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VARIATIONAL ANALYSIS OF HIGH MASS TRANSFER RATES FROM SPHERICAL PARTICLES BOUNDARY-LAYER INJECTION SUCTION CONSIDERATIONS AT LOW PARTICLE REYNOLDS NUMBERS AND HIGH PECLET NUMBERS

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Abstract—A general variational analysis for hydrodynamic, thermal, and diffusional boundary-layer flows previously developed by the authors is adapted here to characterise relatively high mass transfer rates from moving spherical drops, bubbles, or solid particles. This work takes into account the effect of radial velocity at the particle surface (due to the very process of interfacial mass transfer) on the analysis of convective mass transfer rates—an important effect which has been neglected so far in studies of droplet dynamics. The error introduced by neglecting this effect is shown to amount to tens of per cents even when the interfacial radial velocity due to mass transfer is relatively small. In general it is found that the errors introduced in predicting convective transfer rates or boundary-layer thickness with neglected radial velocity at the interface are much more pronounced when circulation inside fluid particles is small. Comparison of our method with existing "exact" or integral methods (for calculating the mass transfer coefficients with a *neglected* radial velocity) clearly demonstrates its general property to extract the "best" trial solution from any assumed family of possible approximate solutions.

NOMENCLATURE

A, functions defined by equation (15);
a_j, coefficients of trial function;
c_i, mass fraction of transferred species;
D, coefficient of diffusion;
F, functional achieving extremal value at steady state;
I, injection coefficient, equation (27);
k, mass transfer coefficient, equation (23);
m, instantaneous mass of the sphere;
$\dot{m}$, mass transferred between the phases per unit time;
Pe, Péclét number;
q, defined by equation (A.7);
Q, defined by equation (15);
R, sphere radius;
r, radial coordinate;
Sh, Sherwood number (mass transfer Nusselt number);
T, nondimensional injection parameter, equation (12);
t, time,
U, velocity (upstream from the sphere);
u_0, tangential velocity at the interface, on the equator,
v, velocity component;
W, defined by equation (A.7);
y, radial variable defined by equation (2);
Y, defined by equation (A.7);
Z, defined by equation (15);
β, function of T;
γ, surface retardation coefficient due to presence of surfactant impurities;

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δ_c, diffusional boundary layer thickness;
θ, tangential coordinate;
μ, viscosity coefficient;
ρ, density;
τ, characteristic time;
ω, mass.

Subscripts
c, refers to diffusional process, or to the continuous phase;
d, dispersed phase;
e, external to the boundary layer;
i, species transferred between the phases;
inj, injection field;
l large internal circulation;
n, negligible internal circulation;
r, refers to radial field;
w, at the interface;
θ, refers to tangential field.

Superscripts
s, steady state;
$\hat{}$, average.

I. INTRODUCTION

THE TRANSFER of mass to or from moving small droplets, bubbles, or solid particles, is a complex phenomenon which occurs in many important processes. Examples are the evaporation of small fuel droplets atomized into pressurized hot gas, condensation or evaporation of water droplets in clouds or fogs, evaporation of liquid insecticides sprayed from the ground or flying aircraft, spray drying, extraction processes, gas absorption, fermentation, emulsion or suspension polymerization, and solid or liquid fluidized beds. In recent years an increasing amount of basic research has been directed to studies of fluid dynamics and mass and heat transfer mechanisms of such processes (see [1] for detailed reviews of the subject). Existing works do not take into account the influence of the radial velocity component, which originates by the very process of interfacial mass transfer. The only attempt to treat this effect [14] includes only the case of particles with no internal circulation. The assumption involved in existing works is that the radial velocity at the interface can be neglected for cases involving very small interfacial mass fluxes. Therefore, the existing solutions available at the literature cannot be applied to many actual cases which involve intermediate or high mass transfer fluxes. Even for small mass fluxes the error introduced by neglecting the radial component at the interface has not been evaluated yet to justify its neglect. The immediate aim of this paper is therefore to evaluate the hydrodynamics, local and total average mass transfer rates to ("suction") or from ("injection") moving small particles. To yield quantitatively useful results by relatively simple analysis we employ here a new variational analysis for hydrodynamic, thermal, and diffusional boundary layers whose general formulation has been recently developed by the authors [4]. Other applications of this method have already demonstrated [7, 15] that many flow and mass transfer problems which cannot be exactly solved analytically may be easily handled by this method. Furthermore this method possesses a general property to extract the "best" solution from any assumed family of possible approximate solutions, in the sense that the dissipation calculated from the obtained solution is closest to the actual (unknown) value.

This method is adapted here for small fluid spherical particles with the possibility of internal circulation. To allow the use of the boundary-layer concept with a simple superposition method we restrict here the illustration of the method to low particle Reynolds numbers and high Péclét numbers. Thus the results obtained would be useful for such processes as liquid–liquid extraction, leaching, or gas-absorption but can serve only as a first approximation for cases involving particles in a continuous gas phase. It should be emphasised, however, that the proposed method is not limited to low Reynolds numbers, spherically shaped boundary layers, or single particles. The effect of neighbouring particles in concentrated systems, for instance, may be taken into account by employing approximate methods [1, 10, 11] for the estimation of the tangential velocity at the interfacial equator.

II. FORMULATION OF THE PROBLEM

The time dependent conservation equation of species i in spherical coordinates for an axisymmetric flow pattern in a nonreactive fluid with negligible thermodynamic coupling and constant diffusivity can be written as

$$\frac{\partial c_i}{\partial t} + v_r \frac{\partial c_i}{\partial r} + \frac{v_\theta}{r}\frac{\partial c_i}{\partial \theta} - \frac{D_i}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial c_i}{\partial r}\right) = 0 \qquad (1)$$

neglecting diffusion in the θ direction. When the Péclét number is large the species concentration distribution around spherical particles is of the boundary layer type, even for Stokes-type flow fields [12]. Therefore, by defining

$$y = r - R\,.\,(0 < y < \delta_c) \qquad (2)$$

for thin diffusional boundary layers equation (1) reduces to

$$\frac{\partial c_i}{\partial t} + v_r \frac{\partial c_i}{\partial y} + \frac{v_\theta}{R}\frac{\partial c_i}{\partial \theta} - D_i \frac{\partial^2 c_i}{\partial y^2} = 0\,. \qquad (3)$$

Employing now our variational analysis for hydrodynamic, thermal and diffusional boundary layers [4] and separating the effects of viscous dissipation according to the Curie–Prigogine principle [4] we obtain the appropriate reduced functional for a control volume in the continuous phase and time-invariant boundary conditions (see Fig. 1):

$$F = \int_0^{\theta}\int_0^{\delta_c} \left(\frac{v_\theta^s}{R}\frac{\partial c_i^s}{\partial \theta} c_i + v_r^s \frac{\partial c_i^s}{\partial y} + D_i \frac{\partial c_i^s}{\partial y}\frac{\partial c_i}{\partial y}\right) \mathrm{d}y R \mathrm{d}\theta. \qquad (4)$$

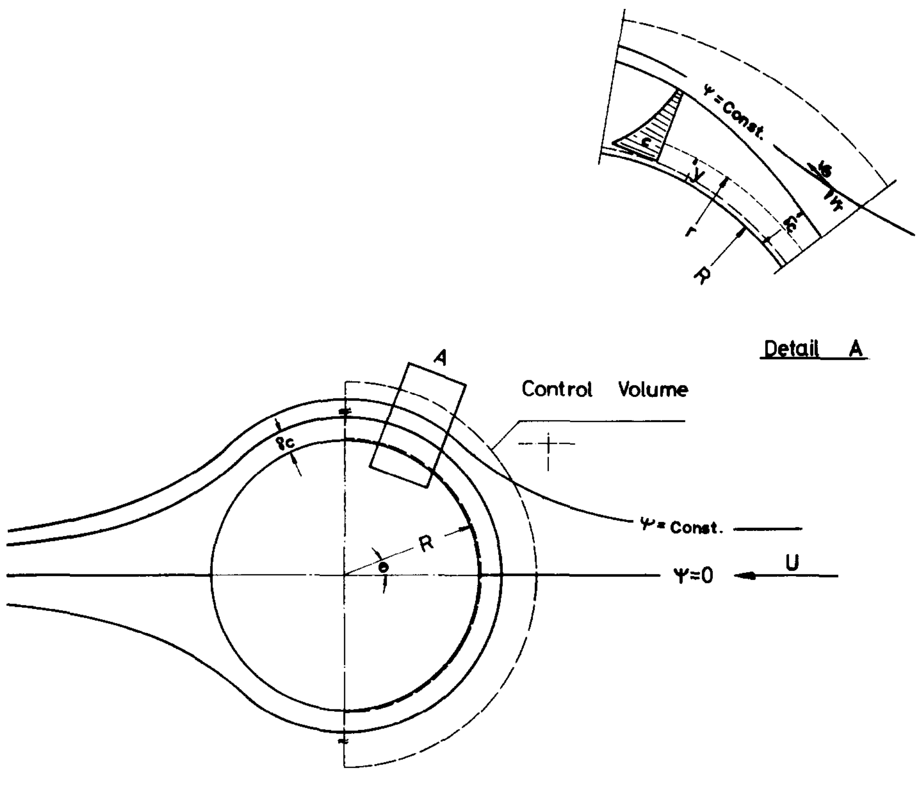

FIG. 1. Coordinate system for viscous flow round spherical particles with appreciable injection or suction convective diffusional rates at the surface.

