CONVECTIVE DIFFUSION TOWARD THE SPHERE IN LAMINAR FLOW AROUND THE SPHERE

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The problem of convective difussion toward the sphere in laminar flow around the sphere is solved by combination of the analytical and net methods for the region of Peclet number $\lambda \geqq 1$. The problem was also studied for very small values λ. Stability of the solution has been proved in relation to changes of the velocity profile.

In our recent studies[1,2] an approximate solution has been obtained of the problem of convective diffusion toward the sphere, around which the liquid flows in laminar flow for large values of the Peclet number λ. Only the analytical solution has been used and thus it was difficult to determine the accuracy and range of validity of the results with respect to the parameter λ and the angle ϑ. The differential equation describing the concentration distribution in the vicinity of the sphere around which the liquid flows has the singularity for $\vartheta = 0°$, which is causing certain complication in the analytical solution. Modern computer technique has made possible the numerical solution of this problem in a very wide range of values λ and also for small angles ϑ. It has been proved advantageous to combine the analytical and numerical solutions so that the analytically obtained terms for the asymptotic case $\lambda \to \infty$ were used as empirical for the numerical evaluation up to the value $\lambda = 1$. The qualitative shape of the diffusion flow has been also derived for very small values of λ.

Solution of the diffusion problem has been based on the approximative velocity profiles derived by Stokes[3]. The actual behaviour of the flowing solution differs more or less from these approximative relations, while it is very difficult to determine quantitatively these differences. Therefore, we have studied stability of the solution with respect to the velocity profiles and we came to the conclusion that the final solution was little affected by their changes.

Analytical Solution of the Differential Equation of Convective Diffusion

The dependence of concentration c of the diffusing compound with the diffusion coefficient D on spherical coordinates r, ϑ, and φ for steady convective diffusion is described by the equation

$$v_r \,.\, \partial c/\partial r + r^{-1} v_\vartheta \,.\, \partial c/\partial \vartheta + r^{-1} \sin^{-1} \vartheta \,.\, \partial c/\partial \varphi =$$
$$= D(\partial^2 c/\partial r^2 + 2r^{-1} \,.\, \partial c/\partial r + r^{-2} \,.\, \partial^2 c/\partial \vartheta^2 + r^{-2} \operatorname{cotg} \vartheta \,.\, \partial c/\partial \vartheta +$$
$$+ r^{-2} \sin^{-2} \vartheta \,.\, \partial^2 c/\partial \varphi^2) \,. \qquad (1a)$$

If the solution flows linearly, in laminar flow around the sphere with the radius a, the concentration is independent of the angle φ *i.e.* $c = c(r, \vartheta)$ at the orientation of the coordinate system according to Fig. 1. The given equation then takes the form

$$v_r \,.\, \partial c/\partial r + r^{-1} v_\vartheta \,.\, \partial c/\partial \vartheta = D(\partial^2 c/\partial r^2 + 2r^{-1} \,.\, \partial c/\partial r +$$
$$+ r^{-2} \,.\, \partial^2 c/\partial \vartheta^2 + r^{-2} \operatorname{cotg} \vartheta \,.\, \partial c/\partial \vartheta) \,. \qquad (1b)$$

If we assume that far from the sphere the concentration of the flowing solution is non-zero and equal to c_0 and that the dissolved compound on the surface of the sphere, due to the very fast chemical reaction or some other very fast operation vanishes, then the boundary conditions have the form

$$c(a, \vartheta) = 0 \,, \quad \lim_{r \to \infty} c(r, \vartheta) = c_0 \,. \qquad (2a), (2b)$$

Some other cases can be transformed by simple modification into the solved problem *e.g.* diffusion of the compound into the pure solvent at constant non-zero concentration on the surface of the sphere in laminar flow around it.

