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CONVECTIVE DIFFUSION TO A PARTICLE IN A FLUID WITH LINEAR KINETICS

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The concentration field in the neighborhood of a solid sphere in a Stokes stream at high Péclet numbers is determined by the method of joining asymptotic solutions. A chemical reaction, whose rate is arbitrarily dependent on the concentration of the diffusing matter close to the surface, takes place at the sphere surface. Dependence of the total diffusing flux at the sphere surface on the chemical reaction rate is determined. The phenomenon of diffusion flux saturation with increasing Péclet number is present, as in the case of first order chemical reaction [1]. Modes of the chemical reaction at the sphere surface are investigated. The distribution of concentration in the diffusion wake region is determined and its structure analyzed. The problem was solved in [1] for the case of linear kinetics.

Convective diffusion at the surface of a reacting particle in a homogeneous stream of viscous fluid was investigated in a number of publications, for instance [2, 3], in which total absorption of reagent by the particle surface and, also, first order heterogeneous chemical reaction were considered [1, 4]. It is interesting to investigate the diffusion of reagent at a moving particle on whose surface a chemical reaction takes place, when the dependence of the reaction rate of reagent close to the surface is more complex or, generally speaking, arbitrary. Problems of this kind appear, for instance, in investigations of reagent diffusion to the particle under conditions in which reaction at the particle surface conforms to the Langmuir kinetics and average coverage of the surface (see, e.g., [5]). Examples of reactions of total order are widely known.

1. Statement of the problem. Concentration distribution in the diffusion boundary layer. Convective diffusion of matter to a solid sphere in Stokes stream of viscous incompressible fluid whose velocity away from the sphere is U is considered. In the spherical system of coordinates r, θ attached to the particle the dimensionless equation of steady convective diffusion and the boundary conditions are of the form

$$\frac{1}{\sin\theta}\left(\frac{\partial\psi}{\partial\theta}\frac{\partial c}{\partial r}-\frac{\partial\psi}{\partial r}\frac{\partial c}{\partial\theta}\right)=\varepsilon^3\left[\frac{\partial}{\partial r}\left(r^2\frac{\partial c}{\partial r}\right)+\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial c}{\partial\theta}\right)\right] \qquad (1.1)$$

$$\left[\frac{\partial c}{\partial r}-kf^*(c)\right]_{r=1}=0, \quad c|_{r\to\infty}\to 1$$

$$[\partial c/\partial\theta]_{\theta=0}=[\partial c/\partial\theta]_{\theta=\pi}=0, \quad \varepsilon^{-3}=P=aU/D$$

$$k=k'a/D, \quad f^*(0)=0$$

where c is the concentration of matter, ψ is the stream function, P is the Péclet number, a is the sphere radius, D is the diffusion coefficient, k' is the constant of the reaction rate, $f^*(c)$ is the dependence of chemical reaction rate on reagent concentration close to the surface, and angle θ is measured relative to the direction of the stream. The sphere radius, stream velocity, and concentration at infinity are used as units in Eq. (1.1).

The dimensionless stream function of the Stokes flow past the particle is of the form

$$\psi = \left(r^2 - \frac{3}{2}r + \frac{1}{2r}\right)\frac{\sin^2\theta}{2} \tag{1.2}$$

Below, the Péclet number is assumed high, i.e. $\varepsilon \ll 1$, as is usual for liquids. An asymptotic analysis of problem (1.1), (1.2) for $\varepsilon \to 0$ was carried out in [1] in the case of linear kinetics $(f^*(c) = c)$.

Several characteristic regions with different mass transfer mechanisms can be distinguished in the particle neighborhood when $\varepsilon \ll 1$ [6]. These are: the outer region e, the region of the leading point b, the diffusion boundary layer d, and the region of the diffusion wake which, in turn, consists of subregions W^i $(i = 1, 2, 3, 4)$. In each of these regions Eq. (1.1) is approximately adjusted by separating principal terms of expansion in the small parameter ε. Agreement between solutions in individual regions is obtained by asymptotic merging at their nominal boundaries.

