ON THE MASS TRANSFER IN A MOVING CHAIN OF ABSORBING DROPS WITH ALLOWANCE FOR THE SATURATION EFFECT

A. D. POLIANIN
(Moscow)

Mass transfer in a chain of absorbing drops at low Reynolds and high Péclet numbers is investigated using the simplest mathematical model of internal diffusion, i.e. of total intermixing inside of drops of the substance diffusing through the drop surface. Present investigation is an extension of results obtained in [1—4] which relate to the limit case of short time intervals. It is shown that the allowance for internal diffusion leads to a substantial alteration of the system kinetics owing to the saturation of drops in time. Thus, when the drops are of equal radii, there always exists a time interval in which a particular drop of the chain absorbs more of the substance diffused in the stream than remaining drops of the chain.

The problem of steady convective diffusion in a chain of reacting particles was previously considered [1—4] under condition of total absorption at their surface at high Péclet numbers. It was shown that the interaction of the diffusion boundary layer of every particle with the diffusion trail of the preceding particle results in a considerable decrease of the total diffusion flux to its surface (in comparison with that without such interaction). Since drops can absorb only a limited quantity of the substance dissolved in the stream, such simplification of the problem formulation is justified whenever it is possible to disregard variations of substance concentration inside drops (for instance in the analysis of weak absorption or comparatively short intervals).

1. Statement of the problem. Let us consider the process of convective diffusion in a chain of absorbing drops of the same radius a equally spaced and moving at the same velocity U. We assume that away from the drops the concentration of substance dissolved in the stream is constant and equal C_0, and that inside drops a complete intermixing of the substance diffusing through their surfaces takes place, i.e. that the concentration inside the drop and on its surface is the same and equal C_k^+ (where k is the ordinal number of a drop in the chain). The substance concentration C outside the drop, and C_k^+ inside it are linked by the law of conservation of mass, i.e. the total change of substance inside the drop in a unit of time $^4/_3 \pi a^3 dC_k^+ / dt$ must be equal the total influx of diffusing substance from the surrounding fluid

$$I_k^* = 2\pi a^2 D \int_0^\pi \left[\frac{\partial C}{\partial r_k} \right]_{r_k=a} \sin \theta_k \, d\theta_k$$

where r_k and θ_k are spherical coordinates of a system attached to the center of the k-th drop. It will be seen that the concentration inside a drop depends on its ordinal number and time.

Below we assume that the Reynolds number $R = aU\nu^{-1}$ of the drop is low while the Péclet number $P = aUD^{-1}$ is high (ν is the kinematic viscosity of the fluid and D is the coefficient of diffusion).

It was shown in [5] that under condition of total absorption on the particle surface $C_1^+ = 0$ and $P \gg 1$ the total diffusion flux $I_1^* \sim aDC_0P^{1/(n+1)}$, where $n = 1$, to it corresponds to drops of moderate viscosity $\beta \leqslant O\,(1)$, while $n = 2$ relates to drops of high viscosity $\beta > O\,(P^{1/3})$, where β is the ratio of viscosities of drops and fluid. The value $n = 2$ also corresponds to the case of moderate viscosity drops when surface-active substances are present [5].

Using the law of conservation of mass and the indicated expression for the total diffusion flux I_1^*, we find that the characteristic time of concentration change inside the drop is $\lambda aU^{-1}P^{n/(n+1)}$; the numerical coefficient $\lambda = O\,(1)$ will be defined later. This time is taken below as the basic unit for conversion to dimensionless time τ.

In dimensionless variables the boundary value problem which determines concentrations c and c_k^+ in- and outside drops may be represented in the form

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

$$\varepsilon^{n+1}\left\{\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\,\frac{\partial c}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial \theta}\left(\sin\theta\,\frac{\partial c}{\partial \theta}\right)\right\}$$

$$r_k = 1, \quad c = c_k^+(\tau); \quad r \to \infty, \quad c \to 1; \quad \tau = 0, \quad c = c^*(r,\theta)$$

$$\frac{dc_k^+}{d\tau} = \frac{3\varepsilon\lambda}{4\pi}\,I_k = \frac{3}{2}\,\lambda\int_0^\pi \varepsilon\left[\frac{\partial c}{\partial r_k}\right]_{r_k=1}\sin\theta_k\,d\theta_k \qquad (1.2)$$

$$c_k^+(0) = 0; \quad \varepsilon = P^{-1/(n+1)}, \quad n = 1,2; \quad k = 1,2,\ldots,M$$

where the system of spherical coordinates r, θ is attached to the center of some particular drop, and the characteristic units are: the drop radius a, its velocity U, the time $\lambda aU^{-1}\varepsilon^{-n}$, and concentration C_0; the stream function ψ is assumed known from the solution of the respective problem of hydrodynamics of flow around a body. The initial condition for concentration $c^*\,(r,\theta)$ is defined by the solution of the stationary $(\partial\,/\,\partial\tau = 0)$ equation (1.1) of convective diffusion under condition $c_1^+ = c_2^+ = \ldots = c_M^+ = 0$ of total absorption on drop surfaces of the substance dissolved in the fluid.