This reduced functional is mathematically equivalent to equation (3) with appropriate boundary conditions, as can be seen by applying the Euler–Lagrange operator [4] on equation (4). The upper limit of integration in the y direction is the edge of the boundary layer, since the concentration of species i is constant in the external flow. The superscript 's' appearing on some of the quantities represents steady-state "values" of the functions, which therefore are not subject to variation. Thus, the velocity components v_r^s and v_θ^s which appear in this form can be taken from other steady-state solutions of the flow field around spherical particles. For illustration of our method we consider here a known steady-state velocity field [11] for $Re \ll 1$.

$$v_r^s = \left[\left(u_0 - \frac{U}{2}\right)\left(1 + \frac{y}{R}\right)^{-3} + \left(\tfrac{3}{2}\,U - u_0\right)\left(1 + \frac{y}{R}\right)^{-1} - U\right]\cos\theta$$

$$v_\theta^s = \left[\left(\frac{u_0}{2} - \frac{U}{4}\right)\left(1 + \frac{y}{R}\right)^{-3} + \left(\tfrac{3}{4}\,U - \frac{u_0}{2}\right)\left(1 + \frac{y}{R}\right)^{-1} + U\right]\sin\theta \qquad (5)$$

in which possible radial velocity distribution at the interface has been neglected. In equation (5) u_0 is the tangential velocity at the equator, and U the undisturbed relative velocity of the continuous phase. These expressions for the velocity components are chosen here since the effects of internal circulation, the presence of surfactants and the influence of other similar spherical particles in concentrated suspensions or emulsions appear only in u_0 and U and can still be taken into account by following the previous mathematical procedures of Gal-Or and Waslo [10] and Yaron and Gal-Or [11]. The velocity field (5) for low Reynolds numbers is linear, and therefore subject to the principle of superposition. Assuming now, that the concentration of the transferred species* is constant on the interface and outside the boundary layer, one can superpose a radial velocity field (the "injection field") on equation (5). (See, however the limitations imposed by the discussion following equation (16) and the Appendix.)

From the equation of continuity

$$(v_r)_{inj} = v_w(\theta)\left(1 + \frac{y}{R}\right)^{-2} \qquad (6)$$

where $v_w(\theta)$ is the radial velocity distribution at the interface due to mass transfer which for spherical particles is a function of the angle θ. Equation (6) actually describes a 3-dimensional source or sink at the center of the sphere, but since the control volume does not include the interior of the sphere no mathematical difficulties are thereby encountered. The velocity field in the diffusional boundary layer can thus be written as

$$v_r^s = \left[\left(u_0 - \frac{U}{2}\right)\left(1 + \frac{y}{R}\right)^{-3} + \left(\tfrac{3}{2}\,U - u_0\right)\times\left(1 + \frac{y}{R}\right)^{-1} - U\right]\cos\theta + v_w\left(1 + \frac{y}{R}\right)^{-2} \qquad .\mathrm{a}$$

$$v_\theta^s = \left[\left(\frac{u_0}{2} - \frac{U}{4}\right)\left(1 + \frac{y}{R}\right)^{-3} + \left(\frac{3}{4}U - \frac{u_0}{2}\right)\times\left(1 + \frac{y}{R}\right)^{-1} + U\right]\sin\theta \qquad .\mathrm{b} \qquad (7)$$

where y is defined only for $0 \leqslant y \leqslant \delta_c$.

Since $y/R \ll 1$, one can neglect higher orders of this ratio in equation (7) and after some manipulations, obtain

$$v_r^s \cong -\left[2u_0\frac{y}{R} + \left(\frac{y}{R}\right)^2\left(\tfrac{3}{2}\,U - 5u_0\right)\right]\cos\theta + v_w\left(1 - 2\frac{y}{R}\right)$$

$$v_\theta^s \cong \left[u_0 + \frac{y}{R}\left(\tfrac{3}{2}\,U - 2u_0\right)\right]\sin\theta \qquad (8)$$

To obtain the desired concentration profiles and the mass transfer coefficients, one must now choose a trial function to approximate the concentration distribution in the diffusional boundary layer. A fourth-order polynomial

* Or alternatively, the temperature.

with one parameter to be varied, is chosen here, as this type gives good results in other similar cases [7]. Thus

$$c_i = \sum_{j=0}^{4} a_j \left(\frac{y}{\delta_c}\right)^j \quad 0 \leqslant y \leqslant \delta_c(\theta)$$

$$c_i = c_{ie} \qquad y \geqslant \delta_c(\theta). \tag{9}$$

The steady-state concentration is then given by

$$c_i^s = \sum_{j=0}^{4} a_j \left(\frac{y}{\delta_c^s}\right)^j \quad 0 \leqslant y \leqslant \delta_c^s(\theta)$$

$$c_i^s = c_{ie} \qquad y \geqslant \delta_c^s(\theta). \tag{10}$$

The diffusional boundary-layer thickness $\delta_c(\theta)$ is therefore the only parameter subject to variation. The coefficients a_j are determined from the boundary conditions:

$$y = 0 \qquad c_i = c_{iw} \tag{11a}$$

$$y = \delta_c(\theta) \qquad c_i = c_{ie} \tag{11b}$$

$$y = 0 \qquad v_w \frac{\partial c_i}{\partial y} = D_i \frac{\partial^2 c_i}{\partial y^2} \tag{11c}$$

$$y = 0 \qquad v_w \frac{\partial^2 c_i}{\partial y^2} = D_i \frac{\partial^3 c_i}{\partial y^3} \tag{11d}$$

$$y = \delta_c(\theta) \qquad \frac{\partial c_i}{\partial y} = 0 \tag{11e}$$

where equations (11a) and (11b) state that the concentrations of species i on the interface and outside of the boundary layer are constant. Equation (11c) is simply equation (3) evaluated on the interface, while equation (11d) is obtained by taking the normal derivative of equation (3) and evaluating it at the interface. Equation (11e) is a smoothness condition at the boundary layer edge. We now introduce a dimensionless parameter for the "injection or suction" intensity

$$T(\theta) = \frac{\delta_c(\theta) v_w(\theta)}{D_i} \tag{12}$$

which has the form of a Péclét number for the injection field. By combining equation (11) with (9) and (12) one obtains

$$c_i(\theta, y) = c_{iw} + \frac{c_{ie} - c_{iw}}{3 + T + T^2/6} \left[4\left(\frac{y}{\delta_c}\right) + 2T\left(\frac{y}{\delta_c}\right)^2 + \frac{2}{3}T^2\left(\frac{y}{\delta_c}\right)^3 - \left(1 + T + \frac{T^2}{2}\right)\left(\frac{y}{\delta_c}\right)^4\right] \tag{13}$$

and an equivalent expression for $c_i^s(\theta, y)$. Substituting these profiles, and the velocity profiles (8) into the functional (4), and performing the first variation, the steady-state case fulfils the condition

$$\frac{\partial F}{\partial \delta_c} = 0. \tag{14}$$

Integrating the expression for the first variation in the direction normal to the surface we have an integral which vanishes for all permissible variations and bounds of integration, so that the integrand itself must be zero. This condition gives a non-linear ordinary differential equation of the first order for δ_c:

