

Mass transfer from a particle suspended in fluid with a steady linear ambient velocity distribution

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This paper is concerned with the rate of transfer of heat or mass from a force-free couple-free particle immersed in fluid whose velocity far from the particle is steady and varies linearly with position. Asymptotic results for both small and large Péclet numbers are considered. There is at least a four-parameter family of different linear ambient velocity distributions, but nevertheless a comprehensive set of results for the transfer rate may be compiled by combining previously published work with some new developments. Some of these are exact results for particular linear ambient flow fields and some are approximate results for classes of linear flow fields.

For small Péclet number (P), the non-dimensional additional transfer rate due to convection is equal to $\alpha N_0^2 P^{\frac{1}{2}}$, where N_0 is the Nusselt number for $P = 0$ and the proportionality constant α is a parameter of the concentration distribution due to a steady point source in the given linear ambient flow field. A general method of determining α is developed, and numerical values are found for some particular linear ambient flow fields. It is estimated that the value of α for any linear ambient flow field in which the vorticity does not dominate the straining motion lies within 10% of 0.34 when P is defined in terms of a particular invariant of the ambient rate-of-strain tensor \mathbf{E} .

At large Péclet number the transfer rate N depends on the velocity distribution near the particle, and attention is restricted to the case of a sphere in low-Reynolds-number flow. For a rigid sphere $N = \beta P^{\frac{1}{2}}$ for any ambient pure straining motion, and the Levich concentration-boundary-layer method may be used to show that $\beta = 0.90$ for both axisymmetric and two-dimensional ambient pure straining, and probably for any other pure straining motion, when P is suitably defined. When the ambient vorticity $\boldsymbol{\omega}$ is non-zero, the sphere rotates, and the Levich method cannot be used. However, it is shown that the part of the velocity distribution that varies sinusoidally with the azimuthal angle around the rotation axis does not affect the transfer rate and that N is asymptotically the same as for an ambient axisymmetric pure straining motion with rate of extension in the direction of the axis of symmetry equal to $E_\omega (= \boldsymbol{\omega} \cdot \mathbf{E} \cdot \boldsymbol{\omega} / \omega^2)$. In the exceptional case $E_\omega = 0$, N approaches a constant as $P \rightarrow \infty$.

It is possible to interpolate between the asymptotic relations for small and large Péclet number with comparatively little uncertainty for any ambient pure straining motion and for any linear ambient flow field in which $\boldsymbol{\omega}$ and E_ω are non-zero.

1. Introduction

Calculation of the rate at which some diffusible quantity is transferred away from a surface at which the intensity is maintained at a constant value is a classical problem in applied mathematics, and appears in a number of different physical forms. The diffusible quantity might be a solute diffusing through a liquid, or a vapour diffusing through a gas, or it might be heat which is being conducted away from a phase boundary at which the temperature is maintained at a constant level. We shall be considering the class of such problems, rather than any one in particular, and will refer to the process as mass transfer (of solute) and to the local intensity as the concentration.

The particular geometry to be studied here corresponds to transfer across the surface of a particle suspended in moving fluid of large extent, with a given difference between the values of the intensity at the particle surface and far from the particle. The purpose of the paper is to investigate theoretically the combined effects of diffusion and convection in the fluid on the rate of transfer from the particle surface.

The concentration of the diffusible quantity in the fluid, to be denoted by C , satisfies the equation

$$\frac{\partial C}{\partial t} + \mathbf{u} \cdot \nabla C = \kappa \nabla^2 C, \quad (1.1)$$

where \mathbf{u} is the local fluid velocity relative to axes such that the particle has zero translational velocity and κ is the diffusivity. The fluid velocity \mathbf{u} will be regarded as determined by specification of the *ambient* flow field, that is, by the velocity \mathbf{U} (relative to the same axes) that the fluid would have in the absence of the particle and to which \mathbf{u} tends as $r (= |\mathbf{x}|) \rightarrow \infty$. The boundary conditions are assumed to be

$$\left. \begin{aligned} C &= C_1 & \text{at the particle surface } A, \\ C &\rightarrow C_0 & \text{as } r \rightarrow \infty, \end{aligned} \right\} \quad (1.2)$$

the origin of the position vector \mathbf{x} being near the particle. The rate of transfer from the particle surface is then

$$Q = - \int_A \kappa \mathbf{n} \cdot \nabla C dS,$$

where \mathbf{n} is the unit outward normal to the surface of integration.

We shall assume that the ambient flow velocity \mathbf{U} is time-independent.

The nature of the solution of equation (1.1) depends of course on the relative magnitude of the convection and diffusion terms, which is measured by the Péclet number P (to be given precise definition later). We shall investigate asymptotic expressions for the mass transfer rate for the two extreme cases, $P \ll 1$ and $P \gg 1$, and it will be seen to be possible, in at least some cases, to draw interpolation curves without much uncertainty. The asymptotic expression that will be found for the case $P \ll 1$ holds for any shape or constitution (i.e. solid or fluid) of the particle and for any Reynolds number of the flow about the particle. On the other hand, the asymptotic results that can be obtained for $P \gg 1$ depend on the form of the fluid motion near the particle, and so in order to be able to obtain explicit results we shall assume in this case that the particle is a sphere and that the Reynolds number of the flow about

the sphere is small enough for the Stokes equation to be applicable (which for practical purposes amounts to requiring that the Reynolds number be smaller than unity). The combination of the assumptions of flow governed by the Stokes equation and of large Péclet number is not without practical relevance, because Prandtl numbers for solutes are typically large (e.g. $\nu/\kappa = 1.0 \times 10^3$ for NaCl in water, and 2.2×10^3 for sugar in water, both at 20 °C).

Two alternative forms of the ambient flow field are suggested by practical transfer problems. If the density of the particle is different from that of the fluid, the particle is subject to gravitational and inertial forces which give it a translational motion relative to the fluid. Provided then that the variation of the ambient fluid velocity over a distance of one particle dimension is small compared with the particle velocity relative to the fluid, the flow near the particle and relative to it is effectively that due to a particle held in a uniform stream. Thus here \mathbf{U} is independent of \mathbf{x} . If on the other hand the particle has the same density as the fluid, it has no translational velocity relative to the fluid. Provided that the linear dimensions of the particle are small compared with distances over which the velocity gradient in the ambient flow field changes significantly, the flow near the particle is then effectively that due to a force-free particle immersed in fluid in which the ambient velocity varies linearly with position. Thus here

$$U_i = G_{ij} x_j.$$

Most published theoretical work on the problem of transfer from a particle in a moving fluid concerns the case of uniform ambient velocity. In this paper we consider the many-sided case of linear variation of the ambient velocity.

Since \mathbf{G} is a second-rank tensor with eight independent components (when $\nabla \cdot \mathbf{U} = 0$), a large number of quite different ambient flow fields are encompassed by this linear variation of \mathbf{U} , some of which are of particular interest. The gradient tensor \mathbf{G} can as usual be written as

$$\mathbf{G} = \mathbf{E} + \mathbf{\Omega},$$

where the symmetrical part \mathbf{E} represents a pure straining motion and the anti-symmetrical part $\mathbf{\Omega}$ represents a rigid-body rotation with angular velocity $\frac{1}{2}\boldsymbol{\omega}$ given by

$$\Omega_{ij} = -\frac{1}{2}\epsilon_{ijk} \omega_k,$$

$\boldsymbol{\omega}$ being the vorticity of the ambient flow. The straining motion can be regarded as specified by three scalar parameters needed to determine the orientation of the three orthogonal principal axes of \mathbf{E} (p_1, p_2, p_3 , say; their precise meanings need not be spelt out) and by the three principal rates of strain, E_1, E_2, E_3 , only two of which are independent because the mass-conservation relation for incompressible fluid requires their sum to be zero. The angular velocity $\frac{1}{2}\boldsymbol{\omega}$ can now be conveniently specified by its three components in the directions of the principal axes of the rate-of-strain tensor \mathbf{E} , to be denoted by $\Omega_1, \Omega_2, \Omega_3$. A representative magnitude of the rate-of-strain tensor, E say, will be used to define the Péclet number, and the remaining quantities needed to specify the ambient flow can then be taken to be

- three orientation parameters p_1, p_2, p_3 ,
- one ratio of principal rates of strain, for example $(E_1 - E_2)/E$,
- and three angular-velocity components $\Omega_1/E, \Omega_2/E, \Omega_3/E$.

The orientation of the ambient flow is of course relevant only when the particle itself has an orientable shape which affects the transfer rate. We shall see that when $P \ll 1$ the transfer rate is independent of particle shape; and in the case of a spherical particle, the particle orientation is irrelevant at all Péclet numbers. Consequently in these cases there is no dependence of the transfer rate on the orientation parameters p_1, p_2, p_3 , and only four parameters remain, aside from the Péclet number. Three of these four remaining parameters disappear for the important class of pure straining motions (when $\Omega_1 = 0, \Omega_2 = 0, \Omega_3 = 0$), leaving one defining parameter unspecified. And in the case of two-dimensional ambient flow, we have

$$E_3 = 0 \quad (\text{and} \quad E_2 = -E_1), \quad \Omega_1 = 0, \quad \Omega_2 = 0,$$

again leaving one defining parameter (*viz.* Ω_3/E_1), with the particular case of simple shearing flow corresponding to $\Omega_3/E_1 = \mp 1$.

An attempt will be made to discuss the transfer problem for all these different kinds of linear ambient flow field systematically, with a brief account of previously published results and methods being given in the appropriate place. This paper is thus in part a survey and in part a presentation of some new developments.

As a final preliminary we define the non-dimensional measure of the transfer rate, the Nusselt number, as Q divided by the product of $\kappa, C_1 - C_0$, and dimensional factors representative of $(C_1 - C_0)^{-1} |\nabla C|$ and of the area of the particle surface. Practice concerning the choice of these last two factors varies. In this paper we choose these two factors as a^{-1} and $4\pi a^2$ respectively, where a is half the maximum diameter of the particle, so that the Nusselt number is

$$N = \frac{Q}{4\pi a \kappa (C_1 - C_0)}. \quad (1.3)$$

The basis for this choice is that for a spherical particle of radius a in stationary fluid the solution is

$$C = C_0 + \frac{a(C_1 - C_0)}{r}, \quad (1.4)$$

giving for the transfer rate at zero Péclet number

$$Q_0 = 4\pi a \kappa (C_1 - C_0), \quad \text{i.e.} \quad N_0 = 1. \dagger$$

The Péclet number P will be defined as a/κ multiplied by a velocity characteristic of the ambient flow. For a linear ambient velocity distribution with gradient of magnitude G (there may be more than one scalar gradient involved, and the choice will be refined later), we may choose this characteristic velocity as aG so that the Péclet number is $a^2 G/\kappa$. And when results for a uniform ambient velocity of magnitude U_0 are mentioned, the Péclet number in use will be aU_0/κ .

† Some authors choose $\frac{1}{2}a^{-1}$ and $4\pi a^2$ for the two-dimensional factors mentioned, in which case $N_0 = 2$ for a sphere; and others choose a^{-1} and a^2 , in which case $N_0 = 4\pi$ for a sphere. It seems to me that since there is no obvious choice for the length factor representative of $(C_1 - C_0)^{-1} |\nabla C|^{-1}$, the arbitrariness is best regulated by choosing this factor to give unity for the Nusselt number in the fundamental and easily remembered case of pure diffusion from a sphere.

2. Mass transfer from a particle at small Péclet number

General argument

Whatever the particle shape, the steady distribution of concentration due to diffusion in stationary fluid (\hat{C} , say) becomes spherically symmetrical at large distances from the particle, and

$$\hat{C} - C_0 \sim \frac{Q_0}{4\pi\kappa r} \quad \text{when } r \gg a.$$

When $P \ll 1$, the effect of motion of the fluid is to modify this distribution at large values of r and to change the transfer rate. Concentration gradients become weaker with increasing distance from the particle, and the convection term $\mathbf{u} \cdot \nabla C$ in the governing equation (1.1) becomes relatively more important as $r/a \rightarrow \infty$. On the assumption that spatial differentiation changes the magnitude of a quantity by a factor r^{-1} , we estimate the ratio of the convection and diffusion terms as of order rU/κ , where U is the magnitude of the ambient velocity at distance r from the particle. Thus at positions near the particle, where rU/κ is of the same order of magnitude as the Péclet number P , diffusion effects are dominant, whereas at large distances such that $rU/\kappa \gg 1$ the local distribution of C is dominated by convection effects.

This problem is of the familiar type for which different asymptotic representations of the concentration are applicable in different parts of the field and a uniformly valid approximation to C may be obtained by matching the two asymptotic representations. Matched asymptotic expansions of this kind and associated expressions for the transfer rate at small Péclet number have been obtained by Acrivos & Taylor (1962) for the cases of a rigid sphere and a rigid circular cylinder in steady translational motion, by Brenner (1963) for a particle of arbitrary shape in steady translational motion, and by Frankel & Acrivos (1968) for the case of a rigid sphere in steady simple shear flow. Here we adopt a simple intuitive procedure which is adequate for the purpose of obtaining the leading term in the expression for the change in the transfer rate due to convection. This leading term depends only on the ambient flow and not on the flow field near the particle, unlike all further terms. The further terms give numerical information which is of limited value for the purpose of extrapolating between results for low and high Péclet number.