Problem (1.1) was thoroughly investigated in [1, 2] in the steady case and $c_1^+ = c_2^+ = \ldots = c_M^+ = \mathrm{const}$ for $n = 1$, and for $n = 2$ it was dealt with in [3, 4]. It was shown, using the method of merging asymptotic expansions in the small parameter ε, that in the stream near each drop there exist several characteristic regions with different mass transfer mechanisms. These are: the external region e, the region $d = \{r - 1 < O\,(\varepsilon),\ O\,(\varepsilon) < \theta\}$ of the diffusion boundary layer (the expression in braces indicate the order of characteristic dimensions of the considered region and subscript k is omitted at spherical coordinates), and the diffusion trail W, which, in turn, consists of four subregions, viz. the convection boundary layer region

$W^{(1)} = \{O\,(\varepsilon) < r - 1 < O\,(\varepsilon^{-1}), O\,(\varepsilon^{n+1}) < \psi < O\,(\varepsilon^n)\}$,   the diffusion trail inner region $W^{(2)} = \{O\,(\varepsilon) < r - 1 < O\,(\varepsilon^{-1}),\quad \psi < O\,(\varepsilon^{n+1})\}$,   the trailing stagnation point region $W^{(3)} = \{r - 1 \ < O\,(\varepsilon),\ \theta < O\,(\varepsilon)\}$, and the mixing region $W^{(4)} = \{O\,(\varepsilon^{-1}) < r - 1,\ \psi \ < O\,(\varepsilon^n)\}$.

As already noted, concentration in the steady case rapidly changes near the drop surface (within the boundary layer) and $[\partial c\,/\,\partial r_k]_{r_k=1} \sim \varepsilon^{-1}$.   Hence the integral in the right-hand side of Eq. (1.2) is of order unity.

In the unsteady problem (1.1), (1.2), as in the steady one, when $\varepsilon \to 0$ the convection term $(v\nabla)\,c$ in regions d, and $W^{(3)}$ is proportional to $O\,(\varepsilon^{n-1})\,c$, and in regions $W^{(1)}$, $W^{(2)}$, and $W^{(4)}$ is proportional to $O\,(1)\,c$, and the order of magnitude of the right-hand side of Eq. (1.1) in the respective regions is as follows: in d, $W^{(3)}$ — $\varepsilon^{n-1}c$, in $W^{(1)}$ — $\varepsilon\,c$, and in $W^{(2)}$ and $W^{(4)}$ — $O\,(1)\,c$. Hence by a successive determination of equations and boundary conditions in a manner similar to that in [1—4] we obtain that in the principle term of expansion in the small parameter ε for concentrations $c_1^+, c_2^+, \ldots, c_k^+; c$ the term $\varepsilon^n \partial c\,/\,\partial\tau$ in Eq. (1.1) is immaterial, while Eqs. (1.2) for concentration inside drops remain in their entirety valid by virtue of $\varepsilon\,[\partial c\,/\,\partial r_k]_{r_k=1} \sim O\,(1)$.

Thus the solution of the input problem (1.1), (1.2) consists of two consecutive stages: 1) solution of the auxilliary problem

$$(v\nabla)\,c = \varepsilon^{n+1}\,\Delta\,c;\ r_k = 1,\ c = c_k^+;\ r = \infty,\ c = 1 \qquad (1.3)$$

for arbitrary c_k^+ in which time τ appears implicitly as a parameter, and 2) solution of the autonomous system of ordinary differential equations

$$4\pi\,dc_k^+\,/\,d\tau = 3\varepsilon\lambda I_k,\ c_k^+\,(0) = 0\ (k = 1, 2, \ldots, M) \qquad (1.4)$$

in which the over-all diffusion flux I_k is calculated by (1.2) using the obtained solution of (1.3) for concentration $c = c\,(r_k, \theta_k;\ c_1^+, \ldots, c_k^+)$.   By virtue of the initial condition for concentration inside drops $c_k^+\,(0) = 0$,   hence the derived solution automatically satisfies the initial condition at $\tau = 0$ in the input problem (1.1),(1.2).

