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ON NONISOTHERMAL CHEMICAL REACTION AT THE PARTICLE SURFACE IN A LAMINAR FLOW

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Abstract—The paper is concerned with the steady-state convective diffusion to a heat-conducting reacting sphere in a laminar translational and shear flow, with a nonisothermal chemical reaction proceeding on its surface at the rate arbitrarily dependent on temperature and concentrations.

It is shown that for the integral heat and mass fluxes of reacting components to a particle (i.e. mean Nusselt, Nu, and Sherwood, Sh_m, numbers) to be determined at small Peclet numbers, it suffices to solve the following universal algebraic (transcendental) system of equations

$$Sh_m = f_m\left(\frac{Nu}{Nu_\infty}, \frac{Sh_1}{Sh_{1\infty}}, \ldots, \frac{Sh_M}{Sh_{M\infty}}\right),$$

$$Nu = \sum_{m=1}^{M} h_m Sh_m,$$

which is appreciably simpler than the initial system of partial differential equations. The system suggested makes it possible to obtain for the Sherwood and Nusselt numbers the first four (shear flow), and the first three (translational flow), terms of the corresponding asymptotic expansion in small Peclet numbers.

An irreversible isothermal 2nd-order reaction and a successive stepwise 1st-order reaction have been studied. The case of nonisothermal surface reaction, which proceeds following the Arrhenius law with one reacting component present in the flow, has been studied in detail. Adsorption on the particle surface, the rate of which is governed by the Langmuir kinetics, is analyzed.

Based on comparison with the earlier results for isothermal reactions at moderate and large Peclet numbers, it is shown that with a suitable choice of the governing parameters $Sh_{m\infty}(Nu_\infty)$ the suggested algebraic system can also be successfully applied for approximate determination of the mean Sherwood numbers within the whole range of Peclet numbers.

The results obtained enable investigation of the inverse mass and heat transfer problem for reacting particles in the flow, i.e. allow one to explicitly determine the dependence of the surface reaction rate on concentrations from the available integral fluxes that can be determined experimentally.

NOMENCLATURE

a, particle radius;
a_e, radius of a volumetrically equivalent sphere;
c_m, dimensionless concentration of mth component;
c_m^*, dimensional concentration;
$c_{m\infty}$, concentration at infinity;
$\hat{c}_m$, solution of auxiliary problem (7.1);
D_m, diffusivity of the mth species in mixture;
E, activation energy;
F_m, dimensional surface reaction rates;
f_m, dimensionless surface reaction rates;
$G_{ij}^*(G_{ij})$, dimensional (dimensionless) elements of the matrix of shear factors;
g, parameter defined in equation (5.3);
H_m, dimensional heat of mth reaction;
h_m, dimensionless heat of mth reaction;
I_m, integral mass flux of the mth reagent to the particle surface;
$I_{m\infty}$, integral mass flux in diffusional mode of reaction;
J, integral heat flux to a particle;
J_∞, parameter defined in equation (3.15);
K, parameters defined in equation (5.9);
k, dimensionless reaction rate constant;
k_0, dimensional reaction rate constant;
$\mathbf{L}$, differential operator defined in equation (3.7);
M, number of reagents participating in reaction;
m, ordinal number of reagent;
Nu, mean Nusselt number;
Nu_∞, mean Nusselt number in diffusional mode of reaction;
P_m, diffusional Peclet numbers ($1 \leq m \leq M$);
P_0, heat transfer Peclet number;
p, parameter defined in equation (5.7);
Q_m, parameters occurring in equation (3.2);
R, universal gas constant;
r, θ, ϕ, spherical coordinate system moving with particle;
S, function defined in equation (6.4);
S_r, surface of sphere with radius r;
Sh_0, mean Sherwood number for a particle at rest;
Sh_∞, mean Sherwood number for diffusional mode of reaction;
Sh_m, mean Sherwood number of the mth components of reacting substance;

s, parameter defined in equation (5.7);
T, dimensionless temperature in liquid (gas) flow;
T^*, dimensional temperature in flow;
T_∞, liquid (gas) temperature at infinity;
t, dimensionless temperature inside a particle;
t^*, dimensional temperature inside a particle;
U, characteristic flow velocity;
U_∞, translational flow velocity at infinity;
$\mathbf{u}$, liquid velocity vector;
u_i, liquid velocity components in Cartesian coordinate system;
x_i, Cartesian coordinate system moving with a particle ($i = 1, 2, 3$);
z, parameter defined in equation (6.3).

Greek symbols

α, numerical factor occurring in equations (3.3);
α_m, parameters occurring in equations (4.4);
β, ratio between dynamic viscosities of a droplet and surrounding liquid; parameter defined in equations (6.3);
Γ, hyperbola (6.7);
γ, parameter defined in equations (6.8);
Δ, Laplace operator;
δ, ratio between thermal conductivities of particle and liquid (gas);
ε, small parameter introduced into equation (3.2);
ζ_m, fundamental solutions of equations for temperature and concentration distributions in the external flow region Ω_∞;
κ, order of reaction;
Λ, shape factor ($\Lambda = 1$ for a sphere);
λ_1, thermal conductivity of a particle;
λ, thermal conductivity of liquid (gas);
μ, dynamic viscosity of liquid;
ξ, dimensionless resistance force of a particle (droplet);
$\mu_m^{(k)}$, coefficients occurring in equations (3.5);
ρ, 'contracted' coordinate;
σ, parameter defined in equation (6.3);
Φ_m, unknown functions, to be determined, that enter into relations (3.3) and (4.2);
χ, thermal diffusivity;
Ψ, unknown function entering into relations (3.3) and (4.2);
ψ, stream function;
Ω_1, internal flow region;
Ω_∞, external region;
ω, dimensionless characteristic parameter defined in equation (6.1).

1. INTRODUCTION

FOR THE first time the problem of heat and mass transfer of a sphere in a steady-state Stokes flow at small Peclet numbers has been studied by Acrivos and Taylor [1] using the method of matched asymptotic expansions. Concentration far from the particle and on its surface was assumed to be constant. The first five terms of asymptotic expansion have been obtained for the mean Sherwood number. Brenner [2] has extended this problem to the case of an arbitrarily shaped particle; a 3-term expansion in Peclet numbers has been obtained for the mean Sherwood number. Rimmer [3, 4] has considered a similar problem for a sphere using the results of Proudman and Pearson [5] for the liquid velocity field which were obtained by the method of matched asymptotic expansions in small Reynolds number.

Frankel and Acrivos [6] have considered convective diffusion to a sphere freely suspended in a simple shear flow. A 2-term expansion has been obtained for the mean Sherwood number. Batchelor [7] has extended these results to the case of a particle of any shape freely suspended in an arbitrary linear flow. On the basis of Batchelor's results [7], Acrivos [8] has obtained for a sphere the first four, and for an arbitrarily shaped particle the first three, terms of the corresponding asymptotic expansion.

Convective diffusion to a sphere and an arbitrarily shaped particle in a uniform translational flow with an isothermal 1st-order reaction occurring on its surface was considered by Gupalo and Ryazantsev [9] and Gupalo, Ryazantsev and Syskov [10]. Taylor [11] has studied mass transfer of a sphere with chemical reactions of the first and second orders occurring on its surface. The case of an arbitrary kinetics of the surface reaction on a sphere in Stokes flow was considered by Gupalo *et al.* [12]. Gupalo, Ryazantsev and Chalyuk [13] have determined the temperature field inside and outside a heat-conducting particle in the case of complete absorption of reagent by the particle surface.

