsorbents,

$$\frac{R_a^2}{D_p} = \frac{R_a^2}{D_a} + \frac{(1-\epsilon_a)K}{[\epsilon_a + (1-\epsilon_a)K]^2} \frac{R_b^2}{D_c} \tag{24}$$

In practical tubular adsorbers, the flow surrounding the adsorbent pellets may not be represented as plug flow, and dispersion in the fluid phase may have to be accounted for. In such cases, the second moments for both diffusion models have additional terms representing the contribution of the dispersion to the moments. But the dispersion terms are independent of the terms related to the internal diffusion (see for example, Haynes and Sarma, 1973), and therefore the relations, Eqs. 23 and 24, still hold for dispersion-type packed-bed adsorbers. In fact, the relations are not dependent upon the phenomena occurring outside of the pellets, such as flow characteristics or external mass transfer, and are thus applicable to any type of flow adsorber.

Discussion

For zeolites, ϵ_b is zero because of the solid diffusion equation, Eq. 11, and if $D_b = KD_c$, Eq. 23 reduces to Eq. 24. This may mean that Eq. 24 is a special case of Eq. 23. Indeed, it can be proved that the pore diffusion model and the solid diffusion model are formally identical, provided that $D_b = KD_c$ and $K \gg 1$.

The adequacy of the formulas is now examined from the numerical comparisons of the two-diffusivity model and the single-diffusivity model. As a measure of the comparison, the integral of the square deviation between C_2 and C_1 is defined as

$$S(p_a, p_b) = \int_0^\infty [m_1 C_2(t; p_a, p_b) - m_1 C_1(t; p)]^2 \, d(t/m_1) \tag{25}$$

where $p_a = \theta D_a/R_a^2$, $p_b = \theta D_b/R_b^2$, and $p = \theta D_p/R_a^2$. In the case of adsorption, thus obtained, $S(p_a, p_b)$ can be seen to be virtually independent of the value of K because $m_1 C_1$ vs. t/m_1 is so whenever $K \gg 1$.

Figure 1 shows contours of $S(p_a, p_b)$ obtained with $K = 200$. The contour plot provides information about the adequacy of the model with the overall effective diffusivity. The magnitude of $S(p_a, p_b)$ increases sharply toward the region where p_a and p_b are small. The deviation between C_1 and C_2 becomes significant as $S(p_a, p_b)$ increases. This is shown in Figure 2 where C_1 and C_2 are compared for several levels of $S(p_a, p_b)$. Roughly, when $S(p_a, p_b)$ is greater than 10^{-3}, the agreement between C_1 and C_2 is less satisfactory, but when $S(p_a, p_b)$ is less than $10^{-3.5}$, the agreement is seen to be almost perfect. This indicates that the model with the overall effective diffusivity may not correctly characterize diffusion in biporous adsorbents in the region where $S(p_a, p_b)$ is greater than 10^{-3}, but in the remaining region the simple single-diffusivity model can be advantageously employed instead of the complex two-diffusivity models.

In the case of adsorption it is seen that ϵ_b has little influence on the predictions of the models and thus on $S(p_a, p_b)$, but the effect of ϵ_a on the predictions as well as on $S(p_a, p_b)$ seems apparent. As implied by Eq. 23, when $S(p_a, p_b)$ was plotted against p_a and $(1 - \epsilon_a)p_b$, the contour maps for different values of ϵ_a were found to coincide. Thus the contour map in Figure 1 can be used for judging the applicability of the overall diffusivity formulas for a wide variety of ϵ_a, ϵ_b, and K. The marked region in the figure denotes the parameter ranges where characterization of the internal diffusion in biporous adsorbents with the overall diffusivity may be inadequate.

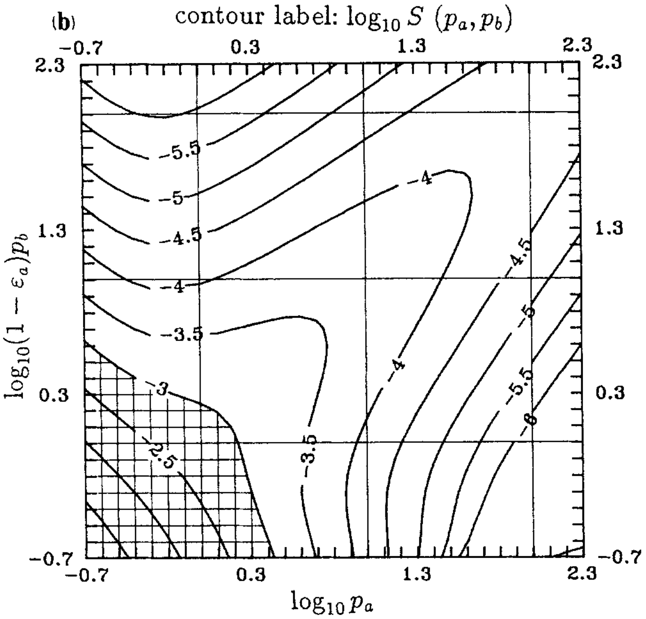

Figure 1. Contours of $S(p_a, p_b)$, adsorption cases.
$\epsilon = 0.4$, $\epsilon_a = 0.3$, $K = 200$, $k_f = \infty$