Investigation of the auxilliary problem (1.3) can be effected as in [1—4].   Here we restrict it to the case in which the dimensionless distance satisfies the inequality

$$O\,(1) < l < O\,(\varepsilon^{-1}) \qquad (1.5)$$

The left-hand side of this inequality is subsequently of little consequence, since it is only required for providing a specific form to the stream function near the surfaces of drops (when $1 \ll l$ in Eq. (1.3) the stream function of an isolated drop may be used, see Sect. 2). The right-hand side of this inequality is important, since it shows that the diffusion boundary layer of every drop interacts with the convection boundary layer region of the diffusion trail of the drop ahead of it [1,4].

The system of Eqs. (1.4) contains total fluxes on the drop surfaces which have to be determined by solving the auxilliary problem (1.3). To determine the dependence of the principal term of expansion (in the small parameter ε) of the total diffusion flux I_k on concentration $c_1^+, c_2^+, \ldots, c_k^+$ it is sufficient to calculate the concentration distribution in the diffusion boundary layer of the k -th drop. The obtained expression for total fluxes $I_k = I_k\,(c_1^+, c_2^+, \ldots, c_k^+)$ must be substituted into the system of differential equations (1.4) that defines the variation of concentration inside

the drop in time.

2. Solution of the auxilliary problem. We derive here
the solution of a more general problem than (1.3), (1.5). Let us consider a chain of
absorbing spherical drops of the same radius in a flow of periodic pattern [2,3], in
which the stream function near the surface of any particle can be represented in a
spherical coordinate system attached to the particle center in the form

$$\psi = (r - 1)^n f(\theta), \quad f \gg 0, \quad n = 1, 2 \ (r \to 1) \tag{2.1}$$

If condition (1.5) is satisfied, formulas $f = \frac{1}{2}(1 + \beta)^{-1} \sin^2 \theta \ (n = 1)$ and
$f = \frac{3}{4} \sin^2 \theta \ (n = 2)$ correspond to a Stokes flow around particles of moderate (β
$\leqslant O(1)$) and high ($\beta > O(P^{1/3})$) viscosities, respectively. Concentration distribu-
tion in the diffusion boundary layer of the k-th sphere is defined by the solution of
the boundary value problem with the boundary condition for concentration constancy
on its surface and the condition for the oncoming flow, which is determined by the
concentration distribution in the diffusion trail of the $(k - 1)$-st sphere [1-4]. If
the distance between spheres satisfies the inequality $l < O(\varepsilon^{-1})$, the condition for
the oncoming flow is determined by the convection boundary layer region of the diffus-
ion trail of the preceding sphere.

The derivation of equations, and initial and boundary conditions for concentration
$c_k^{(d)}$ in the diffusion boundary layer of the $(k - 1)$-st particle is analogous to that
in [1-4] and yields the following boundary value problem:

$$\mathbf{L}_n(\xi, t) c_k^{(d)} = 0, \quad \mathbf{L}_n = \partial / \partial t - \xi^{1-n} \partial^2 / \partial \xi^2 \tag{2.2}$$

$$c_k^{(d)}(0, t) = c_k^+, \qquad c_k^{(d)}(\infty, t) = 1 \quad (0 \leqslant t \leqslant t_0)$$

$$c_k^{(d)}(\xi, 0) = c_{k-1}^{(d)}(\xi, t_0), \quad c_0^{(t)} = 1; \quad t_0 = t(0)$$

$$t = t(\theta) = \frac{1}{n} \int_0^\pi \sin\theta f^{1/n}(\theta) d\theta, \quad \xi = \varepsilon^{-1}\psi^{1/n}, \quad k = 1, 2, \dots, M$$

Problem (2.2) was considered in [1-4] for $c_1^+ = c_2^+ = \dots = c_k^+ = \text{const}$.
To solve (2.2) with arbitrary c_k^+ we use the auxilliary function

$$u(\xi, t) = \Gamma^{-1}\left(\frac{1}{n+1}\right) \gamma\left(\frac{1}{n+1}, \frac{\xi^{n+1}}{(n+1)^2 t}\right), \quad \Gamma(x) = \gamma(x, +\infty) \tag{2.3}$$

where γ is an incomplete gamma function, which is the solution of problem

$$\mathbf{L}_n(\xi, t) u = 0; \quad u(0, t) = 0, \quad u(\infty, t) = 1; \quad u(\xi, 0) = 1$$

Taking into account the properties of function $u(\xi, t)$, we can prove by induction
that the solution of problem (2.2) is of the form

$$c_k^{(d)}(\xi, t) = c_k^+ + \sum_{\alpha=1}^k u(\xi, t + (k - \alpha) t_0)(c_{\alpha-1}^+ - c_\alpha^+) \tag{2.4}$$

$$c_0^+ = 1, \quad k = 1, 2, \dots, M$$

When $c_1^+ = c_2^+ = \dots = c_k^+ = 0$, formula (2.4) yields $c_k^{(d)}(\xi, t) = u(\xi,$
$t + (k - 1) t_0)$ [1-4].