It is necessary to substitute into the differential Eq. (*1b*) the concrete terms for the components of flow v_r and v_ϑ. For very small velocities of the flow around the sphere v, accurately for values of the Reynolds number $\mathrm{Re} = av/\nu$ satisfying the condition $\mathrm{Re} \ll 1$, the equations derived by Stokes[3] hold

$$v_r = v \cos \vartheta (1 - \tfrac{3}{2} a/r + \tfrac{1}{2} a^3/r^3) \,, \qquad (3a)$$
$$v_\vartheta = -v \sin \vartheta (1 - \tfrac{3}{4} a/r - \tfrac{1}{4} a^3/r^3) \,. \qquad (3b)$$

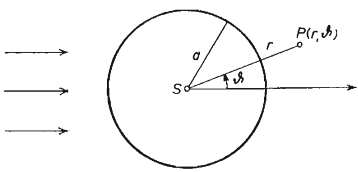

FIG. 1
Orientation of the Coordinate System
The arrows denote the flow direction.

As we demonstrate later in this study, the given relations for v_r and v_ϑ can be used to obtain relatively accurate results up to the values Re ≈ 1.

The main purpose is the calculation of the concentration gradient on the surface of the sphere which is decisive for the magnitude of the diffusion flow of the dissolved compound. For its calculation, we solve at first the boundary problem (*1b*), (*2a,b*). In this case it is advantageous to use dimensionless quantities. Therefore we introduce the relative concentration C, relative distance y from the surface of the sphere and the Peclet number λ by equations

$$C = c/c_0 \,, \quad y = (r - a)/a \,, \quad \lambda = av/D \,.$$

Eq. (*1*) for $C = C(y, \vartheta)$ will have the form

$$\partial^2 C/\partial y^2 + (2(1 + y)^{-1} - \lambda \cos \vartheta \, P_r(y)) \, \partial C/\partial y =$$
$$= -(1 + y)^{-2} \, \partial^2 C/\partial \vartheta^2 - (\lambda \sin \vartheta (1 + y)^{-1} \, P_\vartheta(y) +$$
$$+ \operatorname{cotg} \vartheta (1 + y)^{-2}) \, \partial C/\partial \vartheta \,, \tag{4}$$

where

$$P_r(y) = 1 - \tfrac{3}{2}(1 + y)^{-1} + \tfrac{1}{2}(1 + y)^{-3} \,,$$
$$P_\vartheta(y) = 1 - \tfrac{3}{4}(1 + y)^{-1} - \tfrac{1}{4}(1 + y)^{-3} \,,$$

and the boundary conditions (*2a*) and (*2b*) become

$$C(0, \vartheta) = 0 \,, \tag{5a}$$

$$\lim_{y \to \infty} C(y, \vartheta) = 1 \,. \tag{5b}$$

Similarly as in the study[1] we solve the given boundary problem by the iterative method. At first we introduce the variable u by substitution

$$u = y \, . \, f(\vartheta) \,, \tag{6}$$

so that the looked for function C becomes a function of variables u and ϑ. We would like to stress that the function f of the angle ϑ in substitution (*6*) can be selected so that in the limiting case it would have a physical meaning of a quantity proportional to the concentration gradient on the surface of the sphere as is demonstrated in the Appendix. It is also advantageous to introduce a new function G instead of C by the relation

$$C(u, \vartheta) = \lambda^{1/3} \, . \, G(u, \vartheta) \,. \tag{7}$$

At this choice of the multiplication factor $\lambda^{1/3}$ are values of the gradient of function G on the surface for $\vartheta \neq 0$ in the limiting case $\lambda \to \infty$ finite nad non-zero. By introduction of substitutions (*6*), (*7*) into Eq. (*4*), we obtain for the function G the following differential equation

$$(f^2 + (f')^2 u^2/(f + u)^2)\,\partial^2 G/\partial u^2 + (2f^2/(f + u) - \lambda f \cos\vartheta\, P_r(u/f) +$$
$$+ f\,.\,f''u/(f + u)^2 + f'u/(f + u)\,\lambda \sin\vartheta\, P_\vartheta(u/f) + f\,.\,f'u/(f + u)^2 \operatorname{cotg}\vartheta)\,.$$
$$.\,\partial G/\partial u = -f^2/(f + u)^2\,\partial^2 G/\partial \vartheta^2 - 2f\,.\,f'u/(f + u)^2\,\partial^2 G/\partial u\,\partial\vartheta -$$
$$- (\lambda f \sin\vartheta/(f + u)\, P_\vartheta(u/f) + f^2 \operatorname{cotg}\vartheta/(f + u)^2)\,\partial G/\partial\vartheta\,, \qquad (8)$$