We denote by r_c (which is defined to order of magnitude only) the large value of r at which the convection and diffusion terms in the governing equation are comparable in magnitude; thus rU/κ is of order unity at $r = r_c$ (giving $r_c/a = O(P^{-1})$ for a particle in translational motion and $r_c/a = O(P^{-\frac{1}{2}})$ for a linear ambient velocity field). The transfer of solute from the particle surface to $r = r_c$ is brought about primarily by diffusion, and

$$C - C_0 \approx \frac{Q}{4\pi\kappa r_c} \tag{2.1}$$

at $r = r_c$ in a case where the total rate of transfer is Q . The transfer of solute in the range $r_c < r < \infty$ on the other hand takes place primarily by the much more efficient process of convection, and the associated change in concentration is negligible. The effect of convection on the total rate of transfer is thus equivalent to increase of the overall concentration difference, in a pure diffusion problem, by a fraction

$$\frac{Q}{4\pi\kappa r_c(C_1 - C_0)}, \quad \approx \frac{Q_0}{4\pi\kappa r_c(C_1 - C_0)} \sim \frac{a}{r_c}.$$

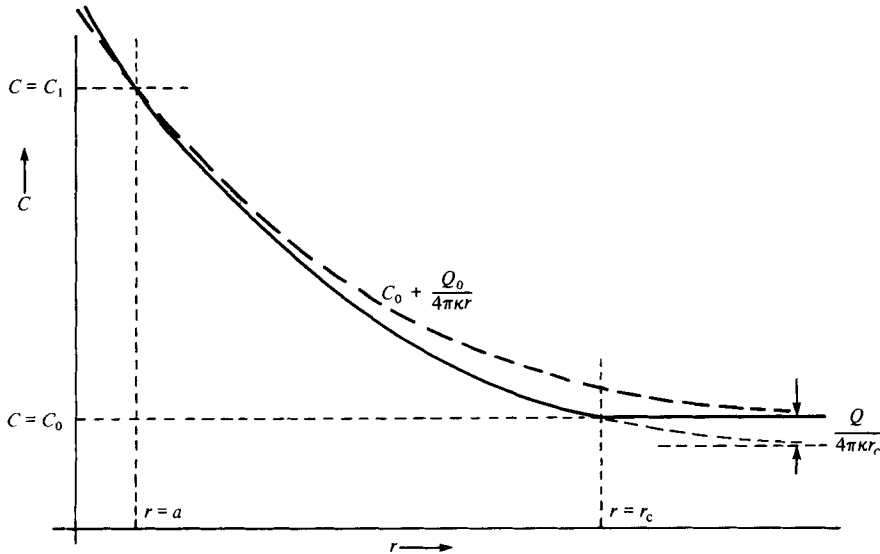


FIGURE 1. The unbroken curve shows schematically the distribution of concentration C at small Péclet number. For $r < r_c$, where diffusion effects are dominant, the curve is of the form

$$C_0 + Q/(4\pi\kappa r) + \text{const.}$$

(except near the surface of a non-spherical particle); and for $r > r_c$, where convection effects are dominant, C is approximately constant. The broken curve shows the concentration distribution at zero Péclet number with the same inner and outer boundary conditions.

And since in the linear pure diffusion problem the rate of transfer is proportional to the overall concentration drop, we see that the fractional increase in the transfer rate due to convection when $P \ll 1$ is

$$\frac{\Delta Q}{Q_0} \sim \frac{a}{r_c}. \quad (2.2)$$

Figure 1 illustrates this simplified description of the distribution of concentration.

This relation (2.2) gives the dependence of ΔQ on the Péclet number (as P for a particle in translational motion and as $P^{1/2}$ for a linear ambient velocity field) but for numerical information we must match the inner and outer asymptotic regions to better than order of magnitude. In the inner region the pure diffusion equation is applicable, with the boundary condition $C = C_1$ at the particle surface A , and in the outer part of this inner region the departure from spherical symmetry of the concentration distribution due to particle shape becomes negligible. In the outer region where convection is important the fluid velocity \mathbf{u} is approximately equal to the ambient value \mathbf{U} and the governing equation is

$$\mathbf{U} \cdot \nabla C = \kappa \nabla^2 C, \quad (2.3)$$

with the boundary condition $C \rightarrow C_0$ as $r \rightarrow \infty$ and an inner boundary condition representing the fact that C is spherically symmetrical and determined by diffusion alone there. The concentration distribution in the outer region is thus the same as if the particle were replaced by a point source of strength Q in the given ambient flow field. In the inner part of this outer region $C - C_0$ is approximately equal to $Q/4\pi\kappa r$,

and an improved approximation at small values of r , assuming analytic dependence on r , will be

$$C - C_0 \approx \frac{Q}{4\pi\kappa r} + \Delta C, \tag{2.4}$$

where the second term ΔC is independent of r but may depend on the direction of \mathbf{x} (this departure from spherical symmetry being a consequence of the effect of convection). This is the outer boundary condition for the distribution of concentration in the inner region.

The additional transfer from the particle due to convection is now obtainable as a consequence of the fact that in the inner region C satisfies the diffusion equation† and an outer boundary condition of the form (2.4) at some large value of r , say $r = R$. We may write C as the sum of two solutions of $\nabla^2 C = 0$, C' and C'' say, where

$$C' = C_1 \text{ for } \mathbf{x} \text{ on } A \quad \text{and} \quad C' = C_0 + \frac{Q}{4\pi\kappa r} + \langle \Delta C \rangle \quad \text{at} \quad r = R$$

and

$$C'' = 0 \text{ for } \mathbf{x} \text{ on } A \quad \text{and} \quad C'' = \Delta C - \langle \Delta C \rangle \quad \text{at} \quad r = R,$$

where $\langle \Delta C \rangle$ denotes the mean of ΔC over all directions of \mathbf{x} . Near $r = R$ the harmonic function C'' can be expressed as a series of spherical harmonics from which those of degree 0 and -1 are excluded, because their means over all directions of \mathbf{x} are non-zero, and so C'' makes no contribution to the transfer across any closed surface. Also we know that if the outer boundary condition for C' were $C' = C_0 + Q/4\pi\kappa R$ at $r = R$ the rate of transfer from the particle surface would be Q_0 . The rate of transfer from the particle surface associated with C' evidently differs from Q_0 in consequence of a change in the overall concentration difference (between the particle surface and infinity) by the amount $-\langle \Delta C \rangle$. The additional rate of transfer due to convection is therefore given by

$$\frac{N - N_0}{N_0} = \frac{\Delta Q}{Q_0} = -\frac{\langle \Delta C \rangle}{C_1 - C_0}. \tag{2.5}$$

Determination of ΔQ is thus reduced to the problem of finding the term ΔC in the expression (2.4) for the concentration at small values of r in the case of a point source in the given ambient flow field.

† More accurately, the equation satisfied by C in the inner region is

$$\kappa \nabla^2 C = \mathbf{u} \cdot \nabla \hat{C},$$

but, as Brenner (1963) showed for the case of a particle in translational motion, the small source term on the right-hand side makes no contribution to the transfer from the particle. For if C_i denotes the solution of the inhomogeneous equation with boundary conditions $C_i = 0$ for \mathbf{x} on A and at $r = R$, we have

$$\nabla \cdot (\mathbf{q}_i + \hat{C} \mathbf{u}) = 0 \quad \text{and} \quad \nabla \cdot (\hat{C} \mathbf{q}_i - C_i \hat{\mathbf{q}} + \frac{1}{2} \hat{C}^2 \mathbf{u}) = 0,$$

where

$$\mathbf{q}_i = -\kappa \nabla C_i \quad \text{and} \quad \hat{\mathbf{q}} = -\kappa \nabla \hat{C};$$

and so

$$\int_A \mathbf{n} \cdot \mathbf{q}_i dS = \int_{r=R} \mathbf{n} \cdot \mathbf{q}_i dS \quad \text{and} \quad C_0 \int_A \mathbf{n} \cdot \mathbf{q}_i dS = C_1 \int_{r=R} \mathbf{n} \cdot \mathbf{q}_i dS,$$

giving

$$\int_A \mathbf{n} \cdot \mathbf{q}_i dS = 0.$$

This expression for $\Delta Q/Q_0$ depends only on N_0 , P and the ambient flow field, and does not otherwise depend on the particle shape or size. Nor does it depend on the distribution of fluid velocity near the particle. Consequently there is no need here for any assumption about the particle Reynolds number. However this latter gain in generality is not of much practical value because solute Prandtl numbers are usually larger than unity and if the Péclet number is small the particle Reynolds number is even smaller.

A particle in steady translational motion through the fluid

The fluid velocity \mathbf{U} in (2.3) is here uniform and equal to \mathbf{U}_0 . Results are already available for this case, but we shall recover them in passing in order to show the applicability of the general method just described.

We require the solution of (2.3) corresponding to a point source of constant strength Q at the origin. By superimposing the distributions of concentration for a sequence of instantaneous point sources in a uniform stream we find

$$C(\mathbf{x}) = C_0 + Q \int_0^\infty \exp\left\{-\frac{(\mathbf{x} - t\mathbf{U}_0)^2}{4\kappa t}\right\} \frac{dt}{(4\pi\kappa t)^{\frac{3}{2}}}. \quad (2.6)$$

The integration in (2.6) can be carried out, giving

$$C(\mathbf{x}) = C_0 + \frac{Q}{4\pi\kappa r} \exp\left(\frac{\mathbf{x} \cdot \mathbf{U}_0 - rU_0}{2\kappa}\right). \quad (2.7)$$

Thus, at small values of r ,

$$C - C_0 \approx \frac{Q}{4\pi\kappa r} + \frac{QU_0}{8\pi\kappa^2} (\cos\theta - 1), \quad (2.8)$$

where θ is the angle between the position vector \mathbf{x} and the free-stream velocity \mathbf{U}_0 .

The second term on the right-hand side of (2.8) can be identified with the quantity ΔC in the general argument, and then from (2.5) we see that the fractional increase in the transfer rate due to the effect of convection is, to leading order,

$$\begin{aligned} \frac{N - N_0}{N_0} &= \frac{\Delta Q}{Q_0} = \frac{Q_0 U_0}{8\pi\kappa^2(C_1 - C_0)} \\ &= \frac{1}{2} N_0 P, \end{aligned} \quad (2.9)$$

where $P = aU_0/\kappa$, as found by Brenner (1963). Brenner showed in addition that for a particle of arbitrary shape and constitution, and for low-Reynolds-number flow, the next term in the expansion is $\frac{1}{2}N_0 f P^2 \log P$, where $6\pi\mu a U_0 f$ is the magnitude of the force exerted across the particle surface. Acrivos & Taylor (1962) also obtained the result (2.9) for the case of a rigid spherical particle, for which $N_0 = 1$, and went on to calculate higher-order terms, in $P^2 \log P$, P^2 and $P^3 \log P$, for low-Reynolds-number flow.

A point source in fluid with a steady linear ambient velocity distribution

Consider now the case in which the fluid velocity in the absence of the particle has the linear form

$$U_i = G_{ij} x_j = (E_{ij} + \Omega_{ij}) x_j, \quad (2.10)$$

where the coefficients satisfy the incompressibility relations

$$G_{ii} = E_{ii} = 0.$$

We require to find the concentration distribution for a maintained point source of material at the origin in fluid whose velocity is \mathbf{U} . This problem has been solved previously for a steady simple shearing motion of the fluid by Novikov (1958) (and again, independently, by Elrick (1962)), and it is not difficult to construct the solution for the rather simpler case of steady pure straining motion.† The method to be used here is applicable to any linear ambient velocity distribution and can be carried to the point of explicit numerical results for a representative set of particular linear ambient velocity fields.

We begin by seeking a solution for the three-dimensional Fourier transform of the concentration distribution in the case of an instantaneous source of strength Q_i at the origin. The concentration $C(\mathbf{x}, t)$ here satisfies the equation

$$\frac{\partial C}{\partial t} + G_{ij} x_j \frac{\partial C}{\partial x_i} = \kappa \nabla^2 C,$$

of which the transform is

$$\frac{\partial \hat{C}}{\partial t} - G_{ij} k_i \frac{\partial \hat{C}}{\partial k_j} = -\kappa k^2 \hat{C}, \tag{2.11}$$

where \hat{C} is defined by

$$\hat{C}(\mathbf{k}, t) = \int e^{-i\mathbf{k} \cdot \mathbf{x}} \{C(\mathbf{x}, t) - C_0\} d\mathbf{x}.$$

Instead of trying to solve (2.11) directly it is simpler to note that the solution must be of the form

$$\hat{C}(\mathbf{k}, t) = Q_i \exp(-\kappa k_i k_j B_{ij}) \tag{2.12}$$

and to choose the symmetric tensor \mathbf{B} as a function of t so as to satisfy the equation. (The form (2.12) follows from the facts (a) that $-G_{ij} k_i$ can be interpreted as a velocity in \mathbf{k} -space, in which event the left-hand side of (2.11) becomes the material derivative of \hat{C} , (b) that the relation between the wavenumber vectors representing the positions of a material element in \mathbf{k} -space at two different times is linear, and (c) that at $t = 0$, when C has a delta-function distribution with magnitude Q_i , \hat{C} is uniform and equal to Q_i .) Now the expression (2.12) satisfies the equation (2.11) provided

$$k_i k_j \frac{dB_{ij}}{dt} = k^2 + 2k_i k_j G_{ij} B_{ij},$$

which requires

$$\frac{dB_{ij}}{dt} = \delta_{ij} + G_{il} B_{jl} + G_{jl} B_{il}. \tag{2.13}$$

All the components of \mathbf{B} can be found as functions of t from this equation and the boundary condition

$$B_{ij} \sim \delta_{ij} t \quad \text{as } t \rightarrow 0, \tag{2.14}$$

but we shall do this explicitly only for certain linear ambient velocity fields.

