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UNSTEADY-STATE EXTRACTION FROM A FALLING DROPLET WITH NONLINEAR DEPENDENCE OF DISTRIBUTION COEFFICIENT ON CONCENTRATION

A. D. POLYANIN
Institute for Problems in Mechanics of the USSR Academy of Sciences, 117526, Moscow, U.S.S.R.

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Abstract—An asymptotic analysis of the problem of unsteady-state heat and mass transfer of a droplet at commensurable phase resistances in the case of high Peclet numbers is carried out. The problem is considered in nonlinear statement when in the boundary condition of phase equilibrium on the droplet surface the concentration of a dispersed phase is arbitrarily dependent on continuous phase concentration. The dynamics of heat and mass transfer is shown to be qualitatively very different over three characteristic time intervals, the boundaries of which depend on Peclet number. The first time interval, $0 \leqslant t < O(\ln Pe)$ (t is the dimensionless time), is characterized by the formation of unsteady-state diffusional boundary layers on both sides of the droplet surface (which are as yet qualitatively identical), with the internal boundary layer generating a diffusional wake near the flow axis. Over the second time interval, $O(\ln Pe) \leqslant t < O(Pe)$, the developed internal diffusional wake starts to interact with the boundary layer and 'smears' it severely (here, the boundary layers outside and inside the droplet are already substantially different), as a result of which the thickness of the internal boundary layer increases considerably in a gradual way. Over the last time interval, $O(Pe) \leqslant t$, a further rearrangement of the concentration field is observed so that the boundary layers are practically no longer present; the concentration outside the droplet becomes constant and equal to the nonperturbed concentration at infinity, while inside the droplet a substantially unsteady-state process occurs when concentration on each fixed streamline has already fully become equalized (at the expense of repeated liquid circulation along the closed streamlines), while mass is transferred by molecular diffusion in the direction normal to the streamlines.

NOMENCLATURE

a	droplet radius
C_1, C_2	concentration in continuous and dispersed phases
C_{10}, C_{20}	concentration in continuous and dispersed phases at time zero
C_h	characteristic concentration, $C_{20}-F_*(C_{10})$
c_1	dimensionless concentration in continuous phase (outside the droplet), $1+(C_{10}-C_1)C_h^{-1}$
c_2	dimensionless concentration in dispersed phase (inside the droplet), $(C_{20}-C_2)C_h^{-1}$
$\bar{c}_i$	Laplace–Carlson transform of concentration
D_1, D_2	diffusion coefficients in continuous and dispersed phases
$F_*(C_1)-C_2 = 0$	condition of phase equilibrium on droplet surface
G	shear coefficient
g	$g_{\psi\psi}g_{\eta\eta}g_{\lambda\lambda}$
$g_{\psi\psi}g_{\eta\eta}g_{\lambda\lambda}$	metric tensor components
n	exponent determining the phase equilibrium law, equation (2)
Pe	Peclet number, aUD_2^{-1}
p	complex parameter of Laplace–Carlson transformation
r, θ, φ	spherical coordinate system associated with droplet
s	continuous 'through' coordinate corresponding to the distance reckoned from the midsection of the droplet along the fixed streamline $\psi^{(2)} = \text{const.}$
T	time for which a liquid element inside the droplet completes a turn along a closed streamline, $\psi^{(2)} = \text{const.}$ (characteristic time of delay)
T_1, T_2	temperature outside and inside a droplet
T_{10}, T_{20}	temperature outside and inside a droplet at time zero
t	dimensionless time, $a^{-1}Ut_*$
t_*	time
t_h	characteristic hydrodynamic time, aU^{-1}
t_d	characteristic diffusional time, $a^2D_2^{-1}$
U	characteristic velocity of translational flow, $(\beta+1)^{-1}U_\infty$
U	characteristic velocity of shear flow, $aG(\beta+1)^{-1}$
U_∞	nonperturbed flow velocity at infinity
$\mathbf{u}_i$	dimensionless liquid velocity vector outside ($i = 1$) and inside ($i = 2$) a droplet
Y	'stretched' boundary layer coordinate, $Pe^{1/2}(1-r)$
z	$r \cos\theta$.

Greek symbols

α	phase equilibrium constant
β	ratio between the dynamic viscosities of droplet and surrounding liquid
ζ	'stretched' stream function, $Pe^{1/2}\psi^{(i)}$

Θ	dimensionless temperature outside ($i = 1$) and inside ($i = 2$) a droplet, $(T_{20} - T_i)/(T_{20} - T_{10})$
κ	ratio of continuous and dispersed phase diffusion coefficients, D_1/D_2
λ	root of equation (18)
λ_i	thermal conductivities of liquid outside ($i = 1$) and inside ($i = 2$) a droplet
μ	$\cos\theta$
ρ	$r \sin\theta$
τ	'slow' time, $Pe^{-1}t$
$\psi^{(i)}$	stream functions corresponding to the flow outside and inside a droplet
ψ	$\psi^{(2)}$
ψ, η, φ	curvilinear orthogonal coordinate system associated with streamlines $\psi = \psi^{(2)} = \text{const.}$
ω	cyclic variable.

1. INTRODUCTION

In the linear formulation the problem of convective mass and heat transfer of a droplet at commensurable phase resistances at large Peclet numbers was investigated earlier in refs. [1–10]. Thin diffusional boundary layers were postulated to exist on each side of the interface [1–8], beyond which the concentration was assumed to be (identically) constant; in refs. [1, 2, 5] the problem was solved in a steady-state formulation. The solutions obtained in refs. [3, 4, 6–8] exhibited a steady-state behaviour corresponding to some inhomogeneous distribution of concentration in boundary layers. It defined the average Sherwood number which was different from zero and proportional to the square root of the Peclet number. In ref. [9], the complete problem of mass transfer of a droplet at commensurable phase resistances was modelled by the equation of transport inside a droplet with the boundary condition on the surface which reflected the nature of mass transfer in a continuous phase. In ref. [10], the concentrational boundary layers and also the diffusional wake originating outside and inside a droplet were described qualitatively. Also shown was the predominance of mass transfer inside a droplet.

The internal problem of the unsteady-state convective mass and heat transfer, when the resistance to transport is completely concentrated inside a droplet, was much investigated earlier by analytical [1–8, 11] and numerical [12–20] means. In refs. [1–8, 11] it was assumed that the Peclet number was large, and two completely different approaches were used to analyse the problem. In ref. [11] (see also ref. [10]), an approximate solution was constructed in the assumption that concentration along the streamlines is constant and mass is transferred by molecular diffusion in the direction normal to the streamlines. In refs. [1–8], the solution of the internal mass and heat transfer problem was obtained as a limiting case of the problem at commensurable phase resistances in the assumption that there were diffusional boundary layers on each side of the interface. These two different approaches to the solution of the internal problem lead to substantially different results for the dimensionless average diffusional flux I on the droplet surface. According to ref. [11], the solution is a substantially unsteady-state one, and the property

$$\lim_{t\to\infty} I = 0$$

is fulfilled, while the results of refs. [1–8] lead to another solution which is nonstationary only over the initial segment and

$$\lim_{t\to\infty} I = O(Pe^{1/2}).$$

The internal problem of convective mass and heat transfer was also investigated numerically using the finite-difference schemes [12–20]. A qualitative disagreement of the results was also observed depending on the scheme selected (compare, e.g. refs. [12–14] and [19, 20]).

The differences observed [1–20] gave rise to a mutual criticism of the methods used and results obtained (see, e.g. refs. [3, 9, 10, 12, 18, 20]). Thus, on the basis of comparison of the experimental [9] and predicted [19, 20] results, the inference was drawn [9, 19, 20] concerning the applicability of the Kronig and Brink model suggested in ref. [11].

In particular, it will be shown in what follows that:

(1) the results of refs. [3, 4, 6–8] are applicable only in the initial, rapidly progressing stage of the process (before the internal diffusional boundary layer starts to interact with the diffusional wake it generates) and that they are inapplicable in most important, intermediate and final, stages of the process;

(2) the Kronig and Brink model [11] and the results of modelling [9] poorly reflect the dynamics of the process of mass and heat transfer inside a droplet during the initial and intermediate stages of the process and reflect it well in the final stage.

2. STATEMENT OF THE PROBLEM

Consider an unsteady-state convective mass and heat transfer between a droplet and a continuous medium in the case of commensurable phase resistances. It is presumed that at the droplet surface the following condition of phase equilibrium holds

$$C_2 = F_*(C_1) \quad (F_*(0) = 0, \quad \mathrm{d}F_*/\mathrm{d}C_1 \geqslant 0). \qquad (1)$$

Here, $F_*(C_1)C_1^{-1}$ is the distribution coefficient [21]. It is usually assumed that the function F_* depends linearly on concentration $F_*(C_1) = \alpha C_1$ [1–8]. It has been shown [9, 22] that in a number of cases the power-law dependence should be used

$$F_*(C_1) = \alpha C_1^n \quad (0.5 \leqslant n \leqslant 2.0), \qquad (2)$$

where, e.g. $n \simeq 0.6$ [22]. Below, a most general case of the arbitrary dependence $F_* = F_*(C_1)$, [equation (1)], will be considered.

It is assumed that at time zero the concentrations outside and inside a droplet are constant and equal to C_{10} and C_{20}, respectively; it is regarded in this case that the requirement for the nontriviality of the problem is met: $C_{20} \neq F_*(C_{10})$.

In dimensionless variables, in the spherical coordinate system r, θ, φ associated with the droplet, the corresponding unsteady-state boundary-value problem for the concentration fields in continuous and dispersed phases is

$$\frac{\partial c_1}{\partial t} + (\mathbf{u}_1 \nabla) c_1 = \frac{\kappa}{Pe} \Delta c_1 \quad (1 < r < \infty), \tag{3}$$

$$\frac{\partial c_2}{\partial t} + (\mathbf{u}_2 \nabla) c_2 = \frac{1}{Pe} \Delta c_2 \quad (0 \leqslant r < 1), \tag{4}$$

$$t = 0, c_1 = 1, c_2 = 0; \quad r \to \infty, c_1 \to 1, \tag{5}$$

$$r = 1, c_2 = F(c_1), \kappa \frac{\partial c_1}{\partial r} = \frac{\partial c_2}{\partial r}, \tag{6}$$

$$c_1 = 1 + \frac{C_{10} - C_1}{C_\mathrm{h}}, \quad c_2 = \frac{C_{20} - C_2}{C_\mathrm{h}};$$

$$F(c_1) = \frac{C_{20} - F_*(C_1)}{C_\mathrm{h}},$$

$$C_\mathrm{h} = C_{20} - F_*(C_{10}), \; Pe = \frac{aU}{D_2}, \; \kappa = \frac{D_1}{D_2}, \; t = \frac{Ut_*}{a}.$$

In the above, the characteristic scales of length and time were chosen to be the quantities a and aU^{-1}. The quantity C_h, as noted in the Nomenclature, is the characteristic concentration [in the linear case corresponding to $n = 1$ in equation (2), $C_\mathrm{h} = C_{20} - \alpha C_{10}$ and $F(c_1) = \alpha c_1 - \alpha + 1$].