or

$$A\,.\,\partial^2 G/\partial u^2 + B\,.\,\partial G/\partial u = F\,, \qquad (9)$$

where the significance of quantities A, B, and F results from comparison of Eq. (*9*) with Eq. (*8*). The boundary conditions (*5a*) and (*5b*) do change to

$$G(0, \vartheta) = 0\,, \qquad (10a)$$

$$\lim_{u \to \infty} G(u, \vartheta) = \lambda^{-1/3}\,. \qquad (10b)$$

The iteration scheme is defined recurrently by the equation

$$A\,.\partial^2 G_n/\partial u^2 + B\,.\,\partial G_n/\partial u = F_{n-1}\,, \quad n \in \mathbb{N}\,, \qquad (11)$$

where

$$F_{n-1} = -f^2/(f + u)^2\,\partial^2 G_{n-1}/\partial\vartheta^2 - 2f\,.\,f'u/(f + u)^2\,\partial^2 G_{n-1}/\partial u\,\partial\vartheta -$$
$$- (\lambda f \sin\vartheta/(f + u)\, P_\vartheta(u/f) + f^2 \operatorname{cotg}\vartheta/(f + u)^2)\,.\,\partial G_{n-1}/\partial\vartheta\,.$$

Moreover each iteration G_n must satisfy the boundary conditions (*10a,b*). The initial iteration G_0 is chosen as a function independent of ϑ *e.g.* in the form

$$G_0(u, \vartheta) = \lambda^{-1/3} u/(1 + u)\,.$$

At this choice is $F_0 = 0$ and for G_1 thus holds

$$A\,.\,\partial^2 G_1/\partial u^2 + B\,.\,\partial G_1/\partial u = 0\,, \qquad (12)$$

which is an ordinary differential equation including the variable ϑ as a parameter. Its solution has at the satisfaction of boundary conditions (*10a,b*) the form

$$G_1(u, \vartheta) = \lambda^{-1/3}\,.\int_0^u \exp\left(-\int_0^w B/A\,\mathrm{d}t\right)\mathrm{d}w\Big/\int_0^\infty \exp\left(-\int_0^w B/A\,\mathrm{d}t\right)\mathrm{d}w\,. \qquad (13)$$

From relations (7) and (6) we obtain

$$(\partial C/\partial y)_{y=0} = (\partial C/\partial u)_{u=0} \cdot \partial u/\partial y = \lambda^{1/3}(\partial G/\partial u)_{u=0} \cdot f(\vartheta) . \tag{14}$$

For the first iteration $G_1(u, \vartheta)$ there holds according to Eq. (13)

$$(\partial G_1/\partial u)_{u=0} \cdot f(\vartheta) = \lambda^{-1/3} \left(\int_0^\infty \exp \left(- \int_0^w B/A \, dt \right) dw \right)^{-1} \cdot f(\vartheta) =$$
$$= a_0(\vartheta) + a_1(\vartheta)\, \lambda^{-1/3} + a_2(\vartheta)\, \lambda^{-2/3} + \ldots \tag{15}$$

(see Appendix). So the choice of function G in Eq. (7) is explained and the statement following the equation is also proved as

$$\lim_{\lambda \to \infty} (\partial G_1/\partial u)_{u=0} \cdot f(\vartheta) = a_0(\vartheta) .$$

From derivation of Eq. (15) results that the expression of the gradient of function G_1 in the form of an infinite series is theoretically justified only for relatively large values of λ and not too small values of angle ϑ. The coefficients in the series (15) depend on the function f whose choice is made in relation with the calculation of the second iteration G_2. For it holds (see (11))

$$A \cdot \partial^2 G_2/\partial u^2 + B \cdot \partial G_2/\partial u = F_1 ,$$

where F_1 is the known function calculated with the use of the first iteration G_1. For gradient of the second iteration G_2 the infinite series is derived analogously as for G_1

$$(\partial G_2/\partial u)_{u=0} \cdot f(\vartheta) = b_0(\vartheta) + b_1(\vartheta)\, \lambda^{-1/3} + b_2(\vartheta)\, \lambda^{-2/3} + \ldots , \tag{16}$$