2. STATEMENT OF THE PROBLEM

Consideration is given to the analysis of convective diffusion to a reacting sphere in a laminar translational and shear flow with a nonisothermal chemical reaction occurring on its surface at the rate arbitrarily dependent on temperature and concentrations. The particle is assumed to be heat-conducting, and the reacting components are thought to be at rather small concentrations so that the presence of surface reaction does not influence the flow and particle parameters. The effect of heat and pressure diffusion is also left out of account.

The dimensionless convective diffusion and heat conduction equations as well as the boundary conditions specifying uniformity of temperature and concentrations far from the particle, temperature continuity, 'reaction law' and heat balance on the particle surface, as well as temperature boundedness at the particle centre are of the form

$$\Delta c_m = P_m(\mathbf{u}\nabla)c_m,\ (1 < r < \infty)\ m = 1, \ldots, M, \tag{2.1}$$

$$\Delta T = P_0(\mathbf{u}\nabla)T,\ (1 < r < \infty), \tag{2.2}$$

$$\Delta t = 0,\ (0 \le r < 1), \tag{2.3}$$

$$r \to \infty,\ c_m \to 0,\ T \to 0, \tag{2.4}$$

$$r = 1,\ T = t, \tag{2.5}$$

$$r = 1,\ -\partial c_m/\partial r = f_m(T, c_1, \ldots, c_M), \tag{2.6}$$

$$r = 1,\ -\frac{\partial T}{\partial r} + \delta\frac{\partial t}{\partial r} = \sum_{m=1}^{M} h_m f_m(T, c_1, \ldots c_M), \tag{2.7}$$

$$r = 0,\ |t| < \infty, \tag{2.8}$$

$$c_m^* = c_{m\infty}(1-c_m),\ T^* = T_\infty(1-T),\ t^* = T_\infty(1-t),$$

$$P_m = \frac{aU}{D_m},\ P_0 = \frac{aU}{\chi},\ \delta = \frac{\lambda_1}{\lambda},\ h_m = \frac{c_{m\infty}H_m D_m}{\lambda T_\infty},$$

$$f_m(T, c_1, \ldots, c_M) \equiv a(c_{m\infty}D_m)^{-1}F_m(T^*, c_1^*, \ldots, c_M^*). \tag{2.9}$$

For the time being, it is assumed that $c_{m\infty} \neq 0\ (m = 1, \ldots, M)$; henceforth these restrictions will be removed.

Equations (2.1) and (2.2) and boundary conditions (2.6) and (2.7) for the case of a non-conducting particle can be found in [14].

3. SHEAR FLOW

In the case of an arbitrary shear flow, the liquid velocity distribution far from a particle will have the following form in dimensionless variables

$$r \to \infty,\ u_i \to G_{ij}x_j,\ G_{ii} = 0\ (i, j = 1, 2, 3),$$

$$(G_{ij} = G_{ij}^* G^{-1},\ G = \max_{i,j}|G_{ij}^*|,\ U = aG).$$

Here and subsequently, the summation is carried out over the repeated indices i and j, G_{ij}^* are the elements of the matrix of shear factors, u_i are the components of liquid velocities in the Cartesian system of coordinates $x_i (i = 1, 2, 3)$.

In particular, in the axisymmetric case ($G_{11} = G_{22} = 1/2$, $G_{33} = -1$ and $G_{ij} = 0$ at $i \neq j$) the liquid velocity distribution, which satisfies the above conditions at infinity, is determined, in Stokesian approximation, by the stream function

$$\psi = \frac{1}{2}\left(r^3 - \frac{5}{2} + \frac{3}{2}r^{-2}\right)\sin^2\theta\cos\theta,$$

$$(\mathbf{u}\nabla)w \equiv \frac{1}{r^2\sin\theta}\frac{\partial(w, \psi)}{\partial(r, \theta)}. \tag{3.1}$$

Here $\partial(w, \psi)/\partial(r, \theta)$ is the Jacobian of the functions w and ψ.

Let us examine the boundary-value problem (2.1)–(2.9) by the method of matched asymptotic expansions in small Peclet numbers. It is assumed that

$$\varepsilon \to 0,\ P_m = \varepsilon Q_m,\ Q_m = O(1),\ m = 0, 1, \ldots, M \tag{3.2}$$

and the entire flow region is divided into the inner, $\Omega_1 = \{1 \le r \le O(\varepsilon^{-1/2})\}$, and the outer, $\Omega_\infty = \{O(\varepsilon^{-1/2}) \le r\}$, subregions (see, e.g. [8]). As usual, a 'contracted' coordinate, $\rho = \varepsilon^{1/2}r$, is introduced in the outer region and the solution in each of the subregions is sought separately in the form of internal and external expansions. In constructing an asymptotic solution in the internal region, the boundary conditions on the particle surface (2.5)–(2.7) are used, while in the outer region, the boundary conditions at infinity (2.4) are employed; the resulting unknown constants are determined by using the matching technique.

The above situation (3.2) is typical of slow gas motions, where $0.5 \lesssim D\chi^{-1} \lesssim 2.0$ [15].

Batchelor [7] and Acrivos [8] have shown that in the case of an arbitrary Stokesian shear flow past a particle, equations (3.1), the temperature and concentration distributions in the outer region Ω_∞ can be expressed by the fundamental solution of the equations

$$\Delta_{\rho_m}\zeta_m = G_{ij}x_i\partial\zeta_m/\partial x_j,\ \rho_m = P_m^{1/2}r\ (m = 0, 1, \ldots, M).$$

Therefore, by virtue of the properties of ζ_m[8], the temperature and concentration distributions at the inner boundary of the outer region Ω_∞ (i.e. at $r \sim \varepsilon^{-1/2}$) can be presented as ($m = 1, \ldots, M$)

$$r = O(\varepsilon^{-1/2}),\ T = \Psi(P_0, P_1, \ldots, P_M) \times \{r^{-1} - \alpha P_0^{1/2} + 0(P_0^2)\},$$

$$r = O(\varepsilon^{-1/2}),\ c_m = \Phi_m(P_0, P_1, \ldots, P_M) \times \{r^{-1} - \alpha P_m^{1/2} + 0(P_m^2)\}. \tag{3.3}$$

Here $\alpha = \alpha(G_{ij})$ is a numerical factor, while Ψ and Φ_m are the unknown functions which are determined in the course of the problem solution. A general expression for determining the value of the parameter α has been obtained by Batchelor [7]. Thus, for an axisymmetrical case, equation (3.1), $\alpha = 0.399$ [7], while for the case of a simple shear (one off-diagonal element of the matrix G_{ij} is equal to unity and the remainder, to zero) $\alpha = 0.258$ [6].

The inner temperature and concentration expansion in the region Ω_1, as well as temperature distribution inside a particle involve the following representation

$$T = \sum_{k=0}^{3} P_0^{k/2}\,T^{(k)} + o(P_0^{3/2}),$$

$$t = \sum_{k=0}^{3} P_0^{k/2}\,t^{(k)} + o(P_0^{3/2}),$$

$$c_m = \sum_{k=0}^{3} P_m^{k/2}\,c_m^{(k)} + o(P_m^{3/2})(m = 1, \ldots, M), \tag{3.4}$$

where the functions $c_m^{(k)}$, $T^{(k)}$ and $t^{(k)}$ satisfy the equations

$$\Delta c_m^{(k)} = 0,\ \Delta T^{(k)} = 0,\ \Delta t^{(k)} = 0 \quad \text{at } k = 0, 1,$$

$$\Delta c_m^{(k)} = -\mu_m^{(k)} r^{-3} u_i x_i,\ \Delta T^{(k)} = -\sigma^{(k)} r^{-3} u_i x_i,\ \Delta t^{(k)} = 0 \quad \text{at } k = 2, 3. \tag{3.5}$$

In deriving the second group of equations (3.5) at $k = 2, 3$ it has been taken into account that the first terms of the inner expansion (3.4) and (3.5) at $k = 0, 1$ depend only on the radial coordinate r and are determined by the binomial of the form $A + Br^{-1}$, where for each $c_m^{(k)}$, $T^{(k)}$ and $t^{(k)}$ $(k = 0, 1)$ there are their own constants A and B determined from the boundary conditions (2.5)–(2.8) and the condition of matching with the solution in the region Ω_∞ (3.3); in (3.5) the summation is carried out over the index i; $\mu_m^{(k)}$, $\sigma^{(k)}$ = const.

