Mass Transfer and Dispersion Around Active Sphere Buried in a Packed Bed

João R. F. Guedes de Carvalho and Manuel A. M. Alves
Faculdade de Engenharia da Universidade do Porto, Dept. de Engenharia Química, 4050-123 Porto, Portugal

Mass transfer around single spheres in granular beds of inerts (packed or fluidized) was analyzed for transport by advection and diffusion/dispersion. Fluid flow in the granular bed around the spheres was assumed to follow Darcy's law; at each point, dispersion of solute was assumed to be determined by the coefficients of transverse and longitudinal dispersion, respectively, in the cross-stream and streamwise directions. The elliptic equation resulting from a differential mass balance was solved numerically over a wide range of the relevant parameters. Resulting values of the Sherwood number are represented accurately by the product of two terms: the solution for advection plus molecular diffusion, which depends only on the Peclet number relative to the active sphere; the enhancement brought about by convective dispersion, which depends only on the Peclet number relative to the inert particles.

Introduction

This article deals with mass transfer from a sphere buried in a packed bed of inert particles, through which a uniform stream of gas flows, as sketched in Figure 1.

Early attempts (see, e.g., Leung and Smith, 1979; Chakraborty and Howard, 1981) to predict the mass-transfer coefficient were based on the analogy with fluid flow past an isolated sphere, for which the correlations of Ranz and Marshall (1952) and Brian and Hales (1969) apply. However, the two situations are very different from a hydrodynamic point of view and, as it happens, the flow field around the buried sphere is easier to treat, since there is no hydrodynamic boundary layer and no fluid separation (and therefore no wake) on the leeward side of the sphere.

Interest in this problem was renewed by the necessity of predicting the rate of combustion of char particles immersed in beds of sand (or ash), where combustion takes place mostly in the dense phase (Avedesian and Davidson, 1973). It has been known (van Heerden et al., 1953) for a long time that mass transfer in the dense phase of such beds occurs at the same rate as in a fixed bed of the same particles, but with gas flowing at the velocity of incipient fluidization. This can be seen as a corollary of the two-phase theory of fluidization (Davidson and Harrison, 1963), and recent experimental evidence supports this view (Prins et al., 1985; Coelho and Guedes de Carvalho, 1988b).

La Nauze et al. (1984) attempted a solution of the mass-transfer problem, proposing a mechanism that combined the processes of transient "dipping" and surface renewal, but their approach did not consider the detailed fluid mechanics of the gas flow. Prins et al. (1985) tried to establish a correlation of dimensionless groups, based on a large amount of experimental data, but since the number of independent variables involved is large, their result has limited applicability.

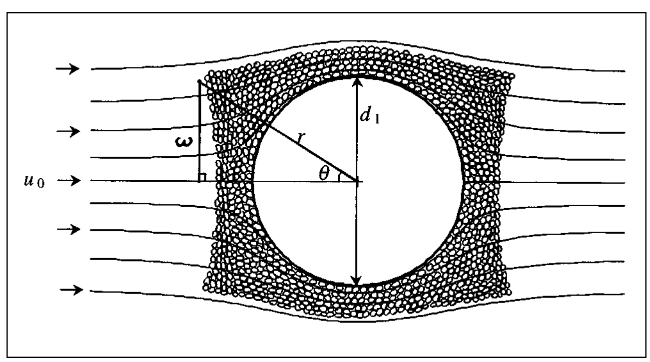

Figure 1. Flow around a sphere immersed in a bed of smaller particles.

Correspondence concerning this article should be addressed to J. R. F. Guedes de Carvalho.

Coelho and Guedes de Carvalho (1988b) were the first to produce a full analytical treatment of the problem, although their analysis was restricted to a thin mass-transfer boundary (i.e., to high Peclet numbers); their approach took into account the important role of transverse dispersion in mass transfer. The resulting asymptotic expression (valid for high Peclet numbers) was

$$\frac{Sh'}{\epsilon} = \left[1.28\,Pe' + 0.141(d/d_1)\,Pe'^2\right]^{1/2}, \tag{1}$$

where the meaning of the variables is given in the Notation list (in primed dimensionless groups, the effective coefficient of molecular diffusion, $D'_m = D_m/\tau$, is used).

This result was then combined with the well-known result, $Sh'/\epsilon = 2$ for molecular diffusion from a buried sphere into a stagnant medium (i.e., at $Pe' = 0$) and the general expression

$$\frac{Sh'}{\epsilon} = \left[4 + 0.576\,Pe'^{0.78} + 1.28\,Pe' + 0.141(d/d_1)\,Pe'^2\right]^{1/2} \tag{2}$$

was proposed. Middleman (1998) quotes this result as an empirical correlation, but it is worth emphasizing that the expressions obtained for the limiting conditions of both small and large Pe' were derived from first principles, without the introduction of any adjustable parameters. It is nevertheless true that for intermediate Pe', Eq. 2 represents an empirical correlation that has been successfully tested against a vast number of experimental data (Coelho and Guedes de Carvalho, 1988b; Pinto and Guedes de Carvalho, 1990; Guedes de Carvalho et al., 1991). In an effort to overcome this limitation, this work gives a full treatment of the mass-transfer problem, valid over the whole range of Pe'.

Theory

Our theory is based on the assumption that the inert particles in the bed are packed with uniform voidage, ϵ, and that the flow of gas may be approximated everywhere by Darcy's law

$$u = -K\,\mathbf{grad}\,p. \tag{3}$$

Furthermore, if the fluid is treated as incompressible, mass conservation leads to

$$\text{div}\,u = 0 \tag{4}$$

so Eqs. 3 and 4 can be combined to give Laplace's equation

$$\nabla^2\phi = 0, \tag{5}$$

where $\phi = Kp$. This result is well known to hydrologists (Scheidegger, 1974), and shows that incompressible Darcy flow through a packed bed follows the laws of potential flow. Darcy's law is strictly valid only for laminar flow through the packing, but according to Bear (1988), it is still a good approximation for values of the Reynolds number (based on superficial velocity) up to ~ 10, which for beds with $\epsilon \sim 0.4$ is equivalent to $Re_p \sim 25$, the upper limit for the validity of this analysis.

