Mass Transfer from a Microsphere to a Fluid in a Fixed Bed

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Predicting particle-to-fluid mass-transfer rates in fixed beds has been the subject of many theoretical and experimental works. Two approaches have been used to model the mechanism of this transfer process. One is related to the concept of "channeling," whose models are represented by the Kuni and Suzuki equation (1967) that predicts $Sh_p \sim Pe_p$ when $Pe_p < 10$. "Cell" models are an example of the other modeling technique. The pioneer work is the Pfeffer model (1964), which predicts $Sh_p \sim Pe_p^{1/3}$ in the low Reynolds number-high Peclet number range. Later, Kawaze and Ulbrecht (1981a,b) used the same concept to describe the mass transfer with non-Newtonian fluids in fixed and fluidized beds. The Nelson and Galloway transfer relationship (1975) could be assigned to the group of "cell" models too. It is applicable to the Reynolds numbers from 0.01 to 100.

In this study, we attempt to develop a new simple model for the mass transfer from a solid microsphere to a fluid moving through a fixed bed. The proposed approximate theoretical expression shows that Sh_p is proportional to Pe_p in the range of high Reynolds and high Peclet numbers. The coefficient of that proportionality depends on Pe_p, specifically on the bed parameters.

Analysis

We examine the mass transfer from a microsphere to a fluid moving through a fixed bed composed of inert and active particles of comparable diameters. We assume that the quantity of inert microspheres is large enough to ensure that each active particle contacts only with inert particles. The idealized surrounding of a single active particle is shown in Figure 1. This structure suggests a macroscopic mass balance expressed as:

$$V_z' \cdot (\bar{c}_1 - \bar{c}_0) \cdot (2d_p)^2 = k \cdot \int_s (c^* - c)\,ds \tag{1}$$

As the first and very simplified approximation, we assume that the active particle surrounding consists of three contiguous cells: inert ideal mixing cell, followed by an active plug flow cell, and then inert ideal mixing cell. This is shown in Figure 1. It is well known that when the ratio between bed length and the particle diameter is higher than 50 and that of bed diameter to particle diameter higher than 10, the flow through the fixed bed is very similar to the ideal plug flow. Thus, for a fixed bed composed of particles with diameter in the order of 1 mm and smaller, the hydrodynamic picture inside the bed can be described with the help of the ideal plug-flow model. When the bed packing is composed of inert and active particles, changes in concentration occur only when the fluid passes near the active surfaces. Thus, for plug flow conditions:

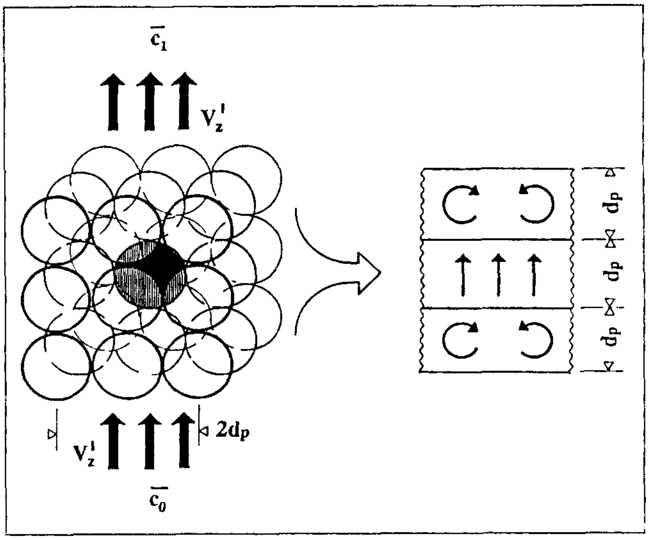

Figure 1. Surrounding of the active particle.

$$c = c^* - (c^* - \bar{c}_0) \cdot \exp[-k \cdot (a/V_z') \cdot z] \tag{2}$$

Expanding the dimensionless concentration in a Taylor series, we obtain:

$$(c^* - c)/(c^* - \bar{c}_0) = \{1 - k \cdot (a/V_z') \cdot z + [k \cdot (a/V_z') \cdot z]^2 \cdot (1/2!) - [k \cdot (a/V_z') \cdot z]^3 \cdot (1/3!) + \dots\} \tag{3}$$

When the fluid passes through the active cell, z changes from

0 to d_p. Then, (z/d_p) varies within the range 0–1, and for k/V_z' $(Sh_p/Pe_p') < 10^{-1}$ the third and the following terms in Eq. 3 can be neglected. For the active cell alone, the specific interfacial area is given by $[4\pi r_p^2/(4d_p^2 \cdot d_p)]$. Thus:

$$\int_s (c^* - c)ds = 2\pi r_p^2 (c^* - \bar{c}_0) \cdot \int_0^{\pi} [1 - (\pi/8) \cdot (k/V_z') \cdot (1 - \cos\theta)] \cdot \sin\theta \cdot d\theta = [c^* - (\bar{c}_1 + \bar{c}_0)/2] \cdot 4\pi r_p^2 \tag{4}$$