and this again for suitable regions of quantities λ and ϑ. In the Appendix it is proved that if the function f is chosen as

$$f(\vartheta) = k^{-1} \cdot (\lambda/3)^{1/3} \cdot \bar{f}(\vartheta) , \tag{17a}$$

where

$$\bar{f}(\vartheta) = \sin \vartheta \Big/ \sqrt[3]{\left(\int_\vartheta^\pi \sin^2 \vartheta \, d\vartheta \right)} \tag{17b}$$

with k as an arbitrary positive constant, it holds $a_0(\vartheta) = b_0(\vartheta)$. This means the optimum choice in the sense that in the assymptotic case $\lambda \to \infty$ already the first iteration gives the accurate result. For finite values λ the difference between the gradients of functions G_2 and G_1 is of the order of magnitude $\lambda^{-1/3}$ *i.e.* for large values of λ it is very small. It is possible to expect that the differences of gradients of the third and higher iterations from the preceding ones will be even smaller by orders of magnitude. Thus we have not continued with the calculation of the next iterations. For gradient of concentration C we thus obtain the approximate formula

$$(\partial C/\partial y)_{y=0} \approx (\partial C_2/\partial y)_{y=0} \approx \lambda^{1/3}(b_0(\vartheta) + b_1(\vartheta)\,\lambda^{-1/3}),$$
$$b_0(\vartheta) = \bar{f}(\vartheta)/(\sqrt[3]{6}\,.\,E) = 0{\cdot}61628\,.\,\bar{f}(\vartheta),$$
$$b_1(\vartheta) = E_1/(4E^2)\,.\,(3 - \cos^2\vartheta/\bar{f}^3) + 0{\cdot}180/(E\,.\,\bar{f})^3\,.\,(-\tfrac{5}{12}\cos\vartheta - \quad (18)$$
$$-\tfrac{1}{4}\bar{f}^3 + \cos^2\vartheta/\bar{f}^3 + \tfrac{1}{4}\sin^2\vartheta/\bar{f}^3) =$$
$$= 1/\bar{f}^3(0{\cdot}53839\bar{f}^3 + 0{\cdot}16917/\bar{f}^3 + 0{\cdot}42561\cos\vartheta + 0{\cdot}50751\,.\,\cos^2\vartheta/\bar{f}^3),$$

where the symbol E_n denotes the integral

$$E_n = \int_0^\infty x^n\,.\,\exp(-x^3)\,dx\,,\quad n = 0, 1, 2, \ldots,\quad E = E_0\,. \qquad (19)$$

The next term $b_2(\vartheta)\,\lambda^{-2/3}$ of the series (*16*) in the approximate formula (*18*) is not given as its values probably change in the third iteration and for the sufficiently large values of λ and not too small ϑ are negligibly small.

It can be seen from Table I that the values of $b_1(\vartheta)$ are unreliable for angles smaller than 80° and thus not suitable for numerical calculations. This is affected by the singularity in the point $\vartheta = 0°$ as $\bar{f}(0) = 0$. The iteration procedure of solution

TABLE I

Values of Functions $\bar{f}$, b_0, b_1 According to (*17b*) and (*18*)

ϑ^0	$\bar{f}(\vartheta)$	$b_0(\vartheta)$	$b_1(\vartheta)$	ϑ^0	$\bar{f}(\vartheta)$	$b_0(\vartheta)$	$b_1(\vartheta)$
10	0·14945	0·09210	—	100	1·15954	0·71460	0·56689
20	0·29510	0·18186	—	110	1·22611	0·75562	0·52668
30	0·43434	0·26767	—	120	1·28363	0·79107	0·50395
40	0·56559	0·34856	—	130	1·33220	0·82100	0·49046
50	0·68795	0·42397	4·95248	140	1·37187	0·84545	0·48217
60	0·80103	0·49366	1·99969	150	1·40268	0·86444	0·47702
70	0·90469	0·55754	1·15181	160	1·42467	0·87799	0·47391
80	0·99894	0·61562	0·79819	170	1·43786	0·88612	0·47223
90	1·08385	0·66795	0·64274	180	1·44225	0·88882	0·47171