When a solid sphere of radius $R = d_1/2$ is immersed in an unbound packed bed of significantly smaller particles, through which fluid flows with uniform interstitial velocity u_0, far from the sphere, the solution to Eq. 5 is (Currie, 1993)

$$\phi = -u_0\left[1 + \frac{1}{2}\left(\frac{R}{r}\right)^3\right] r\cos\theta, \tag{6}$$

and the corresponding stream function is

$$\psi = \frac{u_0}{2}\left[1 - \left(\frac{R}{r}\right)^3\right] r^2 \sin^2\theta. \tag{7}$$

The velocity components will then be

$$u_r = \frac{\partial\phi}{\partial r} = -u_0\cos\theta\left[1 - \left(\frac{R}{r}\right)^3\right] \tag{8}$$

$$u_\theta = \frac{1}{r}\frac{\partial\phi}{\partial\theta} = u_0\sin\theta\left[1 + \frac{1}{2}\left(\frac{R}{r}\right)^3\right]. \tag{9}$$

To formulate the mass-transfer problem we take the saturation concentration of the diffusing species (the solute) to be c^* on the surface of the sphere and the bulk concentration, c_0, a large distance from it, in the approaching stream. The resulting concentration field will have axial symmetry, and the differential equation for mass transfer can be derived from a mass balance on the elementary volume shown in Figure 2. This differential volume is limited by the equipotential surfaces ϕ and $\phi + \delta\phi$ and by the stream surfaces ψ and $\psi + \delta\psi$, and if δS_ψ and δS_ϕ represent incremental lengths

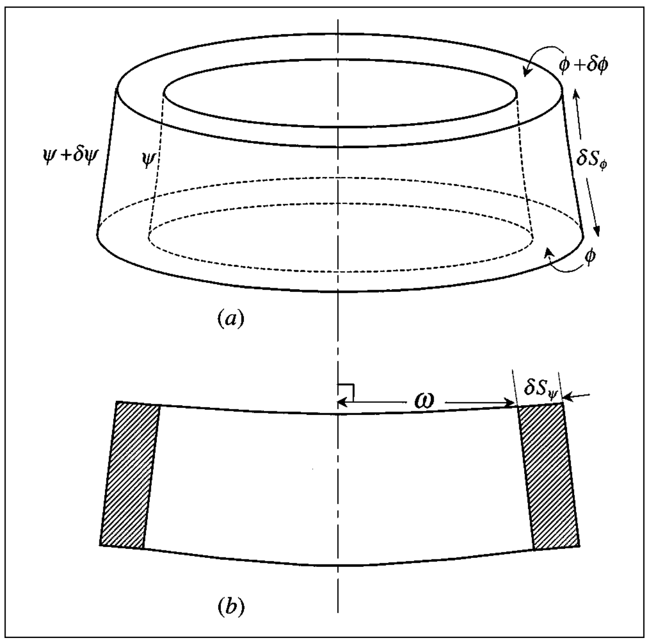

Figure 2. Differential volume for mass balance: (a) perspective; (b) section along the flow axis.

along a potential surface and a stream surface, respectively, two important equalities are

$$\delta S_{\phi} = \frac{1}{u}\delta\phi \tag{10}$$

$$\delta S_{\psi} = \frac{1}{\omega u}\delta\psi . \tag{11}$$

The contributions to solute transfer across the boundary of the volume element will be:

- Net outflow by convection

$$\frac{\partial}{\partial S_{\phi}}(\epsilon\, c 2\pi\delta\psi)\delta S_{\phi} = 2\pi\epsilon\delta\psi\frac{\partial c}{\partial S_{\phi}}\delta S_{\phi}. \tag{12}$$

- Net outflow by longitudinal dispersion

$$\frac{\partial}{\partial S_{\phi}}\left(-\epsilon D_L\frac{\partial c}{\partial S_{\phi}}\frac{2\pi\delta\psi}{u}\right)\delta S_{\phi} = -2\pi\epsilon\delta\psi\frac{\partial}{\partial S_{\phi}}\left(\frac{D_L}{u}\frac{\partial c}{\partial S_{\phi}}\right)\delta S_{\phi}. \tag{13}$$

- Net outflow by transverse dispersion

$$\frac{\partial}{\partial S_{\psi}}\left(-\epsilon D_T\frac{\partial c}{\partial S_{\psi}}2\pi\omega\delta S_{\phi}\right)\delta S_{\psi} = -2\pi\epsilon\frac{\partial}{\partial S_{\psi}}\left(D_T\,\omega\delta S_{\phi}\frac{\partial c}{\partial S_{\psi}}\right)\delta S_{\psi}. \tag{14}$$

In the absence of a chemical reaction, the three contributions must add to zero, and after manipulation of the resulting equation, one obtains

$$\frac{\partial c}{\partial\phi} = \frac{\partial}{\partial\phi}\left(D_L\frac{\partial c}{\partial\phi}\right) + \frac{\partial}{\partial\psi}\left(D_T\,\omega^2\frac{\partial c}{\partial\psi}\right). \tag{15}$$

If the mass transfer boundary layer around the sphere is thin (that is, for high Pe'), the first term on the righthand side of Eq. 15 may be neglected. That situation was considered by Coelho and Guedes de Carvalho (1988b), who arrived at Eq. 1. Here we wish to consider mass transfer for the whole range of Pe', so the term accounting for longitudinal dispersion will have to be considered and Eq. 15 will have to be integrated numerically. An important aspect to consider in the integration of Eq. 15 is the dependence of D_L and D_T on the flow parameters.