For the analysis of equations (3.5), let us use the formula

$$\langle w \rangle \equiv \frac{1}{4\pi r^2} \int_{S_r} w\,ds = \frac{1}{4\pi} \int_0^{2\pi} \int_0^{\pi} \sin\theta\ w\,d\theta\,d\varphi \tag{3.6}$$

to introduce the mean value operation, which has been used by Acrivos [8]. Here S_r is the surface of the sphere with radius r.

Note that the equality $\langle w(r) \rangle = w(r)$ holds for any functions w depending on the coordinate r alone.

Integration of equation (3.5) over the surface of the sphere with radius r and account for the equality $\langle u_i x_i \rangle = 0$ [8] yield

$$\mathbf{L}\langle c_m^{(k)} \rangle = 0,\ \mathbf{L}\langle T^{(k)} \rangle = 0,\ \mathbf{L}\langle t^{(k)} \rangle = 0,$$

$$\mathbf{L} \equiv \frac{1}{r^2} \frac{d}{dr} r^2 \frac{d}{dr},\quad k = 0, 1, 2, 3. \tag{3.7}$$

By virtue of (3.4) and (3.7), the following equations are obtained accurate to $o(\varepsilon^{3/2})$ for the complete mean values

$$\mathbf{L}\langle c_m \rangle = 0,\quad \mathbf{L}\langle T \rangle = 0,\quad \mathbf{L}\langle t \rangle = 0. \tag{3.8}$$

The solution of equations (3.8), which satisfies the condition of matching with the solution in the outer region Ω_∞ (3.3), will, to the same order of accuracy, have the form

$$\langle c_m \rangle = \Phi_m(P_0, P_1, \ldots, P_M)(r^{-1} - \alpha P_m^{1/2}),$$
$$\langle T \rangle = \Psi(P_0, P_1, \ldots, P_M)(r^{-1} - \alpha P_0^{1/2}),$$
$$\langle t \rangle = \Psi(P_0, P_1, \ldots, P_M)(1 - \alpha P_0^{1/2}). \tag{3.9}$$

Here the form of the expression for $\langle t \rangle$ is due to the boundary conditions (2.5) and (2.8).

Since by virtue of equations (3.5) the $c_m^{(k)}$, $T^{(k)}$ and $t^{(k)}$ at $k = 0, 1$ depend only on the distance to the centre of the sphere r, then for any analytical function f the following formula will be valid accurate to $o(\varepsilon^{3/2})$

$$\langle f(T, c_1, \ldots, c_M) \rangle = f(\langle T \rangle, \langle c_1 \rangle, \ldots, \langle c_M \rangle), \tag{3.10}$$

which is proved by direct verification with account for equation (3.4) and (3.6).

The mean Sherwood and Nusselt numbers are determined from

$$Sh_m = -\left\langle \frac{\partial c_m}{\partial r} \right\rangle \Big|_{r=1} = \Phi_m,$$
$$Nu = -\left\langle \frac{\partial T}{\partial r} \right\rangle \Big|_{r=1} = \Psi.$$

Averaging of the boundary conditions (2.6) and (2.7) and use of equations (3.9) and (3.10) will yield the following algebraic (transcendental) system of equations to determine the mean Sherwood and Nusselt numbers

$$Sh_m = f_m\left(\frac{Nu}{Nu_\infty}, \frac{Sh_1}{Sh_{1\infty}}, \ldots, \frac{Sh_M}{Sh_{M\infty}}\right), \quad m = 1, \ldots, M, \tag{3.11}$$

$$Nu = \sum_{m=1}^{M} h_m Sh_m. \tag{3.12}$$

Here, the last of the above equations has been obtained by substituting into the boundary condition (2.7) the expressions for f_m from equation (2.6), while the values of $Sh_{m\infty}$, Nu_∞ correspond to purely diffusional (thermal) mode of reaction (which complies with the boundary conditions on the sphere surface $r = 1, c_m = 1, T = 1$) and have been obtained by Acrivos [8]

$$Sh_{m\infty} = 1 + \alpha P_m^{1/2} + \alpha^2 P_m + \alpha^3 P_m^{3/2} + o(P_m^{3/2}) \approx (1 - \alpha P_m^{1/2})^{-1},$$
$$Nu_\infty = 1 + \alpha P_0^{1/2} + \alpha^2 P_0 + \alpha^3 P_0^{3/2} + o(P_0^{3/2}) \approx (1 - \alpha P_0^{1/2})^{-1}. \tag{3.13}$$

Hence, it is shown that in the case of the surface chemical reaction accompanied by heat release, in order to determine the integral heat and mass inflows of reacting components to the particle at small Peclet numbers, it is sufficient to solve the algebraic (transcendental) system of equations (3.11) and (3.12) which is much simpler than the initial system of partial differential equations (2.1)–(2.9).

It is seen from (3.11) and (3.12) that at small Peclet numbers the ratio between thermal conductivities of the particle and surrounding liquid does not influence the integral characteristics of the process. Variation of the parameter δ [in the boundary condition (2.7)] leads only to redistribution of local thermal and diffusional fluxes over the sphere surface, with the respective total fluxes being unchanged.

The system (3.11) and (3.12) can be written in dimensional form in terms of the integral fluxes

$$I_m = 4\pi a^2 F_m\left[\left(T_\infty - \frac{J}{J_\infty}\right), \left(c_{1\infty} - \frac{I_1}{I_{1\infty}}\right), \ldots, \left(c_{M\infty} - \frac{I_M}{I_{M\infty}}\right)\right], \tag{3.14}$$

$$J = \sum_{m=1}^{M} H_m I_m \quad (m = 1, \ldots, M),$$

$$I_m = 4\pi a D_m \langle \partial c_m^*/\partial r \rangle|_{r=1},\ J = 4\pi a\lambda \langle \partial T^*/\partial r \rangle|_{r=1},$$
$$I_{m\infty} = 4\pi a D_m Sh_{m\infty},\ J_\infty = 4\pi a\lambda Nu_\infty. \tag{3.15}$$

Here I_m and J are total fluxes of reacting substance and heat to the sphere surface; the expressions for $Sh_{m\infty}$ and Nu_∞ are determined by equation (3.13).

Equations (3.14) and (3.15) are more suitable than equation (3.11) and (3.12) in those cases when at least one of the concentrations of reacting components in the incident flow far from the particle is zero, i.e.

$$\text{at} \quad \prod_{m=1}^{M} c_{m\infty} = 0.$$

4. TRANSLATIONAL STOKESIAN FLOW

For a uniform translational Stokesian flow the liquid velocity distribution in dimensionless variables is determined by the stream function

$$\psi = \frac{1}{2}\left(r^2 - \frac{3}{2}r + \frac{1}{2}r^{-1}\right)\sin^2\theta. \tag{4.1}$$

Here the characteristic velocity scale is the flow velocity at infinity $U = U_\infty$; the angle θ is reckoned from the forward stagnation point on the sphere surface.

Let us investigate the problem (2.1)–(2.9) and (4.1) by the method of matched asymptotic expansions in small Peclet numbers. Conditions (3.2) are assumed to be fulfilled. Then, similar to [1], the inner and outer regions are determined as $\Omega_1 = \{1 \le r \le 0(\varepsilon^{-1})\}$ and $\Omega_\infty = \{0(\varepsilon^{-1}) \le r\}$ ("contracted" coordinate $\rho = \varepsilon r$).