Under these conditions, the macroscopic mass balance in terms of dimensionless variables becomes:

$$Sh_p = \beta \cdot Pe_p'$$

with

$$\beta = (4/\pi) \cdot (\bar{c}_1 - \bar{c}_0)/[c^* - (\bar{c}_1 + \bar{c}_0)/2] \tag{5}$$

Due to the presence of adjacent inert particles, it is difficult to predict the velocity distribution of the flow in the neighborhood of the active particle. When the particle diameter is small ($d_p < 1$ mm), void spaces are narrow and it is hard to accept that the hydrodynamic picture could be represented by a suitably modified flow field around one isolated sphere. The sphericity of the particles may not influence the transport process. Assuming that molecular diffusion occurs only in the direction perpendicular to the flow, one can apply the equation of continuity only for the fluid phase in the form:

$$V_z' \cdot (\delta c/\delta z) = D \cdot (\delta^2 c/\delta x^2) \tag{6}$$

As discussed above, we assume $V_z' = \text{const}$. In fact, in a packed bed, a transition occurs from "laminar" to a "turbulent flow regime" at $Re_p > 5$ (Ergun, 1952). At higher Reynolds numbers, a thin velocity boundary layer exists near the particle surface. As the active particle contacts inert particles, the boundary layer deforms, therefore the assumption of a plug-flow regime through the active cell seems to be acceptable as the first approximation. Nevertheless, for high Peclet numbers ($Pe_p \gg 1$), a diffusional boundary layer does exist near the particle surface. The change in concentration occurs entirely in the thin diffusional boundary layer. Outside of it, the concentration remains unchanged and equal to $\bar{c}_0$. It means that the diffusing spaces do not feel the presence of inert neighboring solid surfaces. Thus, the boundary conditions to solve Eq. 6 are:

$$\begin{aligned} &\text{at } x = 0 \quad c = c^* \text{ for all } (z) \\ &\text{at } x \sim \infty \quad c = \bar{c}_0 \text{ for } z > 0 \\ &\text{for } z < 0 \quad c = \bar{c}_0 \text{ for all } (x) \end{aligned} \tag{7}$$

So the solution of the diffusion equation becomes (Bird et al., 1960):

$$(c - \bar{c}_0)/(c^* - \bar{c}_0) = 1 - \text{erf}\,[x/(4Dz/V_z')^{1/2}] \tag{8}$$

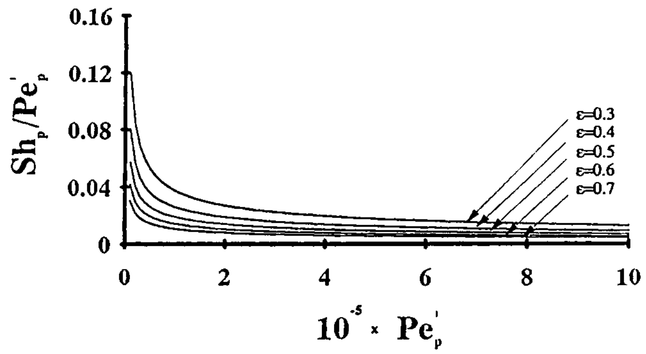

Figure 2. "β" vs. Pe_p' number with "ϵ" as parameter ($d_p = 0.5$ mm).

Since the distance between adjacent particles varies, an average distance L can be introduced and evaluated from:

$$L = 2r_p \cdot [1 - (1-\epsilon)^{1/3}]/(1-\epsilon)^{1/3} \tag{9}$$

Thus, the average concentration $\bar{c}_1$ becomes:

$$\bar{c}_1 = c^* - (c^* - \bar{c}_0) \cdot (1/L) \cdot \int_0^L \text{erf}[(x/2d_p) \cdot (Pe_p')^{1/2}] \cdot dx \tag{10}$$

From Eqs. 5 and 10, one finally obtains:

$$Sh_p = 2{,}54 \cdot \frac{1 - (1/L) \cdot \int_0^L \text{erf}\,[(x/2d_p) \cdot (Pe_p')^{1/2}] \cdot dx}{1 + (1/L) \cdot \int_0^L \text{erf}\,[(x/2d_p) \cdot (Pe_p')^{1/2}] \cdot dx} \cdot Pe_p' \tag{11}$$

A plot of Eq. 11 for different values of the void volume is given in Figure 2. The enhancement of mass transfer with increased fluid velocity is due to the decrease in the diffusional boundary layer thickness δ_c. Beyond the diffusional boundary layer, $c = \bar{c}_0$. Thus, from Eq. 8, $(x)_{c=\bar{c}_0} = \delta_c = 4 \cdot (Dz/V_z')^{1/2}$, or for $z = d_p$, $\delta_c = 4d_p/(Pe_p')^{1/2}$ and the region of validity of Eq. 11 is $4d_p/(Pe_p')^{1/2} \ll L$ or $Pe_p' \gg 16 \cdot \{(1-\epsilon)^{1/3}/[1-(1-\epsilon)^{1/3}]\}^2$.