Except when the adsorber is extremely short in length, or flow through the bed is extremely fast, the set of values $[p_a, (1 - \epsilon_a)p_b]$ for a majority of operating conditions is likely to be located outside of the marked region in the contour map of Figure 1. A brief survey of diffusion data compiled in Table 1 may illustrate this. The listed values are p_a/θ and p_b/θ, and obviously if the space time of the fluid flowing through the bed θ is not excessively small, on the order of one second or less, all of

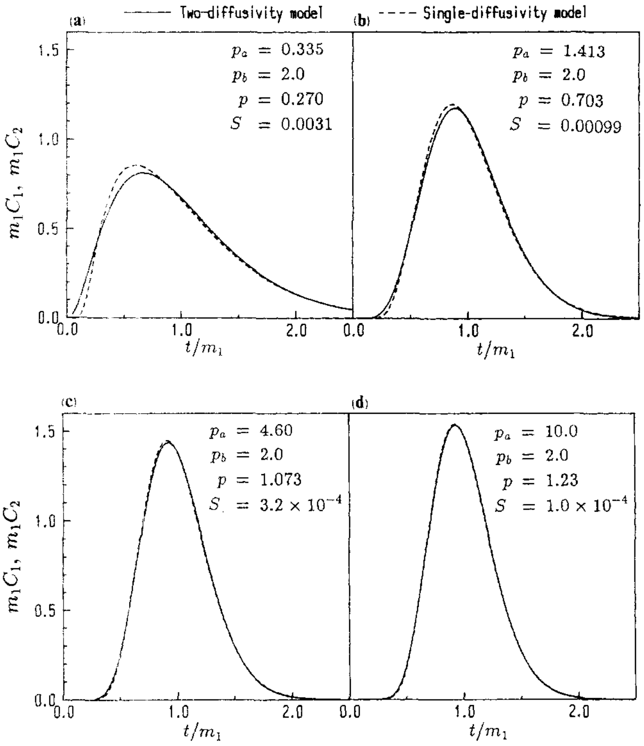

Figure 2. Pulse responses calculated from single-diffusivity model vs. two-diffusivity model.
$\epsilon = 0.4, \epsilon_a = 0.3, K = 200, k_f = \infty$

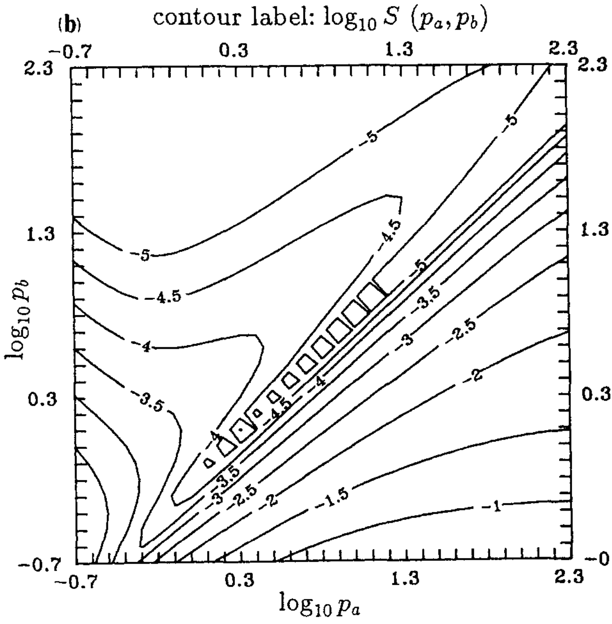

Figure 3. Contours of $S(p_a, p_b)$, no-adsorption cases.
$\epsilon = 0.4, \epsilon_a = 0.3, \epsilon_b = 0.4, K = 0, k_f = \infty$

the data can be well outside the inadequate region for applying the overall effective diffusivity formulas.

Figure 3 shows contours of $S(p_a, p_b)$ when $K = 0$. In contrast to the case of adsorption, deviation between the two-diffusivity model and single-diffusivity model progressively increases with increasing p_a. This means that the single diffusivity as determined by Eq. 23 can not adequately characterize the diffusion of a nonadsorbing gas in biporous solids when micropore diffusion controls. For various sets of (ϵ_a, ϵ_b), different values of $S(p_a, p_b)$ have been obtained, but the trend in S for varying p_a and p_b has been the same as that in Figure 3. A simple criterion for applicability of the formula, Eq. 23, could be stated as:

$$p_b > p_a \tag{26}$$

Table 1. A Survey on p_a/θ and p_b/θ

Adsorbate	Adsorbent	Temp. °C	D_a/R_a^2 sec^{-1}	D_b/R_b^2 sec^{-1}	KD_c/R_b^2 sec^{-1}	K	Reference
n-butane	alumina	30	11.3	12.5		893	Hashimoto and Smith (1974)*
n-butane	alumina	50	13.2	14.5			Hashimoto and Smith (1974)*
n-butane	alumina	75	16.2	18			Hashimoto and Smith (1974)*
ethane	zeolite 5A	25			5.8-11.4	860	Shah and Ruthven (1977)**
propane	zeolite 5A	125			5.4-9.4	720	Shah and Ruthven (1977)**
cyclopropane	zeolite 5A	125			0.53-5.6	1555	Shah and Ruthven (1977)**
propane	zeolite 5A	75			7.5	1300	Chiang et al. (1984)†
propane	zeolite 5A	100			12.2	518	Chiang et al. (1984)†
propane	zeolite 5A	125			17.5	280	Chiang et al. (1984)†
propane	zeolite 5A	150			14	160	Chiang et al. (1984)†
n-butane	zeolite 5A	175			4.8	583	Chiang et al. (1984)†
n-butane	zeolite 5A	200			3.6	318	Chiang et al. (1984)†
n-butane	zeolite 5A	250			3.1	130	Chiang et al. (1984)†
n-butane	zeolite 5A	300			7.7	56	Chiang et al. (1984)†
ethane	zeolite 13X	25	4.9		67	197	Hyun and Danner (1985)††
ethane	zeolite 13X	100	6.28		196	92	Hyun and Danner (1985)††
ethylene	zeolite 13X	25	5.08		34.4	246	Hyun and Danner (1985)††
ethylene	zeolite 13X	50	5.59		47	235	Hyun and Danner (1985)††
ethylene	zeolite 13X	100	6.19		98	185	Hyun and Danner (1985)††

*R_a = 0.0537 cm, R_b = 0.0057 cm
**Data varied with sorbate concentration
$^{\dagger}R_b$ = 0.00015 cm
$^{\dagger\dagger}R_a$ = 0.046 cm, R_b = 0.00015 cm

Except for zeolites, the condition of Eq. 26 would be satisfied in most practical cases. However, it is noted that the case of no adsorption with zeolites is rare.

With the presented formulas, the linear driving force (LDF) approximation can be used to further simplify the description of dynamic adsorption in biporous adsorbents. In fact, the LDF expressions were formally applicable to dynamic adsorption in monoporous adsorbents (Kim, 1989). But the overall effective diffusivity determined by the formulas allows the use of the LDF approximation. The simplest expression from the approximation is

$$\frac{d\overline{C}_p}{dt} = 15\frac{D_p}{R^2_a[\epsilon_p + 1 - \epsilon_a)K]}(C_1 - \overline{C}_p) \qquad (27)$$

where $\overline{C}_p$ *is the volume-averaged* C_p in the adsorbent. With the expression, the mass exchange rate per unit volume of the adsorbent $[\epsilon_p + 1 - \epsilon_a)K]d\overline{C}_p/dt$.

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Notation

C_1 = adsorbate concentration in bed void in the single-diffusivity model
C_2 = adsorbate concentration in bed void in the two-diffusivity model
C_a = adsorbate concentration in macropore
C_b = adsorbate concentration in micropore
C_p = overall adsorbate concentration in adsorbent pellet
D_a = macropore diffusivity based on total void plus and novoid area of absorbent pellet
D_b = micropore diffusivity based on total void plus and novoid area of microparticle in adsorbent
D_c = solid phase diffusivity in zeolite crystal
D_p = overall effective diffusivity based on total void plus and novoid area of adsorbent pellet
K = adsorption equilibrium constant based on microparticle volume
$K' = (1 - \epsilon_a)K$, adsorption equilibrium constant based on adsorbent pellet volume
k_f = mass transfer coefficient (adsorbent-fluid)
m_1 = first moment of the two-diffusivity model for adsorbents except zeolite, defined in Eq. 10
M_2 = second moment of the single-diffusivity model, defined in Eq. 22
m_2 = second moment of the two-diffusivity model for adsorbents other than zeolite, defined in Eq. 9
m_2' = second moment of the two-diffusivity model for zeolites, defined in Eq. 14
$p = \theta D_p/R_a^2$, dimensionless overall diffusivity parameter
$p_a = \theta D_a/R_a^2$, dimensionless macropore diffusivity parameter
$p_b = \theta K D_c/R_b^2$ for zeolites or $\theta D_b/R_b^2$ for other adsorbents, dimensionless micropore diffusivity parameter
Q = adsorbed-phase concentration based on microparticle volume
Q' = adsorbed-phase concentration based on adsorbent pellet volume
R_a = radius of adsorbent pellet
R_b = radius of microparticle in adsorbent pellet
$S(p_a, p_b)$ = integral of square deviation, defined in Eq. 25
t = dimensionless time, real time divided by θ
x = dimensionless radial variable in adsorbent pellet
y = dimensionless radial variable in microparticle
z = dimensionless axial variable in adsorber bed

Greek letters

ϵ = bed porosity
ϵ_a = macropore porosity
ϵ_b = micropore porosity
ϵ_p = overall porosity: ϵ_a for zeolites; $\epsilon_a + (1 - \epsilon_a)\epsilon_b$ for other adsorbents
θ = space time of the flowing phase in the bed

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