The predominant part in the transfer of a dissolved constituent to the particle surface is played by the convective diffusion process in the diffusion boundary layer $d = \{r - 1 < O(\varepsilon),\ O(\varepsilon) < \theta\}$ which consists of convection along the sphere surface and diffusion in the transverse direction.

Substituting variables

$$\xi = \varepsilon^{-1}\psi^{1/2}, \quad t = T(\theta) = {}^1/_8\sqrt{3}\,[\pi - \theta + {}^1/_2 \sin 2\theta]$$

and retaining the principal terms of expansion in parameter ε, from formulas (1.1) and (1.2) we obtain

$$\left(\frac{\partial}{\partial t} - \xi^{-1}\frac{\partial^2}{\partial \xi^2}\right)c^{(d)} = 0 \quad (0 < t \leqslant t_0) \tag{1.3}$$

$$[\eta(t)\,\partial c^{(d)}/\partial\xi - \varepsilon k f^*(c^{(d)})]_{\xi=0} = 0, \quad c^{(d)}|_{\xi\to\infty} \to 1$$

$$c^{(d)}|_{t=0} = 1, \quad t_0 = t(0) = \sqrt{3}\pi/8$$

$$\eta(t) = {}^1/_2\sqrt{3}\sin T^\circ(t), \quad t \equiv T[T^\circ(t)]$$

Solution of the equation of the diffusion boundary layer (1.3) under condition of total absorption of matter at the sphere surface $(k = \infty)$ was obtained in [2]

$$c_*^{(d)}(\xi, t) = \Gamma^{-1}({}^1/_3)\,\gamma({}^1/_3, \xi^3/9t) \tag{1.4}$$

$$\gamma\left(\frac{1}{3}, x\right) = \int_0^x e^{-\tau}\tau^{-2/3}d\tau, \quad \Gamma\left(\frac{1}{3}\right) = \gamma\left(\frac{1}{3}, +\infty\right)$$

As in [1], we carry out the substitution $z = {}^2/_3\,\xi^{3/2}$ and seek the solution of problem (1.3) in the form $c^{(d)} = c_*^{(d)} + u$. For the unknown function u we obtain the equation

$$\frac{\partial u}{\partial t}=\frac{\partial^2 u}{\partial z^2}+\frac{1}{3z}\frac{\partial u}{\partial z} \tag{1.5}$$

$$\left[-z^{1/3}\frac{\partial u}{\partial z}+\left(\frac{2}{3}\right)^{1/3}\frac{k\varepsilon}{\eta(t)}f^*(+u)-\frac{2^{1/3}}{\Gamma(1/3)t^{1/3}}\right]_{z=0}=0$$

$$u\,|_{z\to\infty}\to 0,\quad u\,|_{t=0}=0$$

We seek for it a solution of the form

$$u(z,t)=\frac{2^{-1/3}}{\Gamma(2/3)}\int_0^t \Phi(\lambda)(t-\lambda)^{-2/3}\exp(-\zeta^2)\,d\lambda \tag{1.6}$$

$$\zeta={}^1/_2\, z\,(t-\lambda)^{-1/2}\quad (0<t\leqslant t_0)$$

Function (1.6) satisfies the equation and the last two boundary conditions (1.5) for any kernel $\Phi(t)$ and in the interval $0<t\leqslant t_0$ has the following properties:

$$\lim_{z\to 0} u=\frac{2^{-1/3}}{\Gamma(2/3)}\int_0^t \Phi(\lambda)(t-\lambda)^{-2/3}d\lambda \tag{1.7}$$

$$\lim_{z\to 0}(z^{1/3}\partial u/\partial z)=-\Phi(t)$$

The first of boundary conditions (1.5) and properties (1.7) imply that function $\Phi(x)$ is a solution of the integral equation

$$\eta(x)\,\Phi(x)+K^* f(\mathbf{L}_*\Phi)-\alpha\mu(x)=0 \tag{1.8}$$

$$\mathbf{L}_*\Phi=\int_0^x \Phi(\lambda)(x-\lambda)^{-2/3}d\lambda$$

where the following notation is used:

$$\mu(x)=\eta(x)x^{-1/3},\quad K^*=3^{-1/3}\Gamma^{-1}(2/3)k\varepsilon,\quad \alpha=2^{1/3}\Gamma^{-1}(1/3) \tag{1.9}$$