In what follows, for the sake of definiteness, it will be assumed that the distribution of liquid velocities, $\mathbf{u}_i$, outside and inside a droplet is governed by the Hadamard–Rybczinski solution [23, 24] which corresponds to the steady-state translational Stokes flow around a spherical droplet

$$\psi^{(1)} = \tfrac{1}{4}(1-r)[2(\beta+1)r - \beta - \beta r^{-1}] \sin^2 \theta, \quad 1 \leqslant r < \infty \tag{7}$$

$$\psi^{(2)} = \tfrac{1}{4} r^2 (1 - r^2) \sin^2 \theta, \quad 0 \leqslant r \leqslant 1$$

$$\mathbf{u}_i = (u_r^{(i)}, u_\theta^{(i)}), \; u_r^{(i)} = -\frac{1}{r^2 \sin \theta} \frac{\partial \psi^{(i)}}{\partial \theta},$$

$$u_\theta^{(i)} = \frac{1}{r \sin \theta} \frac{\partial \psi^{(i)}}{\partial r}.$$

In writing equations (3)–(7), the characteristic flow velocity was chosen to be the quantity $U = (\beta + 1)^{-1} U_\infty$. The angle θ is reckoned from the flow direction at infinity (Fig. 1).

It should be noted that the limiting cases of the restricting internal and external phase resistance correspond to the limiting transitions in equations (2)–(6) at $\alpha \to 0$ and $\alpha \to \infty$. In particular, at $\alpha = 0$ in equation (2) the concentration distribution in the dispersed phase ($i = 2$) is determined by solving a purely internal problem, which is described by equation (4) with the initial and boundary conditions

$$t = 0, c_2 = 0; \quad r = 1, c_2 = 1. \tag{8}$$

In what follows, the emphasis will be given to the most important case of commensurable phase resistances which is controlled by the equality $F(c_1) = O(1)$ at $c_1 = O(1)$. This, for example, is always the case in a similar thermal problem (for details see Section 8).

It is furthermore assumed that the Peclet numbers, corresponding to continuous and dispersed phases, are large and of the same order, i.e. the relations $Pe \gg 1$, $\kappa = O(1)$ are valid.

The unsteady-state problem investigated, equations (1)–(6), has two different dimensional characteristic time scales, t_h and t_d

$$t_\mathrm{h} = aU^{-1}, \quad t_\mathrm{d} = a^2 D_2^{-1}, \tag{9}$$

the first of which corresponds to the characteristic time of liquid circulation inside a droplet (hydrodynamic time), and the second to the characteristic time of penetration by diffusion (diffusional time). The equality $t_\mathrm{d}/t_\mathrm{h} = Pe$ yields that at large Peclet numbers the diffusional time is larger than the hydrodynamic time. This results in qualitatively different dynamics of mass transfer inside (and outside) a droplet depending on the

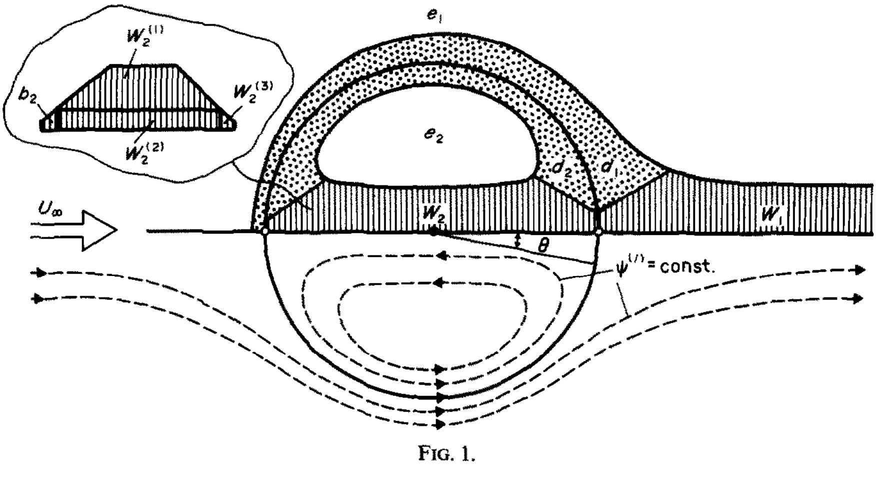

FIG. 1.

ratio between the instantaneous dimensional time t_* and characteristic times t_h and t_d, i.e. it is possible to introduce two dimensionless times in the present problem, $t = t_*/t_h$ and $\tau = t_*/t_d$, which should provide a basis for further asymptotic analysis.

3. INITIAL STAGE OF THE PROCESS

An asymptotic analysis of the problem, equations (1)–(7), at large Peclet numbers $Pe \gg 1$ shows that in the whole flow region outside and inside a droplet it is possible to distinguish the flow cores e_i, the regions of diffusional boundary layer d_i and the regions of diffusional wake W_i; $i = 1, 2$ (Fig. 1).

At $t \ll 1$, the convective term in equations (3) and (4) can be neglected, and the solution here corresponds to a thin 'shock' spherical layer located outside and inside a droplet near its surface, i.e. the layer owing its origin to the disagreement between the initial and boundary conditions, equations (5) and (6), at $F(1) \neq 0$.

At $t = O(1)$, the convective terms in equations (3) and (4) will have the same order as the unsteady-state term; in this case the concentration in the flow core remains equal to the initial concentration, $c_1^{(e)} = 1$ and $c_2^{(e)} = 0$, and diffusional boundary layers develop near the droplet surface

$$d_i = \{|r-1| \leqslant O(Pe^{-1/2}),$$
$$O(Pe^{-1/2}) \leqslant \theta,\ O(Pe^{-1/2}) \leqslant \pi-\theta\},$$

(here and hereafter the inequalities within braces specify the order of the characteristic dimensions of the region considered), which as yet show similar qualitative behaviour on each side of the droplet surface. The introduction of a new 'stretched' boundary layer coordinate Y into equations (1)–(7), initial and boundary conditions, with subsequent separation of higher order terms of the expansion in $Pe^{-1/2}$ leads to the following boundary-value problem

$$\Lambda^{(i)} \cdot c_i^{(d)} = 0,\ \Lambda^{(i)} \equiv \frac{\partial}{\partial t} + Y\mu\frac{\partial}{\partial Y}$$
$$+\tfrac{1}{2}(1-\mu^2)\frac{\partial}{\partial \mu} - \kappa_i\frac{\partial^2}{\partial Y^2}, \quad (10)$$

$$t = 0,\ c_1^{(d)} = 1,\ c_2^{(d)} = 0, \quad (11)$$

$$Y \to -\infty,\ c_1^{(d)} \to 1; \quad Y \to +\infty,\ c_2^{(d)} \to 0, \quad (12)$$

$$Y = 0,\ c_2^{(d)} = F(c_1^{(d)}),\ \kappa\frac{\partial c_1^{(d)}}{\partial Y} = \frac{\partial c_2^{(d)}}{\partial Y}, \quad (13)$$

$$Y = Pe^{1/2}(1-r),\ \mu = \cos\theta; \quad \kappa_1 = \kappa,\ \kappa_2 = 1$$
$$(i = 1, 2).$$

In order to carry out the analysis, equations (10) [25, 26] are rewritten in terms of new variables (x, ζ, ω) which are expressed through former variables (t, Y, μ) by the formulae

$$x = x(\mu) = \tfrac{1}{6}(2-\mu)(1+\mu)^2,$$
$$\zeta = \zeta(Y, \mu) = \tfrac{1}{2}Y(i-\mu^2) = Pe^{1/2}\psi^{(i)}, \quad (14)$$
$$\omega = \omega(t, \mu) = t - \ln\frac{1+\mu}{1-\mu}.$$

As a result, equations (10) take the form

$$\frac{\partial c_i^{(d)}}{\partial x} = \kappa_i\frac{\partial^2 c_i^{(d)}}{\partial \zeta^2} \quad (c_i = c_i(x, \zeta, \omega)). \quad (15)$$

The ordinary parabolic equations with constant coefficients, equations (15), are much simpler than the initial equations with variable coefficients, equations (10), and do not contain (in an explicit form) the variable ω, which is similar to the cyclic variables in analytical mechanics.

The boundary conditions for equations (15) at $\zeta = 0$ and $\zeta \to \pm\infty$ are derived by a simple (formal) substitution of ζ for the coordinate Y in equations (12) and (13).

Taking into account that equations (15) are invariant with respect to the change of the variable $x \to x + X(\omega)$, where $X(\omega)$ is any function of the cyclic coordinate ω, it is not difficult to show that the solution of equations (10)–(13) can be represented in the form

$$c_i^{(d)} = c_i^{(d)}(\mu, \zeta, \omega) = A_i + B_i$$
$$\times \operatorname{erf}\left\{\frac{|\zeta|}{2\kappa_i^{1/2}[x(\mu) - x(-\tanh\omega/2)]^{1/2}}\right\}. \quad (16)$$

Here, constants A_i and B_i are determined by substituting equation (16) into initial and boundary conditions, equations (11)–(13), and are prescribed by the formulae

$$A_1 = 1-\lambda\kappa^{-1/2},\ B_1 = \lambda\kappa^{-1/2},\ A_2 = \lambda,\ B_2 = -\lambda, \quad (17)$$

where the parameter λ can be found by solving the algebraic (transcendental) equation

$$\lambda = F(1-\lambda\kappa^{-1/2}). \quad (18)$$

In the linear case, corresponding to $n = 1$ in equation (2), formulae (16)–(18) go over into the results of refs. [4, 6].

One verifies easily that

$$\frac{Sh}{Sh_0} = \lambda,\ Sh = \tfrac{1}{2}\kappa\int_{-1}^{1}\left(\frac{\partial c_1}{\partial r}\right)_{r=1} d\mu$$
$$= \tfrac{1}{2}\int_{-1}^{1}\left(\frac{\partial c_2}{\partial r}\right)_{r=1} d\mu, \quad (19)$$

where Sh_0 is the mean Sherwood number which is determined by solving a purely internal unsteady-state problem, equations (4) and (8), and which corresponds to the following values of the parameters $A_2 = 1$, $B_2 = -1$ in equation (16).