† See Townsend (1951) for the temperature distribution due to an instantaneous source of heat in a pure straining motion with constant directions of the principal axes of the rate-of-strain tensor, from which the distribution for a maintained source follows by integration.

The Fourier transform of the concentration distribution for a maintained source at the origin of steady strength Q is now found by integrating (2.12) to be

$$\hat{C}(\mathbf{k}) = Q \int_0^\infty \exp(-\kappa k_i k_j B_{ij}) dt. \quad (2.15)$$

The corresponding distribution in physical space is the Fourier transform of (2.15), viz.

$$C(\mathbf{x}) = C_0 + \frac{Q}{(4\pi\kappa)^{\frac{3}{2}}} \int \exp\left(\frac{-x_i x_j b_{ij}}{4\kappa D}\right) \frac{dt}{D^{\frac{3}{2}}}, \quad (2.16)$$

where D denotes the determinant of the matrix \mathbf{B} and b_{ij} is the co-factor of the matrix element B_{ij} . This expression for C can be written as

$$C(\mathbf{x}) = C_0 + \frac{Q}{4\pi\kappa r} + J(\mathbf{x}), \quad (2.17)$$

where

$$J = \frac{Q}{(4\pi\kappa)^{\frac{3}{2}}} \int_0^\infty \left\{ \frac{1}{D^{\frac{3}{2}}} \exp\left(\frac{r^2}{4\kappa t} - \frac{x_i x_j b_{ij}}{4\kappa D}\right) - \frac{1}{t^{\frac{3}{2}}} \right\} \exp\left(\frac{-r^2}{4\kappa t}\right) dt.$$

It follows from (2.14) that

$$tb_{ij}/D \sim \delta_{ij} \quad \text{as } t \rightarrow 0,$$

with an error of order t ; hence, as $r \rightarrow 0$, J approaches a constant, which can be identified with ΔC in (2.4). We have

$$\Delta C = \lim_{r \rightarrow 0} J = \frac{Q}{(4\pi\kappa)^{\frac{3}{2}}} \int_0^\infty (D^{-\frac{3}{2}} - t^{-\frac{3}{2}}) dt. \quad (2.18)$$

In this case of a linear ambient velocity distribution, convection does not cause any departure from spherical symmetry in the term of order r^0 in (2.4).

The additional transfer from the particle due to convection

The fractional increase in the transfer rate due to convection now follows from (2.5) and (2.18), and since Q differs from Q_0 by a small quantity only we have, to leading order,

$$\begin{aligned} \frac{N - N_0}{N_0} &= \frac{\Delta Q}{Q_0} = \frac{Q_0}{(C_1 - C_0)(4\pi\kappa)^{\frac{3}{2}}} \int_0^\infty (t^{-\frac{3}{2}} - D^{-\frac{3}{2}}) dt \\ &= \frac{N_0 a}{(4\pi\kappa)^{\frac{3}{2}}} \int_0^\infty \frac{1 - (t^3/D)^{\frac{1}{2}}}{t^{\frac{3}{2}}} dt. \end{aligned} \quad (2.19)$$

Further progress requires a knowledge of $D(t)$, the determinant of the matrix \mathbf{B} whose elements B_{ij} are given by the equation (2.13). We now find B_{ij} and D for some particular linear ambient velocity distributions.

First, for a steady simple shearing motion with velocity components $(\gamma x_2, 0, 0)$, for which a complete solution for the concentration distribution due to a steady point source was found by Novikov (1958) and Elrick (1962), the only non-zero components of \mathbf{G} and \mathbf{E} are

$$G_{12} = \gamma, \quad E_{12} = E_{21} = \frac{1}{2}\gamma.$$

The equation (2.13) and boundary condition (2.14) are satisfied by

$$B_{11} = t(1 + \frac{1}{3}\gamma^2 t^2), \quad B_{22} = t, \quad B_{33} = t, \quad B_{12} = \frac{1}{2}\gamma t^2, \quad B_{23} = B_{31} = 0. \quad (2.20)$$

Hence

$$D = (B_{11} B_{22} - B_{12}^2) B_{33} = t^3(1 + \frac{1}{12}\gamma^2 t^2), \quad (2.21)$$

and so from (2.19)

$$\frac{N - N_0}{N_0} = N_0 \left(\frac{a^2 \gamma}{4\pi \kappa} \right)^{\frac{1}{2}} \int_0^\infty \frac{1 - (1 + \frac{1}{12}q^2)^{-\frac{1}{2}}}{q^{\frac{3}{2}}} dq. \quad (2.22)$$

This integral can be expressed in terms of tabulated gamma functions. The additional transfer from the particle due to convection, as already found by Frankel & Acrivos (1968) for this case, is

$$\frac{N - N_0}{N_0} = 0.257 N_0 \left(\frac{a^2 \gamma}{\kappa} \right)^{\frac{1}{2}} \quad \text{or} \quad 0.305 N_0 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}}. \quad (2.23)$$

The alternative expression of the result in terms of the quantity E defined as $(E_{ij} E_{ij})^{\frac{1}{2}}$, which here has the value $\gamma/\sqrt{2}$, will be commented on later.

The transfer due to convection may also be calculated readily in the case of an ambient steady pure straining motion, for which $\mathbf{G} = \mathbf{E}$. We use axes coinciding with the principal axes of \mathbf{E} , and the three principal rates of strain will be denoted by E_1, E_2, E_3 . The equations (2.13) and boundary condition (2.14) are here satisfied by

$$B_{11} = \frac{\exp(2E_1 t) - 1}{2E_1}, \quad B_{22} = \frac{\exp(2E_2 t) - 1}{2E_2}, \quad B_{33} = \frac{\exp(2E_3 t) - 1}{2E_3}, \quad (2.24)$$

other components of B_{ij} being zero, and so

$$D = B_{11} B_{22} B_{33} = \frac{\sinh E_1 t \sinh E_2 t \sinh E_3 t}{E_1 E_2 E_3}. \quad (2.25)$$

Hence we find from (2.19)

$$\frac{N - N_0}{N_0} = N_0 \left(\frac{a^2 E}{4\pi \kappa} \right)^{\frac{1}{2}} \int_0^\infty \left[1 - \left\{ \frac{q^3 E_1 E_2 E_3 / E^3}{\sinh(E_1 q / E) \sinh(E_2 q / E) \sinh(E_3 q / E)} \right\}^{\frac{1}{2}} \right] \frac{dq}{q^{\frac{3}{2}}}, \quad (2.26)$$

where $E^2 = E_1^2 + E_2^2 + E_3^2$. Note that the additional transfer due to convection is unchanged by reversal of the signs of E_1, E_2 and E_3 , which is reminiscent of a general theorem due to Brenner (1967).† The integral in (2.26) can be evaluated numerically for particular values of the ratios of the principal rates of strain. The following two cases are the simplest ones:

(a) two-dimensional pure straining motion, for which

$$E_1 = -E_2, \quad E_3 = 0 \quad \text{and} \quad E = \sqrt{2|E_1|},$$

† Brenner's proof that the rate of transfer from a particle is unchanged by reversal of the flow velocity everywhere, regardless of the particle shape or the value of the Péclet number, applies to the case of a particle in translational motion but may be adapted to hold also for a linear ambient flow field. In a later paper Brenner (1970) found that the result holds also for some different forms of the condition satisfied by C at the particle surface.

the result being

$$\frac{N - N_0}{N_0} = 0.428N_0 \left(\frac{a^2 |E_1|}{\kappa} \right)^{\frac{1}{2}} \quad \text{or} \quad 0.360N_0 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}}; \quad (2.27)$$

(b) axisymmetric pure straining motion, for which

$$E_1 = E_2 = -\frac{1}{2}E_3 \quad \text{and} \quad E = \left(\frac{3}{2}\right)^{\frac{1}{2}} |E_3|,$$

the result being

$$\frac{N - N_0}{N_0} = 0.399N_0 \left(\frac{a^2 |E_3|}{\kappa} \right)^{\frac{1}{2}} \quad \text{or} \quad 0.360N_0 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}}. \quad (2.28)$$

The transfer rate for an ambient pure straining motion can be regarded as a function of E and of one parameter which determines the type of straining motion and which we may choose as $(E_1 - E_2)/E$. The whole set of geometrically different pure straining motions are covered if we begin with axisymmetric compression in the x_3 direction (for which $E_1 > 0$, $E_2 > 0$ and $(E_1 - E_2)/E = 0$) and decrease E_2 with E_1 fixed until we reach $E_2 = 0$ and $(E_1 - E_2)/E = 1/\sqrt{2}$, corresponding to two-dimensional motion in the (x_3, x_1) plane, and then decrease E_2 further with E_1 fixed until we reach $E_2 = -\frac{1}{2}E_1$ and $(E_1 - E_2)/E = \sqrt{\frac{3}{2}}$, corresponding to axisymmetric extension in the x_1 direction. But bearing in mind that the transfer rate is invariant to flow reversal, we see that it is sufficient to consider values of $(E_1 - E_2)/E$ in the range from 0 to $1/\sqrt{2}$ and that the value of N is stationary at the two end points of this range. The fact that the transfer rate is the same multiple of $(a^2 E/\kappa)^{\frac{1}{2}}$ at the two end points of this range indicates that the relation

$$\frac{N - N_0}{N_0} = 0.36N_0 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}} \quad (2.29)$$

is likely to give accurate results for any pure straining motion.

A third case for which a solution of (2.13) is obtainable in simple closed form is two-dimensional motion, which we may represent by

$$G_{12} = E_{12} - \Omega, \quad G_{21} = E_{12} + \Omega,$$

all other components of \mathbf{G} being zero. The solution of (2.13) is here

$$B_{11}(E_{12} + \Omega) - \Omega t = B_{22}(E_{12} - \Omega) + \Omega t = \frac{\frac{1}{2}E_{12} \sinh \{2(E_{12}^2 - \Omega^2)^{\frac{1}{2}} t\}}{(E_{12}^2 - \Omega^2)^{\frac{1}{2}}},$$

$$B_{12} = -\frac{\frac{1}{2}E_{12}}{E_{12}^2 - \Omega^2} + \frac{\frac{1}{2}E_{12} \cosh \{2(E_{12}^2 - \Omega^2)^{\frac{1}{2}} t\}}{E_{12}^2 - \Omega^2},$$

$$B_{33} = t, \quad B_{23} = B_{31} = 0.$$

Hence

$$D = \frac{E_{12}^2 t \sinh^2 \{(E_{12}^2 - \Omega^2)^{\frac{1}{2}} t\}}{(E_{12}^2 - \Omega^2)^2} - \frac{\Omega^2 t^3}{E_{12}^2 - \Omega^2}, \quad (2.30)$$

which reduces to (2.20) in the particular case $E_{12} = -\Omega = \frac{1}{2}\gamma$ corresponding to simple shearing motion and to (2.25) in the case $\Omega = 0$ corresponding to pure straining motion with principal rates of strain $E_{12}, -E_{12}, 0$. The transfer rate follows from (2.19), although numerical integration seems to be necessary. It is hardly worth undertaking

since we already have the result for the cases $|\Omega/E_{12}| = 0$ and 1 and will discuss in a moment the case $|\Omega/E_{12}| \gg 1$.

An approximate expression for $(N - N_0)/N_0$ which would hold for an even wider class of linear ambient velocity distribution than pure straining motion or two-dimensional motion would of course be useful. Such an approximation can be found by writing B_{ij} as a power series in t :

$$B_{ij}(t) = t\delta_{ij} + t^2B_{ij}^{(2)} + t^3B_{ij}^{(3)} + \dots$$

and by using the equation (2.13) to determine the first few coefficients. We find

$$B_{ij}^{(2)} = \frac{1}{2}(G_{ij} + G_{ji}) = E_{ij}$$

$$B_{ij}^{(3)} = \frac{2}{3}E_{il}E_{jl} + \frac{1}{3}(E_{il}\Omega_{jl} + E_{jl}\Omega_{il}).$$

If now we choose the axes of reference to coincide with the principal axes of the rate-of-strain tensor \mathbf{E} , the non-diagonal components of $\mathbf{B}^{(2)}$ and $\mathbf{B}^{(3)}$ are zero and so, correct to terms of order t^2 ,

$$\left(\frac{D}{t^3}\right)^{\frac{1}{2}} = \left(\frac{B_{11}B_{22}B_{33}}{t^3}\right)^{\frac{1}{2}} = 1 + \frac{1}{12}E^2t^2, \tag{2.31}$$

where $E = E_{ij}E_{ij}$ as before. The value of $(N - N_0)/N_0$ corresponding to this approximate expression for D is seen from (2.19) to be

$$\frac{N - N_0}{N_0} = N_0 \left(\frac{a^2E}{4\pi\kappa}\right)^{\frac{1}{2}} \int_0^\infty \frac{\frac{1}{12}q^{\frac{1}{2}}}{1 + \frac{1}{12}q^2} dq. \tag{2.32}$$

The integral may be expressed in terms of gamma functions, giving

$$\frac{N - N_0}{N_0} = \frac{\Gamma(\frac{1}{4})\Gamma(\frac{3}{4})}{4(12\pi^2)^{\frac{1}{2}}} N_0 \left(\frac{a^2E}{\kappa}\right)^{\frac{1}{2}} = 0.337N_0 \left(\frac{a^2E}{\kappa}\right)^{\frac{1}{2}}. \tag{2.33}$$

Further terms in the series describe the way in which D/t^3 varies at large t and, provided $D/t^3 \rightarrow \infty$ as $t \rightarrow \infty$ (which amounts to a requirement that the cloud of material is extended appreciably by convection in at least one direction), it is evident that these further terms do not have much effect on the value of the integral in (2.19). The numerical coefficient in (2.33) is 6% too small in the case of pure straining motion and 10% too large in the case of simple shearing. Aside from providing a fair estimate of the transfer for a very wide class of linear ambient velocity distributions, (2.33) reveals E as the primary parameter determining the convective transfer (as had been noticed earlier from the results for different pure straining motions).