$$\frac{A_1(T)}{2} \frac{u_0 \sin\theta}{R} \delta \frac{d\delta}{d\theta} - A_5(T) \frac{v_w}{D_i} \frac{u_0 \sin\theta}{R} \delta^2 \frac{d\delta}{d\theta} + A_2(T) \frac{Q \sin\theta}{R^2} \delta^2 \frac{d\delta}{d\theta} - A_4(T) \frac{Q \sin\theta}{R^2} \frac{v_w}{D_i} \delta^3 \frac{d\delta}{d\theta} + A_1(T) \frac{u_0 \cos\theta}{R} \delta^2 + A_2(T) \frac{Z \cos\theta}{R^2} \delta^3 - \frac{dv_w}{d\theta} \left[A_4(T) \frac{Q}{R} \delta^2 + A_5(T) u_0 \delta\right] \frac{\sin\theta}{R D_i} - A_0(T) v_w \delta + A_1(T) \frac{v_w}{R} \delta^2 - D_i A_3(T) = 0 \tag{15}$$

where

$$Q = \frac{3}{2} U \left(1 - \frac{4}{3}\frac{u_0}{U}\right)$$

$$Z = \frac{3}{2} U \left(1 - \frac{10}{3}\frac{u_0}{U}\right)$$

and

$$A_0 = 3{\cdot}600 + 2{\cdot}93333T + 1{\cdot}18095T^2 + 0{\cdot}24762T^3 + 0{\cdot}023810T^4$$

$$A_1 = 3{\cdot}55555 + 3{\cdot}30160T + 1{\cdot}43492T^2 + 0{\cdot}31746T^3 + 0{\cdot}031746T^4$$

$$A_2 = 1{\cdot}02857 + 1{\cdot}02857T + 0{\cdot}46984T^2 + 0{\cdot}10794T^3 + 0{\cdot}011111T^4$$

$$A_3 = 5{\cdot}14286 + 3{\cdot}08571T + 1{\cdot}08571T^2 + 0{\cdot}20952T^3 + 0{\cdot}019048T^4$$

$$A_4 = -0{\cdot}25714 - 0{\cdot}25397T - 0{\cdot}093121T^2 - 0{\cdot}17857T^3 - 0{\cdot}0007937T^4$$

$$A_5 = -0{\cdot}53968 - 0{\cdot}49735T - 0{\cdot}16984T^2 - 0{\cdot}41640T^3 - 0{\cdot}0013228T^4.$$

Here the superscript s and the subscript c have been omitted, as we now deal only with the steady-state case.

One must note here that by choosing different trial functions only the quantities $A_j (j = 0, 1, \ldots, 5)$ would vary while the form of the equation remains the same. A_0, A_1 are of the same order of magnitude for all values of T, so that the ninth term on the left side of equation (15) can be dropped, using the thin boundary layer assumption. Finally one obtains

$$\frac{u_0 \sin\theta}{R}\left(\frac{A_1}{2} - TA_5\right)\delta\frac{d\delta}{d\theta} + \frac{Q\sin\theta}{R^2}(A_2 - TA_4)\,\delta^2\frac{d\delta}{d\theta} + \frac{u_0\cos\theta}{R}A_1\delta^2 + \frac{Z\cos\theta}{R^2}A_2\delta^2 - \frac{T}{v_w}\frac{dv_w}{d\theta}\frac{\sin\theta}{R} \times \left(u_0A_5 + QA_4\frac{\delta}{R}\right) - D_i(TA_0 + A_3) = 0 \quad (16)$$

which is the general equation describing $\delta_c(\theta)$ for diffusion to or from solid, liquid or gaseous spheres, with injection or suction from the continuous phase, when $Re \ll 1$, and $Pe \gg 1$. The terms including A_4 and A_5 arise from the θ differentiation of T, so that when $T \neq T(\theta)$ (the "self-similar" case) these terms vanish.

We now investigate the range of injection (or suction) velocities v_w for which the above analysis is valid. There appear to be two different criteria limiting the range of validity of this equation: (1) When the mass transferred between the phases during a characteristic time is of the order of the mass of the sphere, the process is no longer time-independent, and the variational analysis is not applicable. (2) When the interfacial radial velocity is large enough the flow-field is altered significantly and the assumed spherical symmetry no longer exists [13]. These criteria are further studied in the Appendix.

Equation (16) can be separated now into two equations; the first describing the case of a sphere with appreciable internal circulation $u_0 = 0(U)$, and the other dealing with negligible circulation ($u_0 \to 0$). Thus

$$\frac{u_0\sin\theta}{R}\left(\frac{A_1}{2} - TA_5\right)\delta\frac{d\delta}{d\theta} + \frac{u_0\cos\theta}{R}A_1\delta^2 - \frac{T}{v_w}\frac{dv_w}{d\theta}\frac{u_0\sin\theta}{R}A_5 - D_i(TA_0 + A_3) = 0 \quad (17)$$

for large internal circulation, and

$$\frac{Q\sin\theta}{R^2}(A_2 - TA_4)\delta^2\frac{d\delta}{d\theta} + \frac{Z\cos\theta}{R^2}A_2\delta^3 - \frac{T}{v_w}\frac{dv_w}{d\theta}\frac{Q\sin\theta}{R^2}A_4\delta - D_i(TA_0 + A_3) = 0 \quad (18)$$

for negligible internal circulation (i.e. solid particles or small drops and bubbles with strong interfacial retardation due to surfactant impurities).

III. RESULTS AND DISCUSSION

Existing treatments of the problem at hand [1, 2, 11, 12] are limited to the case of zero radial velocity at the interface, i.e. $T = 0$ in equations (16)–(18). The zero radial velocity case is solved here first, so as to enable a check of the accuracy of the variational method. Thus when $T = 0$, equation (17) reduces to

$$1{\cdot}77777\, u_0 \delta \frac{d\delta}{d\theta} \sin\theta + 3{\cdot}55555\, u_0 \delta^2 \cos\theta - 5{\cdot}14286\, D_i R = 0. \qquad (19)$$

Demanding that the boundary-layer thickness at the forward stagnation point be finite, the solution of (19) is

$$\frac{\delta_l}{R} = 1{\cdot}3897 \left(\frac{u_0}{U}\right)^{-\frac{1}{2}} Pe^{-\frac{1}{2}} \frac{(\cos^3\theta - 3\cos\theta + 2)^{\frac{1}{2}}}{\sin^2\theta}. \qquad (20)$$

From (18), when $T = 0$ one obtains

$$1{\cdot}02857 \frac{3}{2}\frac{U}{R}\delta^2 \frac{d\delta}{d\theta}\sin\theta + 1{\cdot}02857 \frac{3}{2}\frac{U}{R}\delta^3 \cos\theta - 5{\cdot}14286\, D_i R = 0 \qquad (21)$$

for which

$$\frac{\delta_n}{R} = 1{\cdot}5536\, Pe^{-\frac{1}{3}} \frac{(2\theta - \sin 2\theta)^{\frac{1}{3}}}{\sin\theta}. \qquad (22)$$

This boundary-layer thickness, as a function of the angle θ, appears in Figs. 2 and 3 (see also Table 1).

Total average mass transfer rate from the whole sphere may be estimated by assuming that the boundary layer analysis is valid for the whole surface of the particle. This description does not fit the rear "wake" area, but the contribution of this part of the sphere at low Reynolds numbers is negligible anyway [12]. Thus, defining the average mass transfer coefficient by

$$k = \frac{-D_i \int_0^{\pi} \left(\frac{\partial c_i}{\partial y}\right)_{y=0} 2\pi R^2 \sin^2\theta d\theta}{4\pi R^2 (c_{iw} - c_{ie})} \qquad (23)$$

the average mass transfer Nusselt number (Sherwood number) is given by

$$Sh = \frac{2Rk}{D_i}. \qquad (24)$$

Substituting expressions (20) and (22) in (24) one obtains for large, and negligible internal circulation the required results (see Table 2). Table 2 compares the results obtained by the present method with results of previous works.

Table 1. Nondimensional thickness of the diffusional boundary layer on a single sphere at Re ≪ 1 and negligible radial velocity

Pe number	$\frac{\delta_n}{R}$ ($\theta = 90°$)	$\frac{\delta_l}{R}$ ($\theta = 90°$)*
100	0·492	0·196
500	0·350	0·0880
1000	0·226	0·0621
5000	0·131	0·0278
10000	0·105	0·0196
20000	0·084	0·0139
100000	0·049	0·0062

$*\frac{u_0}{U} = 1$ in this Table.