Figure 2 shows the behaviour of the normalized

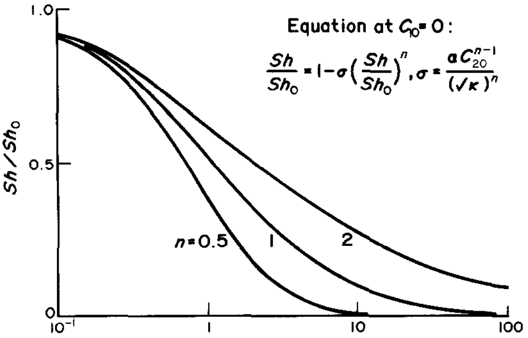

FIG. 2.

mean Sherwood number, equation (19), determined by the solution of the algebraic equation, equation (18), depending on a change in the complex $\sigma = \alpha C_{20}^{n-1}\kappa^{-n/2}$ for the phase equilibrium power law, equation (2), at $C_{10} = 0$ (which corresponds to $C_h = C_{20}$, $F(c_1) = 1-\alpha C_{20}^{n-1}(1-c_1)^n$) for $n = 1/2$, 1, and 2. It is seen that the mean Sherwood number increases monotonically with n and decreases with an increasing phase equilibrium constant α.

4. CONCENTRATION DISTRIBUTION IN A DIFFUSIONAL WAKE. THE REGION OF APPLICABILITY OF THE 'BOUNDARY LAYER APPROXIMATION'

It is not difficult to show that equations (16) become inapplicable in the vicinity of the efflux point $\theta = O(\mu = 1)$, where the formation of the internal and external diffusional wakes W_i occurs. In the external wake, the substance depleted by diffusion is entrained by the flow from the droplet surface to infinity and does not exert the reverse effect on the external diffusional boundary layer, which generates this wake. A much more complex situation is observed in the interior of the droplet. At $t = 0$, the concentration distribution inside of a droplet is uniform, and the diffusional wake W_2 is absent; at $t > 0$, the diffusional wake starts to form and, gradually 'moving' along the flow axis, it sprouts through the entire droplet volume after which it starts to interact with the internal diffusional boundary layer d_2 exerting on it a reverse effect.

The diffusional wake W_2 is substantially nonuniform and consists of the rear stagnation point region $W_2^{(3)} = \{1-r \leqslant O(Pe^{-1/2}), \theta \leqslant O(Pe^{-1/2})\}$, convective boundary-layer wake region $W_2^{(1)} = \{O(Pe^{-1/2}) \leqslant 1-r,\ O(Pe^{-1}) \leqslant \psi(2) \leqslant O(Pe^{-1/2})\}$, internal wake region $W_2^{(2)} = \{O(Pe^{-1/2}) \leqslant 1-r,\ 0 \leqslant \psi^{(2)} \leqslant O(Pe^{-1})\}$ and of the region of the forward stagnation point $b_2 = \{1-r \leqslant O(Pe^{-1/2}),\ \pi-\theta \leqslant O(Pe^{-1/2})\}$ (Fig. 1). The procedure of obtaining the equations and boundary conditions for the distribution of concentration in these regions is completely similar to that used in refs. [27, 28]. The substance coming from the boundary layer is transferred in the main *in the convective-boundary layer region of the diffusional wake* $W_2^{(1)}$, the distribution of concentration in which is determined by

$$\frac{\partial c_2^{(1)}}{\partial t} - \tfrac{1}{2}(1-z^2)\frac{\partial c_2^{(1)}}{\partial z} - \tfrac{1}{2}z\eta\frac{\partial c_2^{(1)}}{\partial \eta} = 0, \quad \eta = Pe^{1/4}\rho. \tag{20}$$

Here, $z = r\cos\theta$, $\rho = r\sin\theta$ is the cylindrical coordinate system associated with the droplet centre; η is the 'stretched' coordinate. Equation (20) is derived by introduction of new variables z and η into equation (4) with the subsequent separation of higher order terms of the expansion in the small parameter $Pe^{-1/2}$. [Here, as always, it is assumed that $\eta = O(1)$, $\partial c_2^{(1)}/\partial\eta = O(1)$, $\partial^2 c_2^{(1)}/\partial\eta^2 = O(1)$.] In terms of the new variables z, ζ, $\omega^{(1)}$, where

$$\zeta = Pe^{1/2}\psi^{(2)} = \tfrac{1}{4}\eta^2(1-z^2), \quad \omega^{(1)} = t + \ln\frac{1+z}{1-z}, \tag{21}$$

equation (20) yields $\partial c_2^{(1)}/\partial z = 0$ for the concentration in the convective boundary-layer region of the diffusional wake, from which it follows that the general solution of equation (20) has the form

$$c_2^{(1)} = \Phi(\zeta, \omega^{(1)}). \tag{22}$$

Here, Φ is an arbitrary function of both arguments ζ and $\omega^{(1)}$.

The condition of the asymptotic matching (agreement) of the solutions in the diffusional boundary-layer region, equations (16)–(18), and in the convective boundary-layer region of the diffusional wake, equation (22), is written as

$$c_2^{(d)}|_{\mu\to 1} = c_2^{(1)}|_{z\to 1}.$$

It determines the explicit form of the function Φ and, consequently, the distribution of concentration in the region $W_2^{(1)}$

$$c_2^{(1)} = c_2^{(d)}(1, \zeta, \omega^{(1)} - \tfrac{1}{2}\ln Pe + \ln\zeta - 4\ln 2). \tag{23}$$

The matching procedure in this case can be much more easily understood if one replaces the two local coordinates μ (in the region d_2) and z (in the region $W_2^{(1)}$) by one continuous 'through' coordinate s (both in the region d_2 and $W_2^{(1)}$) which is reckoned along the streamlines of flow; the origin of the coordinate s is at the droplet midsection $\theta = \pi/2$, with s being the distance from the surface $\theta = \pi/2$ along the fixed streamline $\psi^{(2)} = \text{const}$. It is easy to verify that in the diffusional boundary layer d_2, the local coordinate is expressed in terms of the continuous one as: $\mu = \sin s$ $(0 \leqslant s \leqslant \pi/2)$, while in the convective boundary-layer region of the wake $W_2^{(1)}$, the relationship $z = \pi/2+1-s(\pi/2 \leqslant s \leqslant \pi/2+2)$ is valid. Note further, that the local variables ω and $\omega^{(1)}$ are the characteristics of the truncated equations, equations (10) and (20), at $\partial/\partial Y = \partial^2/\partial Y^2 = \partial/\partial\eta = 0$. Taking into account the equality $(\mathbf{u}_2\nabla)c_2 = |\mathbf{u}_2|\partial c_2/\partial s$, it is not difficult to verify that the local variables ω and $\omega^{(1)}$, corresponding to the local coordinates μ and z, can be substituted by one continuous cyclic variable

$$\bar{\omega} = t - \int_0^s \frac{\mathrm{d}s}{|\mathbf{u}_2|}, \tag{24}$$

with the aid of which the solutions in the diffusional boundary layer and convective boundary-layer region of the wake inside of a droplet can be written as

$$c_2^{(\mathrm{d})} = c_2^{(\mathrm{d})}(\mu, \zeta, \bar{\omega}), \quad c_2^{(1)} = \Phi(\zeta, \bar{\omega}). \tag{25}$$

The matching of expressions (25) [in contrast to equations (16) and (22)] no longer involve any difficulties and can be easily done; in this case a direct computation of $\bar{\omega}$ in the region $W_2^{(1)}$ with subsequent transition from the continuous coordinate s to the local coordinate z leads to formula (23). The presence of logarithmic terms in equation (23) is due to the specific nature of the integrand where the liquid velocity $\mathbf{u}_2$ is close to zero and is substantially inhomogeneous in $\psi^{(2)}$.

Equation (20) corresponds to purely convective substance transfer without any changes along the streamlines. The concentration profile in $W_2^{(1)}$ is governed by the concentration distribution at the 'outlet' from the diffusional boundary layer d_2. It is seen from formula (23) that in the convective boundary-layer region of the diffusional wake $W_2^{(1)}$, there is a substantial delay with a characteristic time $\frac{1}{2}$ ln Pe [as always, it is assumed for $W_2^{(1)}$ that $\zeta = O(1)$] and this delay is different on different streamlines $\zeta =$ const. This dispersion of the delay time is due to a nonuniform distribution of liquid velocities near the critical efflux point $\theta = 0$; in this case, as it follows from equation (23), the substance transport near the flow axis ($\zeta \ll 1$) is practically absent, and the delay is very large. At greater distances from the flow axis, the liquid velocity in the vicinity of the efflux point increases and the delay decreases.

That the diffusional wake could fully develop (i.e. could 'sprout' through the entire droplet volume) and start to interact with the diffusional boundary layer d_2, the continuous coordinate s, after it 'moved' along a fixed streamline near the flow axis and 'passed' the vicinity of the forward stagnation point $\theta = \pi$, must arrive at the region d_2. Then, due to a complete identity between the liquid velocity fields in the vicinities of the forward ($\theta = \pi$) and rear ($\theta = 0$) stagnation points, the time of delay should double.

It follows from the above analysis that the region of applicability of the boundary-layer solution, equations (16)–(18), is restricted by the interval $0 \leqslant t < T$, where T is the characteristic time of delay

$$T = \ln Pe - 2 \ln \zeta + 8 \ln 2 = O(\ln Pe) \quad (Pe \gg 1). \tag{26}$$

The characteristic time of delay T has a simple physical meaning. Namely, determine T from the following formula (K is the full first-kind elliptical integral)

$$T = T(\psi^{(2)}) = \oint \frac{\mathrm{d}s}{|\mathbf{u}_2|} = \frac{8}{\sqrt{(1+\sqrt{\xi})}} K \sqrt{\left(\frac{1-\sqrt{\xi}}{1+\sqrt{\xi}}\right)},$$

$$\xi = 16\psi^{(2)}, \tag{27}$$

which corresponds to the time required for a liquid element inside a droplet to make a complete turn along the closed streamline $\psi^{(2)} =$ const., which passes through the region of the diffusional boundary layer d_2 and also the convective boundary-layer region of the diffusional wake $W_2^{(1)}$, i.e. in the limit at

$$Pe \to \infty, \; \psi^{(2)} = Pe^{-1/2}\zeta, \; \zeta = O(1). \tag{28}$$

As a result of the expansion of $T(\psi^{(2)})$, formulae (27) and (28) yield equation (26).

The results obtained can be interpreted as follows. Since the liquid flow velocity inside of a droplet is finite, then first, at small times $t < T$, a homogeneous concentration, coming from the bulk of the liquid, enters the region of the forward stagnation point b_2. This takes place until the solution, which arrives from the 'end' of the boundary layer d_2 into the diffusional wake W_2 and which has been enriched at the expense of substance influx from outside through the droplet surface, will not arrive at the 'origin' of the diffusional boundary layer after having travelled the whole path near the flow axis. The characteristic time of substance transport in the diffusional wake of the droplet determines the region of applicability of the solution, equations (16)–(18), which at $t > T$ ceases to correctly describe the distribution of concentration in the diffusional boundary layer (owing to a change in the 'flow incidence' conditions).

