Mass Transfer and Concentration Contours around Surfaces Buried in Granular Beds and Exposed to Fluid Flow

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Abstract. This work describes the process of mass transfer which takes place when a fluid flows past a soluble surface buried in a packed bed of small inert spherical particles of uniform voidage. The fluid is assumed to have uniform velocity far from the buried surface and different surface geometries are considered; namely, cylinder in cross flow and in flow aligned with the axis, flat surface aligned with the flow and sphere.

The differential equations describing fluid flow and mass transfer by advection and diffusion in the interstices of the bed are presented and the method for obtaining their numerical solution is indicated. From the near surface concentration fields, given by the numerical solution, rates of mass transfer from the surface are computed and expressed in the form of a Sherwood number (Sh). The dependence between Sh and the Peclet number for flow past the surface is then established for each of the flow geometries.

Finally, equations are derived for the concentration contour surfaces at a large distance from the soluble solids, by substituting the information obtained on mass transfer rates in the equation describing solute spreading in uniform flow past a point (or line) source.

Introduction

There are a number of situations of practical interest in which a fluid flows through a bed of inert particles, packed around a soluble solid mass with which the fluid interacts (e.g. water contamination by buried waste [1], ore leaching [2], fluidised bed combustion [3]).

In such processes there is an interplay between diffusion/dispersion and convection. A detailed physical analysis of the mass transfer process involved was presented in due course by Coelho and Guedes de Carvalho [4,5]. In subsequent work, a detailed numerical analysis was presented and accurate analytical expressions were developed for the calculation of mass transfer coefficients in flow around a sphere, flow along a cylinder and flow past a flat surface [6-9].

Flow around a large soluble sphere is an important model situation in many processes and Guedes de Carvalho and Alves [6] treated the problem numerically. Flow along buried cylindrical and flat surfaces are also important model situations and they were investigated in detail [8,9], yielding results for the whole range of values of Peclet number and aspect ratio of the dissolving solid surface. In all those studies, only the problem of determining the rates of mass transfer from the surfaces was considered.

In the present work the previous research is extended to cover the situation of uniform flow perpendicular to a buried cylinder. Additionally, a simple approximate method is presented to obtain concentration contours plots for solute distribution around and downstream of the buried surfaces (for the different geometries).
Theory

Mass transfer from a buried cylinder exposed to cross flow. Consider a slightly soluble cylinder, of diameter $d_1$, buried in a packed bed of inert spherical particles of diameter $d$ ($d << d_1$) and exposed to fluid flow perpendicular to its axis, with uniform interstitial velocity, $u_0$, at a large distance from the cylinder.

Assuming Darcy’s law to apply, Laplace’s equation is obtained for the flow potential around the cylinder. For a sufficiently long cylinder (assumed to be of “infinite” length) the flow field is bi-dimensional and in polar coordinates $(r, \theta)$, the potential and stream functions are expressed as

$$\phi = -u_0 \left[ 1 + \left( \frac{a}{r} \right)^2 \right] r \cos \theta$$  \hspace{2cm} (1)

$$\psi = u_0 \left[ 1 - \left( \frac{a}{r} \right)^2 \right] r \sin \theta$$  \hspace{2cm} (2)

In the absence of chemical reaction in the fluid and for steady state conditions, the mass balance on the solute crossing the borders of an elementary stream tube, limited by two neighbouring equipotential surfaces, may be performed, to give in the limit [4,5]

$$\frac{\partial c}{\partial \phi} = \frac{\partial}{\partial \phi} \left( D'_m \frac{\partial c}{\partial \phi} \right) + \frac{\partial}{\partial \psi} \left( D'_m \frac{\partial c}{\partial \psi} \right)$$  \hspace{2cm} (3)

The boundary conditions to be observed in the integration of Eq. 3 are that: (i) the solute concentration is equal to the background concentration, $c_0$, far from the cylinder; (ii) the solute concentration is equal to the equilibrium concentration, $c^*$, on the surface of the cylinder and (iii) the concentration field is symmetric about $\psi = 0$, for $r > a$.