$$f(x)\equiv 2^{1/3}\Gamma(2/3)f^*(2^{-1/3}\Gamma^{-1}(2/3)x)$$

Function $\eta(x)$ in Eq. (1.8) has the following properties:

$$x\to 0,\quad \eta(x)\to 3^{2/3}2^{-1/3}x^{1/3}$$

$$x\to t_0,\quad \eta(x)\to 3^{2/3}2^{-1/3}(t_0-x)^{1/3}$$

and in the neighborhood of point $x=0$ may be presented in the form of series

$$\eta(x)=\sum_{n=0}^{\infty}a_n x^{(2n+1)/3};\quad a_0=3^{2/3}2^{-1/3},\quad a_1={}^1/_5,\ldots$$

If function $f(x)$ is continuous, then for $x\to 0$ we have $\Phi(x)\to b_0x^{-1/3}+o(x^{-1/3})$, where b_0 is the root of equation (B is the beta function)

$$H(b_0)=b_0a_0+K^*f(b_0B_0)-\alpha a_0=0,\ B_n=B({}^1/_3,{}^2/_3(n+1)) \tag{1.10}$$

We assume that at point $x_0=b_0B_0$ function $f(x)$ expands in series

$$f(b_0B_0+\tau)=f^\circ+\lambda_1\tau+\sum_{k=2}^{\infty}\lambda_k\tau^k,\quad f^\circ=f(b_0B_0)$$

that is convergent in some interval.

We introduce the following notation:

$$\Omega\,[\eta\,(x),\,\lambda_k,\,p,\,y,\,\mathbf{A}\,(y),\,\mu\,(x)] \equiv \eta\,(x)\,y + K^* p\mathbf{A}\,(y) + F\,(\mu\,(x),\,\lambda_k,\,\mathbf{A}\,(y)) \tag{1.11}$$

$$F\,(\mu\,(x),\,\lambda_k,\,\mathbf{A}\,(y)) = K^* f^\circ\,(1 - a_0^{-1}\mu\,(x)) + K^* \sum_{k=2}^{\infty} \lambda_k\,[\mathbf{A}\,(y)]^k$$

where $\mathbf{A}\,(y)$ is some, so far unspecified linear operator. Equation (1.8) may be written in the form

$$\Omega\,[\eta\,(x),\,\lambda_k,\,\lambda_1,\,y,\,\mathbf{L} * y,\,\mu\,(x)] = 0 \tag{1.12}$$
$$y = y\,(x) = \Phi\,(x) - b_0 x^{-1/3}$$

Its solution is then sought in the form of series

$$y = \sum_{n=1}^{\infty} b_n x^{(2n+1)\,/\,3} \tag{1.13}$$

Substituting (1.13) into Eq. (1.12) and equating coefficients at like powers of x, we obtain for the determination of coefficients b_n the recurrent system

$$b_n = -\,[a_0 + K^*\lambda_1 B_n]^{-1} \left\{ \sum_{k=1}^{n} a_k b_{n-k} + F_n\,(\mu,\,\lambda_k,\,\mathbf{L}{*}y) \right\} \tag{1.14}$$

where $F_n\,(\mu,\,\lambda_k,\,\mathbf{L} * y)$ depends on $b_1,\,b_2,\,\ldots,\,b_{n-1}$ and is determined as the coefficient at $x^{(2n+1)/3}$ in the expansion of $F\,(\mu,\,\lambda_k,\,\mathbf{L} * y)$ in series, after the substitution into it of (1.13), i.e.

$$F\,(\mu,\,\lambda_k,\,\mathbf{L}{*}y) = \sum_{n=1}^{\infty} F_n x^{(2n+1)\,/\,3}$$

Series (1.13), (1.4) is convergent is some neighborhood of zero.