The partial suppression of the effect of convection by strong rotation

There is one circumstance in which the estimate (2.33) will be less accurate, *viz.* when the magnitude of the angular velocity vector ($\frac{1}{2}|\boldsymbol{\omega}|$, = Ω say) is large relative to the components of the rate-of-strain tensor. The effect of a strong rotation is to change the form of the streamlines so that instead of being open curves extending to infinity they resemble closed curves enclosing the rotation axis, and these closed curves become more nearly circular as Ω increases still further, with a consequent suppression of the contribution to the transfer due to the convection associated with some (but not all) of the components of the rate-of-strain tensor.

This may be seen most clearly from the above consideration of two-dimensional ambient motion, in the (x_1, x_2) plane. The components of $\frac{1}{2}\boldsymbol{\omega}$ are here $(0, 0, \Omega)$, and the directions of the x_1 and x_2 axes are such that $E_{11} = 0, E_{22} = 0$. The stream function describing this motion is

$$\psi = -\frac{1}{2}x_1^2(E_{12} + \Omega) + \frac{1}{2}x_2^2(E_{12} - \Omega), \tag{2.34}$$

and the streamlines are hyperbolae when $|\Omega/E_{12}| < 1$ and ellipses when $|\Omega/E_{12}| > 1$. The ratio of the principal diameters of the ellipses tends to unity as $E_{12}/\Omega \rightarrow 0$, and in this limit the increase in the rate of transfer from the point source at the origin due to convection is zero. Analytically, we see from (2.19) and (2.30) that the transfer rate is given by

$$\frac{N - N_0}{N_0} = N_0 \frac{a(\Omega^2 - E_{12}^2)^{\frac{1}{2}}}{(4\pi\kappa)^{\frac{1}{2}}} \int_0^\infty \left\{ 1 - \frac{(\Omega^2 - E_{12}^2)^{\frac{1}{2}} s}{(\Omega^2 s^2 - E_{12}^2 \sin^2 s)^{\frac{1}{2}}} \right\} \frac{ds}{s^{\frac{3}{2}}}$$

when $|\Omega/E_{12}| > 1$. Hence, as $|\Omega/E_{12}| \rightarrow \infty$

$$\begin{aligned} \frac{N - N_0}{N_0} &\sim N_0 \left(\frac{a^2 E_{12}}{\kappa} \right)^{\frac{1}{2}} \left(\frac{E_{12}}{\Omega} \right)^{\frac{3}{2}} \frac{1}{4\sqrt{\pi}} \int_0^\infty \left(1 - \frac{\sin^2 s}{s^2} \right) \frac{ds}{s^{\frac{3}{2}}} \\ &= \frac{2}{15} N_0 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}} \left(\frac{E}{\Omega} \right)^{\frac{3}{2}}, \end{aligned} \tag{2.35}$$

E being equal to $(2E_{12}^2)^{\frac{1}{2}}$ in this case. The suppression of the convective transfer by rotation in this case of two-dimensional motion is thus quite strong.

But on the other hand it is clear that, if an axisymmetric extensional motion, with the axis of symmetry coinciding with the x_3 axis, is combined with rotation about the x_3 axis, the convective transfer due to this axisymmetric extensional motion is unaffected by the rotation.

Consider now the general linear ambient velocity distribution, with the x_3 axis in the direction of $\boldsymbol{\omega}$ and the directions of the x_1 and x_2 axes such that $E_{11} = E_{22}, = -\frac{1}{2}E_{33}$. We shall write $\mathbf{E} = \mathbf{E}^{(0)} + \mathbf{E}^{(1)}$, where

$$\mathbf{E}^{(0)} = \begin{pmatrix} -\frac{1}{2}E_{33} & 0 & 0 \\ 0 & -\frac{1}{2}E_{33} & 0 \\ 0 & 0 & E_{33} \end{pmatrix}, \quad \mathbf{E}^{(1)} = \begin{pmatrix} 0 & E_{12} & E_{13} \\ E_{21} & 0 & E_{23} \\ E_{31} & E_{32} & 0 \end{pmatrix}, \quad \boldsymbol{\Omega} = \begin{pmatrix} 0 & -\Omega & 0 \\ \Omega & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

and we shall suppose that all the components of $\mathbf{E}^{(1)}$ have small magnitude relative to Ω (there being no need for any restriction on E_{33}). The equations determining the components of \mathbf{B} are now obtained by substituting in (2.13). It can be seen without difficulty that they are satisfied by the following perturbation scheme:

$$B_{ij}(t) = t \left\{ B_{ij}^{(0)}(t) + \frac{E'}{\Omega} B_{ij}^{(1)}(T) + O\left(\frac{E'^2}{\Omega^2}\right) \right\}, \tag{2.36}$$

where $T = \Omega t, E'^2 = E_{12}^2 + E_{23}^2 + E_{31}^2$ (E' being a convenient common measure of the magnitudes of E_{12}, E_{23} and E_{31}), and $tB_{ij}^{(0)}(t)$ denotes the solution for a pure straining motion consisting of a principal rate of extension E_{33} in the direction of the x_3 axis and principal rates of extension $-\frac{1}{2}E_{33}$ in two orthogonal directions (so that $tB_{11}^{(0)},$

$tB_{22}^{(0)}, tB_{33}^{(0)}$ are given by (2.24) with $E_1 = E_2 = -\frac{1}{2}E_{33}, E_3 = E_{33}$. The equation for $B_{ij}^{(1)}$ is

$$\frac{dB_{ij}^{(1)}}{dT} + \frac{B_{ij}^{(1)}}{T} = \frac{E_{ii}^{(0)} + \Omega_{ii}}{\Omega} B_{ji}^{(1)} + \frac{E_{ji}^{(0)} + \Omega_{ji}}{\Omega} B_{ii}^{(1)} + \frac{E_{ii}^{(1)}}{E'} B_{ji}^{(0)} + \frac{E_{ji}^{(1)}}{E'} B_{ii}^{(0)}, \quad (2.37)$$

and $B_{ij}^{(1)} = 0$ at $T = 0$ in order to ensure that (2.36) satisfies (2.14). However, there is no need to solve this equation for all the perturbation quantities $B_{ij}^{(1)}$, because we are interested only in the determinant D , which is given by

$$D = t^3 B_{11}^{(0)} B_{22}^{(0)} B_{33}^{(0)} \left\{ 1 + \frac{E'}{\Omega} \left(\frac{B_{11}^{(1)}}{B_{11}^{(0)}} + \frac{B_{22}^{(1)}}{B_{22}^{(0)}} + \frac{B_{33}^{(1)}}{B_{33}^{(0)}} \right) \right\}$$

correct to the first order in the small quantity E'/Ω , and we find immediately that

$$B_{11}^{(1)} + B_{22}^{(1)} = 0, \quad B_{33}^{(1)} = 0.$$

It appears therefore that when the angular velocity Ω is large the rate of transfer from the point source is equal, correct to the first order in the small quantity E'/Ω , to that associated with an axisymmetric pure straining motion with rate of extension E_{33} in the direction of the axis of symmetry, that is, in terms of quantities which are independent of the axes of reference, with rate of extension

$$E_\omega = \boldsymbol{\omega} \cdot \mathbf{E} \cdot \boldsymbol{\omega} / \omega^2 \quad (2.38)$$

in the direction of the axis of symmetry. The formula (2.28) then gives

$$\frac{N - N_0}{N_0} = 0.399 N_0 \left(\frac{\alpha^2 |E_\omega|}{\kappa} \right)^{\frac{1}{2}}. \quad (2.39)$$

The case in which E'/Ω and $|E_\omega|/E$ are both small is clearly exceptional, and

$$(N - N_0)/N_0$$

is then very small although the exact order of smallness is uncertain.

3. Mass transfer from a particle at large Péclet number

At large values of the Péclet number concentration gradients are large near the particle surface, in general, and boundary-layer theory may be applied to the concentration distribution. Furthermore, if the Prandtl number ν/κ is large compared with unity, as we shall assume, velocity gradients change much less rapidly than concentration gradients near the particle surface, and so the velocity gradient is approximately constant across the concentration boundary layer. This is the basis of an extensive body of theory concerning the mass transfer from a stationary rigid particle in a steady flow field (Levich 1962). We shall describe here a generalized version of the theory which applies to cases in which the tangential stress and solute flux density at the particle surface are functions of more than one position co-ordinate, and will then use it to calculate the total transfer rate from a spherical particle immersed in steady ambient pure straining motions of different kinds at small Reynolds number. The theory applies only when the flow relative to a *stationary* particle surface is steady, and so cases of force-free couple-free spherical particles immersed in fluid for which the ambient vorticity is non-zero are excluded, in general, because the

particles then rotate; but we shall see later that, rather surprisingly, the new theory that is needed for such cases is quite simple.

General theory for a stationary particle in a steady flow field

The steady concentration distribution is a solution of the equation

$$\mathbf{u} \cdot \nabla C = \kappa \nabla^2 C,$$

with the boundary conditions

$$C = C_1 \quad \text{at the particle surface,} \quad C \rightarrow C_0 \quad \text{as} \quad |\mathbf{x}| \rightarrow \infty.$$

Within a concentration boundary layer adjoining the particle surface only the contribution to ∇^2 from the normal derivative is significant. The normal derivative of the component of \mathbf{u} parallel to the particle surface will be taken as constant across the concentration boundary layer, as already mentioned; and when the particle is rigid and the fluid velocity is zero at the particle surface, the direction of the fluid velocity is constant across the concentration boundary layer. We introduce orthogonal co-ordinates ξ, η, ζ such that ξ is the normal distance from the particle surface, and the η -co-ordinate line is parallel to the local fluid velocity. The η -co-ordinate lines in the particle surface are parallel to the tangential viscous stress there and define what we shall call the 'surface streamlines'. The above equation thus reduces approximately to

$$u_\xi \frac{\partial C}{\partial \xi} + \frac{u_\eta}{h_\eta} \frac{\partial C}{\partial \eta} = \kappa \frac{\partial^2 C}{\partial \xi^2} \quad (3.1)$$

in the concentration boundary layer, where $h_\xi (= 1), h_\eta, h_\zeta$ are the three metric scale factors and h_η and h_ζ are functions of η and ζ . The velocity components are of the form

$$u_\xi \propto \xi^2, \quad u_\eta = \xi F(\eta, \zeta), \quad u_\zeta = 0, \quad (3.2)$$

where $F(> 0)$ is the magnitude of the local tangential stress divided by the fluid viscosity.

Since $u_\zeta = 0$ everywhere within the boundary layer, we can define a stream function ψ to which the velocity components u_ξ and u_η are related in the usual way, *viz.*

$$u_\xi = -\frac{1}{h_\eta h_\zeta} \frac{\partial \psi}{\partial \eta}, \quad u_\eta = \frac{1}{h_\zeta} \frac{\partial \psi}{\partial \xi},$$

and comparison with (3.2) gives

$$\psi = \frac{1}{2} \xi^2 h_\zeta F. \quad (3.3)$$

If now we write the differential equation (3.1) with ψ, η as independent variables in place of ξ, η (a familiar transformation in boundary-layer theory, introduced first by von Mises (1927)), it becomes

$$\frac{1}{h_\eta h_\zeta} \frac{\partial C}{\partial \eta} = \kappa \frac{\partial}{\partial \psi} \left(\frac{\partial \psi}{\partial \xi} \frac{\partial C}{\partial \psi} \right),$$

where

$$\frac{\partial \psi}{\partial \xi} = \xi h_\zeta F = (2\psi h_\zeta F)^{\frac{1}{2}}.$$

The factors h_η , h_ξ , F , none of which depends on ψ , may be gathered together in a new variable

$$\tau = \kappa \int h_\eta h_\xi (2h_\xi F)^{\frac{1}{2}} d\eta, \tag{3.4}$$

whence the equation becomes

$$\frac{\partial C}{\partial \tau} = \frac{\partial}{\partial \psi} \left(\psi^{\frac{1}{2}} \frac{\partial C}{\partial \psi} \right). \tag{3.5}$$

The boundary conditions to be satisfied by the solution of this equation are

$$C = C_1 \quad \text{at} \quad \psi = 0 \quad (\text{where} \quad \xi = 0), \quad C \rightarrow C_0 \quad \text{as} \quad \psi \rightarrow \infty.$$

We assume that each surface streamline begins (i.e. $\eta = 0$ there) at a point where the surface tangential stress vanishes, and the constant of integration in (3.4) will be chosen so that $\tau = 0$ at $\eta = 0$. There is no boundary condition to be imposed at $\tau = 0$, except the requirement that there be no singularity there. Thus the boundary conditions do not involve any dimensional parameter (C_1 and C_0 are of course not relevant, since they could be removed from the boundary conditions by taking the dependent variable as $(C - C_0)/(C_1 - C_0)$), and the required solution of (3.5) must be a function only of the dimensionless combination

$$\chi = \psi^{\frac{1}{2}} / \tau^{\frac{1}{2}}. \tag{3.6}$$

In terms of this similarity variable the equation reduces to

$$\frac{d^2 C}{d\chi^2} + \frac{4}{3} \chi^2 \frac{dC}{d\chi} = 0, \tag{3.7}$$

of which the required solution is

$$C(\chi) = C_1 - \frac{C_1 - C_0}{1.170} \int_0^\chi \exp(-\frac{4}{9} \chi'^3) d\chi'. \tag{3.8}$$

(This integral is one of the incomplete gamma functions, and 1.170 is its asymptotic value† as $\chi \rightarrow \infty$.) Remarkably, the distribution of concentration with respect to the variable χ is the same at all points on a surface streamline *and* on all surface streamlines; only the relation between χ and position in the concentration boundary layer varies.