Equation (3) has to be solved numerically and the method developed and described in detail by Alves et al. [8] was adapted, to obtain the solute concentration field around the dissolving cylinder.

Rate of mass transfer. Values of the flux of solute leaving the soluble surface were computed from the concentration field obtained numerically and the overall mass transfer rate from the cylinder was calculated and is well expressed (with an error smaller than 3%) by:

$$\frac{S_h}{e} = \left[ \frac{2}{\pi^2} Pe_{cf}^{1/4} + \frac{32}{\pi^5} Pe_{cf} \right]^{1/2}$$  \hspace{2cm} (4)

where $S_h = k d_1 / D_m$, $Pe_{cf} = u_0 d_1 / D'_m$ and $D'_m$ is the effective molecular diffusion coefficient ($D'_m = D_m / \tau$), defined as the ratio between the molecular diffusion coefficient and the tortuosity of the packed bed, $\tau$, with regard to diffusion.

Figure 1 illustrates the excellent agreement between the values of $S_h$ given by the numerical solution and those corresponding to Eq. 4.
Concentration Contour Surfaces. If a slightly soluble cylinder, buried in a packed bed, is exposed to uniform fluid flow with velocity $u_0$, it will then release solute at a rate $n$, given by

$$n = \epsilon k \pi d_1 (c^* - c_0) = \frac{D'_m}{d_1} \epsilon \pi d_1 (c^* - c_0) \left[ \frac{2}{\pi^2} Pe'_{cf}^{1/4} + \frac{32}{\pi^3} Pe'_{cf} \right]^{1/2}$$  \hspace{1cm} (5)$$

with $Sh'_{cf}$ given by Eq. 4 ($b$ being the length of cylinder considered). Under steady state (a useful limiting approximation), the concentration contour surfaces for the solute around the buried cylinder will have shapes as sketched in Figure 2. The exact shapes of the contour surface, for each value of the normalized solute concentration, $C = (c - c_0)/(c^* - c_0)$, may also be found from the concentration field given by the numerical solution of Eq. 3. However, since the concentration field depends on the flow parameters, it would be cumbersome to work out the numerical solution for each flow condition [10]. As an alternative, it is possible to derive an approximate analytical expression for the contour surfaces that converges to the exact solution as the distance to the cylinder increases. Indeed, if the situation sketched in Figure 2 is viewed by an observer placed at a great distance from the cylinder, it will resemble the idealized condition of continuous solute injection at a point $(x = y = 0)$, in plane flow with uniform velocity $u_0$. The rate of solute injection is that given by Eq. 5 and the corresponding concentration field may then be obtained from Wexler [11] as

$$c = \frac{n / b}{2 \epsilon \pi D'_m} \exp \left[ \frac{u x}{2 D'_m} \right] K_0 \left\{ \frac{u^2 (x^2 + y^2)}{4 D'_m^2} \right\}$$  \hspace{1cm} (6)$$

where $K_0$ is the modified Bessel function of second kind and zero order. Substitution of $n / b$ from Eq. 5 into Eq. 6 gives then

$$C = \frac{\left[ \frac{2}{\pi^2} Pe'_{cf}^{1/4} + \frac{32}{\pi^3} Pe'_{cf} \right]^{1/2}}{2} \exp \left[ \frac{Pe'_{cf}}{4 a} \right] K_0 \left\{ \frac{Pe'_{cf}}{16} \left( \frac{x}{a} \right)^2 + \left( \frac{y}{a} \right)^2 \right\}$$  \hspace{1cm} (7)$$
Concentration contours given by Eq. 7 are shown in Fig. 3, for two values of Pe'_cf (note that for \( d = 2 \text{ m}, \ u_0 = 5 \times 10^{-4} \text{ mm/s} \) and \( D' = 10^{-9} \text{ m}^2/\text{s} \), there results \( \text{Pe}'_{\text{cf}} \approx 1000 \)). From a detailed study of a similar nature [10], for flow past a sphere, it is reasonable to expect that values of the downstream reach of each contour surface (defined as the value of \( x/a \), for \( y/a = 0 \), for that surface), predicted by Eq. 7, will differ by less than 10% from the exact value, if \( \text{Pe}' > 500 \) and \( C < 0.05 \). The accuracy of the prediction will improve as \( C \) is decreased.