Similarly to [1] and [12], it can be shown that in the case of translational Stokesian flow around a sphere, equation (4.1), the temperature and concentration distributions over the inner boundary of the outer region Ω_∞ (i.e. at $r \sim \varepsilon^{-1}$) can be presented in the form ($m = 1, \ldots, M$)

$$r = O(\varepsilon^{-1}),\ T = \Psi(P_0, P_1, \ldots, P_M) \times \left\{r^{-1} + \frac{1}{2}P_0(\cos\theta - 1) - \frac{1}{2}P_0^2 \ln P_0 + O(P_0^2)\right\},$$

$$r = O(\varepsilon^{-1}),\ c_m = \Phi_m(P_0, P_1, \ldots, P_M) \times \left\{r^{-1} + \frac{1}{2}P_m(\cos\theta - 1) - \frac{1}{2}P_m^2 \ln P_m + O(P_m^2)\right\}. \tag{4.2}$$

Here, just as in the case of a shear flow (see Section 3), Ψ and Φ_m are unknown functions determined in the course of the problem solution.

In the inner region Ω_1, the temperature and concentration distributions, as well as the temperature field inside a particle, are sought in the form

$$\begin{aligned} T &= T^0 + P_0 T^{(1)} + P_0^2 \ln P_0 T^{(2)} + O(P_0^2), \\ t &= t^0 + P_0 t^{(1)} + P_0^2 \ln P_0 t^{(2)} + O(P_0^2), \\ c_m &= c_m^{(0)} + P_m c_m^{(1)} + P_m^2 \ln P_m c_m^{(2)} + O(P_m^2). \end{aligned} \tag{4.3}$$

Substitution of equation (4.3) into equations (2.1)–(2.3), with account of equation (3.2), and subsequent separation of terms at the same 'powers' of the small parameter ε yield the following equations for the functions $c_m^{(k)}$, $T^{(k)}$ and $t^{(k)}$

$$\begin{aligned} &\Delta c_m^{(k)} = 0,\ \Delta T^{(k)} = 0,\ \Delta t^{(k)} = 0; \\ &\Delta t^{(1)} = 0 \quad (k = 0, 2), \\ &\Delta c_m^{(1)} = \alpha_m \frac{1}{r^2 \sin\theta} \frac{\partial(r^{-1}, \psi)}{\partial(r, \theta)}, \\ &\Delta T^{(1)} = \alpha_0 \frac{1}{r^2 \sin\theta} \frac{\partial(r^{-1}, \psi)}{\partial(r, \theta)} \\ &\qquad (\alpha_m = \text{const};\ m = 0, 1, \ldots, M). \end{aligned} \tag{4.4}$$

In deriving the second group of equations (4.4) (at $k = 1$), it has been taken into account that the zero terms of inner expansion (at $k = 0$) depend only on the radial coordinate r and are determined by the binomial of the form $A + Br^{-1}$.

Now, employing the mean-value operator (3.6), taking into account the equality $\langle \cos\theta \rangle = 0$ and using the same sort of reasoning as with the case of a shear flow, we obtain, for determining the mean Sherwood and Nusselt numbers, the same algebraic system of equations (3.11) and (3.12), where the parameters $Sh_{m\infty}$ and Nu_∞ are determined, according to Acrivos and Taylor [1], as

$$\begin{aligned} Sh_{m\infty} &= 1 + \frac{1}{2}P_m + \frac{1}{2}P_m^2 \ln P_m + O(P_m^2), \\ Nu_\infty &= 1 + \frac{1}{2}P_0 + \frac{1}{2}P_0^2 \ln P_0 + O(P_0^2). \end{aligned} \tag{4.5}$$

It should be noted that at small Peclet numbers, equations (3.2), in the general case of an arbitrary incompressible (both viscous and non-viscous) fluid flow past a sphere, by virtue of the fact that the zero term of the inner expansion is independent of the type of flow and depends on the radial coordinate r alone, the system (3.11) and (3.12) provides a correct result for mean Sherwood and Nusselt numbers at least for the first two terms of the corresponding asymptotic expansion. In this case the parameters $Sh_{m\infty}$ and Nu_∞ correspond to a purely diffusional (thermal) mode of reaction on the sphere surface under the same flow conditions.

In the case of an isothermal reaction, which corresponds to $h_m = 0, m = 1, \ldots, M$, the mean Sherwood numbers are determined from the solution of the system of equations (3.11) at $Nu = 0$. The results reported in [12] can be obtained by solving the first of equations (3.11) and (4.5) at $M = 1, f_1 = kf(c)$.

5. EXAMPLES OF ISOTHERMAL SURFACE REACTIONS

In the solution of specific problems, the system (3.11) and (3.12) can be very conveniently used to analyze the dependence of the mean Sherwood and Nusselt numbers on the reaction kinetics, with the parameters $Sh_{m\infty}$ and Nu_∞ for the shear and translational flows being determined from formula (3.13) and (4.5), respectively.

Now, we shall consider several simple types of chemical reactions occurring on the particle surface.

As is conventional for Frank-Kamenetsky's chemical kinetics [16], the components of the reacting substance will be traditionally denoted by letters A, B, C, ... and so on, while the concentrations of reagents will be labelled by letters instead of numerals ($c_A, c_B, \ldots$ and so on); in the presence of one reacting component in the flow the subscripts on concentration and other parameters are omitted.

Surface reaction of an arbitrary order M = 1

For the κ-order reaction the surface reaction rate, equation (2.9), is determined by the formula

$$F(c^*) = k_0 c^{*\kappa} \quad (\kappa > 0). \tag{5.1}$$

In this case the system (3.11) is reduced to one equation for determining the mean Sherwood number

$$Sh = \frac{ak_0}{D} c_\infty^{\kappa-1} \left(1 - \frac{Sh}{Sh_\infty}\right)^\kappa. \tag{5.2}$$

For a translational flow, the substitution of expression for Sh_∞ (4.5) with subsequent solution of equation (5.2) (by series expansion in small parameter, i.e. Peclet number) for Sh yields the result obtained in [12]. It should be noted, however, that for practical calculations it is more convenient to use directly equations (5.2) and (4.5).

For a shear flow, relation (3.13) should be used in equation (5.2).

Equation (5.2) is easily resolved explicitly at $\kappa = 1/2$, 1 and 2. Thus, for a shear flow, equations (3.1) and (3.13), at $\kappa = 1$ we obtain

$$Sh = g(1 - \alpha g P^{1/2})^{-1} + o(P^{3/2}),$$

$$g = \left(1 + \frac{D}{ak_0}\right)^{-1}. \tag{5.3}$$

At $k_0 \to \infty$ ($g \to 1$), which corresponds to a diffusional mode of reaction, equation (5.3) yields the result obtained by Acrivos [8].

In the case of translational Stokes flow, equations (4.1) and (4.5), at $\kappa = 1$ equation (5.2) provides the first three terms of expansion that have been obtained by Taylor [11] and by Gupalo and Ryazantsev [9].

It follows from equation (5.2) that the mean Sherwood number decreases monotonically with an increase in the reaction order κ (it is assumed that $c_\infty = 1$) and increases monotonically with increase of the rate constant k_0.

Reversible 1st-order reaction (A $\rightleftharpoons$ B)

Consider an isothermal reversible surface 1st-order reaction the rate of which is determined by

$$F_A = -F_B = k_A c_A^* - k_B c_B^*. \tag{5.4}$$

Here k_A and k_B are the reaction rate constants.