Formula (26) allows a remark regarding the qualitative behaviour of the basic characteristics of mass transfer outside and inside a droplet. At very large Peclet numbers, such that ln $Pe > O(1)$, expression (26) yields $T > O(1)$. The dimensionless characteristic relaxation time t_r [3, 6] from the diffusional boundary-layer solution, equations (16)–(18), is $t_\mathrm{r} = O(1)$ and is smaller than the characteristic time of delay, $t_\mathrm{r} < T$. This means that at very large Peclet numbers, the diffusional boundary layer located on each side of the droplet surface, manages to develop earlier than the internal boundary layer starts to interact with the internal diffusional wake generated by this layer [i.e. the steady-state solution

$$c_{i\infty}^{(\mathrm{d})} = \lim_{t \to \infty} c_i^{(\mathrm{d})}(\mu, \zeta, \omega(t, \mu))$$

can be considered as an intermediate asymptotic of the solution for a general unsteady-state problem in the regions d_2]. This attaches a certain meaning to the steady-state solutions obtained in refs. [1, 2, 5]. It follows from what has been said above that the dependence of the mean diffusional flux at the droplet surface on time should have a characteristic flat segment corresponding to the intermediate 'stationary' regime.

5. INTERMEDIATE STAGE OF THE PROCESS

In the intermediate stage of the mass and heat transfer process at $O(\ln Pe) \leqslant t < O(Pe)$, the diffusional boundary layer equations, equation (16), still remain valid, but the solution here, in contrast to equations (16)–(18), is no longer a self-similar one, since it is necessary to take into account the mutual effect of the diffusional wake W_2 and of the boundary layer d_2 inside of a droplet. As a result of interaction, the internal diffusional wake strongly 'smears' the diffusional boundary layer d_2 (here one can notice already a marked qualitative distinction between the boundary layers outside and inside of a droplet) and this leads gradually to a substantial thickening of the internal boundary layer. In order to simplify the presentation and the mathematics, the study is limited to the dynamics of mass transfer process in the intermediate stage $O(\ln Pe) \leqslant t < O(Pe)$ for a purely internal problem, which is described by equation (4) with initial and boundary conditions, equation (8).

The solution of the equation for the distribution of concentration in a diffusional boundary layer, equation (15) [to which the initial equation (10) is reduced after the substitution of variables, equation (14)], which satisfies the boundary and 'initial' conditions

$$\zeta = 0, c_2^{(d)} = 1; \quad \zeta \to \infty, c_2^{(d)} \to 0; \quad x \to 0, c_2^{(d)} \to f(\zeta, \omega) \tag{29}$$

[the first two of which follow from equation (8)], has the form

$$c_2^{(d)} = \int_0^\infty G(\zeta, \zeta^*, x) f(\zeta^*, \omega)\, \mathrm{d}\zeta^* + \operatorname{erfc}\left\{\frac{\zeta}{2[x - x(-\tanh \omega/2)]^{1/2}}\right\},$$
$$G(\zeta, \zeta^*, x) = \frac{1}{2\sqrt{(\pi x)}}\left\{\exp\left[-\frac{(\zeta-\zeta^*)^2}{4x}\right] - \exp\left[-\frac{(\zeta+\zeta^*)^2}{4x}\right]\right\}. \tag{30}$$

The function $f(\zeta, \omega)$ is as yet unknown and will be determined later as part of the problem solution; it is assumed that the second term of equation (30) containing the sign of the additional probability function is, by definition, equal to zero at $x - x(-\tanh \omega/2) \leqslant 0$.

From the results of Section 4 [see equations (23)–(25) with equation (30) taken into account] it follows that the distribution of concentration of the substance (which is dissolved in the liquid and which arrives from the boundary layer) in the convective boundary-layer region of the diffusional wake $W_2^{(1)}$, which corresponds to purely convective transport (without diffusion) along the streamlines, is determined as

$$c_2^{(1)} = \int_0^\infty G(\zeta, \zeta^*, x_0) f(\zeta^*, \bar{\omega})\, \mathrm{d}\zeta^* + \operatorname{erfc}\left\{\frac{\zeta}{2[x_0 - x(-\tanh \bar{\omega}/2)]^{1/2}}\right\},$$
$$x_0 = x(\mu = 1) = 2/3. \tag{31}$$

The solutions obtained, equations (30) and (31), should agree (match) with each other in the vicinity of the forward stagnation point at $\theta \approx \pi$. By satisfying this condition, one can arrive at the following equation to determine the unknown function f

$$f(\zeta, \omega) = \int_0^\infty G(\zeta, \zeta^*, x_0) f(\zeta^*, \omega - T)\, \mathrm{d}\zeta^* + \operatorname{erfc}\left\{\frac{\zeta}{2[x_0 - x(-\tanh (\omega - T)/2)]^{1/2}}\right\}, \tag{32}$$

where $T = T(\zeta)$ is the characteristic time of delay, equation (26).

The integral equation, equation (32), is the equation with a delayed argument in the cyclic variable ω, therefore, for the problem to be correctly formulated, it is necessary to show the 'pre-history' of the function f.

It is clear from the previous analysis (see Section 4) that the function f should satisfy the condition

$$f = 0 \quad \text{at} \quad t - T < 0, \tag{33}$$

which has a clear physical meaning of the finite nature of the velocity of convective substance transport along the streamlines (the fixed liquid element starts again to interact with the boundary layer at the same space point after it has completed its circulation along the closed streamline).

Equation (32) can be presented in a more convenient 'standard' form if the explicit form of the relation $\omega = \omega(t, \mu)$, equation (14), is taken into account, and the unknown function f is redenoted as

$$f(\zeta, \omega) \equiv f_1(\zeta, \mu, t), \quad f(\zeta, \omega - T) \equiv f_1(\zeta, \mu, t - T). \tag{34}$$

In such a presentation, the equation for f_1, equations (32) and (34), depends parametrically on the coordinate μ, while the initial condition, equation (33), is a standard one. For the characteristic delay time T in equations (32)–(34), one should use it as presented in equation (26).

It is seen from equation (32) that at $t \to \infty$, $f \to 1$ [direct verification can easily show that $f = 1$ is an exact specific solution of equation (32) which, however, does not satisfy condition (34)]. This means that as a result of interaction, the internal diffusional wake W_2 strongly 'smears' the diffusional boundary layer d_2 and this leads to a substantial thickening of the boundary layer. Equation (32) remains valid until the linear approximation of the stream function near the droplet surface, and, consequently, equations (10) become already inapplicable for a correct description of the process. Using the procedure described, it is possible in much the same way to obtain a system of integral equations with a delayed argument (to determine the unknown functions f and the surface concentration $g = c_2|_{r=1}$), which would correctly describe the dynamics of mass transfer outside and inside of a droplet in the intermediate stage of the process in the general case of commensurable phase resistances, equations (3)–(6).

6. AGREEMENT BETWEEN THE ASYMPTOTIC EXPANSIONS OF THE TRANSFORM AND INVERSE TRANSFORM FOR THE LAPLACE TRANSFORMATION

In order to determine the asymptotic behaviour of the solution to equations (3)–(7) in the final stage of the process [at $t \geqslant O(Pe)$], one can make use of the following useful remark which allows the detection of many qualitative and quantitative aspects of the asymptotic behaviour of unsteady-state problems with small or large parameters by applying the Laplace–Carlson integral transformation.

Suppose there is an (arbitrary) function $X = X(t, \varepsilon)$, which depends on time and a small (large) parameter ε (to save space, the dependence of X on spatial coordinates is omitted) and which in the limit

$$\varepsilon \to 0, \quad \tau = \delta(\varepsilon)t = O(1), \tag{35}$$

is expanded into an asymptotic series in ε (τ is fixed)

$$\varepsilon \to 0, \quad X \doteq \sum_{n=0}^{\infty} \alpha_n(\varepsilon) X_n(\tau); \quad \lim_{\varepsilon \to 0} \frac{\alpha_{n+1}}{\alpha_n} = 0. \tag{36}$$

The variable τ corresponds to a 'fast' or 'slow' time, depending on a specific function $\delta(\varepsilon)$ selected [a most frequently encountered dependence is $\delta(\varepsilon) = \varepsilon^k$]. Naturally, one can construct as many expansions of the type, equation (36), as one pleases by choosing different $\delta(\varepsilon)$'s. In specific cases, when the function $X = X(t, \varepsilon)$ is the solution of a certain boundary-value problem, the choice of the dependence $\delta = \delta(\varepsilon)$ presents, as a rule, no difficulties [21, 29, 30].

If $X(t, \varepsilon)$ is conformed with the inverse transform $\bar{X}(p, \varepsilon)$ obtained by the Laplace–Carlson transformation

$$\bar{X} = \bar{X}(p, \varepsilon) = \mathbf{L} \cdot X, \quad \mathbf{L} \cdot X = p \int_0^{\infty} \mathrm{e}^{-pt}\, X(t, \varepsilon)\, \mathrm{d}t. \tag{37}$$

In the space of inverse transforms, to series (36) there formally corresponds, by virtue of the properties of the operator $\mathbf{L}$, the series

$$\varepsilon \to 0, \quad \bar{X} \doteq \sum_{n=0}^{\infty} \alpha_n(\varepsilon) \bar{X}_n(q), \quad q = \frac{p}{\delta(\varepsilon)}. \tag{38}$$

Note that the asymptotic expansion of the function $\bar{X} = \bar{X}(p, \varepsilon)$, series (37) at $\varepsilon \to 0$ and $p = \delta(\varepsilon)q$, $|q| = O(1)$ is a unique one [30] for the prescribed asymptotic sequence $\alpha_n(\varepsilon)$ and, consequently, coincides with equation (38). A comparison of series (36) and (38), with the above remark and relations (35) taken into account, shows that to the asymptotic series expansion of the function $\bar{X}$ in the space of inverse transforms at $\varepsilon \to 0$ and $|p| = O(\delta)$, $\delta = \delta(\varepsilon)$, there corresponds the expansions of the transform X at $\varepsilon \to 0$ and $t = O(1/\delta)$, and vice versa. These properties of asymptotic expansions make it possible to easily extend the results, obtained in the space of transforms, to the inverse transform. Some general statements regarding the conformity of convergence of series (36) and (38) can be obtained by simple reformulation of the theorem given in ref. [31]. As applied to the problems of convective heat and mass transfer, the said means that the 'contraction' ('extension') of the complex parameter p in the space of transforms corresponds to 'extension' ('contraction') of the time t in the space of inverse transforms; in this case, the correspondence $t = O(Pe^k) \Leftrightarrow |p| = O(Pe^{-k})$ is fulfilled. (Here, the most simple case of the power-law dependence of δ on Pe is given.)