defined by Eq. (*11*) can be applied also for angles smaller than 80° but in the theoretical calculation of the gradient it is not possible to transfer its expression by the infinite series according to powers $\lambda^{-1/3}$. The direct calculation of gradients would be too complex and cumbersome thus we have used the series (*16*) also for smaller angles with the smaller values of λ as empirical and we have used it for evaluation of coefficients $b_1(\vartheta)$ and $b_2(\vartheta)$ of the results obtained by numerical solution of the given boundary problem (see § Numerical Solution). At the end of this chapter we are giving the values of constant b_0

$$b_0 = \frac{1}{2}\int_0^\pi b_0(\vartheta) \sin \vartheta \, d\vartheta \approx 0{\cdot}62457 \,, \tag{20}$$

which will be needed for calculation of the total diffusion flux toward the sphere around which the liquid flows.

Analytical Solution for Small λ

Expression of the concentration gradient in the form of a series

$$(\partial C/\partial y)_{y=0} = k_0\lambda^{1/3} + k_1 + k_2\lambda^{-1/3} + \ldots$$

resulting from solution of the differential Eq. (*4*) for large values λ looses obviously its substantiation for small λ. The region of very small values of λ has only a small practical significance as it usually concerns only very tiny streaming velocities which are at normal laboratory conditions hindered or even overcome by natural convection. Nevertheless situations can also arise where exist even the very small values of λ without hindering effect of natural convection *e.g.* dissolving of microscopic gaseous bubbles, dissolving of microscopic solid particles. Moreover it is also theoretically interesting to study the shape of the diffusion flux toward the sphere in the whole range of values λ which have physical meaning *i.e.* $\lambda \in \langle 0, \infty)$. For this reason we have started with the study of the boundary problem (*4*), (*5a,b*) for $\lambda \ll 1$.

Thus let us assume that $0 < \lambda \ll 1$. Then it is possible to consider the terms in the differential Eq. (*4*) including λ as perturbation terms and to assume that the solution will be an analytical function of the parameter λ in the right neighbourhood of zero, thus that

$$C(y, \vartheta) = C_0(y, \vartheta) + C_1(y, \vartheta)\,\lambda + C_2(y, \vartheta)\,\lambda^2 + \ldots. \tag{21}$$

By substitution of this series into Eq. (*4*) and by comparison of coefficients with the zeroth and first power of λ we obtain

$$\partial^2 C_0/\partial y^2 + 2(1 + y)^{-1} \,.\, \partial C_0/\partial y = 0 \,, \tag{22}$$

$$\partial^2 C_1/\partial y^2 + (1+y)^{-2} \cdot \partial^2 C_1/\partial \vartheta^2 + 2(1+y)^{-1} \cdot \partial C_1/\partial y + \\ + \operatorname{cotg} \vartheta \cdot (1+y)^{-2} \cdot \partial C_1/\partial \vartheta = P_r(y) \cos \vartheta \cdot \partial C_0/\partial y \,. \tag{23}$$

The function C_0 is thus identical with the function C for $\lambda = 0$ *i.e.* for the zero flow rate. Thus it is independent of angle ϑ and this fact was respected in Eq. (*22*). Its solution under the conditions (*5a,b*) is

$$C_0(y) = 1 - (1+y)^{-1} \,. \tag{24}$$

In Eq. (*23*) we introduce the variable Θ by substitution

$$\Theta = \pi/2 - \vartheta \,. \tag{25}$$

As $\vartheta \in \langle 0, \pi \rangle$ is $\Theta \in \langle -\pi/2, \pi/2 \rangle$. Eq. (*23*) than takes the form

$$\partial^2 C_1/\partial y^2 + (1+y)^{-2} \cdot \partial^2 C_1/\partial \Theta^2 + 2(1+y)^{-1} \cdot \partial C_1/\partial y - \\ - \operatorname{tg} \Theta (1+y)^{-2} \cdot \partial C_1/\partial \Theta = P_r(y) \sin \Theta (1+y)^{-2} \,. \tag{26}$$