Using relations (2.9) and (5.4), from the first two equations of (3.11) we obtain for the mean Sherwood numbers

$$Sh_A = \frac{a}{D_A}\left(k_A - \frac{c_{B\infty}}{c_{A\infty}} k_B\right)\left(1 + \frac{ak_A}{D_A Sh_{A\infty}} + \frac{ak_B}{D_B Sh_{B\infty}}\right)^{-1},$$

$$Sh_B = -(c_{A\infty} D_A / c_{B\infty} D_B) Sh_A. \tag{5.5}$$

Irreversible 2nd-*order reaction* (A + B $\to$ C)

For an irreversible 2nd-order surface reaction occurring at the rate

$$F_A = F_B = k_0 c_A^* c_B^*, \tag{5.6}$$

the system (3.11) yields the following values for the mean Sherwood numbers

$$Sh_A = \frac{1}{2} Sh_{A\infty} \{1 + s + p - [s^2 + 2s(1+p) + (1-p)^2]^{1/2}\}, \tag{5.7}$$

$$Sh_B = \frac{c_{A\infty} D_A}{c_{B\infty} D_B} Sh_A, \quad s = \frac{D_B Sh_{B\infty}}{ak_0 c_{A\infty}}, \quad p = \frac{c_{B\infty} D_B Sh_{B\infty}}{c_{A\infty} D_A Sh_{A\infty}}.$$

Successive stepwise 1st-order reaction $A \xrightarrow[k_A]{} B \xrightarrow[k_B]{} C$

Consider the following reaction occurring on the surface

$$F_A = k_A c_A^*, \quad F_B = -k_A c_A^* + k_B c_B^*, \quad F_C = -k_B c_B^*, \tag{5.8}$$

in which both stages are the 1st-order reactions. We shall assume for simplicity that the concentration of component A far from the particle is equal to unity, while the concentrations of the components B and C are zero, i.e. $c_{A\infty} = 1$, $c_{B\infty} = 0$, $c_{C\infty} = 0$.

By the use of equations (3.14) we obtain the integral mass flux of the final product of reaction C to the particle surface

$$I_C = -\frac{K_A K_B I_{A\infty}}{(K_A + I_{A\infty})(K_B + I_{B\infty})},$$

$$K_{A,B} = 4\pi a^2 k_{A,B}, \left(I_A = \frac{K_A I_{A\infty}}{K_A + I_{A\infty}},\right.$$

$$\left. I_B = -\frac{K_A I_{A\infty} I_{B\infty}}{(K_A + I_{A\infty})(K_B + I_{B\infty})}\right). \tag{5.9}$$

Adsorption occurring by the Langmuir kinetics

When the adsorption rate on the particle surface is determined by the Langmuir isotherm, we have [16]

$$F = k_0 \frac{c^*}{c^* + b} \quad (b = \text{constant}). \tag{5.10}$$

Equations (2.9), (3.11) and (5.10) yield the following expression for the mean Sherwood number on the particle surface

$$Sh = \frac{1}{2} Sh_\infty \left\{d - \left(d - 4\frac{ak_0}{c_\infty D Sh_\infty}\right)^{1/2}\right\},$$

$$d = 1 + \frac{b}{c_\infty} + \frac{ak_0}{c_\infty D Sh_\infty}. \tag{5.11}$$

6. NONISOTHERMAL SURFACE REACTION OCCURRING BY THE ARRHENIUS LAW

Consider now a nonisothermal chemical 1st-order reaction (in the presence of one reacting component in the flow $M = 1$), which occurs by the Arrhenius law on the sphere surface. In this case the surface reaction rate is determined by [16]

$$F = k_0 c^* \exp(-E/RT^*).$$

Here E is the activation energy, R is the universal gas constant, k_0 the reaction rate constant.

The system of equations (3.11) and (3.12) is reduced to the following equation for determination of the mean Sherwood number

$$Sh = \frac{ak_0}{D}\left(1 - \frac{Sh}{Sh_\infty}\right)\exp\left[-\omega\left(1 + h\frac{Sh}{Nu_\infty}\right)^{-1}\right], \tag{6.1}$$

$$\omega = E(RT_\infty)^{-1}, \quad h = -h_1 > 0.$$

To simplify the analysis, equation (6.1) is rewritten in the form of the system

$$q = k\exp(-\omega/z)[1 + k\exp(-\omega/z)]^{-1}, \tag{6.2}$$

$$q = tg\beta(z-1)(z = 1+\sigma q,\ tg\beta = \sigma^{-1}), \tag{6.3}$$

$$q = Sh/Sh_\infty,\ k = ak_0(DSh_\infty)^{-1},\ \sigma = hSh_\infty/Nu_\infty.$$

Assuming now the parameter k to be fixed, let us investigate the number of roots of equation (6.1) depending on variations of the parameters ω, $\sigma \in [0, +\infty]$ (of the angle β, $0 < \beta < \pi/2$). The number of roots of equation (6.1) is determined by the number of intercepts of the straight line (6.3), that passes at the inclination angle β to the axis z on the plane z, q through the point (1,0), with the curve (6.2) (Fig. 1). Depending on the magnitude of the parameter ω the following situations are here possible: (1) at any $\sigma \in [0, \infty]$ the set of equations (6.2) and (6.3) has the single root $q = q(\omega, \sigma)$; (2) there is an interval (σ_1, σ_2) within which at each $\sigma_1 < \sigma < \sigma_2$ the system (6.2) and (6.3) has three roots, while at the end points of the interval $\sigma = \sigma_n (n = 1, 2)$ it has two roots; in this case, for each σ outside this interval $0 \leq \sigma < \sigma_1$ or $\sigma_2 < \sigma$ there corresponds the single root of equation (6.1); $\beta_n = \text{arc } tg\, \sigma_n^{-1}$ (here and henceforth, whenever there are several modes of reaction on the particle surface, the problems of their stability are not investigated).

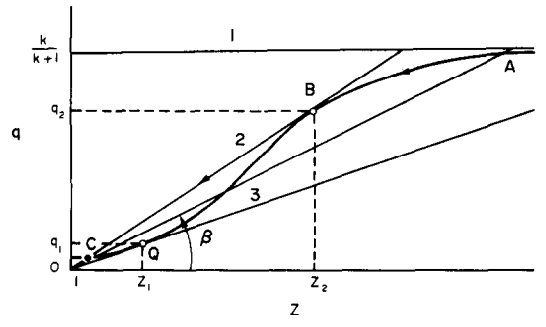

FIG. 1. Qualitative behaviour of the curve (6.2) on the z, q plane; straight line 1 is the asymptote (6.2) at $z \to \infty$. Points B and Q correspond to the points of tangency of the straight line (6.3) with the curve (6.2); the domain of the values of parameter β, for which there are three stationary modes of reaction, is between lines 2 and 3.

It can be shown that the locus of the points of tangency of the straight line (6.3) with the curve (6.2) [which corresponds to two roots of equation (6.1)] on the $\omega\sigma$ plane is prescribed parametrically as

$$\omega(q) = (1-q)y^2S^{-1}(1),\ \sigma(q) = q^{-1}S^{-1}(1),$$

$$y = \ln\{k(q^{-1} - 1)\},$$

$$S(n) = S(q, k, n) = (1-nq)y - n. \tag{6.4}$$

At $\omega \to \infty$ the limiting curve (6.4) has two branches that recede to infinity, with the upper one being asymptotic to the curve

$$\sigma = k^{-1}\omega^{-1}\exp\omega \quad (\omega \to \infty) \tag{6.5}$$

and the lower having the asymptote

$$\sigma = (q_*^{-1} - 1)\omega - 2q_*^{-1} \quad (\omega \to \infty), \tag{6.6}$$

where $q_* < k(k+1)^{-1}$ is the root of the equation $S(q_*, k, 1) = 0$.