Table 2. Comparison of existing methods for calculating average mass transfer coefficient for single spherical particles with negligible radial velocity

Method	$Sh_n(Pe)^{-1/3}$	$Sh_l(Pe)^{-\frac{1}{2}}(u_0/U)^{-\frac{1}{2}}$
"Exact" [12]	0·998	0·923
Present results	0·9956(−0·2%)	0·905(−1·9%)
Integral method [2]	1·037 (+3·9%)	0·895(−3.0%)

The present results are closer to the exact calculations than the classical integral methods such as the momentum integral method when use is made of the same type of approximate

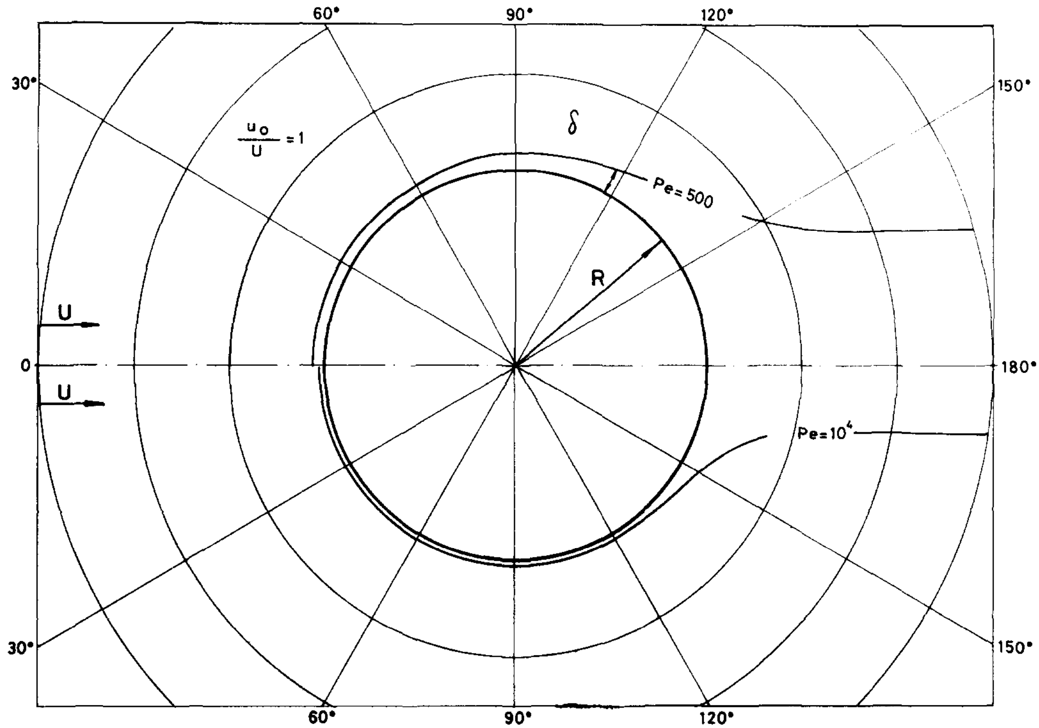

FIG. 2. Diffusional boundary layer for low particle Reynolds number viscous flow past spherical particles with large internal circulation (no radial velocity).

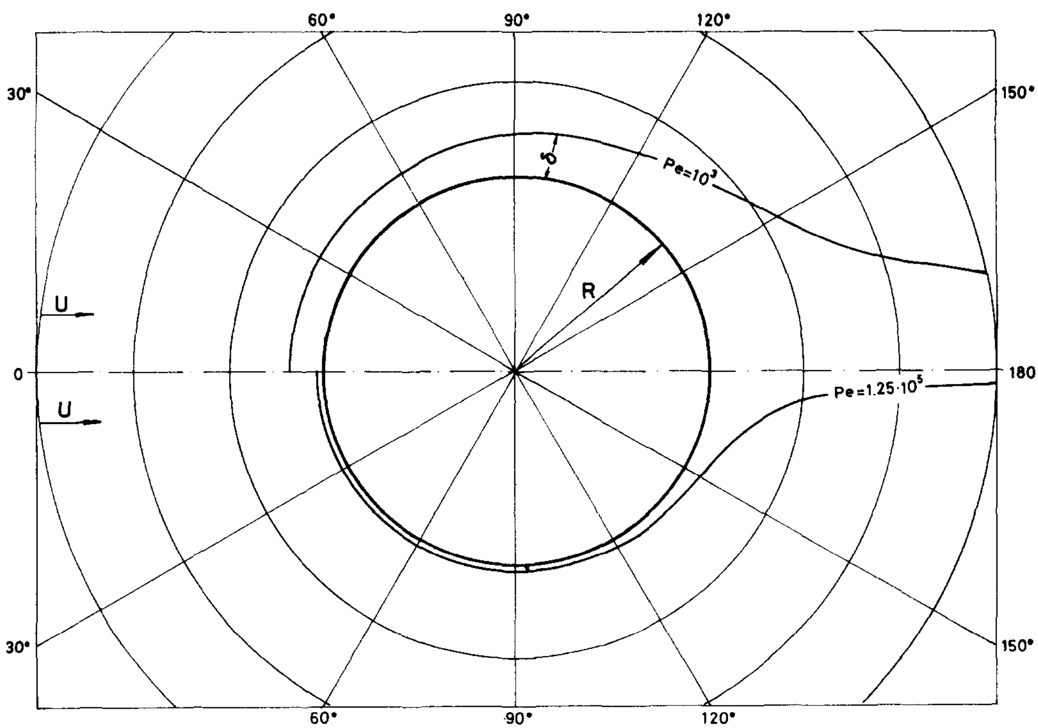

FIG. 3. Diffusional boundary layer for low particle Reynolds number viscous flow past spherical particles with negligible internal circulation (no radial velocity).

function. This has been also the case in hydrodynamic boundary layers [7], and is expected since the variational analysis extracts the "best" solution from any assumed family of possible solutions. The obtained solution is the best, in the sense that if the exact solution minimizes the value of the functional, which in our case is a measure of dissipation, any other solution will, for the same bounds give a higher result. Taking now a n parameter family of trial functions, a larger value of the functional is obtained. The set of values for the parameters which minimize the functional fulfil conditions like equation (14). The minimal value obtained is the one which approaches the exact result most closely, out of the whole family of trial functions.

To characterise now the general case with non-negligible radial velocity at the interface, one has to know the distribution of this velocity, as a function of θ on the sphere. At the absence of accurate experimental information on the functional form of this distribution, the choosing of the form of $v_w(\theta)$, equation (6), and therefore of the injection parameter T, equation (12), becomes more or less arbitrary.

Let us therefore choose first a "self-similar" distribution of v_w, which causes T, equation (12), to be a constant (i.e. not a function of θ). This selection is physically plausible, as the concentration boundary conditions which define the parameter T, do not depend on the angle θ. Here the function A_4, A_5 of equation (16) vanish, and for a large internal circulation one obtains

$$\frac{A_1}{2}\frac{u_0 \sin\theta}{R}\delta\frac{d\delta}{d\theta} + A_1\frac{u_0\cos\theta}{R}\delta^2 - D_i[TA_0 + A_3] = 0 \qquad (25)$$

from which

$$\frac{\delta_l}{R} = \beta_1(T)Pe^{-\frac{1}{2}}\left(\frac{u_0}{U}\right)^{-\frac{1}{2}} \times \frac{(\cos^3\theta - 3\cos\theta + 2)^{\frac{1}{2}}}{\sin^2\theta}$$

where $$\beta_1(T) = \left[\frac{4}{3}\frac{TA_0 + A_3}{A_1}\right]^{\frac{1}{2}} \qquad (26)$$

and the velocity distribution has the form [using also equation (12)]

$$\frac{v_w}{U} = \frac{2I}{\sqrt{Pe}}\left(\frac{u_0}{U}\right)^{\frac{1}{2}}\frac{\sin^2\theta}{(\cos^3\theta - 3\cos\theta + 2)^{\frac{1}{2}}}. \qquad (27)$$

Here I is an injection coefficient independent of material properties, the conditions in the flowfield, and the angle θ.