For the local and total diffusion fluxes on particle surfaces we have

$$j_k(\theta) = [\partial c_k^{(d)} / \partial r]_{r=1} = \varepsilon^{-1}f^{1/n}(\theta)[\partial c_k^{(d)} / \partial \xi]_{\xi=0} \tag{2.5}$$

$$\varepsilon^{-1}\Gamma^{-1}\left(\frac{1}{n+1}\right)(n+1)^{(n-1)/(n+1)}f^{1/n}(\theta)\times$$

$$\sum_{\alpha=1}^{k}[t(\theta)+(k-\alpha)t_0]^{-1/(n+1)}(c_{\alpha-1}^+ - c_{\alpha}^+)$$

$$I_k = 2\pi\int_0^{\pi}j_k(\theta)\sin\theta\,d\theta = I\sum_{\alpha=1}^{k}a_{k\alpha}(c_{\alpha-1}^+ - c_{\alpha}^+)$$

$$I = 2\pi\varepsilon^{-1}\Gamma^{-1}\left(\frac{1}{n+1}\right)(n+1)^{2\sigma}t_0^{\sigma}, \quad c_0^+ \equiv 1$$

$$a_{k\alpha} = (k-\alpha+1)^{\sigma} - (k-\alpha)^{\sigma}, \quad \sigma = n/(n+1)$$

The results obtained with (2.3) can be extended to chains of particles of arbitrary shape in the case of three-dimensional flow and aperiodic pattern of the flow field (in the presence of two stagnation points on the particle surfaces). The concentration distribution in the diffusion boundary layer and the convection boundary layer regions of the diffusion wake of the k-th particle is defined by formulas

$$c_k^{(d)}(\xi, t) = c_k^+ + \sum_{\alpha=1}^{k}u\left(\xi, t+\sum_{v=\alpha}^{k-1}t_{v0}\right)(c_{\alpha-1}^+ - c_{\alpha}^+) \tag{2.6}$$

$$c_k^{(1)}(\xi) = c_k^{(d)}(\xi, t_{k0}), \quad c_0^+ \equiv 1 \quad (0 \leqslant t \leqslant t_{k0}) \quad k = 1, 2, \ldots, M$$

where $\xi = \varepsilon^{-1}\Phi^{1/n}$ (Φ is the analog of the stream function) and the constant t_{v0} and variable t are determined by the local flow field near the particle surfaces [2, 3]. For the total diffusion flux formula (2.5) remains valid. Its coefficients I and $a_{k\alpha}$ can be calculated by formulas in [2,3].

3. **Time dependence of concentration change inside drops.** We consider here an axisymmetric array of drops in a flow field of periodic pattern. Substituting expression (2.5) for I_k into Eq. (1.4) and defining coefficients λ by the formula

$$\lambda = 4\pi(3\varepsilon I)^{-1} = \frac{2}{3}\Gamma(1/(n+1))(n+1)^{-2\sigma}t_0^{-\sigma} \tag{3.1}$$

$$(\lambda_1 = [\pi(\beta+1)/6]^{1/2}, \quad \lambda_2 = 8\Gamma(1/3)(81\pi)^{-2/3})$$

for the concentration $c_1^+, c_2^+, \ldots, c_k^+$ inside drops we obtain the linear system of ordinary differential equations

$$\frac{dc_k^+}{d\tau} = \sum_{\alpha=1}^{k}a_{k\alpha}(c_{\alpha-1}^+ - c_{\alpha}^+), \quad c_0^+ = 1; \quad c_k^+(0) = 0 \tag{3.2}$$

The coefficients λ_1 and λ_2 correspond to a chain of drops of moderate and high viscosity in a Stokes motion mode with condition (1.5) satisfied and coefficients $a_{k\alpha}$ determined by formula (2.5).

Since system (3.2) may also be written in the form

$$dc_k^+/d\tau + c_k^+ = F_k(c_0^+, c_1^+, \ldots, c_{k-1}^+), \quad c_k^+(0) = 0 \tag{3.3}$$

$$F_k = c_{k-1}^+ + \sum_{\alpha=1}^{k-1}a_{k\alpha}(c_{\alpha-1}^+ - c_{\alpha}^+), \quad c_0^+ = 1$$

formulas for concentrations $c_k{}^+$ can be successively determined, commensing with the first drop

$$c_k{}^+(\tau) = e^{-\tau} \int_0^{\tau} e^{\xi} F_k \left(c_0{}^+, c_1{}^+(\xi), \ldots, c_{k-1}^+(\xi)\right) d\xi \qquad (3.4)$$