Much research has been done on the problem of solute dispersion in gas flow through packed beds (Gunn, 1987). There seems to be agreement (Wilhelm, 1962; Gunn, 1968; Coelho and Guedes de Carvalho, 1988a) in taking, as a good approximation for packed beds of nearly spherical mono-sized particles of diameter d,

$$D_L = D'_m + \frac{ud}{Pe_L(\infty)} \tag{16}$$

$$D_T = D'_m + \frac{ud}{Pe_T(\infty)}, \tag{17}$$

with $Pe_L(\infty) \cong 2$ and $Pe_T(\infty) \cong 10$ to 14. We shall take $Pe_T(\infty) = 12$, as suggested by Wilhelm (1962) and Coelho and Guedes de Carvalho (1988a). For dispersion in liquids, Eqs. 16 and 17 are not generally applicable (Gunn, 1987) over the entire range of flow conditions, and therefore the following treatment is strictly restricted to gas flow and certain limiting conditions of liquid flow described below. The work of Guedes de Carvalho and Delgado (1999) considers mass transfer to liquids outside these limiting conditions.

In order to integrate Eq. 15 with auxiliary Eqs. 6, 7, 16 and 17, it is convenient to define the dimensionless variables

$$C = \frac{c - c_0}{c^* - c_0} \tag{18}$$

$$U = \frac{u}{u_0} \tag{19}$$

$$\Re = \frac{r}{R} \tag{20}$$

$$\Phi = \frac{4}{3}\frac{\phi}{u_0 d_1} \tag{21}$$

$$\Psi = \frac{\psi}{u_0 d_1^2} \tag{22}$$

$$Pe' = \frac{u_0 d_1}{D'_m}. \tag{23}$$

Equation 15 can be rearranged to

$$\frac{\partial C}{\partial\Phi} - \frac{\partial}{\partial\Phi}\left[A\frac{\partial C}{\partial\Phi}\right] = \frac{\partial}{\partial\Psi}\left[B\frac{\partial C}{\partial\Psi}\right], \tag{24}$$

with

$$A = \frac{4}{3}\left(\frac{1}{Pe'} + \frac{U}{Pe_L(\infty)}\frac{d}{d_1}\right) \tag{25}$$

$$B = \frac{3}{16}\left(\frac{1}{Pe'} + \frac{U}{Pe_T(\infty)}\frac{d}{d_1}\right)\Re^2\sin^2\theta, \tag{26}$$

and the appropriate boundary conditions

$$\Phi \to -\infty, \quad \Psi \geq 0 \qquad C \to 0 \tag{27}$$

$$\Phi \to +\infty, \quad \Psi \geq 0 \qquad C \to 0 \tag{28}$$

$$\Psi = 0\begin{cases} -1 \leq \Phi \leq 1 & C = 1 \qquad (29a)\\ |\Phi| > 1 & \dfrac{\partial C}{\partial\Psi} = 0 \qquad (29b)\end{cases}$$

$$\Psi \to +\infty, \quad \text{all } \Phi \qquad C \to 0. \tag{30}$$

Discretization

To discretize Eq. 24 we adopted a second-order central difference scheme (CDS) in a nonuniform grid (Ferziger and Peric, 1996) as shown in Figure 3. The discretized equation

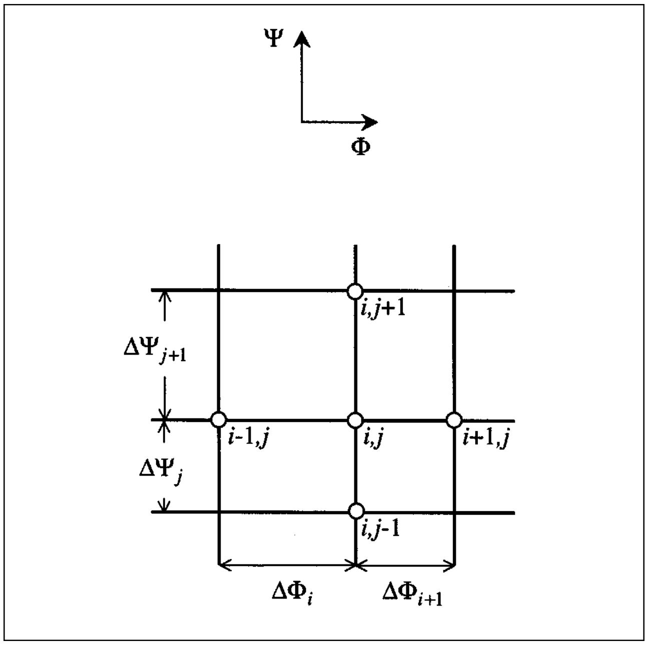

Figure 3. Computational grid.

resulting from the finite difference approximation of Eq. 24 reads

$$\left[1-\left(\frac{\partial A}{\partial \Phi}\right)_{i,j}\right] \times \frac{C_{i+1,j}(\Delta\Phi_i)^2 - C_{i-1,j}(\Delta\Phi_{i+1})^2 + C_{i,j}\left(\Delta\Phi_{i+1}^2 - \Delta\Phi_i^2\right)}{\Delta\Phi_i \Delta\Phi_{i+1}(\Delta\Phi_i + \Delta\Phi_{i+1})} - A_{i,j}\frac{C_{i+1,j}(\Delta\Phi_i) + C_{i-1,j}(\Delta\Phi_{i+1}) - C_{i,j}(\Delta\Phi_i + \Delta\Phi_{i+1})}{\Delta\Phi_i \Delta\Phi_{i+1}(\Delta\Phi_i + \Delta\Phi_{i+1})/2}$$
$$= \left(\frac{\partial B}{\partial \Psi}\right)_{i,j} \frac{C_{i,j+1}(\Delta\Psi_j)^2 - C_{i,j-1}(\Delta\Psi_{j+1})^2 + C_{i,j}\left(\Delta\Psi_{j+1}^2 - \Delta\Psi_j^2\right)}{\Delta\Psi_j \Delta\Psi_{j+1}(\Delta\Psi_j + \Delta\Psi_{j+1})} + B_{i,j}\frac{C_{i,j+1}(\Delta\Psi_j) + C_{i,j-1}(\Delta\Psi_{j+1}) - C_{i,j}(\Delta\Psi_j + \Delta\Psi_{j+1})}{\Delta\Psi_j \Delta\Psi_{j+1}(\Delta\Psi_j + \Delta\Psi_{j+1})/2}, \quad (31)$$

where the indices i and j refer to the potential and stream surfaces, respectively, with i taking the values 1 to n_Φ and j covering the range 1 to n_Ψ. The discretized equation was solved numerically by successive overrelaxation (SOR) (Smith, 1971), starting from an initial estimate obtained by the method outlined below; the relaxation coefficient was evaluated by trial and error. It was found that for low values of Pe', the iterative process was significantly accelerated using relaxation factors in the range 1.5 to 1.9, whereas for high values of Pe' the iterative process should start with a relaxation factor near unity, which was subsequently increased to accelerate convergence.