Similarly, for negligible circulation

$$A_2\frac{Q\sin\theta}{R^2}\delta^2\frac{d\delta}{d\theta} + A_2\frac{Z\cos\theta}{R^2}\delta^3 - D_i(TA_0 + A_3) = 0 \qquad (28)$$

and

$$\frac{\delta_n}{R} = \beta_2 Pe^{-\frac{1}{3}}\frac{(2\theta - \sin 2\theta)^{\frac{1}{3}}}{\sin\theta}$$

where $$\beta_2(T) = \left(\frac{3}{4}\frac{TA_0 + A_3}{A_2}\right)^{\frac{1}{3}} \qquad (29)$$

and the "similar" velocity distribution becomes

$$\frac{v_w}{U} = \frac{2I}{Pe^{\frac{2}{3}}}\frac{\sin\theta}{(2\theta - \sin 2\theta)^{\frac{1}{3}}}. \qquad (30)$$

The "similar" velocity distributions for both ranges of internal circulation assume their largest value at the frontal stagnation point, and gradually taper off to zero at the rear stagnation point. This is in line with Levich's [12] assertion that the rear "wake" area has little influence on the total interphase mass transfer.

The effect of positive or negative v_w on the diffusional boundary layer shape is shown in Figs. 4 and 5, for large and negligible internal circulation, respectively. The effect on single spheres with negligible internal circulation is much larger, as the tangential velocities on the interface (which are much larger than the radial component in the case of internal circulation) "wash" the added mass downstream.

Figures 6 and 7 describe the influence of the injection or suction velocity on the Sherwood number defined by equation (24), as a function of the Péclét number. Positive injection values lower the mass transfer coefficient, since the

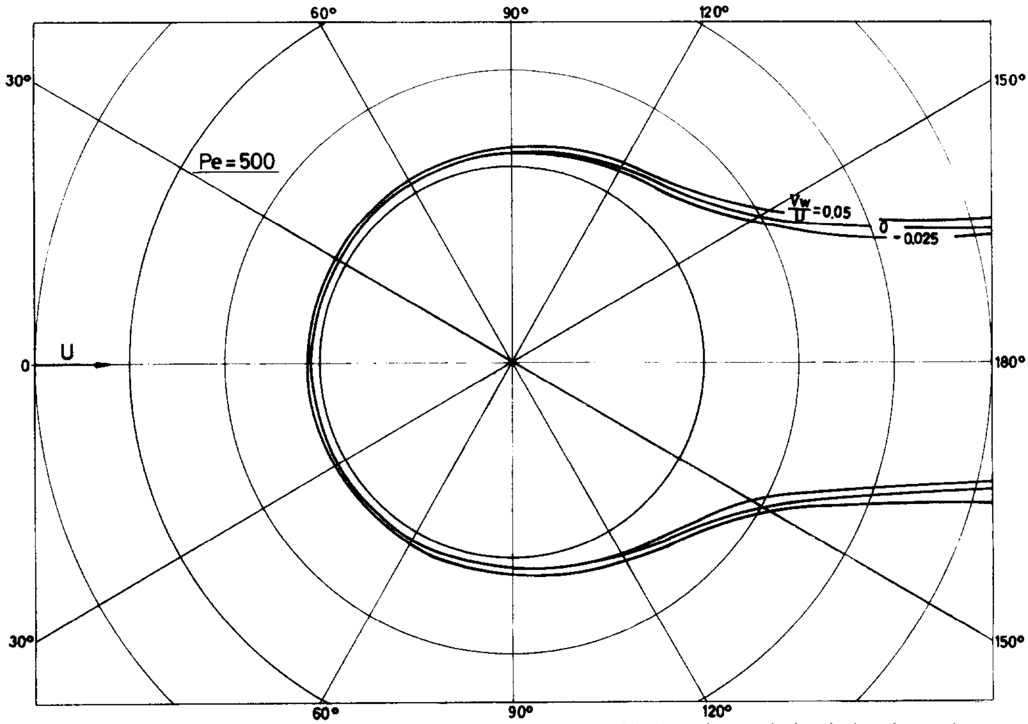

FIG. 4. Diffusional boundary layer around spherical particles with large internal circulation for various injection or suction velocities.

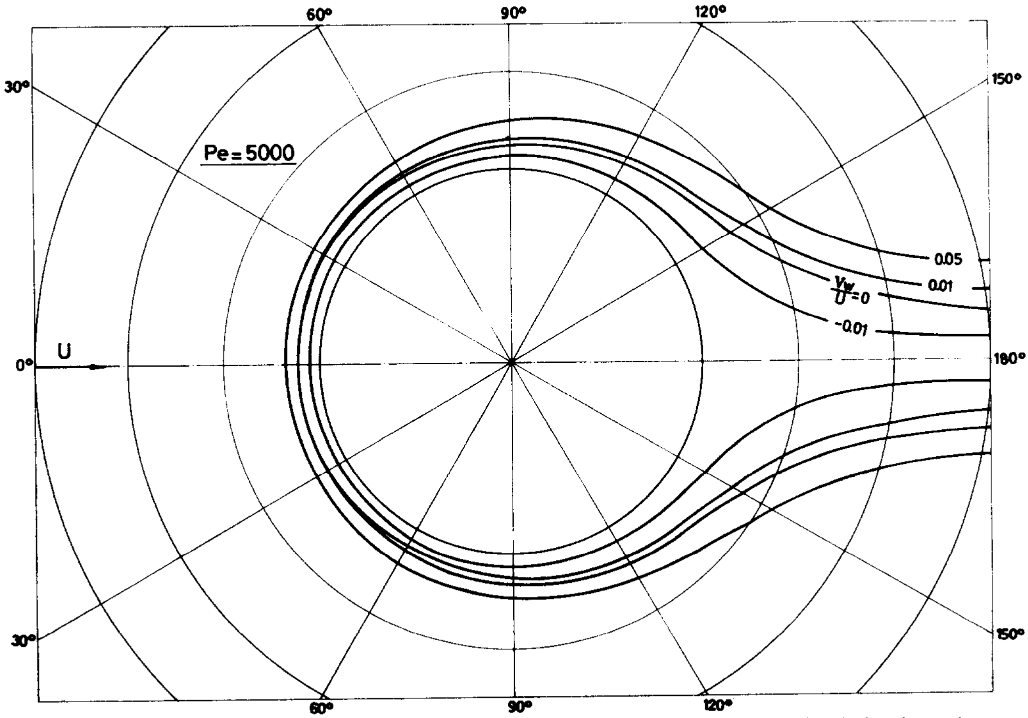

FIG. 5. Diffusional boundary layer around spherical particles with negligible internal circulation for various injection or suction velocities.

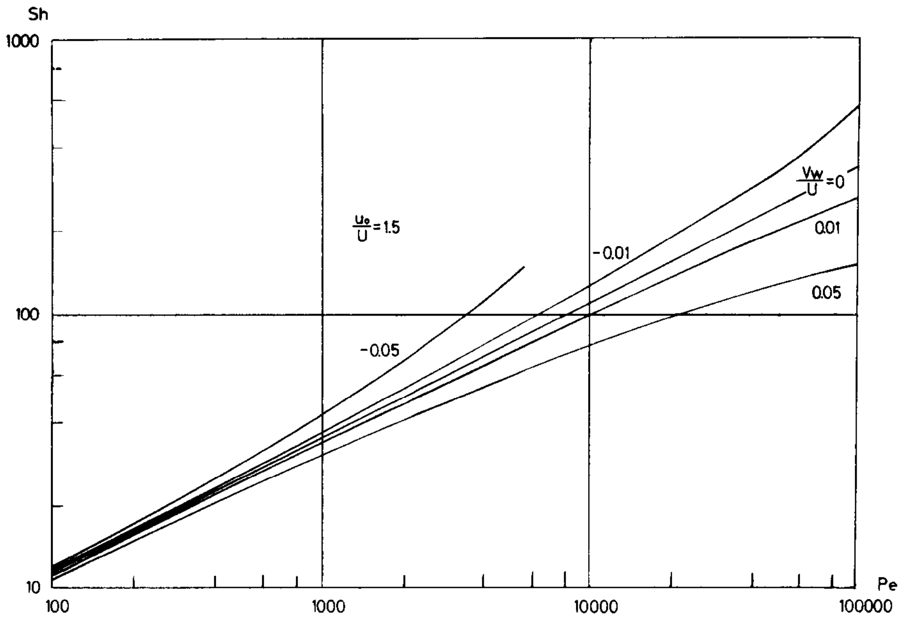

FIG. 6. Average Sherwood number for a sphere with injection or suction vs. Péclét number for $u_0/U = 1{\cdot}5$.