\[
\frac{n}{b} = \frac{D'_m}{L} \varepsilon L (c^* - c_0) \left[ \frac{2}{\pi} \text{Pe}'_{\text{sf}}^{1/4} + \frac{4}{\pi} \text{Pe}'_{\text{sf}} \right]^{1/2}
\]  (8)

and substituting \( n/b \) in Eq. (6) leads to

**Figure 2** – Schematic of physical situation.

**Figure 3** – Concentration contour plots at a large distance from a soluble cylinder exposed to cross flow for \( \text{Pe}'_{\text{cf}} = 10 \) and \( \text{Pe}'_{\text{cf}} = 1000 \).

**Other Geometries**

**Plane 2-D flow.** The method presented above and initially studied in detail for the sphere [10], may be easily adapted to another 2-D flow geometry; namely, flow parallel to a soluble flat surface of length \( L \) (along the flow direction) and “infinite” width. The rate of mass transfer is known to be given as [8]
\[ C = \left[ \frac{2}{\pi} \left( \frac{\text{Pe}_{\text{sf}}}{\text{Pe}_{\text{sf}}} \right)^{1/4} + \frac{4}{\pi} \frac{\text{Pe}_{\text{sf}}}{\text{Pe}_{\text{sf}}} \right]^{1/2} \exp \left[ \frac{\text{Pe}_{\text{sf}} x}{2 L} \right] K_0 \left\{ \frac{\text{Pe}_{\text{sf}}^2}{4} \left[ (x/L)^2 + (y/L)^2 \right] \right\} \]  

(9)

A representation of the corresponding contour surfaces is given in Figure 4(a).

**Axisymmetric flow**

**Sphere.** Uniform flow past a (slightly soluble) buried sphere is possibly the simplest situation with axial symmetry. The detailed numerical treatment of that situation [10] shows that for high Pe' and low C, the shape of the contour surfaces is well approximated by equation

\[ C = \left\{ \frac{4 + \frac{4}{5} \left( \text{Pe}_{\text{s}}^2 \right)^{3/5} + 4 \frac{\text{Pe}_{\text{s}}}{\pi} \right\}^{1/2} \exp \left[ \frac{\text{Pe}_{\text{s}}}{4} \left[ \left( x/a \right) - \left( x/a \right)^2 + (y/a)^2 \right]^{3/2} \right] \]  

(10)

which is obtained by substituting the equation for the rate of mass transfer

\[ n = \frac{D'_{\text{m}}}{d_1} \pi \left( c \ast -c_0 \right) \left[ 4 + \frac{4}{5} \left( \text{Pe}_{\text{s}}^2 \right)^{3/5} + \frac{4}{\pi} \right]^{1/2} \]  

(11)

in the equation for solute spreading by convection/diffusion from a point source in a uniform flow field, given by Wexler [11] as

\[ c = \frac{n}{4 \pi (x^2 + y^2)^{1/2}} \frac{\text{Pe}_{\text{c}}}{D'_{\text{m}}} \exp \left[ u \left( x - (x^2 + y^2)^{1/2} \right) \right] \]  

(12)

A sample representation of contours from Eq. 12 is also shown in Figure 4(b).