7. THE FINAL STAGE OF THE PROCESS

After applying the Laplace–Carlson transformation for time, the problem, equations (3)–(6), takes on the form

$$p(\bar{c}_1 - 1) + (\mathbf{u}_1 \nabla)\bar{c}_1 = \kappa\, Pe^{-1}\, \Delta \bar{c}_1 \quad (1 < r < \infty), \tag{39}$$

$$p\bar{c}_2 + (\mathbf{u}_2 \nabla)\bar{c}_2 = Pe^{-1}\, \Delta \bar{c}_2 \quad (0 \leqslant r < 1), \tag{40}$$

$$r \to \infty, \bar{c}_1 \to 1; \; r = 1, \quad \bar{c}_2 = \mathbf{L} \cdot F(c_1), \quad \kappa \frac{\partial \bar{c}_1}{\partial r} = \frac{\partial \bar{c}_2}{\partial r}, \tag{41}$$

$$\bar{c}_i = \bar{c}_i(\mathbf{r}, p) = \mathbf{L} \cdot c_i(\mathbf{r}, t); \quad i = 1, 2.$$

In order to carry out the analysis of the problem, equations (3)–(6), one can make use of the remark made in Section 6, which establishes the correspondence between the asymptotic expansions of the boundary-value problems for the transform, equations (39)–(41), and for the inverse transform, equations (3)–(6).

Consider the limiting case

$$Pe \to \infty, \quad p = q\, Pe^{-1}, \quad |q| = O(1), \tag{42}$$

which, by virtue of the results obtained in Section 6, will correspond to the behaviour of the inverse transform c_i at large times $t = O(Pe)$.

The solutions in the flow cores are sought in the form of regular expansions in negative powers of Peclet number

$$\bar{c}_i = \bar{c}_{i0}^{(\mathrm{e})} + Pe^{-1} \bar{c}_{i1}^{(\mathrm{e})} + \ldots, \bar{c}_{i0}^{(\mathrm{e})} / \bar{c}_{i1}^{(\mathrm{e})} = O(1),$$
$$\bar{c}_{ij}^{(\mathrm{e})} = \bar{c}_{ij}^{(\mathrm{e})}(r, \theta) \quad (i = 1, 2; j = 0, 1). \tag{43}$$

By substituting equation (43), with equation (42) taken into account, into equations and boundary conditions (39)–(41) and separating the terms at the same powers of Peclet number, one can obtain the following relations for the first terms of the expansion in the flow core

$$(\mathbf{u}_1\nabla)\bar{c}_{10}^{(e)} = 0; \quad r \to \infty, \bar{c}_{10}^{(e)} \to 1, \tag{44}$$

$$(\mathbf{u}_1\nabla)\bar{c}_{11}^{(e)} = \kappa\Delta\bar{c}_{10}^{(e)} - q(\bar{c}_{10}^{(e)} - 1); \quad r \to \infty, \bar{c}_{11}^{(e)} \to 0, \tag{45}$$

$$(\mathbf{u}_2\nabla)\bar{c}_{20}^{(e)} = 0; \tag{46}$$

$$(\mathbf{u}_2\nabla)\bar{c}_{21}^{(e)} = \Delta\bar{c}_{20}^{(e)} - q\bar{c}_{20}^{(e)}. \tag{47}$$

As earlier, a 'stretched' variable Y is introduced in the outer and inner diffusional boundary layers, d_i, and the solution is sought in the form of expansions

$$\bar{c}_i = \bar{c}_{i0}^{(d)} + Pe^{-1/2}\bar{c}_{i1}^{(d)} + \ldots, \bar{c}_{i0}^{(d)}/\bar{c}_{i1}^{(d)} = O(1),$$
$$\bar{c}_{ij}^{(d)} = \bar{c}_{ij}^{(d)}(Y, \mu), \tag{48}$$
$$Y = Pe^{1/2}(1 - r), \quad \mu = \cos\theta \quad (i = 1, 2; j = 0, 1).$$

The substitution of equation (48) into equations (39)–(41) with allowance for equation (42) and the subsequent series expansion in $Pe^{-1/2}$ leads to the following boundary-layer equations and boundary conditions [the differential operator $\Lambda^{(i)}$ has been determined in equation (10)]

$$\Lambda^{(i)} \cdot \bar{c}_{i0}^{(d)} = 0; \quad i = 1, 2, \tag{49}$$

$$\Lambda^{(1)} \cdot \bar{c}_{11}^{(d)} = (1 - \tfrac{3}{2}\beta)\mu Y^2 \frac{\partial \bar{c}_{10}^{(d)}}{\partial Y} - \tfrac{3}{2}\beta Y(1 - \mu^2) \times \frac{\partial \bar{c}_{10}^{(d)}}{\partial \mu} - 2\kappa \frac{\partial \bar{c}_{10}^{(d)}}{\partial Y}, \tag{50}$$

$$\Lambda^{(2)} \cdot \bar{c}_{21}^{(d)} = -\tfrac{1}{2}\mu Y^2 \frac{\partial \bar{c}_{20}^{(d)}}{\partial Y} - \tfrac{3}{2}Y(1 - \mu^2) \times \frac{\partial \bar{c}_{20}^{(d)}}{\partial \mu} - 2\frac{\partial \bar{c}_{20}^{(d)}}{\partial Y}, \tag{51}$$

$$Y = 0, \bar{c}_{20}^{(d)} = \mathbf{L} \cdot F(c_{10}^{(d)}), \bar{c}_{21}^{(d)} = \mathbf{L} \cdot (\gamma c_{11}^{(d)}),$$
$$\gamma = \left.\frac{\partial F}{\partial c_1}\right|_{c_1 = c_{10}^{(d)}}, \tag{52}$$

$$Y = 0, \kappa \frac{\partial \bar{c}_{ij}^{(d)}}{\partial Y} = \frac{\partial \bar{c}_{2j}^{(d)}}{\partial Y}; \quad j = 0, 1.$$

In the convective boundary-layer region of the diffusional wake $W_2^{(1)}$ inside of a droplet the solution is sought in the form of the expansion

$$\bar{c}_2 = \bar{c}_{20}^{(1)} + Pe^{-1/2}\bar{c}_{21}^{(1)} + \ldots, \bar{c}_{20}^{(1)}/\bar{c}_{21}^{(1)} = O(1),$$
$$\bar{c}_{2j}^{(1)} = \bar{c}_{2j}^{(1)}(z, \eta), \eta = Pe^{1/4}\rho, \tag{53}$$

the terms of which satisfy the following equations

$$(1 - z^2)\frac{\partial \bar{c}_{20}^{(1)}}{\partial z} + \eta z \frac{\partial \bar{c}_{20}^{(1)}}{\partial \eta} = 0, \tag{54}$$

$$(1 - z^2)\frac{\partial \bar{c}_{21}^{(1)}}{\partial z} + \eta z \frac{\partial \bar{c}_{21}^{(1)}}{\partial \eta} = 2\eta^2 \frac{\partial \bar{c}_{20}^{(1)}}{\partial z} - 2\frac{\partial^2 \bar{c}_{20}^{(1)}}{\partial \eta^2}.$$

Equations and boundary conditions (44)–(47), (49)–(52), (54) should be supplemented with the conditions of the asymptotic matching of solutions on general boundaries [21, 29, 30], i.e. the solutions in the cores of the outer and inner flow e_i should match the solutions of the outer and inner diffusional boundary layer d_i; the solution in the convective boundary-layer region of the inner diffusional wake $W_2^{(1)}$ should match the solution of the boundary layer d_2 and the solution in the flow core inside of a droplet e_2. For the main terms of the asymptotic expansions (43), (48), and (53) the conditions of matching are written as

$$\begin{aligned}
&\bar{c}_{10}^{(e)}(r \to 1) = \bar{c}_{10}^{(d)}(Y \to -\infty),\\
&\qquad\bar{c}_{20}^{(e)}(r \to 1) = \bar{c}_{20}^{(d)}(Y \to +\infty),\\
&\bar{c}_{20}^{(e)}(\rho \to 0) = \bar{c}_{20}^{(1)}(\eta \to \infty),\\
&\qquad\bar{c}_{20}^{(d)}(\mu \to \pm 1) = \bar{c}_{20}^{(1)}(z \to \pm 1).
\end{aligned} \tag{55}$$

It follows from equations (44) and (46) that the zero terms of the expansion in the outer and inner flow core depend only on the stream function

$$\bar{c}_{i0}^{(e)} = \bar{c}_{i0}^{(e)}(\psi^{(i)}); \quad i = 1, 2. \tag{56}$$

The asymptotic solution of the problem, equations (39)–(41), will be constructed in succession starting from the core of the outer flow

$$e_1 \to d_1 \to d_2 \to e_2 \rightleftarrows W_2^{(1)}.$$

The boundary conditions at infinity, equations (44) and (45), and equations (45), with equation (56) taken into account, yield the first terms of the expansion in the flow core outside of a droplet in the form

$$\bar{c}_{10}^{(e)} = 1, \quad \bar{c}_{11}^{(e)} = 0. \tag{57}$$

A direct verification shows that the higher term of expansion of the outer diffusional boundary layer, being the solution of equation (49) at $i = 1$ and satisfying the condition of matching with the solution in the flow core, equations (55) and (57), has the form

$$\bar{c}_{10}^{(d)} = 1. \tag{58}$$

Taking into account the equality $c_{10}^{(d)} = 1$ [which follows from equation (58)], it is not difficult to verify in a similar way that the main term of the expansion of the inner diffusional boundary layer, which is the solution of equation (49) at $i = 2$ and which satisfies the boundary conditions on the droplet surface, equation (52), is determined by the expression

$$\bar{c}_{20}^{(d)} = F(1). \tag{59}$$

The distribution of concentration in the convective boundary-layer region inside of a droplet $W_2^{(1)}$ is determined by the solution of equation (54) with the boundary condition $|z| \to 1, \bar{c}_{20}^{(1)} = F(1)$ [which follows from the condition for the matching of the solutions $\bar{c}_{20}^{(d)}$ and $\bar{c}_{20}^{(d)}$, equation (55)] and has the form

$$\bar{c}_{20}^{(1)} = F(1). \tag{60}$$

The condition for the matching of solutions in the regions d_2, $W_2^{(1)}$ and e_2, equation (55), by virtue of equations (59) and (60), specifies the boundary condition in the explicit form for the equation in the flow core inside of a droplet