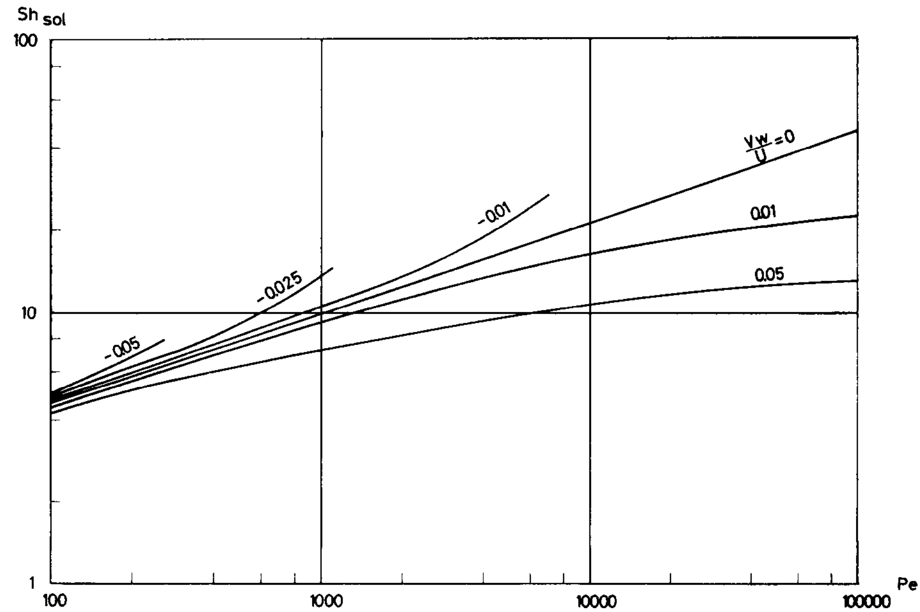

FIG. 7. Average Sherwood number for a sphere with injection or suction vs. Péclét number for negligible internal circulation.

direction of mass transfer as defined is positive towards the sphere, equation (11). When the interfacial velocity is negative (i.e. into the sphere) the solution exists only for a limited range of the injection coefficient, so that for every negative value of v_w/U there is a maximum *Pe* number for which the solution is valid.

Figure 8 shows the effect of internal circulation on the Sherwood number, (as a function of the *Pe* number), again pointing to the augmentation of mass transfer due to the internal circulation.

A different distribution, where $v_w(\theta)$ = const. has been also studied, to check the influence of

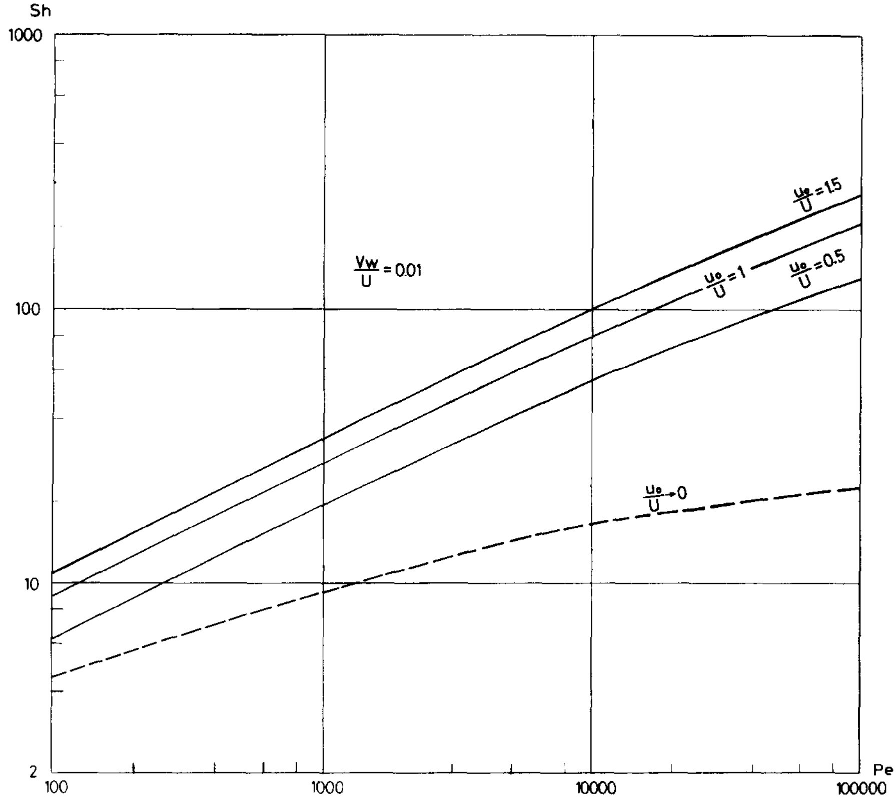

FIG. 8. Average Sherwood number for a sphere with injection vs. Péclét number for various internal circulation intensities.

Increasing the intensity of internal circulation moves this limit towards higher values of suction rates. Similar behaviour was found by Merk [8] for diffusional boundary layers on flat plates at high Reynolds numbers, and by Weihs [7] for hydrodynamic boundary layers. A possible explanation is that at higher suction velocities the boundary layer assumptions do no longer apply.

the choice of distribution on total average mass transfer to or from the sphere. Such a distribution may occur when a droplet is suspended from a much smaller (in diameter) pipe, through which mass is continually added to compensate for the mass lost by transfer to the surrounding phase. In this case the concentration profiles are no longer self-similar, and one has to use the local-similarity method [9] of boundary-layer theory, calculating the profiles at several stations around the sphere. The total average mass transfer for this case as a function of the injection coefficient I appears in Fig. 9, which shows that difference between the two distributions is relatively small. Again no solution is feasible below certain values of the suction coefficient.

solution from any assumed family of possible approximate solutions.

From the mass transfer point of view the following aspects are important.

(i) Estimation of total average mass transfer rate to (suction) or from (injection) particles is strongly affected by radial velocity components

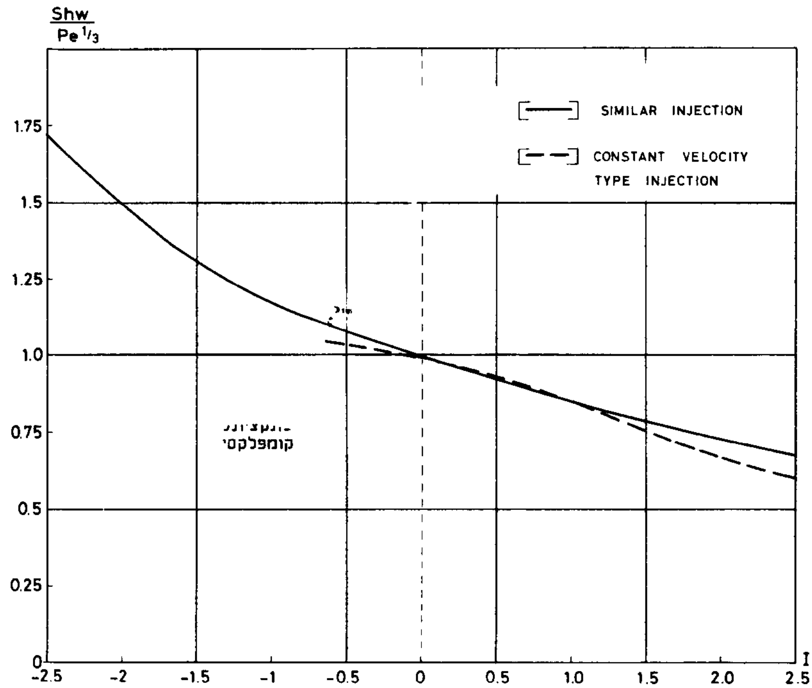

FIG. 9. Normalized average Sherwood number on the particle surface vs. injection-suction coefficient (small internal circulation).

IV. CONCLUSIONS

The proposed variational analysis for treating convective mass (or heat) transfer to or from moving solid or fluid particles, when properly treated, can yield quantitatively useful results. Many flow and mass transfer problems that cannot be exactly solved analytically may be easily handled by this method. Comparison of the present method with existing "exact" and integral methods (for calculating average mass transfer coefficients for single spherical particles with negligible radial velocity) clearly demonstrates its general property to extract the "best" at the interface which are due to the very process of interfacial mass transfer.

(ii) Available analyses for interfacial mass and heat transfer to or from particles (which do not take into account this radial velocity) introduce a considerable error into their predictions. This error may be tens of per cents even when the interfacial velocity due to mass transfer is very small. For instance, when $v_w/U = 0{\cdot}01$, the mass transfer from a solid sphere is changed by 20 per cent ($Pe = 6000$), and when $v_w/U = 0{\cdot}05$, the change may reach 45 per cent (see figures).