Using (3.4) it is possible to show by induction that for the concentration inside drops the following formula is valid:

$$c_k{}^+(\tau) = 1 - e^{-\tau} + e^{-\tau} P_{k-1}(\tau), \quad P_0 = 0 \qquad (3.5)$$

$$P_{k-1}(\tau) = \sum_{\beta=1}^{k-1} A_{\beta}^{(k)} \tau^{\beta}, \quad A_{\beta}^{(\beta)} = 0, \quad A_1^{(k)} = -1 + a_{k1}$$

$$A_{\beta}^{(k)} = \beta^{-1} \left\{ A_{\beta-1}^{(k-1)} + \sum_{\alpha=\beta}^{k-1} a_{k\alpha} \left(A_{\beta-1}^{(\alpha-1)} - A_{\beta-1}^{(\alpha)} \right) \right\}; \quad \beta = 2, \ldots, k-1$$

The concentration $c_k{}^+$ in the first three drops changes in conformity with the law

$$c_1{}^+(\tau) = 1 - e^{-\tau}, \quad c_2{}^+ = 1 - e^{-\tau} - (2 - 2^{\sigma}) \tau e^{-\tau} \qquad (3.6)$$

$$c_3{}^+(\tau) = 1 - e^{-\tau} - \left\{ (1 + 2^{\sigma} - 3^{\sigma}) \tau + \frac{1}{2} (2 - 2^{\sigma})^2 \tau^2 \right\} e^{-\tau}$$

The concentration distribution in the diffusion boundary layer and in the convection boundary layer region of the diffusion trail of the k-th drop is determined by formula (2.4), where $c_1{}^+(\tau), \ldots, c_k{}^+(\tau)$ are defined by formula (3.5). A direct substitution of expressions derived in this way for $c_k{}^{(d)}$ and $c_k{}^{(1)}$ into the input equation (1.1) shows that the presence of the unsteady term in (1.1) does not lead to the appearance of irregularities in the expansion in the small parameter ε, as $\tau \to \infty$ (which theoretically may occur). This represents a further substantiation of the feasibility of reducing the complete problem (1.1), (1.2) to the consecutive solution of Eqs. (1.3) and (1.4).

For the total diffusion fluxes on the drop surfaces we have

$$I_k{}^{\circ} = e^{-\tau} \left\{ a_{k1} - A_{k-1}^{(k)} \tau^{k-1} + \sum_{\beta=1}^{k-1} \sum_{\alpha=\beta+1}^{k} a_{k\alpha} \left(A_{\beta}^{(\alpha-1)} - A_{\beta}^{(\alpha)} \right) \tau^{\beta} \right\} \qquad (3.7)$$

$$I_k{}^{\circ} = \frac{dc_k{}^+}{d\tau} = \frac{I_k}{I}, \quad A_k^{(k)} = 0, \quad A_{k-1}^{(k)} = -\frac{(2 - 2^{\sigma})^{k-1}}{(k-1)!}$$

where the expressions for coefficients $A_{k-1}^{(k)}$ are derived by induction from (3.5). Formula (3.7) shows that $I_k{}^{\circ} \to a_{k1} = k^{\sigma} - (k-1)^{\sigma}$ as $\tau \to 0$, which corresponds to the steady case under condition of total absorption on the surfaces of drops [1—4], and when $\tau \to \infty$ we have $I_k{}^{\circ} \to 0$, which corresponds to "saturation" of drops $(c_k{}^+ \to 1)$. For the total diffusion fluxes we have the limiting relation

$$\lim_{\tau \to 0} \frac{I_k}{I_{k-1}} = \frac{k^{\sigma} - (k-1)^{\sigma}}{(k-1)^{\sigma} - (k-2)^{\sigma}}, \quad \lim_{\tau \to \infty} \frac{1}{\tau} \frac{I_k}{I_{k-1}} = \frac{2 - 2^{\sigma}}{k-1}$$

which shows that $I_k{}^{\circ}(0) - I_{k-1}^{\circ}(0) < 0$ and $I_k{}^{\circ}(\tau) - I_{k-1}^{\circ}(\tau) > 0$ when $\tau \gg 1$. This implies that initially the total flux on the $(k-1)$-st drop is greater than on the k-th drop which follows it in its diffusion trail, then after expiry of time $\tau = \tau_{k,k-1}$ these fluxes become equal, and this is followed by the total flux on the k-th drop becoming greater.

For the first three drops we have

$$I_1{}^\circ = e^{-\tau}, \quad I_2{}^\circ = \{2^\sigma - 1 + (2-2^\sigma)\,\tau\}\,e^{-\tau} \tag{3.8}$$
$$I_3{}^\circ = \{3^\sigma - 2^\sigma + (5\cdot2^\sigma - 2^{2\sigma} - 3^\sigma - 3)\,\tau + {}^{1}/_{2}\,(2-2^\sigma)^2\tau^2\}e^{-\tau}$$

which shows that the diffusion fluxes on the surface of the first two drops become equal at $\tau = \tau_{21} = 1$.