The local density of solute flux across the particle surface is thus

$$\begin{aligned} -\kappa \left(\frac{\partial C}{\partial \xi} \right)_{\xi=0} &= -\kappa (2h_\xi F)^{\frac{1}{2}} \left(\psi^{\frac{1}{2}} \frac{\partial C}{\partial \psi} \right)_{\psi=0} = -\frac{\kappa (\frac{1}{2} h_\xi F)^{\frac{1}{2}}}{\tau^{\frac{1}{2}}} \left(\frac{dC}{d\chi} \right)_{\chi=0} \\ &= \frac{\kappa (C_1 - C_0)}{1.170} \frac{(\frac{1}{2} h_\xi F)^{\frac{1}{2}}}{\tau^{\frac{1}{2}}}. \end{aligned} \tag{3.9}$$

The total rate of transfer across the portion of the particle surface that is traversed by surface streamlines emanating from a position of zero tangential stress where

† It is given incorrectly as 1.15 on page 84 of the book by Levich (1962), and this small slip affects some of the numbers on subsequent pages.

$\eta = 0$ (and $\tau = 0$) and ending at another position where $F = 0$ ($\eta = \eta_1$ and $\tau = \tau_1$, say) follows from integration with respect to η and ζ :

$$\begin{aligned} Q &= \frac{C_1 - C_0}{1.170} \int \left\{ \int_0^{\eta_1} \frac{\kappa (\frac{1}{2} h_\zeta F)^{\frac{1}{2}}}{\tau^{\frac{1}{2}}} h_\eta h_\zeta d\eta \right\} d\zeta \\ &= \frac{C_1 - C_0}{2.340} \int \left\{ \int_0^{\tau_1} \frac{d\tau}{\tau^{\frac{1}{2}}} \right\} d\zeta \\ &= 0.808 \kappa^{\frac{3}{2}} (C_1 - C_0) \int \left\{ \int_0^{\eta_1} F^{\frac{1}{2}} h_\zeta^{\frac{3}{2}} h_\eta d\eta \right\}^{\frac{2}{3}} d\zeta. \end{aligned} \quad (3.10)$$

We see that, regardless of the choice of length and velocity scales in the definitions of the Nusselt and Péclet numbers, N is proportional to $P^{\frac{1}{3}}$, as of course was evident from the way in which ξ and κ enter into the relations (3.1) and (3.2).

This theory for mass transfer at high Péclet number has been available for many years in various particular forms appropriate to cases in which C does not depend on the lateral co-ordinate ζ . It appears to have been given first by V. G. Levich in the early nineteen-forties (see Levich 1962, chap. 2) for the case of transfer from a thin flat plate immersed in a steady uniform stream parallel to the plate at large Reynolds number with $\nu/\kappa \gg 1$. Independently Lighthill (1950) found a series solution of the von Mises equation (3.5) for the case of transfer from a two-dimensional body in a stream with an arbitrary steady distribution of tangential stress at the body surface (provided F does not change sign), and, although he did not obtain the concentration distribution in the closed form (3.8) (this further step being taken by Acrivos (1960)), he was able to find the total rate of transfer from the plate. Morgan & Warner (1956) also noted that the total rate of transfer is proportional to $P^{\frac{1}{3}}$ for various two-dimensional or axisymmetric steady flow fields at large Reynolds number in which the concentration boundary layer is much thinner than the velocity boundary layer. The transfer rate has also been calculated for various axisymmetric flow fields, as will be mentioned in a moment.

We note from (3.10) that if the flow velocity is reversed everywhere the roles of the two ends of each surface streamline ($\eta = 0$ and $\eta = \eta_1$) are interchanged but the total transfer rate Q is unaffected. This again extends the conditions of the result found by Brenner (1967), and it shows also that when $P \gg 1$ the unit for which the total transfer is invariant to flow reversal is not the whole body but the portion of the body surface traversed by streamlines joining two points of zero tangential stress. The result is surprising, because as Brenner noted, the distribution of mass flux density at the particle surface may be quite different when the flow is reversed.

Our concern in this paper is with specific results for a spherical particle immersed in the steady linear ambient velocity distribution $U_i = G_{ij} x_j$. The sphere is assumed to be couple-free, so it rotates with the same angular velocity $\frac{1}{2}\boldsymbol{\omega}$ as the fluid. Since the theory applies only to a particle with a stationary surface, we must suppose that $\boldsymbol{\omega} = 0$ for the moment, leaving only the pure straining motion $U_i = E_{ij} x_j$. The velocity distribution in low-Reynolds-number flow due to a rigid sphere of radius a immersed in a pure straining motion characterized by the rate-of-strain tensor \mathbf{E} is known (Batchelor 1967, p. 249) to be

$$\mathbf{u} = \mathbf{x} \cdot \mathbf{E} \left(1 - \frac{a^5}{r^5} \right) - \frac{5}{2} \mathbf{x} \frac{\mathbf{x} \cdot \mathbf{E} \cdot \mathbf{x} a^3}{r^2} \frac{a^3}{r^3} \left(1 - \frac{a^2}{r^2} \right). \quad (3.11)$$

Near the surface of the sphere the fluid velocity is

$$\mathbf{u} \approx 5\xi(\mathbf{1} \cdot \mathbf{E} - \mathbf{1}\mathbf{1} \cdot \mathbf{E} \cdot \mathbf{1}), \tag{3.12}$$

where $\mathbf{1} = \mathbf{x}/r$ and $\xi (= r - a) \ll a$. In terms of spherical polar co-ordinates (r, θ, ϕ) , with $\theta = 0$ in the direction of the principal axis of \mathbf{E} associated with the principal rate of strain E_3 and $\theta = \frac{1}{2}\pi$, $\phi = 0$ in the direction of the principal axis associated with E_1 , the θ and ϕ components of the velocity near the sphere surface are

$$\left. \begin{aligned} u_\theta &= \frac{5}{2}\xi \sin 2\theta \{-3E_3 + (E_1 - E_2) \cos 2\phi\} \\ u_\phi &= \frac{5}{2}\xi \sin \theta \sin 2\phi (E_2 - E_1), \end{aligned} \right\} \tag{3.13}$$

where the three principal rates of strain satisfy the relation

$$E_1 + E_2 + E_3 = 0.$$

The tangential stress at the surface vanishes at the six points (i) $\theta = 0$, (ii) $\theta = \pi$, (iii) $\theta = \frac{1}{2}\pi$, $\phi = 0$, (iv) $\theta = \frac{1}{2}\pi$, $\phi = \pi$, (v) $\theta = \frac{1}{2}\pi$, $\phi = \frac{1}{2}\pi$, (vi) $\theta = \frac{1}{2}\pi$, $\phi = \frac{3}{2}\pi$, and in general surface streamlines emanate from the two or four points associated with negative principal rates of strain and end at the four or two points associated with positive principal rates of strain.

Steady axisymmetric flow about a spherical particle

We choose the origin of the polar angle θ to be the direction of the axis of symmetry, so that the surface streamlines are longitudinal lines $\phi = \text{const}$. On making the substitutions

$$\eta = \theta, \quad \zeta = \phi, \quad h_\eta = a, \quad h_\zeta = a \sin \theta$$

in the formula (3.10) we find for the rate of transfer from the portion of the sphere surface swept by surface streamlines joining points of zero tangential stress at $\theta = 0$ and $\theta = \theta_1$

$$Q = 1.616\pi\kappa^{\frac{2}{3}}a^{\frac{4}{3}}(C_1 - C_0) \left\{ \int_0^{\theta_1} \sin^{\frac{2}{3}} \theta F^{\frac{1}{3}} d\theta \right\}^{\frac{3}{2}}. \tag{3.14}$$

The first specific result of this kind to be published concerns a rigid sphere immersed in a steady uniform stream of speed U_0 at small Reynolds number (Levich 1962), for which

$$\theta_1 = \pi, \quad F(\theta) = \frac{3}{2} \frac{U_0}{a} \sin \theta \quad (U_0 > 0).$$

The non-dimensional rate of transfer from the sphere here is

$$N = \frac{Q}{4\pi\kappa a(C_1 - C_0)} = 0.625 \left(\frac{aU_0}{\kappa} \right)^{\frac{1}{2}}.$$

More relevant to the subject of this paper is the result obtained by Gupalo & Ryazantsev (1972) for a rigid sphere immersed in a steady ambient axisymmetric

pure straining motion.† There are here two sets of surface streamlines, each one joining one of the poles $\theta = 0, \pi$ to the 'equator' $\theta = \frac{1}{2}\pi$, and

$$\theta_1 = \frac{1}{2}\pi, \quad F(\theta) = -\frac{15}{4}E_3 \sin 2\theta \quad (E_3 < 0), \quad E_1 = E_2$$

in the notation of (3.13). The rate of transfer from the whole sphere surface is thus

$$N = \frac{2Q}{4\pi\kappa a(C_1 - C_0)} = 0.808 \left(\frac{15}{2}\right)^{\frac{1}{2}} J^{\frac{2}{3}} \left(\frac{a^2 |E_3|}{\kappa}\right)^{\frac{1}{3}},$$

where

$$J = \int_0^{\frac{1}{2}\pi} \cos^{\frac{1}{2}} \theta \sin^2 \theta d\theta = \frac{\pi^{\frac{1}{2}} \Gamma(\frac{7}{4})}{3 \Gamma(\frac{9}{4})} = 0.4794;$$

that is

$$N = 0.968 \left(\frac{a^2 |E_3|}{\kappa}\right)^{\frac{1}{3}}. \quad (3.15)$$

If $E_3 > 0$, the surface streamlines are reversed, and emanate from the equator and flow towards one of the two poles. We can here see explicitly from (3.9) that the mass flux density does not have the same distribution over the particle surface as in the case $E_3 < 0$ (being of order unity near $\theta = 0$ for $E_3 < 0$ and of order θ near $\theta = 0$ for $E_3 > 0$), but the total transfer is unchanged.

It is worth noting in passing that we may also calculate from (3.14) the transfer from a rigid sphere in translational motion (parallel to the direction $\theta = 0$) through fluid whose ambient motion is a pure straining with symmetry about $\theta = 0$. In this case we have

$$\begin{aligned} F(\theta) &= \frac{3}{2} \frac{U_0}{a} \sin \theta - \frac{15}{4} E_3 \sin 2\theta \quad (U_0 > 0) \\ &= \frac{3}{2} \frac{U_0}{a} \sin \theta (1 - \beta \cos \theta), \end{aligned}$$

where $\beta = 5aE_3/U_0$. Points of zero tangential stress occur at $\theta = 0$ and $\theta = \pi$, and also at $\cos^{-1} \beta^{-1}$ if $|\beta| > 1$. Thus, if $|\beta| \leq 1$, each surface streamline goes from pole to pole and we may express the total transfer as

$$Q = 1.616\pi\kappa^{\frac{2}{3}} a^{\frac{1}{3}} (C_1 - C_0) \left(\frac{9}{4} \frac{U_0^2}{a^2} + \frac{225}{4} E_3^2\right)^{\frac{1}{3}} I^{\frac{2}{3}},$$

where

$$I(\beta) = \int_0^{\pi} \sin^2 \theta \frac{(1 - \beta \cos \theta)^{\frac{1}{2}}}{(1 + \beta^2)^{\frac{1}{2}}} d\theta.$$

† Gupalo & Ryazantsev remark that any linear ambient flow field may be regarded as a superposition of three axisymmetric pure straining motions and a rigid-body rotation, and that the latter can be removed by the use of rotating axes; and they appear to imply that the transfer from a particle in a general steady linear ambient flow field can consequently be calculated from a knowledge of the transfer from a particle in a steady axisymmetric pure straining motion. This is not so, first because the concentration distributions associated with two or more ambient axisymmetric pure straining motions cannot be superimposed, and second because relative to rotating axes the pure straining component of the linear ambient flow field is no longer steady.