**Flow parallel to cylinder.** Uniform flow parallel to a slightly soluble cylinder with length L and diameter d_1 is the other situation with axial symmetry which has been studied in detail [8], but only with regard to the prediction of the mass transfer rate. A very accurate prediction is provided by the following equation [8]

\[ n = \frac{D'_{\text{m}}}{L} \pi d_1 L \left( c \ast -c_0 \right) \left[ \frac{2}{\pi} \text{Pe}_{\text{c}}^{1/4} + \frac{4}{\pi} \text{Pe}_{\text{c}}^4 \right]^{1/2} + \left( \frac{4 L}{3 d_1} \right)^{3/2} + \frac{5}{3} \text{Pe}_{\text{c}}^{5/9} \left( \frac{L}{d_1} \right)^{1/3} - \text{Pe}_{\text{c}}^{2/9} \left( \frac{2 L}{d_1} \right)^{1/3} \]  

(13)

Again, if this is coupled with Eq. 12, an equation is obtained for the concentration contour surfaces, which reads:
A sample representation of contours from Eq. 14 is also shown in Figure 4(c).

In all the cases considered above, the approximate predictions, corresponding to Eq. 7, Eq. 9, Eq. 10 and Eq. 14, will tend to the exact contour surfaces as $C$ is decreased (and accordingly, the distance to the soluble surface is increased).

Figure 4 – Concentration contour plots at a large distance from a soluble solid mass with different shapes: (a)-flat slab; (b)-sphere and (c)-cylinder in alignment with the flow.

Conclusions

The problem of mass transfer between a buried solid (with different shapes) and the fluid flowing along it in a granular bed, lends itself to a full theoretical analysis, under an appropriate set of conditions. The elliptic equations resulting from a differential mass balance were described and their numerical solutions over a wide range of values of the relevant parameters are presented in
terms of correlations of Sherwood number as a function of relevant parameters (Peclet number and aspect ratio of the soluble mass).

A theoretical analysis of continuous injection of a solute at a point/line source is used, and coupling with the correlations obtained for the mass transfer rate, helps establish analytical expressions for the spatial distribution of the solute concentration around the soluble mass. This approximate analysis is more accurate for points far away from the soluble mass, and this has important applications, as in the prediction of the transport of pollutants underground.

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Notation

- \( a \): Radius of soluble cylinder or sphere
- \( A \): Area of soluble particle
- \( b \): Thickness of soluble mass in the “neutral” direction
- \( c \): Solute concentration
- \( c_0 \): Bulk concentration of solute
- \( c^* \): Equilibrium concentration of solute
- \( C \): Dimensionless solute concentration \( = (c - c_0)/(c^* - c_0) \)
- \( d \): Diameter of inert particles
- \( d_1 \): Diameter of soluble sphere or cylinder
- \( D_m' \): Effective molecular diffusion coefficient \( = D_m/\tau \)
- \( K_0 \): Modified Bessel function of second kind and zero order
- \( k \): Mass transfer coefficient
- \( L \): Length of solid slab or cylinder
- \( n \): Mass transfer rate \( = kA(c^* - c_0) \)
- \( r \): Spherical radial coordinate
- \( u \): Absolute value of interstitial velocity
- \( x, y, z \): Cartesian co-ordinates

Greek letters

- \( \varepsilon \): Bed voidage
- \( \phi \): Potential function
- \( \omega \): Cylindrical radial coordinate (distance to the axis)
- \( \psi \): Stream function

Dimensionless groups

- \( \text{Pe}_c' \), \( \text{Pe}_{sf} \): Peclet number based on length of cylinder or flat slab \( = uL/D_m' \)
- \( \text{Pe}_p' \): Peclet number based on diameter of inert particles \( = ud/D_m' \)
- \( \text{Pe}_s' \), \( \text{Pe}_{cif} \): Peclet number based on diameter of sphere or cylinder in cross flow \( = ud_1/D_m' \)
- \( \text{Sh}_c' \), \( \text{Sh}_{cif} \): Sherwood number of cylinder or slab flat \( = kL/D_m' \)
- \( \text{Sh}_s' \), \( \text{Sh}_{cif} \): Sherwood number of the sphere or cylinder in cross flow \( = kd_1/D_m' \)

References