4. Further generalizations. We consider here the general case of mass transfer in a chain of drops of arbitrary shape in the field of flow around drops of an aperiodic pattern (the flow field away from the particle is assumed steady). The diffusion problem reduces to solving the following dimensionless system of equations:

$$\varepsilon^n\lambda^{-1}\partial c \,/\, \partial\tau + (\mathbf{v}\nabla)\,c = \varepsilon^{n+1}\Delta c; \quad c\big|_{\Gamma_k} = c_k{}^+, \quad c\big|_{\Gamma_\infty} = 1 \tag{4.1}$$

$$V_k\frac{dc_k{}^+}{d\tau} = \lambda I_k = \varepsilon\lambda \iint\limits_{\Gamma_k}\left[\frac{\partial c}{\partial n}\right]_{\Gamma_k} d\Gamma_k, \quad c_k{}^+(0) = 0 \tag{4.2}$$

(Below, the all characteristic dimensions of drops are assumed to be of the same order). In these equations the dimensionless time τ is the same as defined in Sect. 1; Γ_k and V_k are, respectively, surface areas and volumes of drops, and the initial condition for (4.1) is determined by the solution of the respective stationary problem under condition of total absorption on the drop surface.

Using investigations [2,3] and a reasoning similar to that in Sect. 1, it is possible to show that when $\varepsilon \to 0$ the solution of problem (4.1), (4.2) is reduced to: 1) solving the steady equation (4.1) for any arbitrary $c_k{}^+$, 2) using that solution for calculating the total diffusion flux $I_k = I_k(c_1{}^+, c_2{}^+, \ldots, c_k{}^+)$ in the right-hand side of Eq. (4.2), and 3) solving system (4.2) thus obtained for concentrations $c_k{}^+$.

For calculating the principal term of the expansion of I_k in parameter ε it is sufficient to know the concentration in the diffusion boundary layer of every drop. Assuming that the distance between drops is smaller than the characteristic length of their respective regions of the convection boundary layer trail, we use the remark in Sect. 2 and formula (2.6) for calculating the total flux I_k. Formula (2.6) used for the determination of the right-hand side of Eq. (4.2) generally yields for the concentration inside drops the system of equations

$$\frac{dc_k{}^+}{d\tau} = \sum_{\alpha=1}^{k}\gamma_{k\alpha}\,(c_{\alpha-1}{}^+ - c_\alpha{}^+), \quad c_0{}^+ = 1; \quad c_k{}^+(0) = 0 \tag{4.3}$$

where $\gamma_{k\alpha} > 0$ are some, generally arbitrary, coefficients that can be calculated by (2.6) and [formulas in] [2,3].

If specific values of coefficients $a_{k\alpha}$ in (2.5) are disregarded, system (4.3) is the same as Eq. (3.2), except for the notation. Values of concentrations $c_k{}^+$ can be obtained successively, beginning with the first

$$c_k{}^+(\tau) = \exp(-\gamma_{kk}\tau)\int_0^\tau \exp(\gamma_{kk}\xi)\,F_k{}^*(c_0{}^+, c_1{}^+(\xi), \ldots, c_{k-1}{}^+(\xi))\,d\xi \tag{4.4}$$

$$F_k{}^* = \gamma_{kk}c_{k-1}{}^+ + \sum_{\alpha=1}^{k-1}\gamma_{k\alpha}\,(c_{\alpha-1}{}^+ - c_\alpha{}^+), \quad c_0{}^+ = 1$$

$$c_k^+(\tau) = 1 + \sum_{\alpha=1}^{s(k)} \exp(-\gamma_{\alpha\alpha}^* \tau) E_\alpha^{(k)}(\tau) \tag{4.5}$$

$$E_\alpha^{(k)}(\tau) = \sum_{\beta=0}^{r_\alpha-1} B_{\alpha\beta}^{(k)} \tau^\beta, \quad 1 + \sum_{\alpha=1}^{s(k)} B_{\alpha 0}^{(k)} = 0$$