$$\psi^{(2)} = 0, \quad \bar{c}_{20}^{(e)} = F(1). \tag{61}$$

Equation (46), the general solution of which has the form of equation (56), and boundary condition (61) turn to be insufficient for the determination of concentration field in the flow core. In order to obtain the required additional information about the function $\bar{c}_{20}^{(e)}$, one can make use of the equation for the next term of expansion (47). For convenience, introduce a new orthogonal system of coordinates $\psi^{(2)}, \eta, \varphi$, which is associated with the streamlines (the fixed curves $\eta = \text{const.}$ are orthogonal to the streamlines $\psi^{(2)} = \text{const.}$) and which, accurate to stretching along $\psi^{(2)}(\psi^{(2)} \to \xi = 16\psi^{(2)})$, was used in ref. [11]

$$\psi \equiv \psi^{(2)} = \tfrac{1}{4}r^2(1-r^2)\sin^2\theta, \quad \eta = \frac{r^4\cos^4\theta}{2r^2-1},$$

$$g_{\psi\psi} = \frac{4}{r^2\sin^2\theta\Xi}, \quad g_{\eta\eta} = \frac{(2r^2-1)^4}{16r^6\cos^6\theta\Xi},$$

$$g_{\phi\phi} = r^2\sin^2\theta,$$

$$\Xi = \Xi(r,\theta) = (1-r^2)^2\cos^2\theta + (2r^2-1)^2\sin^2\theta, \tag{62}$$

$$g = g_{\psi\psi}g_{\eta\eta}g_{\phi\phi}$$

$$(0 \leqslant \psi \leqslant 1/16, -\infty < \eta < +\infty).$$

Equation (47) in the system of coordinates, equation (62), takes on the form

$$\frac{\partial\bar{c}_{21}^{(e)}}{\partial\eta} = \left[\frac{\partial}{\partial\psi}\left(\frac{\sqrt{g}}{g_{\psi\psi}}\frac{\partial\bar{c}_{20}^{(e)}}{\partial\psi}\right) + \frac{\partial}{\partial\eta}\left(\frac{\sqrt{g}}{g_{\eta\eta}}\frac{\partial\bar{c}_{20}^{(e)}}{\partial\eta}\right)\right] - q\sqrt{g}\bar{c}_{20}^{(e)}. \tag{63}$$

The procedure which will be used is similar to a successive elimination of the secular (increasing) terms in the method of many scales [29, 30]. Due to the continuity of the solution of the initial problem, equations (3)–(6), the following equality should hold along the fixed streamline $\psi = \text{const.}$

$$c_2(\psi, \eta = -\infty) = c_2(\psi, \eta = +\infty), \tag{64}$$

which means that different points $(\psi, -\infty)$ and $(\psi, +\infty)$ correspond, in the new coordinate system ψ, η, to the same point $(1/\sqrt{2}, \theta)$ in the former spherical coordinate system r, θ. The integration of equation (63) over η: $-\infty < \eta < +\infty$, with equality (64) taken into account (the consequences of which are similar relations for all the terms of expansion of the solution transform in the flow core), leads to the following ordinary differential equation

$$\frac{\mathrm{d}}{\mathrm{d}\psi}\left[J(\psi)\frac{\mathrm{d}\bar{c}_{20}^{(e)}}{\mathrm{d}\psi}\right] - qT(\psi)\bar{c}_{20}^{(e)} = 0; \quad \psi = 0,$$

$$\bar{c}_{20}^{(e)} = F(1), J(\psi) = \tfrac{1}{6}\sqrt{(1+\sqrt{\xi})}\left[(4-3\xi)E\sqrt{\left(\frac{1-\sqrt{\xi}}{1+\sqrt{\xi}}\right)} - (4\sqrt{\xi}-3\xi)K\sqrt{\left(\frac{1-\sqrt{\xi}}{1+\sqrt{\xi}}\right)}\right]. \tag{65}$$

Here, E is the full elliptical second-kind integral, $T = T(\psi)$ is the characteristic time of delay, equation (27), $\xi = 16\psi$.

The coefficients of equation (65) have the following singularities: $\psi \to 0$, $T(\psi) = O(\ln\psi)$; $\psi \to 1/16$, $J(\psi) = O(1/16-\psi)$. The local analysis of equation (65) shows that the first derivative $\bar{c}_{20}^{(e)}$ is finite at zero, while the second is infinite and has a logarithmic singularity; the second singularity at the point of generacy $\psi = 1/16$ determines two linearly independent solutions, one of which is infinite at $\psi \to 1/16$. This means that, besides the boundary condition, equation (61), the condition of the solution limitness at $\psi = 1/16$ should be used for equation (65).

The expansion of the solution of equation (65) at $\psi \to 0$ has the form $\bar{c}_{20}^{(e)} = F(1) + A\psi + O(\psi)$ [where $A(q)q^{-1}$ is the transform of the mean concentration in the flow core]. Therefore, with regard for equation (58) and for the second equality in equation (57), the boundary conditions on the droplet surface, equation (52), $(j = 1)$ and at infinity (corresponding to the asymptotic matching in the regions e_1, d_1 and e_2, d_2) for the second terms of expansions in the regions of the inner and outer diffusional boundary layer have the form

$$Y = 0, \quad \bar{c}_{21}^{(d)} = \gamma\bar{c}_{11}^{(d)}, \quad \kappa\frac{\partial\bar{c}_{11}^{(d)}}{\partial Y} = \frac{\partial\bar{c}_{21}^{(d)}}{\partial Y}$$

$$\times\left(\gamma = \left.\frac{\mathrm{d}F}{\mathrm{d}c_1}\right|_{c_1=1}\right), \quad Y \to -\infty, \quad \bar{c}_{11}^{(d)} \to 0;$$

$$Y \to +\infty, \quad \bar{c}_{21}^{(d)} \to \tfrac{1}{2}AY(1-\mu^2). \tag{66}$$

Equations (50) and (51), the RHSs of which vanish because of equations (58) and (59) and boundary conditions (66), determine the second terms of the expansion in the outer and inner boundary layer of the droplet.

It should be noted that the higher term of expansion in the flow core, which is determined by solving equation (65) is, due to equalities (59) and (60), uniformly adequate (in large Peclet number) over the entire flow region inside of a droplet. For this reason, $c_{20}^{(e)}$ will be further replaced simply by c_{20}.

Using the inverse transform, one obtains the following unsteady-state equation for the concentration distribution inside of a droplet

$$T(\psi)\frac{\partial c_{20}}{\partial\tau} = \frac{\partial}{\partial\psi}J(\psi)\frac{\partial c_{20}}{\partial\psi}; \quad \tau = Pe^{-1}t \equiv a^{-2}D_2t_*, \tag{67}$$

$$\tau = 0, \quad c_{20} = 0; \quad \psi = 0, \quad c_{20} = F(1)(|c_{20}| < \infty).$$

Equation (67) at $F(1) = 1$, after the replacement $\xi = 16\psi$ and $c = 1-c_{20}$, coincides with the Kronig–Brink mathematical model [11] suggested to describe the mass transfer process dynamics inside of a droplet at the limiting resistance of the dispersed phase, equations (4) and (8). Therefore, for the mean concentration, corresponding to equation (67), one has

$$\langle c_{20}\rangle = F(1)\left[1-\sum_{m=1}^{\infty}\alpha_m\exp(-\lambda_m\tau)\right] \quad (68)$$

$$\alpha_1 = 0.6831,\quad \alpha_2 = 0.0981,\quad \alpha_3 = 0.0813,$$

$$\alpha_4 = 0.0618,\quad \alpha_5 = 0.0057,\quad \lambda_1 = 26.844,$$

$$\lambda_2 = 137.91,\quad \lambda_3 = 315.66,\quad \lambda_4 = 724.98,$$

$$\lambda_5 = 1205.2.$$

Here, the coefficients α_m and λ_m are such as given in ref. [9]; at $m = 1, 2$ the values of these coefficients, close to the above, are calculated in ref. [11].

It follows from the above analysis that the results of ref. [11] [corresponding to the solution of the internal problem, equations (4) and (8)] are applicable only in the final stage of the process at $t \geqslant O(Pe)$ and are inapplicable in the initial $[0 \leqslant t < O(\ln Pe)]$ and intermediate $[O(\ln Pe) \leqslant t < O(Pe)]$ stages.

Remark. In deriving equation (67), only the convective boundary-layer region of the diffusional wake $W_2^{(1)}$ was considered. Similarly, one can investigate the distribution of concentration in the regions of the rear and forward stagnation points $W_2^{(3)}$ and b_2, and also of the inner region of the wake $W_2^{(2)}$. The equations for the main terms of asymptotic expansions of the concentration field in these regions in appropriate variables coincide with those obtained in ref. [26] and can be presented in terms of the sum of the first and second derivatives with the factors depending on coordinates. Therefore, all of these equations can have a particular solution equal to $F(1)$, which satisfies the conditions of matching with all the main terms of expansions in the regions e_2, d_2 and $W_2^{(1)}$: This means that the solution of equation (67) is uniformly adequate in Peclet number over the entire region inside of a droplet (including also all the regions of the wake) at $t \geqslant O(Pe)$.

8. SOME GENERALIZATIONS

The results of the asymptotic analysis carried out above can be easily extended to the general case of convective mass and heat transfer of a droplet of any shape in an arbitrary stationary two-dimensional flow of incompressible (ideal, viscous, etc.) liquid at commensurable phase resistances.

The initial stage of the process $0 \leqslant t < O(\ln Pe)$

Here, one can show that the distribution of concentration in diffusional boundary layers on each side of the droplet surface is prescribed as

$$c_i^{(\mathrm{d})} = A_i + B_i\,\mathrm{erf}\left\{\frac{|\zeta|}{2\kappa_i^{1/2}[x(s)-x(\sigma(\omega))]^{1/2}}\right\} \quad (i = 1, 2), \quad (69)$$

$$\zeta = Pe^{1/2}\,\psi^{(i)} = Pe^{1/2}\,\xi u_s,\quad x = x(s) = \int_0^s u_s\,\mathrm{d}s,$$

$$\omega = \omega(t, s) = t - \int_{s_0}^{s}\frac{\mathrm{d}s}{u_s};\quad s \equiv \sigma(\omega(0, s)).$$

Here ξ, s is the local system of coordinates naturally connected with the droplet surface $\psi^{(2)} = 0$ [the coordinate ξ is directed along the normal and is reckoned from the droplet surface, while s is directed along the surface and is reckoned from the critical point of flow incidence $s = s^- = O(u_s(s^-) = 0)$, in the vicinity of which the liquid velocity is directed toward the droplet surface]; $u_s = u_s(s) \geqslant 0$ is the liquid velocity on the interface surface; the last identity in the system of equations (69) allows the determination of the functional dependence $\sigma = \sigma(s)$; as earlier, the coefficients A_i and B_i are prescribed by relations (17) and (18); $u_s(s_0) \neq 0$.