(iii) In general, the errors introduced in predicting convective mass transfer rates or boundary-layer thickness with a neglected radial velocity at the interface are much more pronounced when circulation inside fluid spheres is small. Since for a given fluid–fluid system the degree of internal circulation depends on the degree of purity of the fluids with respect to surfactant contaminants (which are present even in "normally" distilled fluids), the last have a pronounced effect on interfacial mass transfer rates. Predictions of mass transfer rates from solid spheres are however more sensitive to very small radial velocity components than those for highly pure drop or bubble fluid systems. Decreasing internal circulation in a given fluid–fluid system (due, say, to surfactant contamination) decreases the potential of the boundary layer to carry mass efflux downstream into the wake. This increases the influence of small radial mass transfer fluxes on the predictions of total average interfacial transfer.

(iv) In the analytical estimation of total average mass transfer rate the choice of a particular radial velocity distribution along the interface may have small influence on the final results. However, the last conclusion is based only on two types of distributions which have been considered; the "similar" injection-suction distribution, and the constant (in θ) injection-suction velocity.

(v) Up to positive values of $v_w/U = 0(10^{-2})$ the separating streamline $\Psi = 0$ has an approximately spherical shape. For the extremely large value of $v_w/U = 0$ (1) interfacial mass transfer takes place between the continuous fluid phase and a type of Rankine-body shaped "phase" which is composed of species i only (i.e. the interfacial transfer no longer takes place on the surface of the sphere). The last case is expected to characterize some of the extremely high rates of evaporation and combustion of small fuel droplets injected into very hot gas. Numerical solutions characterizing such Rankine-body shaped "phases" for evaporation and combustion of fuel droplets are now in progress and the results will be reported in due time.

ACKNOWLEDGEMENT

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APPENDIX

Criteria for Validity of the Spherical-Symmetric, Steady-State Analysis

For the mass transfer to be quasi-steady it must be subject to the condition

$$\dot{m}\tau \ll m \qquad \text{and} \qquad m = \tfrac{4}{3}\pi R^3 \rho_d \tag{A.1}$$

where $\dot{m}$ is the mass transferred between the phases per unit time, m—the instantaneous mass of the sphere, and τ a characteristic time defined by

$$\tau = \frac{2R}{U}. \tag{A.2}$$

Since

$$\dot{m} = 4\pi R^2 \rho_i \hat{v}_w \tag{A.3}$$

where ρ_i is the density of the transformed material in the continuous phase, and $\hat{v}_w$ the average radial velocity on the surface of the entire sphere, the condition (A.1) becomes

$$\frac{\hat{v}_w}{U} \ll \frac{1}{6}\frac{\rho_d}{\rho_i}. \tag{A.4}$$

When the two densities are of the same order of magnitude, then, for example,

$$\frac{\hat{v}_w}{U} \cong O(10^{-2}). \tag{A.5}$$

Following now Gal-Or and Waslo [10] and adding the injection terms, the stream-functions for flow around and within a sphere with or without internal circulation and

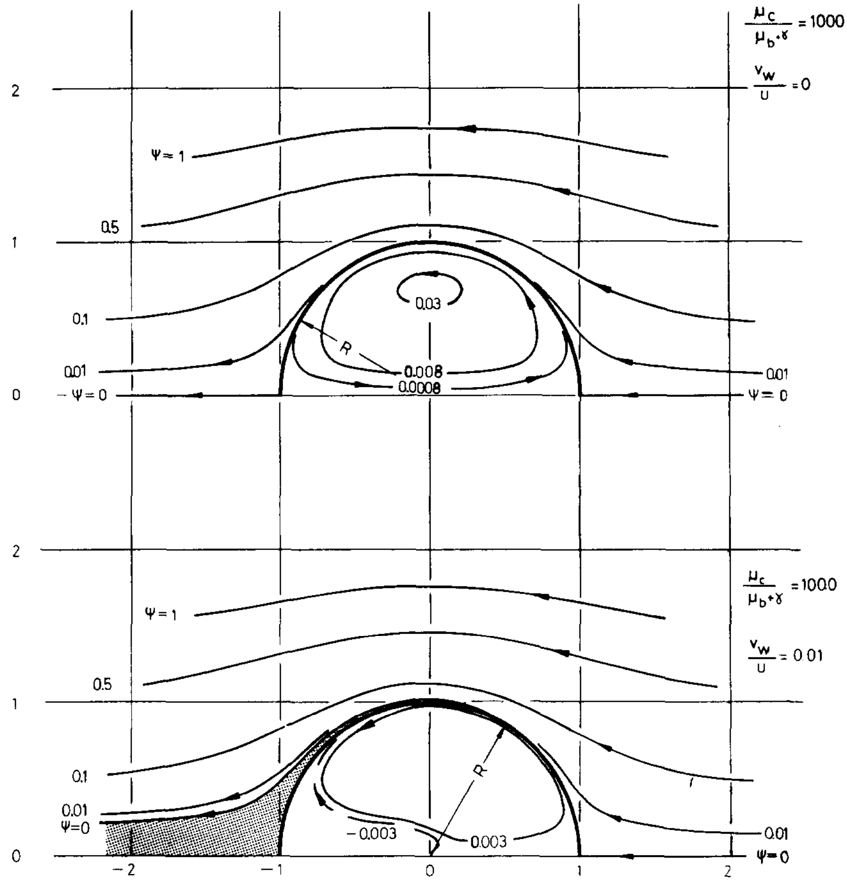

FIG. A1. The flow field around a spherical drop with and without radial velocity (v_W) on its surface, equation (A.6).

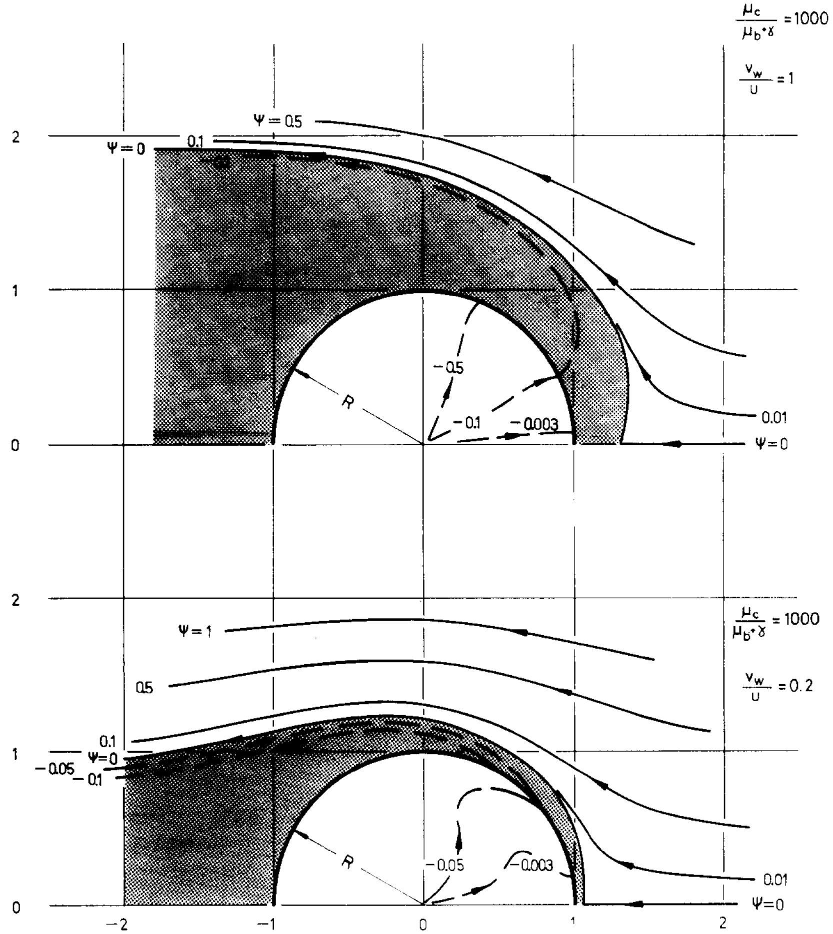

FIG. A2. The flow field around a spherical particle with extremely large radial velocity on its surface, equation (A.6).

surfactant impurities may be approximated by

$$\Psi_c = \frac{3}{4}\frac{R^2U}{W}\left(\frac{R}{r} - W\frac{r}{R} + Y\frac{r^2}{R^2}\right)\sin^2\theta + v_w(1-\cos\theta)$$

$$\Psi_d = \frac{3}{4}\frac{qUr^2}{W}\left(1 - \frac{r^2}{R^2}\right)\sin^2\theta - v_w(1-\cos\theta) \qquad (A.6)$$

where

$$Y \equiv 2(1+q) \qquad (A.7)$$

$$W \equiv 3(1+\tfrac{2}{3}q) \qquad (A.8)$$

$$q = \frac{\mu_c}{\mu_d + \gamma}. \qquad (A.9)$$

Substituting in (A.6) various values of v_w/U and q, we obtain Figs. A.1 and A.2 which show that for values of $v_w/U = 0(10^{-2})$ the separating streamline $\psi = 0$ has an approximately spherical shape (except for the "wake" region). For extremely large values of $v_w/U = 0[1]$ the mass-transfer surface is of the Rankine-body type and therefore can no longer be treated by using spherical-symmetrical analysis. The effect of interphase viscosity ratio on the flow-field shape is shown in Fig. A.3.