Every result was numerically computed in successively finer meshes, until the corresponding computed values, in two subsequent solutions, were coincident within less than 1%.

Boundary conditions

For a given Pe', the domain of integration was chosen so that $C < 10^{-4}$ along the boundary lines corresponding to $\Phi = \Phi_{\min}$ and $\Psi = \Psi_{\max}$, that is, $C_{1,j} < 10^{-4}$ for all j and $C_{i,n_\Psi} < 10^{-4}$ for all i; this is a satisfactory substitute for the boundary conditions corresponding to Eqs. 27 and 30.

The boundary condition corresponding to Eq. 28 was not used as such, for that would have required the use of very large values of $\Phi_{\max}$ (see Figure 4). Instead, a moderate value of $\Phi_{\max}$ was adopted (typically $\Phi_{\max} = 5$), with the corresponding grid value of C being estimated from the three neighboring concentrations (to the left) obtained in the previous iteration. A second-order interpolating polynomial was used for that purpose. Finally, the specification of the condition of symmetry, given as Eq. 29b, was approximated by means of a second-order interpolating polynomial (Anderson, 1995), so the concentration on the nodes along the axis of symmetry (j index equal to 1) is

$$C_{i,1}^{(n+1)} = \frac{C_{i,2}^{(n)}(\Psi_3 - \Psi_1)^2 - C_{i,3}^{(n)}(\Psi_2 - \Psi_1)^2}{(\Psi_3 - \Psi_1)^2 - (\Psi_2 - \Psi_1)^2}, \quad (32)$$

where the index (n) represents the iteration number.

Initial estimate

The initial estimate of $C_{i,j}$ was obtained by numerically solving the parabolic equation that results when the second term on the lefthand side of Eq. 24 is neglected; the numerical solution adopted followed the implicit Crank-Nicholson method (Anderson, 1995). The term neglected in Eq. 24 represents transport by longitudinal dispersion and for high Pe', its contribution is expected to be negligible; accordingly, under those circumstances, integration of the full equation gave virtually the same concentration distribution as was obtained

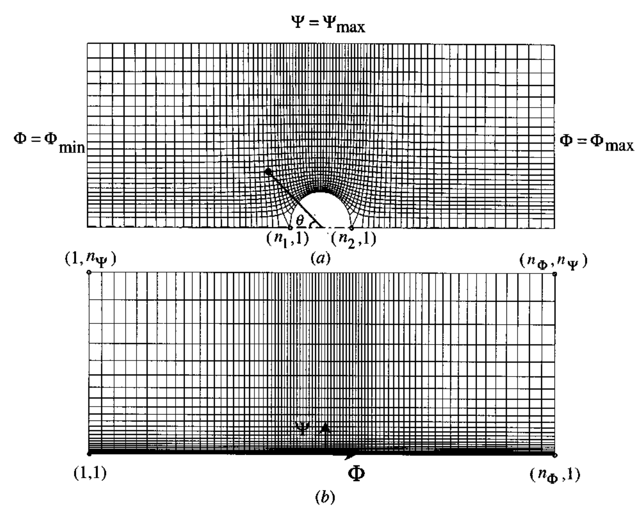

Figure 4. Domain of computation: (a) physical domain; (b) computational mesh.

In reality the mesh was more refined than represented in the sketch, and the values of $\Phi_{\min}$, $\Phi_{\max}$, and $\Psi_{\max}$ were chosen in each case as a function of Pe' and d/d_1.

from the simplified equation. For lower Pe' the solution of the parabolic equation merely gives an initial estimate for the iterative process.

Results

The numerical procedure adopted yields values of $C_{i,j}$, from which the overall mass-transfer rate from the sphere, $\dot{n}$, is to be calculated and expressed by means of the average Sherwood number

$$Sh' = \frac{kd_1}{D'_m} = \left[\dot{n}/\left(\pi d_1^2\right)(c^* - c_0)\right] d_1/D'_m. \tag{33}$$

With reference to Figure 4, the value of $\dot{n}$ will have to be equal to the net amount of solute leaving the domain of integration along the line joining the grid points $(n_\Phi,1)$ and (n_Φ,n_Ψ), since $c \cong c_0$ all along the upper boundary of that domain. In the discretized form this can be expressed as

$$\dot{n} \equiv 2\pi\epsilon\left\{\left(c - D_L\frac{\partial c}{\partial \phi}\right)_{n_\Phi,1}\frac{\Delta\psi_2}{2} + \sum_{j=2}^{n_\Psi - 1}\left[\left(c - D_L\frac{\partial c}{\partial \phi}\right)_{n_\Phi,j}\frac{\Delta\psi_{j+1} + \Delta\psi_j}{2}\right]\right\} \tag{34}$$

or, after some manipulation,

$$\frac{Sh'}{\epsilon} = Pe'\left\{\left(C - A\frac{\partial C}{\partial \Phi}\right)_{n_\Phi,1}\Delta\Psi_2 + \sum_{j=2}^{n_\Psi - 1}\left[\left(C - A\frac{\partial C}{\partial \Phi}\right)_{n_\Phi,j}\left(\Delta\Psi_{j+1} + \Delta\Psi_j\right)\right]\right\}. \tag{35}$$