This integral could be evaluated numerically for several values of β but it is sufficient here to notice that $I^{\frac{2}{3}}$ varies smoothly between the values

$$\{I(\mp 1)\}^{\frac{2}{3}} = 1.183, \quad \{I(0)\}^{\frac{2}{3}} = 1.351$$

at the two ends of the range. On the other hand, if $|\beta| > 1$, there are two sets of surface streamlines and the total transfer is

$$Q_1 + Q_2 = 1.616\pi\kappa^{\frac{2}{3}}a^{\frac{2}{3}}(C_1 - C_0) \left(\frac{9}{4} \frac{U_0^2}{a^2} + \frac{225}{4} E_3^2 \right)^{\frac{1}{6}} (I_1^{\frac{2}{3}} + I_2^{\frac{2}{3}}),$$

where

$$I_1(\beta) = \int_0^{\cos^{-1}\beta^{-1}} \sin^2\theta \frac{|1 - \beta \cos\theta|^{\frac{1}{2}}}{(1 + \beta^2)^{\frac{1}{2}}} d\theta, \quad I_2(\beta) = \int_{\cos^{-1}\beta^{-1}}^{\pi} \dots$$

We already have the result

$$\{I_1(\mp \infty)\}^{\frac{2}{3}} + \{I_2(\mp \infty)\}^{\frac{2}{3}} = 2\{I_1(\mp \infty)\}^{\frac{2}{3}} = 2J^{\frac{2}{3}} = 1.225.$$

It appears therefore that the ratio of the total transfer to $(\frac{9}{4}U_0^2/a^2 + \frac{225}{4}E_3^2)^{\frac{1}{6}}$ does not vary by more than about 14% over the whole range of values of the parameter β measuring the relative strengths of the translational and ambient pure straining motions. (The variation could be made even less by choosing this dimensional factor as $\{(\frac{3}{2}U_0/a)^{2n} + (\frac{15}{2}E_3)^{2n}\}^{1/6n}$ with n having a value larger than 1.)

Gupalo & Ryazantsev (1972) also calculated the transfer from a spherical liquid drop immersed in a steady axisymmetric pure straining motion by a variant of the above method. In the governing equation (3.1) (in which $u_\eta = 0$) the velocity component u_ξ here does not vanish at $\xi = 0$ and is approximately constant across the concentration boundary layer, from which it follows that the concentration boundary-layer thickness is proportional to $P^{-\frac{1}{2}}$ and the rate of transfer from the sphere surface is proportional to $P^{\frac{1}{2}}$. But we shall not investigate the transfer from a liquid drop in other linear ambient flow fields.

More recently, Gupalo, Polyanin & Ryazantsev (1976) have obtained the expression for the mass transfer from a stationary particle in an arbitrary steady axisymmetric ambient flow field, and have calculated in particular the transfer from a spheroid immersed in a stream parallel to the axis of symmetry at low Reynolds number.

Two-dimensional pure straining motion

The type of pure straining motion that is next in order of simplicity appears to be two-dimensional motion ($E_1 = -E_3, E_2 = 0$). We see from (3.13) that in this case the velocity near the sphere surface is given by

$$\left. \begin{aligned} u_\theta &\approx \frac{5}{4}\xi E_1 \sin 2\theta (3 + \cos 2\phi), & = \xi F_\theta \quad \text{say,} \\ u_\phi &\approx -\frac{5}{2}\xi E_1 \sin \theta \sin 2\phi, & = \xi F_\phi \quad \text{say.} \end{aligned} \right\} \quad (3.16)$$

When $E_1 > 0$, one set of surface streamlines emanate from the pole $\theta = 0$ and divide into two sets one of which flows into the point $\theta = \frac{1}{2}\pi, \phi = 0$ and the other of which flows into the point $\theta = \frac{1}{2}\pi, \phi = \pi$; and there is a mirror image set emanating from the opposite pole $\theta = \pi$. The equation to these surface streamlines is

$$\frac{d\theta}{\frac{1}{2} \sin 2\theta (3 + \cos 2\phi)} = \frac{\sin \theta d\phi}{-\sin \theta \sin 2\phi},$$

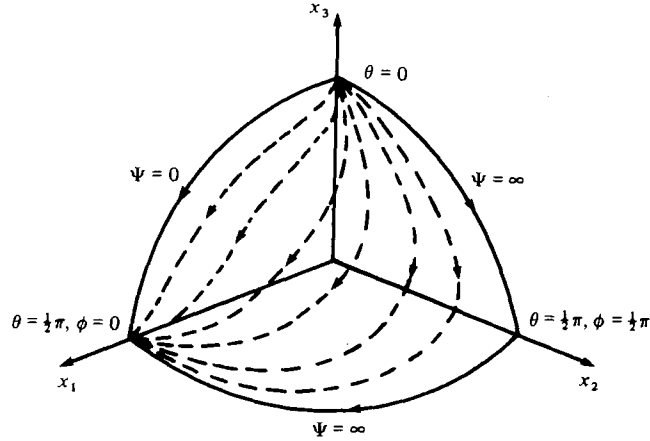


FIGURE 2. The surface streamlines on one octant of a sphere in a two-dimensional pure straining motion at low Reynolds number (schematic only). The x_1 and x_3 axes are principal axes of the rate-of-strain tensor. The arrows apply to a case in which $E_1 > 0$.

of which the solution is

$$\tan^2 \theta \tan^3 \phi \sin 2\phi = \Psi. \tag{3.17}$$

The surface streamlines covering the octant $0 \leq \theta \leq \frac{1}{2}\pi, 0 \leq \phi \leq \frac{1}{2}\pi$ are given by values of the constant Ψ in the range from 0 to ∞ (see figure 2), and it is sufficient to consider the transfer from this portion of the surface of the sphere.

The η -co-ordinate lines coincide with these surface streamlines, and we can identify ζ , the orthogonal co-ordinate, with Ψ . To make use of the formula (3.10) we need to find the metric scale factor h_ζ . Now the square of the length of a line element on the surface of the particle may be expressed in terms of the two alternative orthogonal co-ordinate systems, giving

$$a^2 \delta\theta^2 + a^2 \sin^2 \theta \delta\phi^2 = h_\eta^2 \delta\eta^2 + h_\zeta^2 \delta\zeta^2.$$

We also have

$$\delta\eta = \frac{\partial\eta}{\partial\theta} \delta\theta + \frac{\partial\eta}{\partial\phi} \delta\phi, \quad \delta\zeta = \frac{\partial\zeta}{\partial\theta} \delta\theta + \frac{\partial\zeta}{\partial\phi} \delta\phi,$$

from which it follows that

$$h_\zeta^2 = \frac{a^2 \left(\frac{\partial\eta}{\partial\phi}\right)^2 - a^2 \sin^2 \theta \left(\frac{\partial\eta}{\partial\theta}\right)^2}{\left(\frac{\partial\zeta}{\partial\theta}\right)^2 \left(\frac{\partial\eta}{\partial\phi}\right)^2 - \left(\frac{\partial\zeta}{\partial\phi}\right)^2 \left(\frac{\partial\eta}{\partial\theta}\right)^2}. \tag{3.18}$$

The fact that the η -co-ordinate lines coincide with the surface streamlines gives

$$\frac{\partial\eta}{\partial\theta} \bigg/ \frac{1}{\sin \theta} \frac{\partial\eta}{\partial\phi} = F_\theta / F_\phi,$$

whence

$$h_\zeta^2 = \frac{a^2 \sin^2 \theta (F_\phi^2 - F_\theta^2)}{\sin^2 \theta F_\phi^2 (\partial\zeta/\partial\theta)^2 - F_\theta^2 (\partial\zeta/\partial\phi)^2}. \tag{3.19}$$

On substituting from (3.17) (with $\Psi \equiv \zeta$) and using (3.16), we find

$$h_\zeta^2 = \left(\frac{5E_1 a}{8F}\right)^2 \sin^2 \theta \sin^2 2\theta \sin^2 2\phi,$$

where $F = (F_\theta^2 + F_\phi^2)^{\frac{1}{2}}$ is the magnitude of the tangential stress at the sphere surface, divided by the viscosity, introduced earlier.

The integral in (3.10) along the length of a surface streamline is thus

$$\begin{aligned} \int_0^{\eta_1} F^{\frac{1}{2}} h_\zeta^{\frac{3}{2}} h_\eta d\eta &= \int_0^{\frac{1}{2}\pi} \left(F^{\frac{1}{2}} h_\zeta^{\frac{3}{2}} \frac{aF}{F_\theta}\right)_{\zeta \text{ const.}} d\theta \\ &= \frac{\sqrt{5} |E_1|^{\frac{1}{2}} a^{\frac{5}{2}}}{4\zeta^{\frac{3}{2}}} \int_0^{\frac{1}{2}\pi} \sin^2 \theta \cos^{\frac{1}{2}} \theta \left(\frac{\sin^{\frac{3}{2}} 2\phi}{3 + \cos 2\phi}\right)_{\zeta \text{ const.}} d\theta \\ &= \frac{\sqrt{5} |E_1|^{\frac{1}{2}} a^{\frac{5}{2}}}{8 \cdot 2^{\frac{1}{2}}} \int_0^{\frac{1}{2}\pi} \frac{\sin \theta \cos^{\frac{3}{2}} \theta \{(1 + 8\zeta^{-1} \tan^2 \theta)^{\frac{1}{2}} - 1\}^{\frac{3}{2}}}{\zeta(1 + 8\zeta^{-1} \tan^2 \theta)^{\frac{1}{2}}} d\theta. \end{aligned} \quad (3.20)$$

The rate of transfer Q from one octant of the sphere surface is now obtained from (3.10), with the range of integration with respect to ζ being from 0 to ∞ , and the non-dimensional rate of transfer from the whole surface is

$$N = \frac{8Q}{4\pi\kappa a(C_1 - C_0)} = 0.808 \left(\frac{a^2 |E_1|}{\kappa}\right)^{\frac{1}{2}} \frac{5^{\frac{1}{2}}}{2^{\frac{3}{2}}\pi} \int_0^\infty \{L(\zeta)\}^{\frac{3}{2}} d\zeta, \quad (3.21)$$

where $L(\zeta)$ denotes the integral in (3.20). Numerical integration is needed to give L as a function of ζ . A second numerical integration then gives the value of the integral in (3.21) as 5.16, whence we have

$$N = 1.009 \left(\frac{a^2 |E_1|}{\kappa}\right)^{\frac{1}{2}}. \quad (3.22)$$

This case of transfer from a sphere immersed in a two-dimensional pure straining motion has been considered by Poe (1975), in a Ph.D. dissertation describing work done at the Stanford University under the supervision of Professor A. Acrivos. Poe's method was quite different in detail and involved the finding of a function $g(\theta, \phi)$ such that there exists a solution of the governing equation for C (with ξ, θ, ϕ as independent variables) which is a function of $\xi/\kappa^{\frac{1}{2}}g$ alone, there being no explicit reference to the surface streamlines and no use of the von Mises transformation. Our generalized Levich method is more direct inasmuch as it shows that such a similarity solution must exist (and I do not think this could have been anticipated), and as a consequence of relating the similarity solution to the development of the concentration boundary layer along a streamline we are able to make the integration with respect to τ analytically in the step preceding (3.10), thereby giving the total transfer as a double integral in place of Poe's triple integral. But Poe's calculation, which led to exactly the same numerical result (3.22) despite the quite different analytical forms, has the undeniable merit of having been made first.

Purcell (1978) has recently described measurements of the heat transfer from a solid sphere maintained at a constant temperature and immersed in fluid subjected to a steady two-dimensional pure straining motion. The Reynolds number of the motion was small and the Péclet number $a^2 |E_1|/\kappa$ was varied over the range 0.33 to

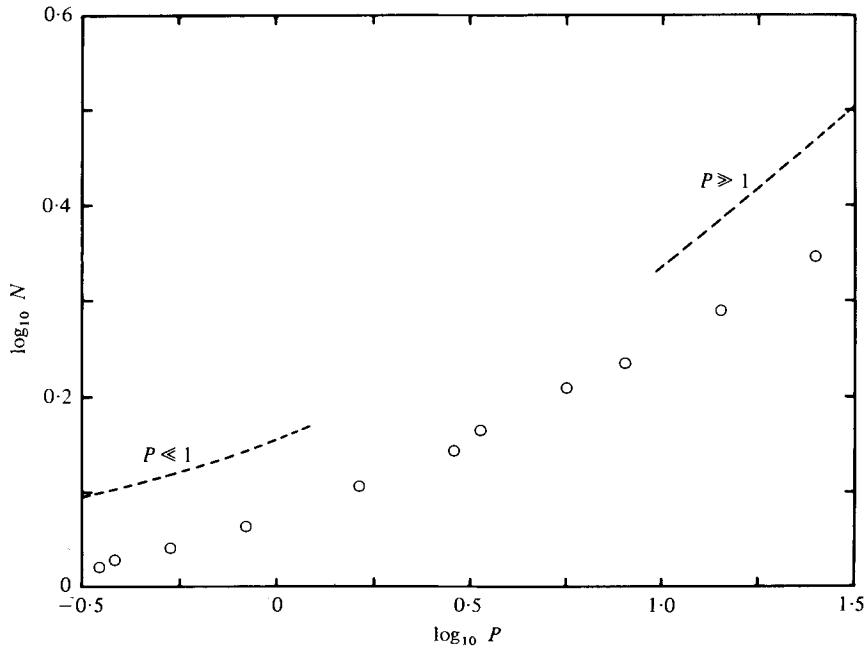


FIGURE 3. The rate of transfer from a sphere immersed in a steady two-dimensional pure straining motion with principal rates of strain E_1 , $-E_1$. N is the Nusselt number ($= 1$ for $P = 0$). P is the Péclet number $a^2|E_1|/\kappa$. The points marked \circ are measurements made by Purcell (1978) in a bounded flow field. The curve marked $P \gg 1$ is the asymptotic relation (3.22). The curve marked $P \ll 1$ is the asymptotic relation (2.27) which applies to an unbounded flow field.