Formula (4.5) indicates the form of the solution of (4.3); $s(k)$ is the number of coefficients $\gamma_{\alpha\alpha}^*$ ($\alpha = 1, 2, \ldots, s(k)$) differing in magnitude in the set of quantities γ_{nn} $n \leqslant k$, $r_\alpha = r_\alpha(k)$ is the multiplicity of root $\gamma_{\alpha\alpha}^*$. When $\gamma_{11} = \gamma_{22} = \ldots = \gamma_{kk} = 1$ the coefficients $B_{\alpha\beta}^{(k)} = 0$, $\alpha \geqslant 2$, and $B_{1\beta}^{(k)} = A_\beta^{(k)}$ where $A_\beta^{(k)}$ is determined by formulas (3.5) after the respective substitution of $\gamma_{k\alpha}$ for $a_{k\alpha}$. The other limit case when $\gamma_{kk} \neq \gamma_{nn}$, and $k \neq n$ leads to the following relations in (4.5):

$$r_\alpha = 1, \quad s(k) = k, \quad \gamma_{\alpha\alpha}^* = \gamma_{\alpha\alpha}, \quad E_\alpha^{(k)}(\tau) = B_{\alpha 0}^{(k)} = \text{const} \tag{4.6}$$

Note that the exponents $\gamma_{\alpha\alpha}^*$ in formulas (4.5) can be obtained from solutions of the system of equations that corresponds to the absence of diffusion interaction between drops (a system of drops in which only one drop is absorbing, while all remaining are neutral and do not absorb the substance dissolved in the stream; to which corresponds the boundary condition $[\partial c / \partial n]_{\Gamma_k} = 0$ at their surface), i.e. of system (4.3) in which we must set $\gamma_{k\alpha} = 0$ with $\alpha \neq k$ and $c_{k-1}^+ = 1$.

As an example, let us consider a chain of spherical drops of various radii $a(k)$, $k = 1, 2, \ldots, M$, $a(k)/a(1) = O(1)$, moving one after another in a Stokes flow mode at the same velocity U, and satisfying condition (1.5). Taking the radius of the first drop as the basic unit of length, we determine coefficients I and λ using formulas (2.5) and (3.1), and obtain for the exponents $\gamma_{k\alpha}$ in (4.6) the following expressions:

$$\gamma_{\alpha\alpha} = [a(1) / a(\alpha)]^{(2n+1)/1(n+1)}; \quad n = 1, 2 \tag{4.7}$$

Using formulas (2.6) and (4.2) and taking into account the results in [2,3], we shall indicate the expressions for coefficients $\gamma_{k\alpha}$ in Eqs. (4.3) and adduce the final expressions for the total diffusion fluxes I_k° on the surface of the first two drops ($\varkappa \neq 1$) $\varkappa = a(2) a^{-1}(1)$

$$\gamma_{11} = 1, \quad \gamma_{22} = \varkappa^{-(2n+1)/(n+1)}, \quad \gamma_{21} = [(\varkappa^{(n+2)/n} + 1)^\sigma - 1] \varkappa^{-3} \tag{4.8}$$

$$I_1^\circ = e^{-\tau}, \quad I_2^\circ = \varkappa^3 (1 - \gamma_{21})(1 - \gamma_{22}^{-1})^{-1}[e^{-\tau} - e^{-\gamma_{22}\tau}] + \varkappa^3 \gamma_{21} e^{-\tau}$$

Note that although formula (4.8) for I_2° was obtained for $\varkappa \neq 1$, it becomes (3.8) when $\varkappa \to 1$ and $\tau = \text{const} > 0$.

5. Discussion of results. Formulas (3.5) and (4.5) defining concentrations show that as $\tau \to \infty$, the drops become "saturated", $c_k^+ \to 1$, and total diffusion fluxes I_k on their surfaces tend to vanish. If the chain consists of spherical drops of the same radius, the effect of saturation process on the interaction between diffusion boundary layers and particle trails is that in the beginning the total diffusion flux on the first drop is higher than on the second, then owing to saturation the concentration inside the first drop becomes higher than in the second. This, in turn, weakens the diffusion trail of the first drop (i.e., the difference between concentrations in the oncoming stream and in the trail diminishes) and, as shown in Sect. 3, at the instant

of time

$$t = [{}^{1}/_{6}\pi\,(\beta + 1)\,P]^{1/2}aU^{-1}, \quad \beta \leqslant O\,(1) \quad (n = 1)$$

$$t = 8\Gamma\,({}^{1}/_{3})(81\pi)^{-2/3}\,P^{1/3}aU^{-1}, \quad \beta > O\,(P^{1/3}) \quad (n = 2)$$

the diffusion. fluxes on the surfaces of the first two drops become equal; then the second drop begins to absorb more of the dissolved substance than the first. Some time after that the total fluxes on the second and third drops become equal, and so on.