An alternative route to the evaluation of $\dot{n}$ is by integration of the diffusional flux all around the surface of the sphere

$$\dot{n} = \epsilon\sum_{i=n_1+1}^{n_2-1} - D_T R\sin(\theta_i)\frac{3}{2}u_0\sin(\theta_i)\left(\frac{\partial c}{\partial \psi}\right)_{i,1} \cdot 2\pi R^2\left[\frac{\cos(\theta_{i-1}) - \cos(\theta_{i+1})}{2}\right], \tag{36}$$

which in the dimensionless discretized form reads

$$\frac{Sh'}{\epsilon} = -\frac{3}{8}\sum_{i=n_1+1}^{n_2-1}\left[1 + \frac{3}{2}\frac{Pe'(d/d_1)}{Pe_T(\infty)}\sin(\theta_i)\right] \cdot \sin^2(\theta_i)\left(\frac{\partial C}{\partial \Psi}\right)_{i,1}\left[\frac{\cos(\theta_{i-1}) - \cos(\theta_{i+1})}{2}\right]. \tag{37}$$

In our calculations, for any chosen grid, the value of Sh'/ϵ was calculated by both of the preceding methods, and if the results differed by more than 1%, the grid was refined until satisfactory agreement was reached. This gives additional confirmation that the numerical solutions had been refined until grid independence was achieved. Numerical solutions were worked out for many pairs of Pe' and d/d_1, in the ranges $0.1 \le Pe' \le 10^5$ and $0 \le d/d_1 \le 0.2$, and it is important to start by considering those situations for which the value of the Peclet number is low, based on the particles in the packing $Pe'_p = (d/d_1)Pe'$, since they correspond to $D_T = D_L = D'_m$.

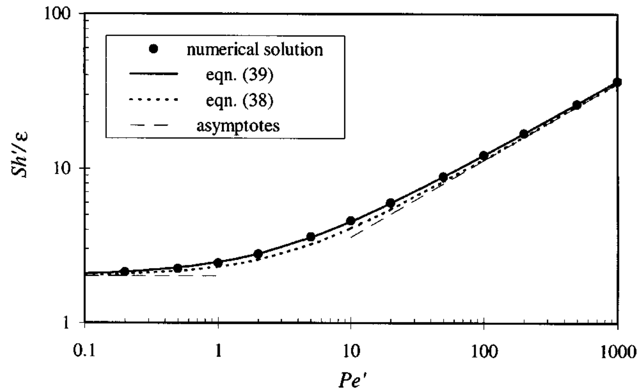

Figure 5. Dependence of Sh'/ϵ on Pe' when $D_T = D_L = D'_m$ throughout.

Case I: $Pe'_p < 0.1$ (that is, $D_T = D_L = D'_m$). Equations 16 and 17 show that D_T and D_L tend to D'_m as $u_0 d/D'_m$ ($= Pe'_p$) tends to zero, and for $Pe'_p < 0.1$, the error in taking $D_T = D_L = D'_m$ is already negligible. The numerical solutions for this condition were worked out taking $d/d_1 = 0$ in Eqs. 24, 35 and 37, and the values obtained are shown as dots in the plot of Figure 5.

As expected $Sh'/\epsilon \to 2$ when $Pe' \to 0$, whereas for high Pe', the asymptote is observed for convection with molecular diffusion across a thin boundary layer, $Sh'/\epsilon = [4Pe'/\pi]^{1/2}$, and this coincides with the result of La Nauze et al. (1984), corrected by Guedes de Carvalho and Coelho (1986).

The quadratic mean of the two asymptotes is

$$\frac{Sh'}{\epsilon} = \left[4 + \frac{4}{\pi}Pe'\right]^{1/2}, \tag{38}$$

and values of Sh'/ϵ calculated from this equation differ at most by 12% from the corresponding numerical solution obtained in the present work. An improved approximation, inspired by the form of Eq. 38, is

$$\frac{Sh'}{\epsilon} = \left[4 + \frac{4}{5}(Pe')^{2/3} + \frac{4}{\pi}Pe'\right]^{1/2}, \tag{39}$$

which is represented as a full line in Figure 5, and gives values of Sh'/ϵ within less than 1% from those obtained by the numerical solution described earlier. It should be stressed that Eq. 39 is a general result, valid for a gas or liquid flowing around a sphere buried in a packed bed of inerts, under conditions for which molecular diffusion dominates over convective dispersion. For that reason, we shall use below the symbol Sh'_{md} to represent the value of Sh' given by Eq. 39.

Case II: $Pe'_p > 0.1$. When $Pe'(d/d_1) > 0.1$, values of Sh'/ϵ depend both on Pe' and d/d_1, as shown by the points plotted

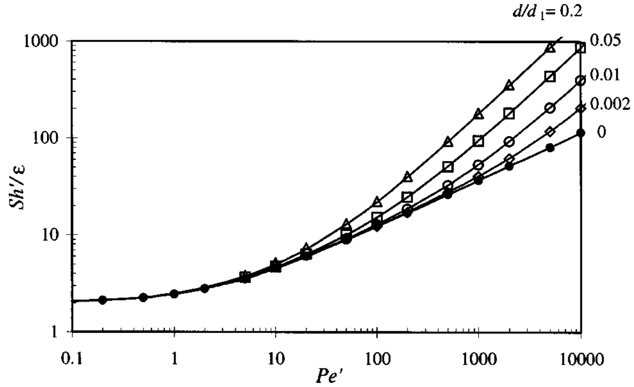

Figure 6. Dependence of Sh'/ϵ on Pe' for different values of d/d_1; the points were obtained from numerical solutions and the solid lines from Eq. 41.

in Figure 6, which were obtained from the numerical solution of Eq. 24.

This is the consequence of convective dispersion becoming a significant (or even dominant) mechanism of cross-stream transport of solute. The plot shows that Sh'/ϵ increases with d/d_1, at constant Pe', which means that the rate of mass-transfer increases with the size of the inert particles, other things being equal. Equation 39 therefore gives a lower limit for the rate of mass transfer from a buried sphere, and it is interesting to calculate the ratio between the value of Sh'/ϵ observed under any given set of conditions and the value of Sh'_{md}/ϵ that would result for the same value of Pe', if d were sufficiently low for molecular diffusion to be dominant; this ratio, $\eta = Sh'/Sh'_{md}$, represents the enhancement factor due to convective dispersion. A curious aspect, illustrated in Figure 7, is that η turns out to be a unique function of Pe'_p, which is not very surprising considering that, for gases, D_L/D'_m and D_T/D'_m are unique functions of Pe'_p, as shown by Eqs. 16 and 17.