To prove this we consider the series with coefficients

$$b_n{}^* = a_0^{-1} \left\{ \sum_{k=1}^{n} |\,a_k\,|\, b_{n-k}^* - F_n\,(\mu^*,\,\lambda_k,\,\mathbf{L}{*}y^*) \right\} \tag{1.15}$$
$$b_0{}^* = b_0$$

The terms of this series are determined as the formal solution of the nonlinear integral equation

$$\Omega\,[\eta^*\,(x),\,\lambda_k,\,0,\,y^*,\,\mathbf{L}{*}y^*,\,\mu^*\,(x)] = 0 \tag{1.16}$$
$$y^* = \sum_{n=1}^{\infty} b_n{}^* x^{(2n+1)\,/\,3}, \quad \mu^*\,(x) = \sum_{n=0}^{\infty} |\,a_n\,|\, x^{2n\,/\,3}$$
$$\eta^*\,(x) = a_0\,x^{-1/3} - \sum_{n=1}^{\infty} |\,a_n\,|\, x^{(2n+1)\,/\,3}$$

Let us consider together with (1.16) the series

$$b_n{}^\circ = a_0^{-1} \left\{ \sum_{k=1}^{n} |\,a_k\,|\, b_{n-k}^\circ - F_n\,(\mu^*,\,-\,|\,\lambda_k\,|,\,B_0 y^\circ) \right\}, \quad b_0{}^\circ = b_0 \tag{1.17}$$

where $B_0 y \equiv B_0 \mathrm{E}(y)$, E is an identic operator, $B_0 = B(^1/_3,\,^2/_3)$; the coefficients of series (1.17) can be obtained as the formal solution of equation

$$\Omega\ [\eta^*(x), -|\lambda_k|, 0, y^\circ, B_0 y^\circ, \mu^*(x)] = 0 \qquad (1.18)$$

$$y^\circ = \sum_{n=1}^{\infty} b_n^\circ x^{(2n+1)/3}$$

which unlike (1.12) and (1.16) is algebraic with respect to the variable y°. Introduction of the new variable $\tau = x^{1/3}$ yields $[\partial\Omega / \partial\tau]_{\tau=0,\, y=0} \neq 0$ and by the Cauchy theorem there exists some neighborhood of point $x = 0$ at which series (1.17) is convergent.

Let $f(x) \geqslant 0$ for $x \geqslant 0$ and $f'(x) \geqslant 0$ for $-\infty < x < \infty$, then Eq. (1.10) has a single positive root $b_0 \geqslant 0$. Hence all coefficients of series (1.17), (1.18) are positive. Noting that $B_n \leqslant B_0$ when $n \geqslant 1$ and $\lambda_1 \geqslant 0$, we obtain by induction that $b_n \leqslant b_n^\circ$. Consequently series (1.17), (1.18) majorates (1.13), (1.14) and the latter is convergent.

The complete solution in the diffusion boundary layer may be written, using function Φ, in the form

$$c^{(d)}(\xi, t) = \Gamma^{-1}\left(\frac{1}{3}\right)\gamma\left(\frac{1}{3}, \frac{\xi^3}{9t}\right) + \qquad (1.19)$$

$$\frac{1}{2^{1/3}\Gamma(2/3)}\int_0^t \Phi(\lambda)(t-\lambda)^{-2/3}\exp\left[-\frac{\xi^3}{9(t-\lambda)}\right]d\lambda$$

2. **Distribution of concentration in the diffusion wake.** Region of the diffusion wake whose boundary corresponds to $\theta \sim \varepsilon$ contributes relatively little, $\sim \varepsilon$, to the over-all diffusion flux to the particle surface. The concentration field in the wake has, however, a significant effect on the particle mass transfer in particles moving in the wake of the first [8, 9].

For convenience we introduce in the diffusion wake region W the supplementary condition (of symmetry) $[\partial c / \partial\theta]_{\theta=0} = 0$ which in this case is equivalent to the condition of concentration boundedness.