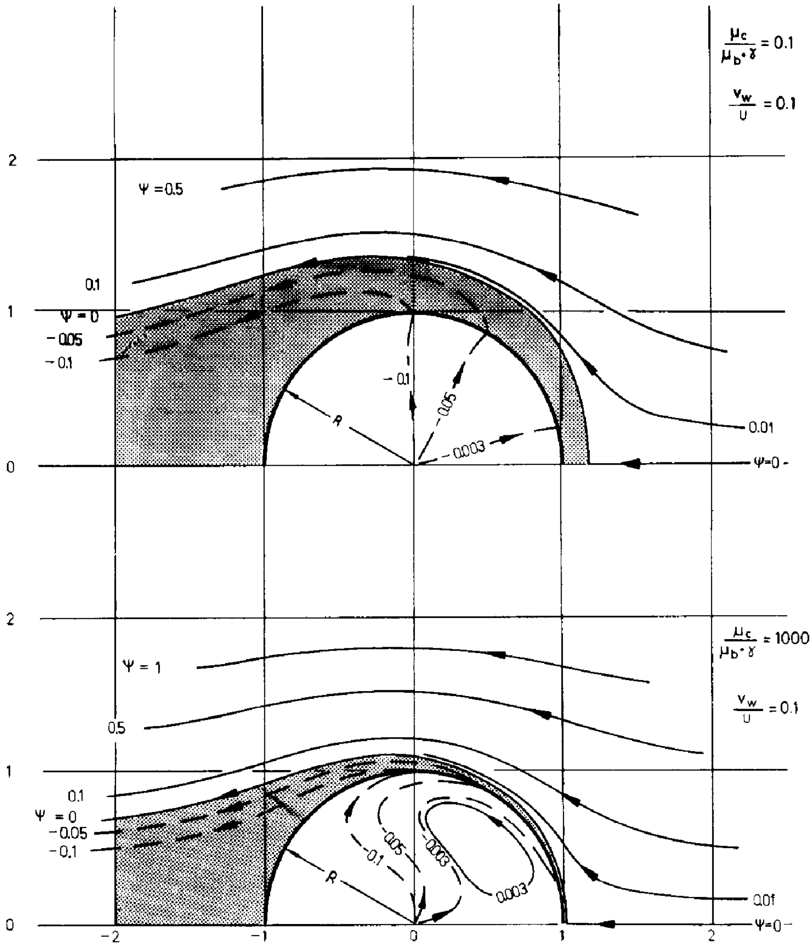

FIG. A3. Influence of the interphase viscosity ratio q and surface-active impurities on the flow around a spherical drop, equation (A.6).

ANALYSE VARIATIONNELLE DE HAUTS FLUX MASSIQUES A PARTIR DE PARTICULES SPHERIQUES. COUCHE LIMITE AVEC INJECTION-SUCCION POUR DES PETITS NOMBRES DE REYNOLDS DE PARTICULE ET DES NOMBRES DE PECLET ELEVES

Résumé—Une analyse variationnelle générale pour des écoulements à couches limites hydrodynamiques thermiques et diffusionnels antérieurement développée par les auteurs est adaptée ici pour caractériser des flux massiques relativement grands à partir de gouttes sphériques, de bulles ou de particules solides mobiles. Ce travail tient compte de l'effet de vitesse radiale à la surface de la particule (dû au processus de transfert massique interfacial) dans l'analyse de flux massiques convectifs. Un effet important qui a été négligé jusqu'ici dans les études dynamiques de gouttes. L'erreur introduite en négligeant cet effet atteint 10% même lorsque la vitesse radiale interfaciale due au transfert massique est relativement faible. En général on trouve que les erreurs introduites en estimant les flux convectés ou l'épaisseur de la couche limite en négligeant la vitesse radiale à l'interface sont beaucoup plus prononcées quand la circulation au sein des particules fluides est petite. Une comparaison de notre méthode avec les méthodes "exactes" ou intégrales existantes (pour le calcul des coefficients de transfert de masse avec une vitesse radiale négligée) démontre clairement sa propriété générale d'extraire la "meilleure" solution d'essai d'une famille quelconque de solutions approchées possibles.

VARIATIONSANALYSE VON GROSSEN MASSENTRANSPORTVERHÄLTNISSEN BEI KUGELFÖRMIGEN TEILCHEN

Zusammenfassung—Eine allgemeine Variationsanalyse für hydrodynamische, thermische und Diffusionsgrenzschichten wurde früher von den Autoren entwickelt. Sie wird hier angewandt, um die relativ grossen Massentransportverhältnisse bewegter kugelförmiger Tröpfchen, Blasen oder fester Teilchen zu charakterisieren.

Diese Arbeit berücksichtigt auch die Wirkung der Radialgeschwindigkeit an der Teilchenoberfläche (wegen des eigentlichen Stoffaustausches an der Trennfläche) auf die Analyse der konvektiven Massentransportverhältnisse, ein wichtiger Effekt, der bis jetzt in Abhandlungen über Tröpfchendynamik vernachlässigt wurde. Es zeigt sich, dass der Fehler, der durch diese Vernachlässigung entsteht, bis zu 10% beträgt, auch dann, wenn die Radialgeschwindigkeit an der Trennfläche in bezug auf den Massentransport relativ gering ist. Allgemein findet man, dass die Fehler, die entstehen, wenn man den konvektiven Massentransport oder die Grenzschichtdicke bestimmt, ohne die Radialgeschwindigkeit an der Trennfläche zu berücksichtigen, stark zunehmen, wenn die Zirkulation innerhalb der Flüssigkeit klein ist. Ein Vergleich unserer Methode mit vorhandenen "exakten" oder integralen Methoden (zur Berechnung des Massentransportkoeffizienten bei vernachlässigter Radialgeschwindigkeit) zeigt ihre allgemeine Eigenschaft, aus einer Reihe möglicher Näherungslösungen die "beste" auszusuchen.

ВАРИАЦИОННЫЙ АНАЛИЗ БОЛЬШИХ СКОРОСТЕЙ ПЕРЕНОСА МАССЫ ОТ СФЕРИЧЕСКИХ ЧАСТИЦ

Вдув-отсос пограничного слоя при малых числах Рейнольдса и больших числах Пекле для частиц

Аннотация—Общий вариационный анализ гидродинамических, тепловых и диффузионных течений с пограничным слоем, ранее разработанный авторами, используется здесь для характеристики относительно больших скоростей переноса массы от движущихся сферических капель, пузырьков или твердых частиц. В данной работе принимается во внимание влияние радиальной скорости частиц (благодаря самому процессу межфазного массообмена) на интенсивность конвективного переноса массы — эффект, который до сих пор не учитывался при изучении динамики капель. Показано, что погрешность, вносимая из-за пренебрежения этим эффектом, достигала десятков процентов даже тогда, когда межфазная радиальная скорость за счет переноса массы была относительно небольшой. В общем, установлено, что ошибки, вносимые в расчет скоростей конвективного переноса или толщины пограничного слоя в пренебрежении радиальной скоростью на границе раздела гораздо более существенны при малой скорости внутри жидкости. Сравнение нашего метода с существующими «точными» или интегральными методами (для расчета коэффициентов массообмена в пренебрежении радиальной скоростью) показывает, что с его помощью можно получить наиболее точное решение.