The points in Figure 7 were calculated from values of Sh'/ϵ obtained in the numerical computation, and they all fall on

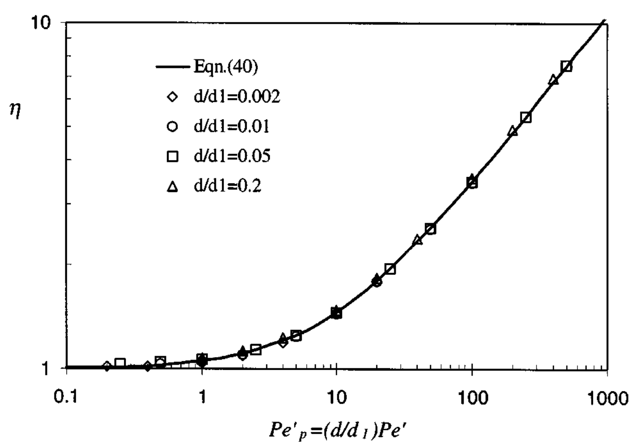

Figure 7. Dependence of η on Pe'_p (the points were obtained from the numerical solutions).

the line

$$\eta = \sqrt{1 + Pe'_p/9} \tag{40}$$

with an error of less than 2%. A general expression for mass transfer around large spheres buried in packed beds of inerts is then

$$\frac{Sh'}{\epsilon} = \left[4 + \frac{4}{5}(Pe')^{2/3} + \frac{4}{\pi}Pe'\right]^{1/2}\left(1 + \frac{1}{9}Pe'_p\right)^{1/2}, \tag{41}$$

which reduces to Eq. 39, with an error of less than 0.6% in Sh'/ϵ, when $Pe'_p < 0.1$ (for Pe'_p up to 1, Eq. 39 gives Sh'/ϵ with an error of up to 5%).

Several restrictions were introduced in the derivation of the results condensed in Eq. 41, and it is important to recall them. First, Darcy's equation holds strictly only up to $Re_p \sim 1$, the value above which the turbulent contribution to pressure drop starts to be significant. However, as pointed out in the text, Darcy's law is a reasonable approximation up to $Re_p \sim 25$, and indeed the experimental results used suggest that our theory is a good approximation even at higher Re_p. Second, we always took $d/d_1 \leq 0.2$, since for higher values of this ratio the packed bed around the sphere may not be treated as a *continuum* (an approximation implicit throughout). In terms of mass transfer, the assumption of a continuum would require that the concentration boundary layer around the sphere be of significant thickness compared to d. Following Coelho and Guedes de Carvalho (1988a), this is observed if $d/d_1 < 2/Pe'_p$ (or equivalently $d/d_1 \ll 1/Sh'$, as suggested by one of the referees). Curiously, the experimental results agree with the predictions of Eq. 41 well beyond the range defined by this criterion. Finally, using Eqs. 16 and 17 restricts the preceding analysis to gas flow, even though some of the results (namely Eq. 39) apply to liquid flow. These are the main limitations imposed on the theoretical treatment just presented, and it is interesting to see how the theoretical predictions compare with experimental measurements.

Comparison of theory with experiment

Measurements on mass transfer may be used to test different aspects of the preceding theory. As a first test, Figure 8 considers experimental data for which $Pe'_p = Pe'(d/d_1) < 1$, and agreement with Eq. 39 is seen to be excellent. Data for liquids are not available at such low values of Pe'_p.

Figure 9 is for $Pe'_p > 1$, and the individual values of η were calculated by dividing the actual Sh'/ϵ (obtained from the experiment) by the value of Sh'_{md}/ϵ, as given by Eq. 39, for the same Pe'. The data are again only for gas flow and the agreement between theory and experiment is very good; liquids are not expected to follow Eq. 40, and so they are not represented in the plot.

In experiments where Pe'_p is large, Eq. 40 shows that $\eta \cong (1/3)Pe'^{1/2}_p$ and if $Pe' > 50$ (approximately), Eq. 39 can be replaced by $Sh'_{md} \cong (2/\sqrt{\pi})Pe'^{1/2}$. As a result, $Sh'/\epsilon = (2/3\sqrt{\pi})(Pe'Pe'_p)^{1/2}$, and this is equivalent to

$$\frac{k}{\epsilon u_0} = \frac{2}{3\sqrt{\pi}}\left(\frac{d}{d_1}\right)^{1/2}, \tag{42}$$

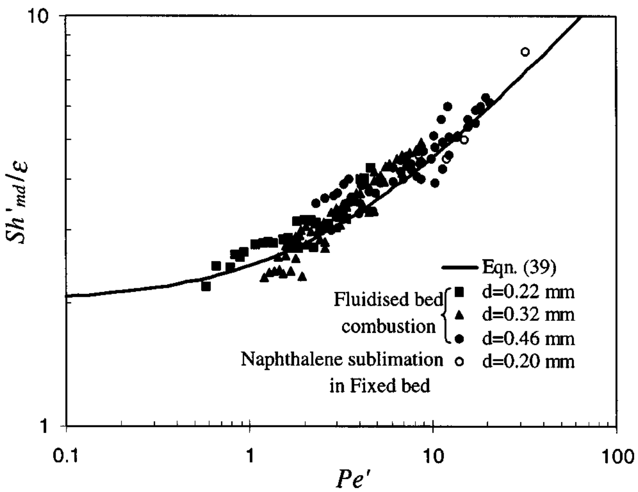

Figure 8. Theory (Eq. 39) vs. experimental data (Pinto and Guedes de Carvalho, 1990) for $Pe'_p < 1$.

an important result in that it shows the independence between k and D_m in the range of variables considered. Figure 10, which is largely borrowed from Coelho and Guedes de Carvalho (1988b) shows that measurements obtained in fixed and fluidized beds follow very closely the theoretical prediction of Eq. 42. Again the data refer only to gas flow, because no experimental values for liquids are available in the appropriate range; however, Eq. 42 is expected to apply also to liquids, for very high values of Pe'_p, since in that case Eqs. 16 and 17 will hold.