The estimate of individual terms of Eq. (1.1), (1.2) in the boundary layer convective region $W^{(1)} = \{O(\varepsilon) < r - 1,\ O(\varepsilon^3) < \psi < O(\varepsilon^2)\}$ of the wake shows that there the right-hand side of the equation can be neglected. Hence concentration there depends only on the stream function and is constant along streamlines and equal to that at the exit from the diffusion boundary layer. Formula for the concentration in $W^{(1)}$ is obtained by joining with solution (1.19) and is of the form

$$c^{(1)}(\xi) = c^{(d)}(\xi, t(\theta))\,|_{\theta\to 0,\ \xi=\mathrm{const}} = c^{(d)}(\xi, t_0) \qquad (2.1)$$

In the diffusion wake inner region $W^{(2)} = \{O(\varepsilon) < r - 1 < O(\varepsilon^{-1}), \psi < O(\varepsilon^3)\}$ the radial transfer is insignificant. Equations and boundary conditions for $\zeta = 0$ and $s \to \infty$ ($\zeta = \varepsilon^{-3}\psi$) coincide with those obtained in [1] for the case of linear kinetics. The condition of joining with the solution in the convective boundary layer region $W^{(1)}$ (for $\zeta \to \infty$) implies that $c^{(2)} \sim \sqrt{\varepsilon}$. Consequently for the boundary condition for $y \to 0$ and $\theta = \mathrm{const}$ in the case of a reaction of order $\varkappa$ we obtain

$$[\partial v / \partial y] - \lambda_0 v^\varkappa = 0, \quad \lambda_0 = k\varepsilon^{(\varkappa-1)/2}, \quad v = \varepsilon^{-1/2}c^{(2)} \qquad (2.2)$$

Since $\varepsilon \ll 1$ some simplifications are possible:

1) if $k \gg \varepsilon^{(1-\varkappa)/2}$ $(\lambda_0 \gg 1)$, the first term in the boundary condition (2.2) can be neglected from which for $c^{(2)}$ we have

$$c^{(2)}(y=0)=0 \tag{2.3}$$

2) when $k \ll \varepsilon^{(1-\varkappa)/2}$ $(\lambda_0 \ll 1)$, the second term in (2.2) is immaterial, and for the concentration we have

$$[\partial c^{(2)} / \partial y]_{y=0,\ \theta=\mathrm{const}} = 0 \tag{2.4}$$

If $k\varepsilon^{(\varkappa-1)/2} \sim 1$ it is necessary to take into account both terms in formula (2.2). The most interesting case of the diffusion boundary layer is when $k\varepsilon \sim 1$. Then for $\varkappa < 3$ boundary condition (2.3) is valid for $c^{(2)}$, and for $\varkappa > 3$ it is condition (2.4). For $\varkappa = 3$ the problem of concentration in the inner zone must be solved with the total boundary condition (2.2).

Below we restrict our investigation to the case of $0 < \varkappa < 3$.

The solution for concentration distribution in $W^{(2)}$ is of the form [1]

$$c^{(2)} = (2\varepsilon)^{1/2}\,\Gamma(^3/_2)\,Ay^{1/2}\Phi(-^1/_2, 1, -\zeta/(2y)) \tag{2.5}$$

$$\Phi(a,c,x) = 1 + \sum_{k=1}^{\infty} \frac{a(a+1)\ldots(a+k-1)}{c(c+1)\ldots(c+k-1)}\frac{x^k}{k!}$$

where $\Phi(a, c, x)$ is a degenerate hypergeometric function.

The region of the trailing stagnation point $W^{(3)} = \{\theta < O(\varepsilon),\ r-1 < O(\varepsilon)\}$ in which radial and tangential transfer takes place is not considered here. We would only point out that the contribution of $W^{(3)}$ to the total diffusion flux on the sphere is of order ε. A similar problem of total absorption of the dissolved matter on the particle surface $(k = \infty)$ was investigated in [6, 10] by numerical methods.

Diffusion along streamlines in the mixing region $W^{(4)} = \{O(\varepsilon^{-1}) < r,\ \psi < O(\varepsilon)\}$ can be neglected. Omitting intermediate results, which are obtained similarly to [1], we present the final formula for concentration distribution in that region

$$c^{(4)}(\xi,\rho) = \int_0^\infty \frac{\xi^*}{2\rho}\exp\left\{-\frac{\xi^2+\xi^{*2}}{4\rho}\right\} I_0\left(\frac{\xi\xi^*}{2\rho}\right) c^{(1)}(\xi^*)\,d\xi^* \tag{2.6}$$

where I_0 is a modified Bessel function and $c^{(1)}(\xi)$ is defined by formula (2.1).