Equation (69) is valid up to the critical runoff points $s^+ : u_s(s^+) = 0$, in the vicinity of which the normal liquid velocity is directed from the droplet surface. If the interface surface has several critical incidence points s_k^- $(k = 1, \ldots, K)$, then there will be the same number of diffusional boundary layers located on each side of the droplet surface. The distribution of concentration in each of these boundary layers will also be determined by formulae (69), where the function u_s and the lower limits in the integrals, which determine the variables x and ω, should be replaced by $|u_s|$, s_k^- and s_{k0}, respectively (s_{k0} is any nonsingular point in the interior of the kth boundary layer $u_s(s_{k0}) \neq 0$); in this case, each boundary layer will have its own function σ.

Using formulae (69), it is not difficult to obtain the distribution of concentration in diffusional boundary layers in the initial stage of the process in the case of convective mass transfer of a spherical droplet in an axisymmetric linear shear flow, when the liquid velocity field outside and inside of a droplet is determined by the following dimensionless expressions for the stream function [32]

$$\psi^{(1)} = -\tfrac{1}{4}[2(\beta+1)r^3-(5\beta+2)+3\beta r^{-2}] \times \sin^2\theta\cos\theta,\quad 1 \leqslant r < \infty, \quad (70)$$

$$\psi^{(2)} = \tfrac{3}{4}r^3(1-r^2)\sin^2\theta\cos\theta,\quad 0 \leqslant r \leqslant 1.$$

The characteristic velocity scale in equation (70) was chosen to be $U = aG(\beta+1)^{-1}$, where G is the shear factor.

The flow field, equation (70), is characterized by the critical plane of incidence $\theta = \pi/2$, along which the streamlines arrive at the droplet surface from infinity, and by two runoff trajectories $\theta = 0$ and π, which 'emerge' from isolated singular points on the droplet surface. The diffusional boundary layers, originating in the vicinity of the flow incidence line $\theta = \pi/2, r = 1$, are located on both sides near the droplet surface.

Omitting the intermediate calculations (which are analogous to those used in ref. [33] for the limiting resistance of the continuous phase), one obtains that at commensurable phase resistances, equations (3)–(6), the distribution of concentration in diffusional boundary layers in the initial stage of the process in the case of linear shear flow around a spherical droplet, equation (70), is determined by the formula

$$c_i^{(d)} = A_i + B_i \operatorname{erf}\left\{\left(\frac{3Pe}{2\kappa_i}\right)^{1/2} \times \frac{|r-1|\,|\cos\theta|}{[(\sin^2\theta + e^{-3t}\cos^2\theta)^{-2} - 1]^{1/2}}\right\}. \quad (71)$$

Here, the following notation is introduced:

$$Pe = a^2 G(\beta+1)^{-1} D_2^{-1}, \quad t = G(\beta+1)^{-1} t_*;$$

the coefficients A_i and B_i are prescribed by expressions (17) and (18).

The use of relations (19) and (71) yields the following formula for the mean Sherwood number

$$Sh = \lambda \left[\frac{3Pe}{2\pi} \coth\left(\tfrac{3}{2}t\right)\right]^{1/2}, \quad (72)$$

where the parameter λ is the root of equation (18).

The intermediate stage of the process $O(\ln Pe \leqslant t < O(Pe)$

The situation originating in this case is determined by the interaction of the diffusional boundary layer with the wake inside of a droplet (Fig. 1). For a purely internal problem, equations (4) and (29), when the flow field inside of a droplet has the same qualitative aspects as the Hadamard–Rybczinski flow, equation (7) (an axisymmetric case characterized by the presence of only two singular isolated critical points $s^- = 0$ and s^+ and by the absence of critical lines on the droplet surface), it can be shown in a general case that the distribution of concentration in the diffusional boundary layer is prescribed by the expression

$$c_2^{(d)} = \int_0^\infty G(\zeta, \zeta^*, x) f(\zeta^*, \omega)\, d\zeta^* + \operatorname{erfc}\left\{\frac{\zeta}{2[x - x(\sigma(\omega))]^{1/2}}\right\}. \quad (73)$$

Here, the variables ζ, x, ω and the function σ are the same as in formula (69), while the core of the integral operator $f(\zeta, \omega)$ is found by solving the following equation with a delayed argument

$$f(\zeta, \omega) = \int_0^\infty G(\zeta, \zeta^*, x_0) f(\zeta^*, \omega - T)\, d\zeta^* + \operatorname{erfc}\left\{\frac{\zeta}{2[x_0 - x(\sigma(\omega - T))]^{1/2}}\right\}, \quad (74)$$

$$T = \left(\oint \frac{ds}{|\mathbf{u}_2|}\right)_{\psi^{(2)} = Pe^{-1/2}\zeta}, \quad x_0 = x(s^+).$$

It should be taken into account in solving equation (74) that the function f should satisfy the condition (34), while the characteristic time of delay $T = O(\ln Pe)$ is determined by the higher terms of the expansion of $T = T(\psi^{(2)})$ in the limiting case, equation (28).

The final stage of the process $O(Pe) \leqslant t$

In this stage, the distribution of concentration outside of a droplet is virtually uniform and is governed by the respective value at infinity, while inside of a droplet there is a substantially unsteady-state process described by the boundary-value problem, equation (67), with the following expressions for equation coefficients

$$T(\psi) = \oint \sqrt{g}\, d\eta \equiv \oint \frac{ds}{|\mathbf{u}_2|}, \quad J(\psi) = \frac{g}{g_{\psi\psi}}\, d\eta,$$

$$g = g_{\psi\psi} g_{\eta\eta} g_{\phi\phi}. \quad (75)$$

Here, ψ, η, φ is the orthogonal coordinate system connected with the stream function $\psi = \psi^{(2)}$, which determines the flow inside of a droplet; the curvilinear coordinate η, orthogonal to ψ, is found by solving the equation $(\nabla\psi \cdot \nabla\eta) = 0$ (for a plane case it should be assumed that $g_{\phi\phi} = 1$, while in the axisymmetric case —that $g_{\phi\phi} = r^2 \sin^2\theta$). In those cases when inside of a droplet there are several independent regions Ω_j $(j = 1, \ldots, n)$ with completely closed streamlines and these regions are limited by singular (critical) streamlines or streamsurfaces which close on the droplet surface (in particular, in the case of linear shear flow around a spherical droplet, equation (70), one has $j = 2$; the flow at $j = 4$ was considered in refs. [7, 8]), equation (67) should be written and solved independently in all the regions Ω_j (in this case, each region Ω_j has its own functions T_j and J_j), taking into account the fact that at $\tau \geqslant O(1)$ the same amount of concentration, equal to $F(1)$, is transported from the droplet surface to the singular streamlines.

9. SOME CONCLUSIONS AND REMARKS

Summarizing the results of the aforegoing asymptotic analysis, it is necessary to point out the following:

(1) The boundary-layer solution, equations (16)–(18), which for the linear case $F(c_1) = \alpha c_1$ was obtained in refs. [3, 4, 6], is valid only in the initial stage of the process (before the diffusional boundary-layer starts to interact with the wake inside of a droplet) at $0 \leqslant t < O(\ln Pe)$ (this very interval of times determines also the region of applicability of the results of refs. [7, 8]) and is inapplicable at $t \geqslant O(\ln Pe)$.

(2) In the intermediate stage of the process $O(\ln Pe) \leqslant t < O(Pe)$, characterized by intensive interaction between the inner boundary layer and the wake, the distribution concentration in the diffusional boundary layer (in the case when the resistance to transport is fully concentrated inside of a droplet) is prescribed by formula (31), where the function f is determined by the solution of the integral equation with a delayed argument, equation (33).

(3) Equation (67), corresponding to the unsteady-state mass exchange between a droplet and a continuous medium at commensurable phase resistances, equations (3)–(7), is valid only in the final stage of the process at $t \geqslant O(Pe)$ [and is inapplicable at $t < O(Pe)$] and in form [at $F(1) = 1$] coincides with the

Kronig–Brink equation [11] obtained for a purely internal problem.

The first and the third points mean that the characteristic dimensional time scale, which corresponds to equations (16)–(18), is the quantity t_h, and that corresponding to equation (67), is the quantity t_d, equation (9).

It is clear from the above why the results of many experimental investigations carried out at the limiting resistance of the dispersed phase are in a good agreement with the solution presented in ref. [11] and fail to agree with the solution suggested in refs. [3, 4, 6] (this comparison is carried out in ref. [8]). This is due to the fact that as yet there is no reliable experimental technique to investigate the initial (rapidly progressing) stage of the process, and the experiment correctly 'catches hold' of only the final (slow) stage of the process, which is described by equation (67).

The asymptotic analysis carried out should be used as a basis for the development of adequate finite-difference computational methods of investigation of similar unsteady-state problems with a small parameter at higher derivatives. The results of numerical calculations carried out in refs. [12–20] can hardly be considered convincing, as the mesh steps of the grid used for the region near the flow axis were not made finer (moreover, the grid should be nonuniform in time), which is necessary to correctly account for the interaction between the diffusional boundary layer and the wake inside of a droplet.

In view of the initial condition $\tau = 0$, $c_{20} = 0$, the asymptotics of the solution of equation (67) is a 'through' one in time t in the sense that in order to obtain a correct result at $t = O(Pe)$ there is no need to know the previous nonuniform distribution of concentration in the flow at smaller times $t = O(Pe^{\delta})$, $0 \leqslant \delta < 1$. It should be noted that the equation and boundary condition on the droplet surface, equation (67), at $t = O(Pe)$ can be obtained directly from the analysis of the unsteady-state problem, equations (3)–(7), without resorting to the Laplace–Carlson transformation. But then, the problem would have remained unsolved regarding a correct specification of the initial condition for concentration distribution in the flow core at $t = O(Pe)$, which is unknown at sufficiently large times $O(\ln Pe) \leqslant t < O(Pe)$ and may differ substantially from the initial uniform distribution $c_2 = 0$ [in principle, it follows from nowhere that the initial condition for the 'contracted' variable $\tau = Pe^{-1}t$ can be 'carried' throughout the entire region $0 \leqslant t < O(Pe)$].

The validity of equation (67) at $t \geqslant O(Pe)$ means physically that at sufficiently large times ($t \geqslant O(Pe)$) a steady-state regime actually develops outside of the droplet, which corresponds to a uniform distribution of concentration in the flow, while inside of a droplet there is a substantially unsteady-state process with constant concentration on its surface determined by the phase equilibrium condition (1) with a stationary uniform concentration field outside of the droplet (by this time the diffusional boundary layer and the diffusional wake have been actually smeared out and ceased to exist). Moreover, it follows from equality (60) that concentration on the flow axis inside of a droplet at $t \geqslant O(Pe)$ has already become equal to that on its surface (due to multiple circulation of dissolved substance along the closed streamlines).