A significant number of experimental points are also available for systems in which $d/d_1 > 0.2$, and although the theory was developed for lower values of d/d_1, it is interesting to see how the predictions of Eq. 41 compare with the experiment. That comparison is partly made in Figure 10, and especially in Figure 11, where the measurements from Hsiung and Thodos (1977) refer to a number of experiments in which the active particles have the same diameter as the inerts. The

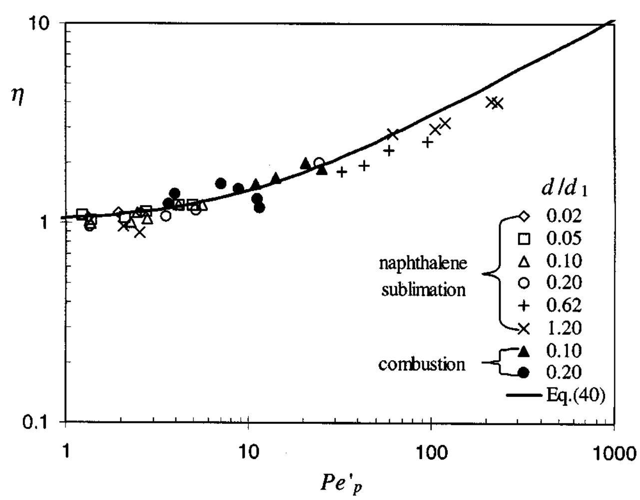

Figure 9. Experimental (Pinto and Guedes de Carvalho, 1990) vs. theoretical values of enhancement factor for $Pe'_p > 1$.

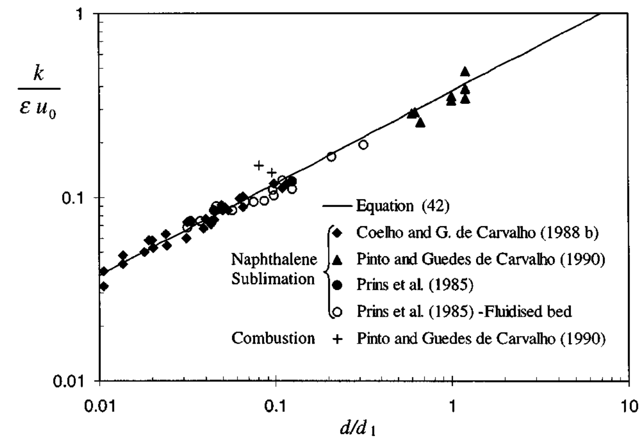

Figure 10. Theory vs. experiment for large Pe' and large Pe'_p.

agreement is again remarkable, even though it may be fortuitous.

Conclusions

The problem of mass transfer around a sphere buried in a granular bed (be it packed or incipiently fluidized) lends itself to a simple full theoretical analysis, under an appropriate set of conditions. If Darcy flow is considered in the packing, with dispersion coefficients given by $D_T = D'_m + ud/Pe_T(\infty)$ and $D_L = D'_m + ud/Pe_L(\infty)$, the differential equation describing mass transfer may be integrated numerically and the results are accurately described by

$$\frac{Sh'}{\epsilon} = \left[4 + \frac{4}{5}(Pe')^{2/3} + \frac{4}{\pi}Pe'\right]^{1/2}\left(1 + \frac{1}{9}Pe'_p\right)^{1/2}.$$

The second term on the righthand side of this equation represents the enhancement factor brought about by convective dispersion of the solute in the interstices of the granular bed. The preceding equation was tested against a very large number of experimental data and the agreement was seen to be very good.

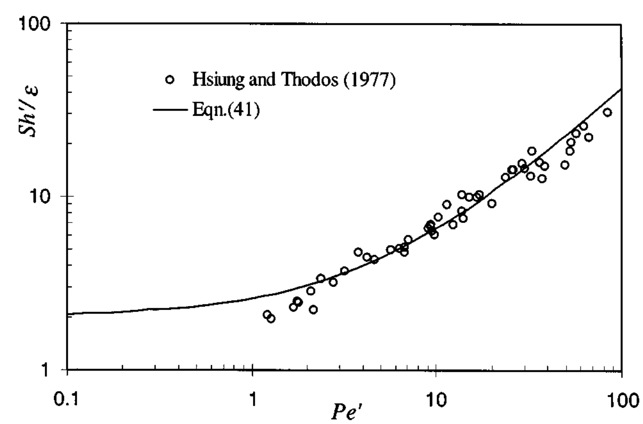

Figure 11. Predictions of Eq. 41 vs. experimental data for $d = d_1$.

Notation

A = dimensionless parameter, Eq. 25
B = dimensionless parameter, Eq. 26
c = solute concentration
C = dimensionless solute concentration, Eq. 18
d_1 = diameter of active sphere
D_L = longitudinal dispersion coefficient
D_m = molecular diffusion coefficient
D'_m = effective molecular diffusion coefficient ($= D_m/\tau$)
D_T = transverse (radial) dispersion coefficient
K = permeability in Darcy's law
k = average mass-transfer coefficient
n_Φ = number of grid points along a stream surface (Figure 4)
n_Ψ = number of grid points along a potential surface (Figure 4)
p = pressure
$Pe_L(\infty)$ = asymptotic value of Pe_L when $Re_p \to \infty$
$Pe_T(\infty)$ = asymptotic value of Pe_T when $Re_p \to \infty$
$\Re$ = dimensionless spherical radial coordinate ($= r/R$)
r = spherical radial coordinate (distance to the origin)
S_Φ = length coordinate along a stream surface
S_Ψ = length coordinate along a potential surface
U = dimensionless interstitial velocity ($= u/u_0$)
u = absolute value of interstitial velocity
$\mathbf{u}$ = interstitial velocity (vector)
u_r, u_θ = components of fluid velocity
δ = increment
Φ = dimensionless potential function, Eq. 21
ϕ = potential function, Eq. 6
$\Phi_{\min}$ = minimum value of dimensionless potential function used in a given mesh
η = enhancement factor due to convective dispersion, Eq. 40
μ = fluid viscosity
θ = spherical angular coordinate (Figure 1)
ρ = fluid density
τ = tortuosity
ω = cylindrical radial coordinate (distance to the axis, Figure 1)
Ψ = dimensionless stream function, Eq. 22
ψ = stream function, Eq. 7
$\Psi_{\max}$ = maximum value of dimensionless stream function used in a given mesh