The curve (6.4) on the $\omega\sigma$ plane has a sharp peaked singularity (ω_0, σ_0), which is its absolute minimum and is located on the hyperbola

$$\Gamma(\omega, \sigma) = \sigma\omega - 4\sigma - 4 = 0. \tag{6.7}$$

It is specified by the value of the parameter $q = q_0$, where q_0 is the root of the equation $S(q_0, k, 2) = 0$.

The local behaviour of the curve (6.4) in the vicinity of the singular point is prescribed by

$$\sigma - \sigma_0 = tg\gamma(\omega - \omega_0),\ \omega \geqslant \omega_0;\ tg\gamma = \frac{1}{2}q_0^{-1} - 1,$$

$$\omega_0 = \omega(q_0) = 4(1-q_0)(1-2q_0)^{-1},$$

$$\sigma_0 = \sigma(q_0) = q_0^{-1} - 2. \tag{6.8}$$

Figure 2 shows qualitatively the curve (6.4) on the $\omega\sigma$ plane. In the hatched region Ω, located between the

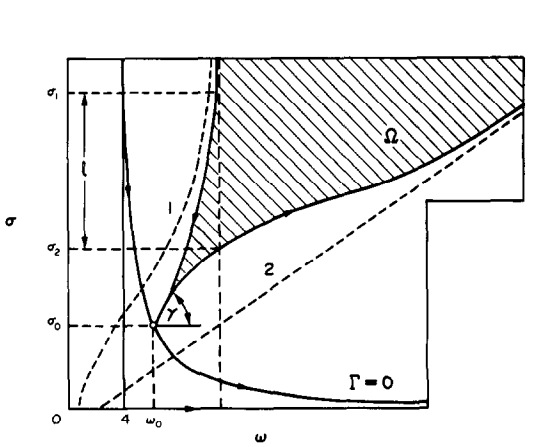

FIG. 2. Qualitative behaviour of the curve (6.4). Hatched region corresponds to those values of the determining parameters ω and σ at which there are three modes of reaction on the particle surface. Curves 1 and 2 correspond to the asymptotes (6.5) and (6.6).

two branches of the curve (6.4) (which are determined by the ranges of the parameter $q: 0 < q < q_0$ and $q_0 < q < q_*$, respectively), at fixed $\omega > \omega_0$ the segment $l(\omega) = \{\sigma_1(\omega) \leq \sigma \leq \sigma_2(\omega)\}$ (solid line in Fig. 2) specifies those values of σ at which equation (6.1) has three roots.

It can be shown that $\partial q_*/\partial k > 0$, $\partial q_0/\partial k > 0$. Therefore, with a change in the parameter k the whole region $\Omega = \Omega(k)$ undergoes deformation due to equations (6.5)–(6.8) as follows: with an increasing parameter k the singular point located on the hyperbola (6.7) moves downward and to the right, with the angle of inclination γ to the axis ω becoming smaller; the asymptotic curve (6.5), which corresponds to the maximum possible values of the parameter σ comes closer to the axis ω in inverse proportion to k, while its point of intersection with the axis ω, still remaining on the right-hand semi-plane, moves to the right of the zero.

In Fig. 2, dashed lines I and II correspond to the asymptotic curves (6.5) and (6.6); the arrows at the boundary of the region show the direction of the points of curve (6.4) into which they move with an increase in the parameter k; the arrows on the hyperbola $\Gamma(\omega, \sigma) = 0$ show the direction of the singular point (ω_0, σ_0) moving with an increase in the parameter k; the arrows on the axis ω show the direction of the points of intersection between the asymptote (6.6) and the axis ω with increasing k, the intersection point coordinate in this case being always above 2.

It is seen from the system of equations (6.2) and (6.3) that the following limiting relations are valid

$$\sigma \to 0,\ q \to (1 + k^{-1} e^{\omega})^{-1};$$

$$\sigma \to \infty,\ q \to k(k+1)^{-1}. \quad (6.9)$$

With decreasing particle or shear flow velocities the mean Sherwood and Nusselt numbers, corresponding to a purely diffusional mode of reaction, equations (3.13) and (4.5), decrease. In this case, as follows from equation (6.1), the point A, corresponding to a high-temperature mode of reaction, moves to the left along the curve (6.2) up to the point B (Fig. 1). On having passed B, the high-temperature mode can no longer exist, hence the point B "jumps over" from the upper part of the curve to the lower one, to the point C, which already corresponds to the low-temperature mode of reaction on the particle surface.

7. INTERPOLATION FORMULA FOR MEAN SHERWOOD NUMBERS AT ARBITRARY PECLET NUMBERS, SOME CORRELATIONS

Comparison with the results [17] obtained for the other limiting case of large Peclet numbers (it should be noted that in the isothermal case at $M = 1$ equation (3.11) coincides, to the accuracy of transformations, with the interpolation equation (40) derived intuitively in [17]), suggests the possibility of using the system (3.11) and (3.12) as interpolation equations for approximate determination of mean Sherwood and Nusselt numbers within the whole range of Peclet numbers $0 \leq P_m < \infty$ (at $\delta \ll 1$) in the presence of not very large number of reagents in the flow and simple reaction kinetics. Then, as the parameters $Sh_{m\infty}$, Nu_∞ entering into the system of equations (3.11) and (3.12), one should use the values of the mean Sherwood and Nusselt numbers obtained by solving an auxiliary, linear and much more simple, problem which corresponds to a purely diffusional mode of reaction on the sphere surface ($m = 0, 1, \ldots, M$)

$$Sh_{m\infty} = -\langle \partial \hat{c}_m/\partial r \rangle|_{r=1},\ Nu_\infty = -\langle \partial \hat{T}/\partial r \rangle|_{r=1},$$

$$\Delta \hat{c}_m = P_m(\mathbf{u}\nabla)\hat{c}_m;\ r \to \infty,\ \hat{c}_m \to 0;$$

$$r = 1,\ \hat{c}_m = 1;\ \hat{c}_0 \equiv \hat{T}. \quad (7.1)$$

At present there is a fairly large number of solutions of the problem (7.1) obtained for different patterns of flow by numerical, analytical and approximate methods.

At large Peclet numbers (in the diffusional boundary layer approximation) for an isothermal reaction of order $\kappa = 1/2, 1, 2$ the testing for applicability of equation (5.2) was carried out for the whole range of the parameter $K = ak_0 D^{-1} c_\infty^{\kappa-1}$ by comparing its root Sh with the accurate results obtained for the Sherwood number by numerical integration of respective integral equations in the case of a translational Stokesian flow around a sphere [18], circular cylinder [19], droplet and bubble [20]. In these cases, the characteristic length scale is expressed in terms of the radii of the sphere, cylinder and droplet, and the characteristic velocity, by that of the flow at infinity U_∞ (for a droplet and bubble the quantity $U_\infty(\beta + 1)^{-1}$ has been taken as the characteristic velocity, where β is the ratio of dynamic viscosities of the droplet and surrounding liquid; $\beta = 0$ corresponds to a gas bubble). The results of comparison between the accurate and approximate values of the Sherwood number show that the maximum deviation of the root of equation (5.2) from the mean Sherwood number [18–20] is observed at $K = 1.5$–5.0 and does not exceed 7–9%.

The adequacy of the approximate equation (5.2) for application at intermediate Peclet and Reynolds numbers in translational flow past a sphere has been checked by comparison with the known results of numerical solution of a respective problem for the 1st-order reaction $\kappa = 1$ on the sphere surface at $P = 10$, 20 and 50. It follows from Table 13 presented in [21] that the use of equation (5.2) yields very good results, with the error not exceeding 1.5%.

The above comparison provides good reason to believe that in the case of isothermal reactions, in order to approximately determine mean Sherwood numbers within the whole range of Peclet numbers, it is a sound plan to use the interpolation system of equations (3.11) and (3.12) provided there is a small number of reagents in the flow and the kinetics of reactions is simple enough (with the only one mode of reaction on the sphere surface).