Formulas for concentration distribution in regions $W^{(2)}$ and $W^{(3)}$ of the diffusion wake show that the distribution in these differs from that in the case of total absorption $(k = \infty)$ only by the proportionality coefficient A [1] which contains the additional term with $\Phi(t_0)$, and this results in an increase of concentration in these regions in compraison with the limit case of $k = \infty$.

3. **Diffusion flux on the sphere surface.** Using the integral equation (1.8) for function Φ, we obtain for the diffusion flux j on the sphere surface the equation

$$j(t) = [\partial c^{(d)} / \partial r]_{r=1} = \varepsilon^{-1}\eta(t)\,[\partial(c_*^{(d)} + u)/\partial\xi]_{\xi=0}$$

From formula (1.4) for c_*^{d} with allowance for properties (1.7) we obtain the following relation between functions $\Phi(t)$ and $j(t)$:

$$\Phi(t) = \frac{2^{1/3}}{\Gamma(1/3)\, t^{1/3}} - \varepsilon\left(\frac{2}{3}\right)^{1/3} \eta^{-1}(t)\, j(t) \tag{3.1}$$

Substituting this expression for $\Phi(t)$ into Eq. (1.8) we obtain the nonlinear integral equation for the local diffusion flux j on the sphere surface

$$j(t) = \frac{k}{2^{1/3}\Gamma(2/3)} f\left(\frac{2^{1/3}B_0}{\Gamma(1/3)} - \varepsilon\left(\frac{2}{3}\right)^{1/3} G{*}j\right) \tag{3.2}$$

$$G{*}j = \int_0^t j(\lambda)\, \eta^{-1}(\lambda)\, (t-\lambda)^{-2/3}\, d\lambda$$

Let us investigate two limit cases: 1) $k\varepsilon \gg 1$ and 2) $k\varepsilon \ll 1$.

For the local flux j the first case corresponds to a fixed ε and $k \to \infty$ while the second relates to fixed k and $\varepsilon \to 0$.

In the first case, because of condition $f(0) = 0$, we obtain in the zero approximation the equation

$$G{*}j^{\circ} = \frac{3^{1/3}B_0}{\varepsilon\Gamma(1/3)} \quad (k \to \infty,\ \varepsilon = \text{const})$$

whose solution is of the form

$$j^{\circ} = \varepsilon^{-1}\frac{3^{1/3}}{\Gamma(1/3)}\, \eta(t)\, t^{-1/3} \tag{3.3}$$

which corresponds to the limit diffusion flux that is determined by the concentration distribution $c_*^{(d)}$ (1.4).

When function $f(x)$ corresponds to a reaction of order $\varkappa$, i.e.

$$f(x) = x^{\varkappa}, \quad \varkappa > 0 \tag{3.4}$$

the next approximation with respect to parameter k is determined by the Abel equation

$$G{*}j^1 = -\sigma\,[j^{\circ}]^{1/\varkappa}, \quad \sigma = \varepsilon^{-1}3^{1/3}\Gamma(2/3)\, k^{-1/\varkappa} \tag{3.5}$$

from which in conformity with [11] we obtain with an accuracy to $O(k^{-2/\varkappa})$ the formula for the diffusion flux

$$j = j^{\circ} - k^{-1/\varkappa}\frac{\sqrt{3}\sigma}{2\pi}\, \eta(t)\frac{d}{dt}\int_0^t [j^{\circ}(\lambda)]^{1/\varkappa}(t-\lambda)^{-1/3}\, d\lambda, \quad k \to \infty \tag{3.6}$$

where $j = j^{\circ}(t)$ is determined by formula (3.3).

Formula (3.6) shows that the diffusion flux increases with increasing k, while the increase of index $\varkappa$ in the "reaction law" results in its decrease.

Determination of the integral in (3.6) yields for the total flux the formula

$$I = I_0\,[1 - C(\varkappa)\, K^{*-1/\varkappa}] \tag{3.7}$$

where I_0 is the total flux for $k = \infty$. In the case of linear kinetics $(\varkappa = 1)$ we have $C(1) \approx 0.46$ [1].