From the form of differential Eq. (*26*) results, that function C_1 is an odd function of angle Θ. Also the gradient $(\partial C_1/\partial y)_{y=0}$ has this property and thus the contribution to the total diffusion flux originating from the function C_1 equals zero. This means physically that this contribution to the total diffusion flux on the impact part of the sphere is just compensated by the loss on the turned away part of the sphere. Therefore the non-zero contribution to the diffusion flux in comparison to the standstill state can be caused by the function C_2. For the total flux Q then it holds

$$Q = 4\pi a^2 c_0 D \cdot \left(1 + (\partial C_2/\partial y)_{y=0} \, \lambda^2 + \ldots\right) . \tag{27}$$

From here results that the convective component of the diffusion flux Q is at very small velocities negligible. This is because if it is expressed as a function of parameter $\omega = \lambda^{1/3}$ which plays an important role in the region $\lambda > 1$ (see *e.g.* (*16*)) its value is approximatelly proportional to ω^6.

Dependence of Diffusion Flux on Velocity Profile

In our previous computations we have assumed that the velocity profile of the forced flow in the vicinity of the sphere around which the liquid flows is determined by Eqs (*3a,b*). This assumption is well satisfied for very small Reynolds numbers from which in standard situations results that also the Peclet number λ is relatively small. This is because it holds

$$\lambda = av/D = av/\nu \cdot \nu/D = \mathrm{Re} \cdot \nu/D \,, \tag{28}$$

where ν is the dynamic viscosity of flowing solution. For Re numbers close to one the relations derived by Stokes (*3a,b*) are already not satisfactory. There are known more accurate solutions[4-6] whose application for solution of the diffusion problem would lead to great complications. These more precise solutions have the advantageous property that in close vicinity of the surface they merge with the Stokes solution. This results in the natural question how great is the effect of coefficients in the differential Eq. (*4*) including the velocity profile *i.e.* functions P_r, P_ϑ on solution or how the solution of this equation will change if we proceed from the velocity profile (*3a,b*) to the new one characterized by functions $\bar{P}_r$ and $\bar{P}_\vartheta$ in agreement with the conditions

$$\lim_{y\to 0} \bar{P}_r(y)/P_r(y) = \lim_{y\to 0} \bar{P}_\vartheta(y)/P_\vartheta(y) = 1\,. \qquad (29)$$

Thus we have solved the given diffusion problem at the hypothetic profiles

$$\bar{P}_r(y) = \tfrac{3}{2}z^2 - (\tfrac{1}{2} + \delta_1)\, z^3 + \delta_1 z^4\,,$$
$$\bar{P}_\vartheta(y) = \tfrac{3}{2}z - (\tfrac{3}{4} + \delta_2)\, z^2 + (\tfrac{1}{4} + \delta_2)\, z^3\,, \qquad (30)$$

where

$$z = y/(1 + y)\,.$$

The functions $\bar{P}_r$ and $\bar{P}_\vartheta$ were formed by modification of functions P_r and P_ϑ given after the formula (*D3*) so that there were satisfied both conditions (*29*) and conditions $\lim_{y\to\infty} \bar{P}_r(y) = \lim_{y\to\infty} \bar{P}_\vartheta(y) = 1$.

The functions $\bar{P}_r$ and $\bar{P}_\vartheta$ are not characterizing the solution of the hydrodynamic problem of flow around the sphere but they satisfy the basic physical requirements expressed by the given limiting conditions.

Applying the same procedure as in the analytical solution with the Stokes profiles we have obtained

$$(\partial C_2/\partial y)_{y=0} \approx \lambda^{1/3}(\bar{b}_0(\vartheta) + \bar{b}_1(\vartheta)\,\lambda^{-1/3})\,, \qquad (31)$$

where (J is introduced in (*D27*))

$$\bar{b}_0(\vartheta) = b_0(\vartheta)\,,\quad \bar{b}_1(\vartheta) = b_1(\vartheta) + (\Delta b_1)_r + (\Delta b_1)_\vartheta\,,$$
$$(\Delta b_1)_r = \delta_1(9/(E\mathcal{J})^3\,.\,(\tfrac{4}{3}\cos\vartheta + (1 + 3\cos^2\vartheta)/\mathcal{J}^3)\,J -$$
$$-\tfrac{3}{4}E_1/E^2\,.\,(1 + 3\cos^2\vartheta)/\mathcal{J}^6),$$
$$(\Delta b_1)_\vartheta = \delta_2(-9/(E\mathcal{J})^3\,.\,(\mathcal{J}^3/9 + \tfrac{5}{3}\cos\vartheta + (1 + 3\cos^2\vartheta)/\mathcal{J}^3)\,.\,J +$$
$$+\tfrac{3}{4}E_1/E^2\,.\,\mathcal{J}^{-6}\,.\,(1 + 3\cos^2\vartheta + \mathcal{J}^3/3\,.\cos\vartheta - \mathcal{J}^6/3))\,. \qquad (32)$$