Some generalizations for the case of an arbitrarily shaped particle

It can be shown that for an arbitrarily shaped particle freely suspended in a shear flow (G_{ij}) in the case of isothermal reaction of the first order $\kappa = 1$ [equation (5.1)] the following formula is valid for the mean Sherwood number

$$\frac{Sh}{Sh_0} = 1 + \alpha Sh_0 P^{1/2} + \alpha^2 Sh_0^2 P + O(P^{3/2}), \quad \alpha = \alpha(G_{ij}). \quad (7.2)$$

Here, the mean Sherwood number Sh_0 corresponds to mass transfer between a reacting particle (at $F = k_0 c^*$) and a stationary medium (i.e. at $P = 0$), while the coefficient α and the Peclet number P are determined according to [7] see also Section 3, equation (3.3)].

Equation (7.2) is derived in the same manner as has been used in [8] to investigate the diffusional mode of reaction ($k_0 \to \infty$) and just as a corresponding extension of work [2] was made in [10] to a translational flow. Equation (7.2) is a natural generalization of the results of work [8]. In the case of a solid sphere, $Sh_0 = g$, equation (7.2) coincides with (5.3) accurate to $O(P^{3/2})$.

Consider now the case of an arbitrary kinetics of surface chemical reaction. For simplicity, let us restrict ourselves to the study of the case of an isothermal reaction with the presence in the flow of one reacting component ($M = 1$); it is also assumed that there is the only one mode of surface reaction within the whole range of governing parameters.

Having chosen the radius of a volumetrically equivalent sphere a_e, for the characteristic particle dimension we obtain the relation

$$\Lambda = Sh^{-1} f(Sh/Sh_\infty). \quad (7.3)$$

By virtue of (3.11), $\Lambda = 1$ for the sphere; in the general case $\Lambda = \Lambda(f, \tau, P)$, where τ is the shape parameter. At $P \to 0$, $\Lambda \to Sh_0^{-1} f(Sh_0/Sh_{0\infty})$, where Sh_0 is the mean Sherwood number for a fixed particle at the prescribed kinetics f, while $Sh_{0\infty}$ is for a fixed particle reacting in a purely diffusional mode. Taking into account this limiting relation and assuming that at small Peclet numbers for an arbitrarily shaped particle, just as for a sphere, one can restrict oneself to the asymptotic expansion of Λ at $P \to 0$ on the RHS of equation (7.3), we obtain the following equation to determine the mean Sherwood number Sh

$$\frac{Sh}{Sh_0} = \frac{f(Sh/Sh_\infty)}{f(Sh_0/Sh_{0\infty})}. \quad (7.4)$$

This equation allows an easy calculation of the Sherwood number for a moving reacting particle once the parameters Sh_∞, Sh_0, $Sh_{0\infty}$ are known. Thus, for the 1st-order reaction ($F = k_0 c^*$) equation (7.4) yields

$$\frac{1}{Sh} = \frac{1}{Sh_0} + \frac{1}{Sh_\infty} - \frac{1}{Sh_{0\infty}}. \quad (7.5)$$

In order to check the above calculation procedure for the convective mass transfer of droplets and arbitrarily shaped particles let us use an analytical solution of the problem for the 1st-order surface reaction obtained in [10] for a translational Stokesian flow by the method of matched asymptotic expansions in small Peclet number

$$Sh = Sh_0 + \frac{1}{2} Sh_0^2 P + \frac{1}{2} \xi Sh_0^2 P^2 \ln P + O(P^2). \quad (7.6)$$

Here ξ is the magnitude of the dimensionless hydrodynamic resistance of particle at prescribed orientation in the flow (it is equal to the dimensional force related to the Stokesian resistance of the sphere of radius a_e, i.e. $6\pi\mu U_\infty a_e$; μ is the dynamic fluid viscosity).

The use of equation (7.6) will obviously yield, accurate to the terms of order P^2, that

$$\frac{1}{Sh} = \frac{1}{Sh_0} - \frac{1}{2} P - \frac{1}{2} \xi P^2 \ln P = \frac{1}{Sh_0} + \frac{1}{Sh_\infty} - \frac{1}{Sh_{0\infty}},$$

which coincides exactly with equation (7.5).

It can be shown in a similar way that equation (7.2) obtained at small Peclet numbers for an arbitrary shear flow past an arbitrarily shaped particle can be expressed in the form of equation (7.5). Bearing this in mind and taking into account that at any f equation (7.4) at $P = 0$ leads to an accurate result for the particle of any shape, while for a spherical particle it gives several first terms of asymptotic expansion for the mean Sherwood number at small Peclet numbers and can be successfully used also for approximate determination of Sh at any P's ($0 \le P < \infty$), it is thus to be expected that equation (7.4) can also be applied for approximate determination of the mean Sherwood number on an arbitrarily shaped particle surface within the whole range of Peclet numbers at simple enough kinetics of surface reaction.

It should be noted that equation (7.4) can be written in the form of equation (3.11) on having chosen $a_e[\Lambda]^{-1}_{P=0}$ as the characteristic length scale.

General boundary conditions

In some problems on heat and mass transfer between particles (droplets) and the flow (2.1)–(2.9) one can encounter other boundary conditions than those given by equations (2.6) and (2.7). Thus, for vapour adsorption on the liquid droplet surface, being in the saturation state, the following boundary condition is used: $r = 1, c^* = dT^* + b$ (d and b = constants) [22].

It can be shown that at small Peclet numbers equation (3.2), in the general case of the boundary conditions on the particle surface ($m = 1, 2, \ldots, M + 1$)

$$r = 1, f_m\left(T, t, c_1, \ldots, c_M, \frac{\partial T}{\partial r}, \frac{\partial t}{\partial r}, \frac{\partial c_1}{\partial r}, \ldots, \frac{\partial c_M}{\partial r}\right) = 0 \quad (7.7)$$

the mean Sherwood and Nusselt numbers in the problem (2.1)–(2.5), (2.8) and (2.9) are determined from solution of the system

$$f_m\left(\frac{Nu}{Nu_\infty}, \frac{Nu}{Nu_\infty}, \frac{Sh_1}{Sh_{1\infty}}, \ldots, \frac{Sh_M}{Sh_{M\infty}}, -Nu, O, -Sh_1, \ldots, -Sh_M\right) = 0, \quad (7.8)$$

where the parameters Nu_∞ and $Sh_{m\infty}$ for the shear and translational Stokesian flow past a sphere are prescribed by equations (3.13) and (4.5), respectively.

The system (7.8) is also valid for a purely external problem (2.1), (2.2), (2.4), (7.7) and (2.9), in which the functions f_m are independent of t and $\partial t/\partial r$. In this case, for a translational Stokesian flow past a spherical droplet the governing parameters occurring in the system (7.8) [or in (3.11) and (3.12)] are of the form [2]

$$Sh_{m\infty} = 1 + \frac{1}{2}P_m + \frac{1}{6}\frac{2+3\beta}{1+\beta}P_m^2 \ln P_m + O(P_m^2),$$

$$Nu_\infty = 1 + \frac{1}{2}P_0 + \frac{1}{6}\frac{2+3\beta}{1+\beta}P_0^2 \ln P_0 + O(P_0)^2.$$

The inverse problem

The results obtained make it possible to study the inverse problem of mass transfer of reacting particles in the flow and allow determination of the surface chemical reaction rate from the known integral fluxes. Thus, in the case of an isothermal reaction at $M = 1$ the experimental data on Sh at various Pe numbers (i.e. at various velocities of flow past the particle, U_∞) can be used to construct, by equation (3.11), the points with the coordinates $f = Sh$ and $x = Sh/Sh_\infty$ (experimental values of Sh_∞ are well known see, e.g. [21]), that determine the relation $f = f(x)$ which is sought for.