In the second limit case $(\varepsilon \to 0)$ the integral equation (3.2) shows that in the principal approximation with respect to ε the local diffusion fluxes over the whole sphere surface (except the trailing stagnation point neighborhood $\sigma = \{| t_0 - t | < O(e^{-1/\varepsilon})\}$) are the same, and that for a reaction of order $\varkappa$

$$j(t) = kf^*(c)|_{r\to\infty} = k \quad (\varepsilon \to 0, k = \text{const}) \tag{3.8}$$

which means that for $k \ll P^{1/3}$ the reaction process is close to the kinetic mode over the whole sphere surface.

Since $G * 1 \to \infty$ when $t \to t_0$, a region of the type of boundary layer $\sigma = \{| t_0 - t | < O(e^{-1/\varepsilon})\}$ in which the local diffusion flux rapidly decreases from unity to zero appears near the trailing stagnation point. The contribution of region σ to the total diffusion flux is insignificant. Hence

$$I = 4\pi\, kf^*(1) \quad (\varepsilon \to 0, \ k = \text{const}) \tag{3.9}$$

It will be seen that that the expression for diffusion fluxes is independent of the Péclet number (ε). This means that in this case, as in that of first order reaction [1], saturation of the diffusion flux takes place. This phenomenon is related to that for any finite rate surface reaction the Sherwood number approaches the constant value (3.9) determined by the surface reaction kinetics, as the Péclet number is increased.

The obtained results make it possible to investigate the course of surface reaction at the particle surface, as was done in [1]. It appears that, as in the case of first order reaction [1], a region of the diffusion reaction mode always exists near the trailing stagnation point, while near the leading stagnation point the kinetic mode is generally absent.

Note that the local diffusion flux in the small neighborhood of the trailing stagnation point exceeds the local flux under conditions of total absorption of matter by the particle surface. This is explained by that a region of the diffusion mode of reaction (i.e. $c \to 0$, as $\theta \to 0$, and $r = 1$) is always present, while the stream of fluid in that region is less starved than in the case of total absorption.

Let us investigate the dependence of diffusion of matter in the neighborhood of the leading stagnation point $b = \{r - 1 < O(\varepsilon)$ and $\pi - \theta < O(\varepsilon)\}$ in a reaction of order $\varkappa$. For $kP^{-1/3} = O(1)$ for the local diffusion flux in the neighborhood of $\theta = \pi$ we have

$$j_\pi = \frac{3^{1/3} a_0}{\varepsilon \Gamma(1/3)} \left(1 - \Gamma\left(\frac{1}{3}\right) \frac{b_0}{2^{1/3}}\right) \tag{3.10}$$

Let us consider the behavior of the first coefficient of series (1.13) depending on variation of parameters $\varkappa$ and λ. The equation for b_0 may be written in the form

$$H(x, \lambda, \varkappa) = 0, \quad H(x, \lambda, \varkappa) = x + \lambda x^{\varkappa} - 1 \tag{3.11}$$

$$b_0 = 2^{1/3}\Gamma(2/3) B_0^{-1} x, \quad \lambda = {}^1/_3\, k\varepsilon 2^{1/2}\Gamma(1/3)$$

For $x = 0$ $H(0, \lambda, \varkappa) < 0$ and for $x = 1$ $H(1, \lambda, \varkappa) > 0$. Since $H_x' > 0$, hence for $x \geqslant 0$ Eq. (3.11) has a single root in the interval $[0, 1]$.

Let $\varkappa_1 < \varkappa_2$ and x_1 be a root of Eq. (3.11) for $\varkappa = \varkappa_1$ and x_2 for $\varkappa = \varkappa_2$ (λ is fixed in both cases). The inequality $x_1 < x_2$ is then satisfied, as follows from the inequality $H(x_1, \lambda, \varkappa_2) = x_1 + \lambda x_1^{\varkappa_2} - 1 = \lambda x_1^{\varkappa_1}(x_1^{\varkappa_2 - \varkappa_1} - 1) < 0$. Similarly, if $\lambda_1 < \lambda_2$ and x_1^* is the root of Eq. (3.11) when $\lambda = \lambda_1$, or x_2^* is such root for $\lambda = \lambda_2$, then (for fixed $\varkappa$) $x_1^* > x_2^*$.