Dimensionless groups

Pe' = Peclet number based on diameter of active sphere ($= u_0 d_1/D'_m$)
Pe'_p = Peclet number based on diameter of inert particles ($= u_0 d/D'_m$)
Re_p = Reynolds number based on diameter of inert particles ($= \rho u d/\mu$)
Sh'_{md} = Sherwood number when $D_T = D_L = D'_m$ (that is, $Pe'_p < 0.1$), Eqs. 16 and 17

Subscripts and superscripts

n = iteration number
$'$ = a prime is used in D'_m and in all dimensionless groups containing D'_m

Literature Cited

Anderson, J. D., *Computational Fluid Dynamics*, McGraw-Hill, New York, p. 137 (1995).

Avedesian, M. M., and J. F. Davidson, "Combustion of Carbon Particles in a Fluidised Bed," *Trans. Inst. Chem. Eng.*, **51**, 121 (1973).

Bear, J., *Dynamics of Fluids in Porous Media,* Dover, New York, p. 177 (1988).

Brian, P. L. T., and H. B. Hales, "Effects of Transpiration and Changing Diameter on Heat and Mass Transfer to Spheres," *AIChE J.*, **15**, 419 (1969).

Chakraborty, R. K., and J. R. Howard, "Combustion of Single Carbon Particles in Fluidized Beds of High-Density Alumina," *J. Inst. Energy*, **54**(418), 55 (1981).

Coelho, M. N., and J. R. Guedes de Carvalho, "Transverse Dispersion in Granular Beds: I. Mass Transfer from a Wall and the Dispersion Coefficient in Packed Beds," *Chem. Eng. Res. Des.*, **66**, 165 (1988a).

Coelho, M. N., and J. R. Guedes de Carvalho, "Transverse Dispersion in Granular Beds: II. Mass Transfer from Large Spheres Immersed in Fixed or Fluidised Beds of Small Inert Particles," *Chem. Eng. Res. Des.*, **66**, 178 (1988b).

Currie, I. G., *Fundamental Mechanics of Fluids*, McGraw-Hill, New York, p. 150 (1993).

Davidson, J. F., and D. Harrison, *Fluidised Particles*, Cambridge University Press, Cambridge, p. 19 (1963).

Ferziger, J. H., and M. Peric, *Computational Methods for Fluid Dynamics*, Springer-Verlag, Berlin, p. 42 (1996).

Guedes de Carvalho, J. R. F., and M. A. N. Coelho, "Comments on Mass Transfer to Large Particles in Fluidized Beds of Smaller Particles," *Chem. Eng. Sci.*, **41**, 209 (1986).

Guedes de Carvalho, J. R., A. R. Pinto, and C. T. Pinho, "Mass Transfer Around Carbon Particles Burning in Fluidised Beds," *Trans. Inst. Chem. Eng.*, **69** (Part A), 63 (1991).

Guedes de Carvalho, J. R. F., and J. M. Delgado, "Mass Transfer from a Large Sphere Buried in a Packed Bed Along which Liquid Flows," *Chem. Eng. Sci.*, **54**, 1121 (1999).

Gunn, D. J., "Mixing in Packed and Fluidised Beds," *Chem. Eng. Lond.*, CE153 (1968).

Gunn, D. J., "Axial and Radial Dispersion in Fixed Beds," *Chem. Eng. Sci.*, **42**, 363 (1987).

Hsiung, T. H., and G. Thodos, "Mass-Transfer Factors from Actual Driving Forces for the Flow of Gases Through Packed Beds ($0.1 < Re < 100$)," *Int. J. Heat Mass Transfer*, **20**, 331 (1977).

La Nauze, R. D., K. Jung, and J. Kastl, "Mass Transfer to Large Particles in Fluidized Beds of Smaller Particles," *Chem. Eng. Sci.*, **39**, 1623 (1984).

Leung, L. A., and I. W. Smith, "Role of Fuel Reactivity in Fluidized-Bed Combustion," *Fuel*, **58**, 354 (1979).

Middleman, S., *An Introduction to Mass and Heat Transfer*, John Wiley, New York, p. 292 (1998).

Pinto, A. R., and J. R. Guedes de Carvalho, "Transverse Dispersion in Granular Beds: III. Mass Transfer Around Particles Dispersed in Granular Beds of Inerts and the Combustion of Carbon Particles in Beds of Sand," *Trans. Inst. Chem. Eng.*, **68**(Part A), 503 (1990).

Prins, W., T. P. Casteleijn, W. Draijer, and W. P. M. Van Swaaij, "Mass Transfer from a Freely Moving Single Sphere to the Dense Phase of a Gas Fluidized Bed of Inert Particles," *Chem. Eng. Sci.*, **40**, 481 (1985).

Ranz, W. E., and W. R. Marshall, Jr., "Evaporation from Drops," *Chem. Eng. Prog.*, **48**(Pt. I), 141, and (Pt. II), 173 (1952).

Scheidegger, A. E., *The Physics of Flow Through Porous Media*, Univ. of Toronto Press, Toronto, p. 100 (1974).

Smith, G. D., *Numerical Solutions of Partial Differential Equations*, Oxford Univ. Press, London, p. 149 (1971).

van Heerden, C., A. P. P. Nobel, and D. W. Krevelen, "Mechanism of Heat Transfer in Fluidised Beds," *Ind. Eng. Chem.*, **45**, 1237 (1953).

Wilhelm, R. H., "Progress Towards the a Priori Design of Chemical Reactors," *Pure Appl. Chem.*, **5**, 403 (1962).

Manuscript received Dec. 17, 1998, and revision received June 11, 1999.