24. The thickness of the thermal boundary layer at large Péclet number is roughly $a(a^2|E_1|/\kappa)^{-\frac{1}{2}}$, and so the Péclet number would need to be considerably larger than 24 before the thermal boundary-layer thickness is small compared with the sphere radius. Nevertheless a comparison between Purcell's measurements and the theoretical result (3.22) should have some value as an indication of the error in the asymptotic results at these Péclet numbers. It appears from figure 3 that the observations are rather far, surprisingly far, from the theoretical large- P curve at values of P near 20. It was partly the discrepancy between these measurements and the large- P asymptotic relation calculated by Poe that led me to make the independent calculation described above as a check. Either the asymptotic relation provides a very poor guide to the magnitude and the trend of the transfer rate at values of P below 30, or the measurements made by Purcell were affected in some way by the presence of an outer boundary to the flow field. Whatever the explanation, there is a need for more experiments, with this and other linear ambient velocity distributions.

Also shown on figure 3 is the theoretical result (2.27) found earlier for small Péclet number. The disagreement between this theoretical small- P curve and the observations is not unexpected, because the ambient pure straining motion in Purcell's experiment was disturbed by boundaries at distances from the sphere of about eight radii. The outer parts of the flow field are of course relevant to the transfer at small Péclet number, and the additional transfer due to convection in an enclosed flow field would be an analytic function of the Péclet number and so would vary as P^2 as $P \rightarrow 0$ (not

as the first power, because that would give a decrease in N when the flow was reversed), instead of as $P^{\frac{1}{2}}$ in an unbounded flow field.

Other ambient pure straining motions

It is possible that the rate of transfer from the sphere at large Péclet number could be calculated by the method described above for other pure straining motions, but the general similarity of the results obtained for axisymmetric and two-dimensional pure straining motions suggests there is little need to do so. Although there is not the same analytical reason for believing that the transfer depends primarily on the parameter $E(= (E_1^2 + E_2^2 + E_3^2)^{\frac{1}{2}})$ as in the case of small Péclet number, the two available results become very close numerically when expressed in terms of E , for we then have

(a) for steady axisymmetric pure straining given by $E_1 = E_2 = -\frac{1}{2}E_3, E = \sqrt{\frac{3}{2}} |E_3|,$

$$N = 0.968 \left(\frac{a^2 |E_3|}{\kappa} \right)^{\frac{1}{2}} \quad \text{or} \quad 0.905 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}}; \tag{3.23}$$

(b) for steady two-dimensional straining given by $E_1 = -E_3, E_2 = 0, E = \sqrt{2} |E_1|,$

$$N = 1.009 \left(\frac{a^2 |E_1|}{\kappa} \right)^{\frac{1}{2}} \quad \text{or} \quad 0.899 \left(\frac{a^2 E}{\kappa} \right)^{\frac{1}{2}}. \tag{3.24}$$

For given E , which can be regarded as specifying the intensity of the motion and the Péclet number, pure straining motions form a one-parameter family. The defining parameter can conveniently be chosen as $(E_1 - E_2)/E$, and, for exactly the same reasons as in the case of transfer at small Péclet number, it is sufficient to consider the transfer for values of $(E_1 - E_2)/E$ in the range from 0 (corresponding to axisymmetric straining about the x_3 -axis) to $1/\sqrt{2}$ (corresponding to two-dimensional motion in the (x_3, x_1) -plane). Moreover the transfer rate has a stationary value at these two values of $(E_1 - E_2)/E$. Since the non-dimensional transfer rate has been found above to have almost the same value, for given E , at these two ends of the range, there is very little scope for variation of N at intermediate values of $(E_1 - E_2)/E$.

The relation

$$N = 0.90(a^2 E/\kappa)^{\frac{1}{2}} \tag{3.25}$$

is thus likely to apply accurately at large Péclet number for *any* ambient flow field which is a steady pure straining motion.

Ambient velocity distributions with non-zero vorticity

We consider now steady linear ambient velocity distributions which are a combination of a pure straining motion characterized by \mathbf{E} and a rigid-body rotation with angular velocity $\frac{1}{2}\boldsymbol{\omega}$ (and $\frac{1}{2}|\boldsymbol{\omega}| = \Omega$). The force-free couple-free sphere here rotates with the fluid with angular velocity $\frac{1}{2}\boldsymbol{\omega}$, and we cannot calculate the transfer by the method used above since it requires a stationary particle surface relative to axes such that the flow is steady.

In the case of transfer at small Péclet number we saw that, when Ω is large relative to the components of the rate-of-strain tensor \mathbf{E} , the effect of the rotation is to suppress the contribution to the convective transfer from all components of \mathbf{E} except those associated with axisymmetric extension or compression in the direction of the axis of rotation, essentially because the strong rotation causes the streamlines of motion in the plane normal to the rotation axis to be nearly circular and so ineffective in

carrying material away from the particle. It is evident that a similar effect will occur at large Péclet number. Furthermore, whereas at small Péclet number a *strong* rotation is needed, we may expect that at large Péclet number this suppression of the convective transfer due to certain components of the pure straining motion will occur at *any* non-small value of Ω , because within the thin concentration boundary layer the contribution to the fluid velocity due to the ambient pure straining motion is proportional to distance from the sphere surface and so is small. This will lead us to the remarkable and paradoxical conclusion that calculation of the transfer rate at large Péclet number is simpler when the ambient vorticity is non-zero than when it is zero.

The fluid velocity is here a superposition of a rigid-body rotation with angular velocity of magnitude Ω and the contribution (3.11) from the ambient pure straining motion. We choose the x_3 axis to be in the direction of the ambient vorticity. The velocity near the sphere is then (see (3.12)) approximately

$$\mathbf{u} = \Omega r \mathbf{i} \times \mathbf{l} + 5\xi(\mathbf{l} \cdot \mathbf{E} - \mathbf{l} \mathbf{l} \cdot \mathbf{E} \cdot \mathbf{l}), \quad (3.26)$$

where \mathbf{i} is a unit vector in the direction of the x_3 axis, $\mathbf{l} = \mathbf{x}/r$ and $\xi (= r - a) \ll a$. The corresponding velocity components in the directions of the θ - and ϕ -co-ordinate lines, where r, θ, ϕ are spherical polar co-ordinates with $\theta = 0$ in the direction of the x_3 axis and $\theta = \frac{1}{2}\pi, \phi = 0$ in the direction of the x_1 axis, are

$$u_\theta = \xi \left\{ -\frac{15}{4} E_{33} \sin 2\theta + \frac{5}{4} \sin 2\theta (E_{11} \cos 2\phi - E_{22} \cos 2\phi + 2E_{12} \sin 2\phi) \right. \\ \left. + 5 \cos 2\theta (E_{31} \cos \phi + E_{23} \sin \phi) \right\}, \quad (3.27)$$

$$u_\phi = \Omega(a + \xi) \sin \theta + \xi \left\{ \frac{5}{2} (E_{22} - E_{11}) \sin \theta \sin 2\phi \right. \\ \left. + 5E_{12} \sin \theta \cos 2\phi + 5 \cos \theta (E_{23} \cos \phi - E_{31} \sin \phi) \right\}, \quad (3.28)$$

the component u_r near the sphere being found from

$$u_r = \frac{\xi}{2a \sin \theta} \left\{ -\frac{\partial(u_\theta \sin \theta)}{\partial \theta} - \frac{\partial u_\phi}{\partial \phi} \right\}. \quad (3.29)$$

To a first approximation, when $\xi \ll a$, a material element of fluid moves on a circular orbit about the x_3 axis with speed $a\Omega \sin \theta$. The most important consequence of the small departures from uniform motion in a circular orbit is that the average value of the θ component of velocity of a material element over one revolution is non-zero. This pole-ward drift velocity is approximately equal to the azimuthal average of (3.27), that is, to

$$\langle u_\theta \rangle = \frac{1}{2\pi} \int_0^{2\pi} u_\theta(\xi, \theta, \phi) d\phi = -\frac{15}{4} \xi E_{33} \sin 2\theta. \quad (3.30)$$

Thus a material element migrates to one of the two poles ($\theta = 0, \pi$) when $E_{33} > 0$ or to the equator ($\theta = \frac{1}{2}\pi$) when $E_{33} < 0$ and, as the same average procedure on (3.29) shows, moves away from the sphere surface there. The streamline path is a helix with wavy irregularities, the pitch of the helix being of order $\xi |E_{33}| / a\Omega$ and so smaller for streamlines closer to the surface. Since at large Péclet number the transfer from the sphere takes place by fluid elements being brought close enough to the sphere surface to receive material by direct diffusion and then being carried far away from the sphere, it is clear that the transfer rate is determined here primarily by the migration to or from the poles and that it is approximately the same as for an axisymmetric

pure straining motion with rate of extension E_{33} in the direction of the axis of symmetry.

Analytical confirmation that the wavy irregularities on the helical path of a material element cause only a small perturbation of the transfer rate may be obtained by considering the azimuthal-average concentration $\langle C \rangle$. We write

$$C = \langle C \rangle + C', \quad \mathbf{u} = \langle \mathbf{u} \rangle + \mathbf{u}',$$

where $\langle u_\phi \rangle = \Omega(a + \xi) \sin \theta$, $\langle u_\theta \rangle$ is given by (3.30), and C' and \mathbf{u}' are periodic functions of ϕ with zero mean, and substitute in the boundary-layer equation

$$\mathbf{u} \cdot \nabla C = \kappa \frac{\partial^2 C}{\partial \xi^2}. \tag{3.31}$$

An average of all terms in (3.31) then gives

$$\langle u_r \rangle \frac{\partial \langle C \rangle}{\partial \xi} + \frac{\langle u_\theta \rangle}{a} \frac{\partial \langle C \rangle}{\partial \theta} + \langle \mathbf{u}' \cdot \nabla C' \rangle = \kappa \frac{\partial^2 \langle C \rangle}{\partial \xi^2}. \tag{3.32}$$

Our object is to show that the concentration fluctuation C' is small by comparison with $C_1 - C_0$. The equation determining C' is found by subtracting (3.32) from (3.31) to be

$$\langle \mathbf{u} \rangle \cdot \nabla C' + \mathbf{u}' \cdot \nabla C' - \langle \mathbf{u}' \cdot \nabla C' \rangle - \kappa \frac{\partial^2 C'}{\partial \xi^2} = -\mathbf{u}' \cdot \nabla \langle C \rangle, \tag{3.33}$$

and the boundary conditions on C' are

$$C' = 0 \quad \text{at} \quad \xi = 0 \quad (\text{where } C = C_1)$$

and

$$C' \rightarrow 0 \quad \text{as} \quad \xi \rightarrow \infty \quad (\text{where } C \rightarrow C_0).$$

Since C' vanishes outside the concentration boundary layer, the appropriate position variable in (3.33) is ξ/δ , where δ is a measure of the boundary-layer thickness (and $\delta/a = O(P^{-1/2})$), and we therefore write (3.33) as

$$\left(1 + \frac{\xi}{a}\right) \frac{\Omega}{E} \frac{\partial C'}{\partial \phi} + \frac{1}{E} \left(\langle u_r \rangle \frac{\partial C'}{\partial \xi} + \frac{\langle u_\theta \rangle}{a} \frac{\partial C'}{\partial \theta} + \mathbf{u}' \cdot \nabla C' - \langle \mathbf{u}' \cdot \nabla C' \rangle \right) - \frac{a^2}{\delta^2} \frac{1}{P} \frac{\partial^2 C'}{\partial (\xi/\delta)^2} = -\frac{\mathbf{u}' \cdot \nabla \langle C \rangle}{E}, \tag{3.34}$$

where E is a measure of the ambient rate of strain and $P = a^2 E/\kappa$. Now $\partial C'/\partial \theta$ and $\partial C'/\partial \phi$ are of the same order of magnitude as C' , and $|\langle u_\theta \rangle|/Ea$ and $|\mathbf{u}'|/Ea$ are both of order δ/a at points within the boundary layer. Thus when $P \gg 1$ the dominant term on the left-hand side of (3.34) is the first term (provided Ω/E is not small), and the equation reduces approximately to

$$\frac{\Omega}{E(C_1 - C_0)} \frac{\partial C'}{\partial \phi} = -\frac{\mathbf{u}' \cdot \nabla \langle C \rangle}{E(C_1 - C_0)} = O(\delta/a).$$

C' can be found explicitly in terms of $\langle C \rangle$ by integration, but it is sufficient to observe that $C'/(C_1 - C_0)$ is of order $P^{-1/2}$.

The fluctuation of the concentration about its azimuthal mean is thus small when $P \gg 1$, and to leading order the equation (3.32) for the mean concentration reduces to that satisfied by C in the case of a steady axisymmetric ambient pure straining motion with rate of extension E_{33} (that is, $\boldsymbol{\omega} \cdot \mathbf{E} \cdot \boldsymbol{\omega}/\omega^2 = E_\omega$, when expressed in a way

which is independent of the co-ordinate system) in the direction of the axis of symmetry. Hence the asymptotic expression for the total transfer rate is found from (3.15) to be

$$N = 0.968 \left(\frac{a^2 |E_\omega|}{\kappa} \right)^{\frac{1}{2}}. \quad (3.35)$$

This result holds for a very wide class of steady linear ambient velocity distributions, *viz.* all except those for which either $|\omega| \ll E$ or $|E_\omega|$ is small in some sense. The above argument concerning equation (3.34) is not valid when $|\omega|/E$ is as small as $P^{-\frac{1}{2}}$ (because then the first term is not dominant), and as $|\omega|/E \rightarrow 0$ the expression for N changes from (3.35) to that appropriate to a steady ambient pure straining motion represented by the rate-of-strain tensor E_{ij} . If $E_\omega = 0$ the poleward migration that is responsible for the convective transfer vanishes and the expression for the transfer is quite different, as we shall see in a moment.