These properties and formula (3.10) for the diffusion flux imply that in the neighborhood of the leading stagnation point the local flux increases with increasing k and diminishes with increasing reaction order.

For intermediate values of $k\varepsilon$ the solution of integral equation (1.8) was obtained by numerical methods. Function $j(t)$ for $\varkappa = {}^1/_2$, 1.0, and 2.0 is shown in Fig. 1 by dash, solid, and dash-dot lines, respectively. Curves *1*, *2* and *3* correspond to $K^* = 0.1$, 1 and 10. Dependence of the total flux on a particle on K^* is shown in Fig. 2 for the same values of $\varkappa$. In this case I_0 is the limit flux on a sphere and corresponds to $k \to \infty$.

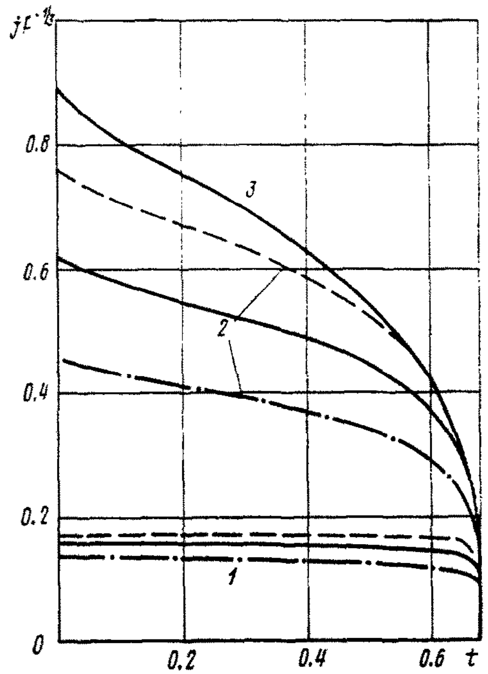

Fig. 1

Note that the representation of function $\Phi(t)$ in the form of series (1.13) makes possible the representation of the local flux in the form of series

$$j(t) = j_\pi + a_1\Pi j_\pi^{(2\varkappa-1)/\varkappa}\,[a_0^2 3^{1/3}\Gamma({}^1/_3) + a_0\Pi j_\pi^{(\varkappa-1)/\varkappa}]^{-1} t^{2/3} + \ldots, \tag{3.12}$$

$$\Pi = \varepsilon k^{1/\varkappa}\varkappa B_1$$

The numerical solution of the integral equation (3.2) shows that for fairly high K^* series (3.12) can be restricted to its first two terms

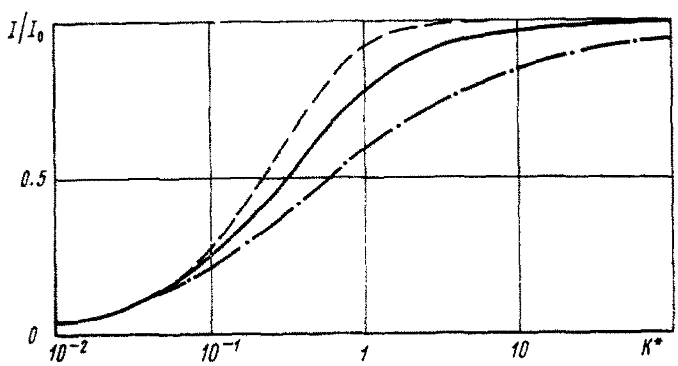

Fig. 2

for calculating the total flux. Thus, for example, for $\varkappa = {}^1/_2$ and $K^* = 0.1$ the contribution of the residual term is about 15% and rapidly diminishes with an increase of K^*; for $\varkappa = 2$ and $K^* = 0.1$ this contribution is about 20%.

Dependence of the total flux on the velocity constant can, thus, be represented in a wide range of K^* values by the approximate formula

$$\frac{I}{I_0} = \frac{\alpha - b_0(K^*)}{\alpha} + \frac{b_1(K^*)}{\alpha} t_0^{2/3}$$

In concluding we would point out that the above results can be extended to more complex flow fields (see, e. g., [12]).

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