We shall first analyze the general case of chains in flow fields of aperiodic pattern on the example of two drops of different radii. For this we investigate the sign of function

$$\Delta_{2i}\,(\tau) = e^{\tau}\{I_2{}^{\circ}\,(\tau) - I_1{}^{\circ}\,(\tau)\} =$$
$$\varkappa^3\,(1 - \gamma_{21})(1 - \gamma_{22}^{-1})^{-1}[1 - \exp\{(1 - \gamma_{22})\,\tau\}] + \varkappa^3\gamma_{21} - 1$$

where the dependence of coefficients γ_{21} and γ_{22} on the drop radii is defined in (4.8). Since $d\Delta_{21}\,(\tau)/d\tau$ does not change its sign when $\tau \geqslant 0$, the equation $\Delta_{21}\,(\tau) = 0$ has not more than one root. Three cases are possible: 1) when $\varkappa > \varkappa_2 = (2^{1/\sigma} - 1)^{n/(n+2)}$, then $\Delta_{21}\,(\tau) > 0$, i.e. the total diffusion flux on the second drop is always greater than on the first; 2) when $\varkappa < \varkappa_1$, where $\varkappa_1$ is the root of equation

$$\Omega\,(\varkappa_1) = 0, \quad \Omega\,(\varkappa) = \varkappa^3 - 1 + [1 - \varkappa^3\gamma_{21}\,(\varkappa)]\,\gamma_{22}^{-1}\,(\varkappa)$$

then $\Delta_{21}\,(\tau) < 0$, i.e., the total diffusion flux on the first drop in the course of saturation is always greater than on the second; and 3) when $\varkappa_1 < \varkappa < \varkappa_2$, the total diffusion flux on the first drop is initially greater than on the second, then at some time $\tau_{21} = \tau_{21}\,(\varkappa)$, $\Delta_{21}\,(\tau_{21}) = 0$ the two fluxes become equal, and at $\tau > \tau_{21}$ the total flux on the second drop becomes the greater. The following relations hold: $0 < \varkappa_1 < 1 < \varkappa_2 < 2$ and $\tau_{21}\,(1) = 1$; $\tau_{21}\,(\varkappa_2) = 0$; $\varkappa \to \varkappa_1$, and $\tau_{21} \to \infty$.

Let us now investigate the behavior of the ratio of total diffusion fluxes $I_2{}^{\circ}\,/\,I_1{}^{\bullet}$ as $\tau \to \infty$. Using the expressions in (4.8) we obtain

$$\varkappa \geqslant 1, \quad \lim_{\tau \to \infty} I_2{}^{\circ}\,/\,I_1{}^{\circ} = +\infty$$

$$\varkappa < 1, \quad \lim_{\tau \to +\infty} I_2{}^{\circ}\,/\,I_1{}^{\circ} = \varkappa^3\,[1 - \gamma_{21}\,(\varkappa)\,\gamma_{22}^{-1}\,(\varkappa)][1 - \gamma_{22}^{-1}\,(\varkappa)]^{-1}$$

The limiting expressions show that when the radius of the first drop is smaller than that of the second, then for fairly considerable times the total diffusion flux on the first is negligibly small in comparison with the flux on the second. If the radius of the first drop is greater than that of the second, the total diffusion fluxes on both drops are of the same order when $\tau \to \infty$.

In the case of a chain of drops $k = 1, 2, \ldots, M$ of various radii the basic contribution during a fairly long time τ to the over-all diffusion flux (on all drops of the chain) is from drops with ordinal numbers $k_m \leqslant k \leqslant M$, where the number k_m belongs to the drop of maximum radius. This means that the drop of maximum radius determines the law of the diminution with time of total diffusion fluxes on all drops moving in the diffusion trail, and when $\tau \to \infty$, the total diffusion fluxes on drops preceding it can be neglected.

The following conservation laws apply:

$$\int_0^{\infty} I_k{}^{\circ}\,(\tau)\,d\tau = a^3\,(k), \quad a\,(1) = 1; \quad k = 1, 2, \ldots, M$$

REFERENCES

1. Gupalo, Iu. P., Polianin, A. D., and Riazantsev, Iu. S., On the diffusion in a chain of drops (bubbles) at high Péclet numbers. Izv. Akad. Nauk SSSR, MZhG, No. 1, 1978.

2. Polianin, A. D., On the diffusion interaction between drops in a fluid. Izv. Akad. Nauk SSSR, MZhG, No. 2, 1978.

3. Polianin, A. D., On diffusion interaction of solid particles at high Péclet numbers. PMM, Vol. 42, No. 2, 1978.

4. Gupalo, Iu. P., Polianin, A. D., and Riazantsev, Iu. S., On mass transfer of particles lying on the stream axis at high Péclet numbers. Izv. Akad. Nauk SSSR, MZhG, No. 2 1977.

5. Levich, V. G., Physicochemical Hydrodynamics. (English translation) Prentice-Hall, Englewood Cliffs, New Jersey, 1962.

Translated by J. J. D.