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REACTION CHIMIQUE NON ISOTHERME A LA SURFACE DE LA PARTICULE DANS UN ECOULEMENT LAMINAIRE

Résumé—On considère la convection stationnaire pour une sphère conductrice de la chaleur en réaction dans un écoulement laminaire, parallèle, avec une réaction chimique non isotherme sur sa surface avec une vitesse dépendant arbitrairement de la température et des concentrations.

On montre que pour les flux globaux de chaleur et de masse des composants (nombres moyens de Nusselt Nu et de Sherwood Sh_m) à déterminer pour les petits nombres de Péclet, il suffit de résoudre le système d'équations algébriques universel (transcendant):

$$Sh_m = f_m\left(\frac{Nu}{Nu_\infty}, \frac{Sh_1}{Sh_{1\infty}}, \ldots, \frac{Sh_M}{Sh_{M\infty}}\right),$$

$$Nu = \sum_{m=1}^{M} h_m Sh_m,$$

qui est notablement plus simple que le système initial d'équations aux dérivées partielles. Le système suggéré permet d'obtenir pour les nombres de Sherwood et de Nusselt les quatre premiers (écoulement à cisaillement) et les trois premiers termes (écoulement de transition) du développement asymptotique en petits nombres de Péclet.

On étudie une réaction irréversible, isotherme du seconde ordre et une réaction du premier ordre à échelons successifs. On a considéré en détail le cas d'une réaction en surface non isotherme qui suit la loi d'Arrhenius avec un composant réactant présent dans l'écoulement. On analyse l'adsorption sur la surface de la particule, gouvernée par la cinétique de Langmuir.

En comparaison avec des résultats antérieurs pour des réactions isothermes à des nombres de Péclet modérés ou grands, on montre qu'avec un choix judicieux des paramètres $Sh_{m\infty}$ (Nu_∞), le système algébrique proposé peut être appliqué avec succès à la détermination approchée des nombres de Sherwood moyens dans tout le domaine des nombres de Péclet.

Les résultats obtenus permettent l'étude du problème inverse de transfert de chaleur et de masse pour des particules en réaction dans l'écoulement, permettent par exemple de déterminer explicitement la dépendance de la vitesse de réaction en surface aux concentrations à partir des flux globaux que l'on peut déterminer expérimentalement.

NICHTISOTHERME CHEMISCHE REAKTION AN EINER TEILCHENOBERFLÄCHE BEI LAMINARER STRÖMUNG

Zusammenfassung—Der Bericht befaßt sich mit der stationären konvektiven. Diffusion an eine wärmeleitende, chemisch reagierende Kugel in einer laminaren Translations- und Scherströmung bei nichtisothermer chemischer Reaktion an ihrer Oberfläche, deren Geschwindigkeit von Temperatur und Konzentration abhängt. Es wird gezeigt, daß es für die integralen Wärme- und Massenströme der reagierenden Komponenten an ein Teilchen (d. h. mittlere Nusselt-Zahl Nu und mittlere Sherwood-Zahl Sh_m), die für kleine Peclet-Zahlen bestimmt werden sollen, genügt, das folgende universelle algebraische (transzendente) Differentialgleichungssystem zu lösen

$$Sh_m = f_m\left(\frac{Nu}{Nu_\infty}, \frac{Sh_1}{Sh_{1\infty}}, \ldots, \frac{Sh_m}{Sh_{m\infty}}\right),$$

$$Nu = \sum_{m=1}^{m} h_m Sh_m,$$

das wesentlich einfacher ist als das ursprüngliche System partieller Differentialgleichungen. Das vorgeschlagene Gleichungssystem ermöglicht es, für die Sherwood-Zahl und die Nusselt-Zahl die ersten vier Terme (Scherströmung) und die ersten drei Terme (Längsströmung) der entsprechenden asymptotischen Reihenentwicklung bei kleinen Peclet-Zahlen zu bestimmen. Es wurden eine irreversible isotherme Reaktion zweiter Ordnung und eine sukzessive, schrittweise Reaktion erster Ordnung untersucht. Der Fall der nichtisothermen Reaktion an der Oberfläche, die nach dem Arrhenius-Gesetz abläuft, mit einer reagierenden Komponente in der Strömung, wurde im Detail analysiert. Die Adsorption an der Teilchenoberfläche, deren Geschwindigkeit von der Langmuir-Kinetik bestimmt wird, wird untersucht. Auf der Grundlage eines Vergleichs mit früheren Ergebnissen für isotherme Reaktionen bei mittleren und großen Peclet-Zahlen wird gezeigt, daß mit einer geeigneten Vorgabe der bestimmenden Parameter $Sh_{m\infty}$ (Nu_∞) das vorgeschlagene algebraische Gleichungssystem auch erfolgreich für die näherungsweise Bestimmung der mittleren Sherwood-Zahlen innerhalb des genannten Peclet-Zahlenbereichs angewandt werden kann. Die erhaltenen Ergebnisse ermöglichen die Untersuchung der inversen Wärme- und Stoffübergangsprobleme von chemisch reagierenden Teilchen in einer Strömung, d.h. sie ermöglichen es, anhand vorhandener integraler Ströme, die experimentell bestimmt werden können, die Abhängigkeit der Konzentrationen von der Oberflächenreaktions-Geschwindigkeit explizit zu bestimmen.

О НЕИЗОТЕРМИЧЕСКОЙ ХИМИЧЕСКОЙ РЕАКЦИИ НА ПОВЕРХНОСТИ ЧАСТИЦЫ В ЛАМИНАРНОМ ПОТОКЕ

Аннотация — Рассматривается стационарная конвективная диффузия к теплопроводной реагирующей сфере, обтекаемой ламинарным поступательным и сдвиговым потоком при протекании на ее поверхности неизотермической химической реакции, скорость которой произвольным образом зависит от температуры и концентраций.

Показано, что при малых числах Пекле для определения интегральных притоков тепла и вещества реагирующих компонент к частице (т. е. для определения средних чисел Нуссельта Nu и Шервуда Sh_m) достаточно решить следующую универсальную алгебраическую (трансцендентную) систему

$$Sh_m = f_m\left(\frac{Nu}{Nu_\infty}, \frac{Sh_1}{Sh_{1\infty}}, \ldots, \frac{Sh_m}{Sh_{m\infty}}\right),$$

$$Nu = \sum_{m=1}^{m} h_m Sh_m,$$

которая существенно проще исходной системы уравнений в частных производных. Предлагаемая система позволяет получить в случае сдвигового потока четыре, а в случае поступательного — три первых члена соответствующего асимптотического разложения по малым числам Пекле для чисел Шервуда и Нуссельта.

Исследована необратимая изотермическая реакция второго порядка и последовательная ступенчатая реакция первого порядка. Подробно рассмотрен случай неизотермической поверхностной химической реакции, протекающей по закону Арениуса при наличии в потоке одной реагирующей компоненты. Анализируется адсорбция на поверхности частицы, скорость которой определяется лангмюровской кинетикой.

На основе сопоставления с ранее известными результатами для изотермических реакций при умеренных и больших числах Пекле показано, что при соответствующем выборе определяющих параметров $Sh_{m\infty}(Nu_\infty)$, полученная алгебраическая система может быть с успехом использована также для приближенного определения средних чисел Шервуда во всем диапазоне чисел Пекле.

Полученные результаты дают возможность исследовать обратную задачу о массотеплообмене реагирующих частиц в потоке — т. е. позволяют в явном виде определить зависимость скорости поверхностной химической реакции от концентраций по известным интегральным потокам, которые могут быть определены из эксперимента.