The result (3.35) is striking in its simplicity and generality, and it would be useful to undertake an experimental check. It is worth noting in this context that there is an alternative and possibly more convenient way of generating the relevant part of the required flow field, *viz.* by setting up a steady ambient pure straining motion and by applying a couple to the sphere and causing it to rotate with angular velocity $\frac{1}{2}\omega$. The flow field generated in this way at small Reynolds number is identical with that considered above at all points near the sphere surface and so the asymptotic expression for the transfer should be the same in the two cases. The kind of set-up described by Purcell (1978) could be used to generate a two-dimensional pure straining motion, for instance, and rotation of the sphere about an inclined axis could perhaps be produced by suitable suspension of the sphere.

The case $E_\omega = 0$

We see from (3.27) and (3.28) that when $E_{33} = 0$ the pole-ward displacement of a fluid element in one circuit about the rotation axis is zero, to the order of ξ , showing that the streamlines near the sphere surface are closed nearly circular paths. Convection consequently makes little contribution to the transfer in the neighbourhood of the sphere, and there is no basis for the Levich method of calculating the transfer rate. The resistance to transfer from the particle may be expected to be dominated by diffusion across the region of closed streamlines enclosing the particle, with the consequence that $N \rightarrow \text{const.}$ as $P \rightarrow \infty$. Thus here the suppression of convective transfer by ambient vorticity is more drastic, and reduces the order of magnitude of N . Problems of transfer across a region of closed streamlines have recently been tackled by Poe & Acrivos (1976), using an approximate method which involves representation of C as a polynomial in a measure of distance from the sphere surface and which had been developed earlier by Acrivos (1971) for the particular case of a sphere in a simple shearing motion. These authors considered a two-dimensional ambient flow field represented by the vorticity $(0, 0, 2\Omega)$ and principal rates of strain E_1 and E_2 ($= -E_1$) in the (x_1, x_2) plane. The transfer from the sphere is here a function of the Péclet number $a^2|E_1|/\kappa$ and of the ratio E_1/Ω (with $E_1/\Omega = \mp 1$ in the case of simple shearing motion). An ambient flow field for which $E_\omega = 0$ is not necessarily two-dimensional (two-dimensionality would require $E_{23} = 0$ and $E_{31} = 0$ as well as $E_{33} = 0$, when the vorticity is in the direction of the x_3 axis), but no detailed results for three-dimensional ambient flow fields with $E_\omega = 0$ are yet available.

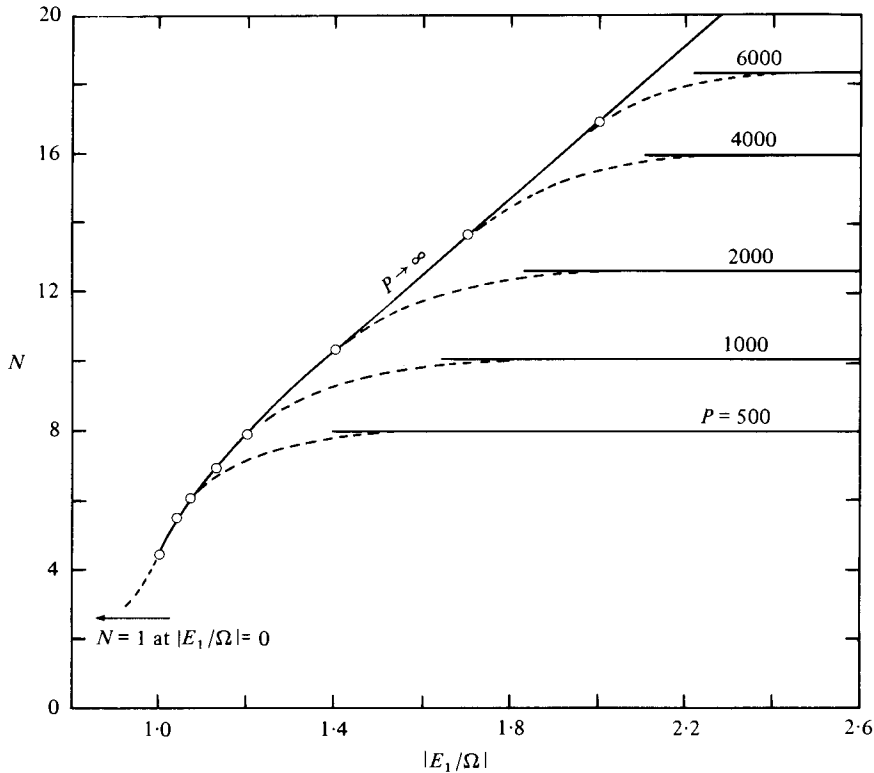


FIGURE 4. The rate of transfer from a sphere in a steady two-dimensional ambient flow field at small particle Reynolds number and large Péclet number. The angular velocity in the ambient flow is Ω and the principal rates of strain are $E_1, -E_1$. The Péclet number P is defined as $a^2|E_1|/\kappa$. The points marked \circ represent an approximate calculation of the transfer in the limit, as $P \rightarrow \infty$, by Poe & Acrivos (1976). The lines parallel to the abscissa show the asymptotic values of N as $|E_1/\Omega| \rightarrow \infty$, for given (large) P , as found from (3.22). The broken curves are suggested interpolations.

Poe & Acrivos (1976) found that the constant to which N tends as $P \rightarrow \infty$ increases as the value of $|E_1/\Omega|$ is increased from unity. The increase is asymptotically linear, as would be expected from the fact that the region of closed streamlines across which the transfer takes place by diffusion becomes a shell of (non-uniform) thickness proportional to $|E_1/\Omega|^{-1}$ (as may be seen from (3.28)). Some of their numerical results are shown in figure 4. It should be noted that these results apply only when the Péclet number is so large that the concentration boundary layer at the surface does not extend beyond the region of closed streamlines. The quantitative significance of this restriction can be seen from the horizontal lines that I have added to figure 4 in order to show the values (obtained from (3.22)) to which the Nusselt number must tend as $|E_1/\Omega| \rightarrow \infty$ for given values of $a^2|E_1|/\kappa$. The Nusselt number presumably varies monotonically with $|E_1/\Omega|$ for given Péclet number, and the broken curves in figure 4 are my suggested interpolation curves. Péclet numbers for particles suspended in a fluid in motion are usually less than 2000 when the particle Reynolds number is less than unity.

Poe & Acrivos were not able to obtain any results for $0 < |E_1/\Omega_2| < 1.0$, when the streamlines far from the sphere are similar ellipses. However, we do know that $N = 1$

when $E_1/\Omega = 0$ and all the streamlines are circular, and it seems certain that the constant value to which N tends as $P \rightarrow \infty$ lies between 1 and 4.5 for values of $|E_1/\Omega|$ in this range.

4. Summary of the main results for $P \ll 1$ and $P \gg 1$ and interpolations between them

The theoretical results for the transfer rate described in the two previous sections are asymptotically valid for small and large values of the Péclet number. In this section we restate the main results for a particle in various steady linear ambient flow fields. Where possible we shall show the results for $P \ll 1$ and for $P \gg 1$ on the one diagram in order to see the degree of uncertainty about the rate of transfer in the intermediate range of Péclet number.

For the additional rate of transfer from a particle due to convection at small Péclet number, we have the following three results which together cover virtually all kinds of linear ambient flow field.

(a) For any pure straining motion,

$$\frac{N - N_0}{N_0} = 0.36N_0 \left(\frac{\alpha^2 E}{\kappa} \right)^{\frac{1}{2}}, \quad (4.1)$$

where $E = E_{ij}E_{ij}$; the numerical coefficient is likely to be correct to within 3%.

(b) For a general linear ambient velocity distribution in which the ratio of the vorticity magnitude to E is not large compared with unity,

$$\frac{N - N_0}{N_0} = 0.34N_0 \left(\frac{\alpha^2 E}{\kappa} \right)^{\frac{1}{2}}; \quad (4.2)$$

the numerical coefficient here is approximate but is unlikely to be in error by more than 10%.

(c) For a general linear ambient velocity distribution in which the vorticity magnitude is significantly larger than E (or, more precisely, significantly larger than $(E^2 - \frac{3}{2}E_\omega^2)^{\frac{1}{2}}$),

$$\frac{N - N_0}{N_0} = 0.40 \left(\frac{\alpha^2 |E_\omega|}{\kappa} \right)^{\frac{1}{2}}, \quad (4.3)$$

where $E_\omega = \boldsymbol{\omega} \cdot \mathbf{E} \cdot \boldsymbol{\omega} / \omega^2$.

The results for the rate of transfer from a rigid spherical particle at large Péclet number and with small Reynolds number of the flow around the particle are as follows.

(A) For any pure straining motion,

$$N = 0.90 \left(\frac{\alpha^2 E}{\kappa} \right)^{\frac{1}{2}}, \quad (4.4)$$

with an accuracy of about 1%.

(B) For a general linear ambient velocity distribution in which $\boldsymbol{\omega} \neq 0$ and $E_\omega \neq 0$,

$$N = 0.97 \left(\frac{\alpha^2 E_\omega}{\kappa} \right)^{\frac{1}{2}}. \quad (4.5)$$

(C) When $E_\omega = 0$, N approaches a constant as $P \rightarrow \infty$. Values of this constant have been calculated by Poe & Acrivos (1976) for the case of a two-dimensional linear ambient velocity distribution with $|E_1/\Omega| \geq 1$ and are shown in figure 4. For simple shearing motion ($|E_1/\Omega| = 1$), $N \rightarrow 4.5$ as $P \rightarrow \infty$.

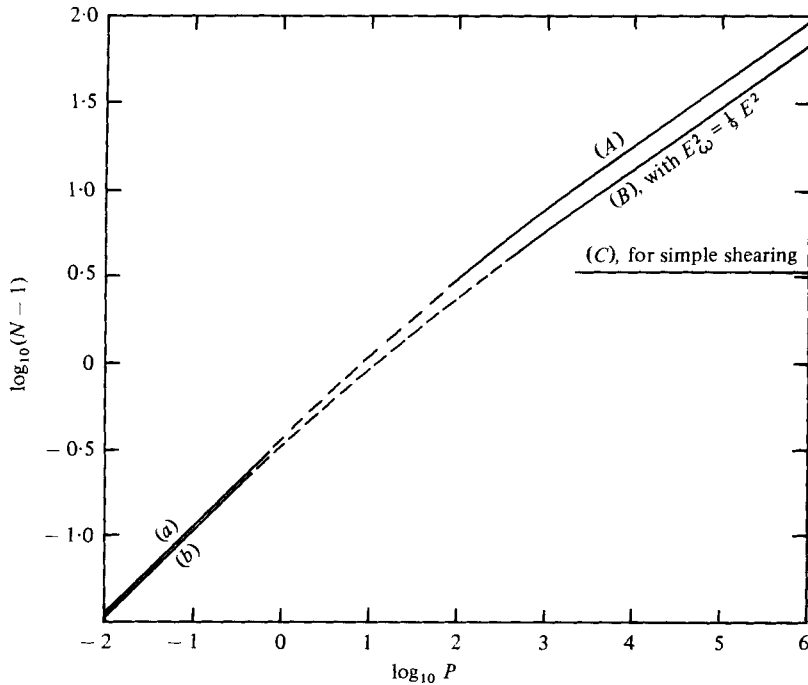


FIGURE 5. The rate of transfer from a sphere as a function of Péclet number $\alpha^2 E/\kappa$ (where $E = E_{ij} E_{ij}$) for different steady ambient flow fields. The unbroken curves are theoretical asymptotic relations and the broken curves are suggested interpolations. Curves (a) and (A) apply to any pure straining motion. Curve (b) applies approximately to any linear ambient velocity distribution in which Ω/E is not large; and (B) applies to any linear ambient velocity distribution in which Ω/E is not small and $E_\omega^2 = \frac{1}{9} E^2$. Curve (C) applies to simple shearing motion in the limit $P \rightarrow \infty$.

The results (a) and (A) for small and large values of P in the case of a rigid sphere (for which $N_0 = 1$) immersed in a pure straining motion are shown in figure 5. The two asymptotic relations run together so smoothly that there cannot be much error in the suggested interpolation curve. At $\alpha^2 E/\kappa = 30$, the small- P asymptotic form (4.1) gives $N = 2.97$ and the large- P asymptotic form (4.4) gives $N = 2.80$, a difference of only 6%.

The results (b) and (B) apply to very wide classes of linear ambient velocity distribution, and they *both* apply to any linear ambient flow field such that Ω/E is neither large nor small compared with unity and $E_\omega \neq 0$. These two results give N in terms of different parameters of the linear velocity distribution, so it is not possible to put the two asymptotic results on the same figure and interpolate between them unless the form of the linear velocity distribution is given. As a representative example we may suppose $E_\omega^2 = \frac{1}{9} E^2$ (on the grounds that there are 9 terms like E_ω^2 in the expression for E^2), and the result (B) is shown in figure 5 for this case. Again there is not much uncertainty about the suggested interpolation curve. If $\Omega/E \gg 1$ the results (c) and (B) are applicable, and again interpolation is possible (but is not shown in figure 5). We thus have here reasonably accurate theoretical results over the whole range of values of the Péclet number for almost any linear ambient velocity distribution.

As an example of the rather different result which applies at large Péclet number when $E_\omega = 0$, we show in figure 5 the constant value of N appropriate to the case of simple shearing motion. This constant value is the asymptote for a curve of the form (2.23) at small Péclet number (which differs from the curve (b) in figure 5 by being displaced vertically from it by only -0.05), but interpolation between the two asymptotic curves is not straightforward.

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