It should be noted that though equations and boundary conditions, equations (3)–(6), have been formulated for a concentrational problem, all of the results obtained can easily be extended to a corresponding thermal problem which is described by equations and initial conditions, equations (3)–(5), at $c_1 \equiv \Theta_1$ and $c_2 \equiv \Theta_2$ with the following boundary conditions on the droplet surface

$$r = 1, \quad \Theta_1 = \Theta_2, \quad \left(\frac{\lambda_1}{\lambda_2}\right)\frac{\partial \Theta_1}{\partial r} = \frac{\partial \Theta_2}{\partial r}$$

$$\times \left(\Theta_i = \frac{T_{20} - T_i}{T_{20} - T_{10}}; \quad i = 1, 2\right),$$

where T_i, T_{i0} and λ_i are the instantaneous temperature, the temperature at the initial time and thermal conductivity of liquid outside ($i = 1$) and inside ($i = 2$) of the droplet, respectively. In particular, for the mean temperature distribution inside of a droplet at $t \geqslant O(Pe)$, formula (68) remains to be valid at $\langle c_2 \rangle \equiv \langle \Theta_2 \rangle$ and $F(1) = 1$.

Investigate now the behaviour of the solution of equations (3)–(7) depending on a change in the droplet and surrounding liquid viscosity ratio β. The analysis carried out corresponded to the case $\beta = O(1)$ and, therefore, the final results did not depend on the parameter β. In the limiting case $\beta \gg 1$, the flow inside of the droplet is practically absent ($\beta = \infty$ corresponds to a solid particle). It is possible to show that at large Peclet numbers $Pe_{\infty} = aU_{\infty}D_1^{-1} \gg 1$ [note that Pe_{∞} differs by a factor $\kappa(\beta + 1)$ from the Pe number used in equations (3)–(6)] and $\tau = a^{-2}D_2t_{*} \geqslant O(1)$, the distribution of concentration (temperature) in the flow outside the particle is uniform (in the main approximation, of course) $c_1 = 1$, while the distribution of concentration inside the particle is determined by the ordinary unsteady-state heat conduction equation [which corresponds to $\mathbf{u}_2 = 0$ in equation (4)]

$$\frac{\partial c_2}{\partial \tau} = \Delta c_2; \quad \tau = 0, c_2 = 0; \quad r = 1, c_2 = F(1). \quad (76)$$

The solution of equation (76) leads to the following expression for the mean concentration inside a droplet [9]

$$\langle c_2 \rangle = F(1)\left[1 - \frac{6}{\pi^2}\sum_{m=1}^{\infty}\frac{1}{m^2}\exp(-\pi^2 m\tau)\right]. \quad (77)$$

A comparison between formulae (68) and (77) shows that the order of the characteristic times required for the development of the unsteady-state heat and mass transfer process for a droplet with moderate ($\beta = O(1)$)

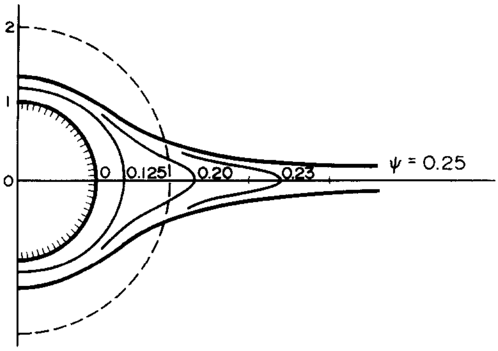

FIG. 3.

and extremely high ($\beta \gg 1$) viscosity is the same and is determined by the quantity $\sim a^2 D_2^{-1}$. However, the decrements of solution decay at $\tau \rightarrow \infty$ differ (substantially) by a factor of 2.72. This means that the presence of liquid circulation inside of a droplet at $\beta = O(1)$ enhances the process of convective mass and heat transfer as compared to a 'solid' droplet ($\beta = \infty$), where the substance transport by circulation is absent. It should also be noted that even a substantial increase in the external flow velocity $U_\infty(Pe \rightarrow \infty)$ exerts a weak effect on the degree of substance extraction from the droplet which is limited from above at $Pe \rightarrow \infty$ and is proportional to the decrement of mean concentration decay, equation (68).

Remark. Equation (67) can also be used for the description of unsteady-state heat and mass transfer in one final stage of the process at $t \geqslant O(Pe)$ and in some external problems, where the regions of closed circulation are formed near the body surface which are similar to the region shown in Fig. 3. In particular, this is the case when a circular cylinder freely rotates in the simplest linear shear flow [34] (see also ref. [35]); in this case at $t \geqslant O(Pe)$ equation and initial condition (67) remain valid in the region with closed streamlines, but these should be supplemented with the boundary condition on the cylinder surface (at $r = 1$). In Fig. 3, the solid line shows a specific limiting streamline $\psi = 1/4$, which is the boundary of the regions with closed $\{0 \leqslant \psi < 1/4\}$ and open $\{1/4 \leqslant \psi < \infty\}$ streamlines. In the region with open streamlines at $t \geqslant O(Pe)$, the concentration is practically uniform and is determined by its initial value $c = 0$.

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EXTRACTION VARIABLE D'UNE GOUTTELETTE TOMBANTE AVEC UNE DEPENDANCE NON LINEAIRE DU COEFFICIENT DE DISTRIBUTION EN FONCTION DE LA CONCENTRATION

Résumé—On présente une analyse asymptotique du problème de transfert variable de chaleur et de masse d'une gouttelette avec des résistances de phase commensurables dans le cas des nombres de Péclet élevés. Le problème est considéré non linéaire puisque dans la condition d'équilibre entre phases à la surface de la goutte, la concentration d'une phase dispersée dépend arbitrairement de la concentration de la phase continue. La dynamique du transfert de chaleur et de masse est qualitativement très différente selon trois intervalles de temps caractéristiques pendant lesquels les frontières dépendent du nombre de Péclet.

INSTATIONÄRE EXTRAKTION AUS EINEM FALLENDEN TROPFEN MIT NICHTLINEARER ABHÄNGIGKEIT DES VERTEILUNGSKOEFFIZIENTEN VON DER KONZENTRATION

Zusammenfassung—Das Problem des instationären Wärme- und Stoffübergangs an einem Tropfen wurde mit Hilfe eines Näherungsverfahrens untersucht, und zwar für den Fall, daß die Phasenwiderstände gleichartig und die Peclet-Zahlen groß sind. Das Problem wird als nichtlinear angesehen, wenn in der Randbedingung des Phasengleichgewichts an der Tropfenoberfläche die Konzentration der dispersen Phase beliebig von der Konzentration der kontinuierlichen Phase abhängt. Es zeigt sich, daß das dynamische Verhalten des Wärme- und Stoffübergangs in drei charakteristischen Zeitintervallen qualitativ sehr unterschiedlich ist, wobei die zeitlichen Grenzen von der Peclet-Zahl abhängen. Das erste Zeitintervall, $0 \leqslant t < O(\ln Pe)$ (t ist die dimensionslose Zeit), ist durch die Ausbildung instationärer Diffusions-Grenzschichten auf beiden Seiten der Tropfenoberfläche (die zunächst qualitativ identisch sind), gekennzeichnet. Die innere Grenzschicht erzeugt einen Diffusionsstrom in der Nähe der Strömungsachse. Während des zweiten Zeitintervalls, $O(\ln Pe) \leqslant t < O(Pe)$ beginnt der im Innern entwickelte Diffusionsstrom die Grenzschicht zu beeinflussen; er "verschmiert" sie. Hier unterscheiden sich die Grenzschichten an der Innen- und an der Außenseite des Tropfens bereits ganz wesentlich. In der Folge nimmt die Dicke der inneren Grenzschicht allmählich zu. Während des letzten Zeitintervalls, $O(Pe) \leqslant t$, ist eine weitere Umordnung des Konzentrationsfeldes zu beobachten, wonach die Grenzschichten praktisch nicht mehr existieren. Die Konzentration außerhalb des Tropfens wird konstant und nimmt den Wert der ungestörten Konzentration in großer Entfernung an, während sich im Inneren des Tropfens ein instationärer Vorgang abspielt. Die Konzentration auf einer jeden Stromlinie hat sich bereits vollständig ausgeglichen (infolge der wiederholten Zirkulation der Flüssigkeit entlang der geschlossenen Stromlinien), während senkrecht zu den Stromlinien ein Massentransport durch molekulare Diffusion stattfindet.

НЕСТАЦИОНАРНАЯ ЭКСТРАКЦИЯ ИЗ ПАДАЮЩЕЙ КАПЛИ ПРИ НЕЛИНЕЙНОЙ ЗАВИСИМОСТИ КОЭФФИЦИЕНТА РАСПРЕДЕЛЕНИЯ ОТ КОНЦЕНТРАЦИИ

Аннотация—В работе проведен асимптотический анализ задачи о нестационарном массо- и теплообмене капли при соизмеримых фазовых сопротивлениях в случае больших чисел Пекле. Задача рассматривается в нелинейной постановке, когда в граничном условии фазового равновесия на поверхности капли концентрация дисперсной фазы произвольным образом зависит от концентрации сплошной фазы. Показано, что динамика процесса массо- и теплопереноса протекает с существенными качественными отличиями на трех характерных интервалах времени, границы которых зависят от числа Пекле. На первом интервале времени $0 \leq t < O(\ln Pe)$ (t—безразмерное время) происходит формирование нестационарных диффузионных пограничных слоев по обе стороны от поверхности капли (которые пока качественно аналогичны друг другу), при этом внутренний пограничный слой порождает диффузионный след, расположенный вблизи оси потока. Во втором интервале времени $O(\ln Pe) \leq t < O(Pe)$ развитый внутренний диффузионный след начинает взаимодействовать с пограничным слоем и сильно "размывает" его (здесь уже погранслои, расположенные вне и внутри капли, существенно различаются), в результате чего толщина внутреннего погранслоя постепенно значительно увеличивается. На последнем интервале времени $O(Pe) \leq t$ происходит дальнейшая перестройка поля концентрации, так что погранслои практически уже прекратили свое существование; при этом вне капли концентрация становится постоянной и равной невозмущенной концентрации на бесконечности, а внутри капли протекает существенно нестационарный процесс, когда на каждой фиксированной линии тока концентрация уже полностью выравнялась (за счет многократной циркуляции жидкости по замкнутым линиям тока), а массопередача осуществляется путем молекулярной диффузии в направлении, перпендикулярном линиям тока.