Figure 3 compares Eq. 11 with available experimental data

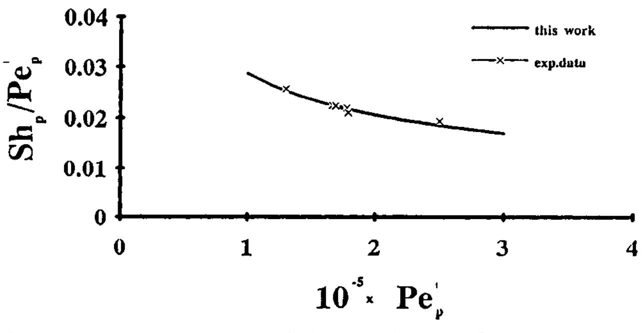

Figure 3. Comparison of the predicted (Eq. 11) and experimental values of β.

Data from Doytcheva et al. (1988): dissolution of polystyrene beads ($d_p = 0.782$ mm) in ethyl acetate ($\epsilon = 0.37$, $T = 25$°C).

(Doytcheva et al., 1988). The values of the polymer diffusivity were evaluated by minimizing the sum of square deviations of the Sh_p/Pe_p' ratio, calculated using Eq. 11, from the experimental k/V_z' ratio. The assumed D value is 2.2×10^{-10} m^2/s. ($M \sim 10^4$, $T = 25°$C). This value is quite reasonable compared to 2.64×10^{-11} m^2/s. ($M \sim 7.7 \times 10^5$, $T = 27°$C) (Polymer Handbook, 1975). Thus, Eq. 11 provides a qualitatively good description of the mass transfer between a microsphere and a fluid under the above conditions.

Hershey and Brodkey (1988) show that in the case of creeping flow, $Sh \sim Pe^{1/3}$ and for high Reynolds numbers $Sh \sim Re^{1/2} Sc^{1/3}$. Our predictions differ from these classical relations. The mass-transfer coefficients calculated from Eq. 11 are much higher. Similar abnormally high values were recently discussed by Brodkey et al. (1991). They showed that the general magnitudes of the real particle-to-fluid heat-transfer coefficients in fluidized beds of fine particles ($d_p \sim 1$ mm) surpassed about 100 times those expected on the basis of the known correlations for Reynolds numbers of the order of 10. As noted by Brodkey et al., the effect of adjacent particles on the transfer process from a given particle is not well established, but it is agreed that the presence of neighboring particles facilitates the transfer. On the other hand, at high Reynolds numbers, the flow around a single sphere separates at $\theta = 109.6°$ (Schlichting, 1979), and the mass-transfer process is disturbed. In a fixed bed composed of fine particles due to the presence of the neighboring particles, all active surfaces may become almost equally accessible to the transported species. This would result in an enhancement of the mass-transfer rate.

Our analysis is restricted to the active plug-flow cell. This enables us to avoid discussion of the influence of tortuosity by solving an exact boundary value problem for the fluid phase alone. Of course, the geometry, and the hydrodynamics of the system of interest are oversimplified. Our analysis, however, provides the useful prediction that the mass transfer from a microsphere to a fluid in an assembly of inert microspheres is accomplished more rapidly compared with the mass transfer from a single microsphere to a fluid moving with uniform velocity. In consequence, models based on the concept of flow around a submerged particle cannot adequately describe the particle-to-fluid mass transfer in a fixed bed composed of inert and active microspheres. The comparison with the experimental data shows that Eq. 11 is applicable to Reynolds numbers in the order of 10 and higher.

Unfortunately, we were unable to find other mass-transfer data to check our predictions. Further work is underway in this direction.

Notation

a = specific active interfacial area (m^2/m^3)
c = solute concentration (mol/m^3)
$\bar{c}_0$ = average inlet solute concentration (mol/m^3)
$\bar{c}_1$ = average outlet solute concentration (mol/m^3)
c^* = solute solubility (mol/m^3)
d_p = particle diameter (m)
D = diffusivity (m^2/s)
k = particle-to-fluid mass-transfer coefficient (m/s)
L = average distance defined in Eq. 9
M = molecular weight (g/mol)
r_p = particle radius (m)
s = active interfacial area (m^2)
V_z = superficial velocity (m/s)
V_z' = actual average fluid velocity [$V_z' = V_z/$] (m/s)
x, y, z = independent coordinates (m)

Greek letters

β = coefficient defined in Eq. 5
δ = diffusional boundary layer thickness (m)
ϵ = void fraction
θ = angle in spherical coordinates
μ = viscosity (kg/m·s)
ρ = density (kg/m^3)

Dimensionless numbers

Pe_p = particle Peclet number [$V_z \cdot d_p/D$]
Pe_p' = particle Peclet number [$V_z' \cdot d_p/D$]
Re_p = particle Reynolds number [$V_z \cdot d_p/D$]
Sh_p = particle Sherwood number [$k \cdot d_p \cdot /D$]

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