From these equations it results that the chosen changes of the velocity profile affect only the second and next terms in formula (*18*) while changes of the second term are proportional to quantities δ_1, δ_2. In the numerical evaluation of these changes, we have chosen δ_1 and δ_2 so that the maximum deviations of modified profiles (*30*) from the Stokes ones accounted for 10% which occurs for $\delta_1 = 0{\cdot}6$, $\delta_2 = 0{\cdot}5$.

In Table II are given values of $(\Delta b_1)_r$ and $(\Delta b_1)_\vartheta$ and the percentage changes p_r and p_ϑ of the gradient C caused by the changes of the coefficient b_1 for $\lambda = 1000$.

From the results given in Table II results a serious fact that solution of Eq. (*4*) with respect to changes in the velocity profile is very stable as the 10% change in this profile is causing in average 8times smaller changes in the gradient for $\lambda = 1000$. For greater values λ the percentage changes would be even smaller. This fact seems to us very significant also from a more general point of view as a similar situation occurs in studies of various operations described by differential equations with the approximatively determined coefficients, resp. boundary or initial conditions. For these reasons we have performed the numerical solution of the given problem by the Stokes approximation of the velocity profile.

Numerical Solution of the Differential Equation for Convective Diffusion

For numerical solution of the differential Eq. (*4*) with the boundary conditions (*5a,b*) we have chosen the net method. The given equation has been approximated by the difference equation so that we have substituted the corresponding partial

TABLE II

Changes of Some Quantities Resulting from Changes in the Velocity Profile

ϑ^0	$-(\Delta b_1)_r$	$-(\Delta b_1)_\vartheta$	$-p_r$	$-p_\vartheta$	$-(p_r + p_\vartheta)$
90	0·0638	0·0316	0·888	0·440	1·328
100	0·0689	0·0306	0·902	0·400	1·302
110	0·0786	0·0246	0·976	0·306	1·282
120	0·0882	0·0181	1·050	0·216	1·266
130	0·0993	0·0123	1·107	0·142	1·249
140	0·1027	0·0077	1·149	0·086	1·235
150	0·1074	0·0043	1·177	0·047	1·224
160	0·1107	0·0019	1·195	0·020	1·215
170	0·1126	0·0005	1·205	0·005	1·210
180	0·1132	0	1·208	0	1·208

derivations by difference terms with the total accuracy having the order of magnitude h^2 where h is the step of the net. The condition (*5b*) was approximated by the conditions

$$C(y_0, \vartheta) = 1 \quad \text{for} \quad 80° \leqq \vartheta \leqq 180°\,,$$
$$C(y, 80°) = 1 \quad \text{for} \quad y_0 \leqq y \leqq 2y_0\,,$$
$$C(2y_0, \vartheta) = 1 \quad \text{for} \quad 30° \leqq \vartheta \leqq 80°\,,$$
$$C(2y_0, 20°) = 0{\cdot}9576\,,$$
$$C(2y_0, 10°) = 0{\cdot}6307\,,$$

where $y_0 = 3{\cdot}6\,\lambda^{-1/3}$. The value y_0 was chosen so that the absolute difference between the selected boundary values and the approximate theoretical values resulting from the formula (*D11*) for the asymptotic case $\lambda \to \infty$ would be smaller than 10^{-3}. So we have substituted for the infinite region on which the problem (*4*), (*5a,b*) is solved by the finite region according to Fig. 2. Moreover we have chosen

$$C(y, 0°) = 0 \quad a \quad \partial C/\partial\vartheta(y, 180°) = 0$$

in agreement with the physical model. The described finite region was covered by the net with the steps $h_y = y_0/24$, $h_\vartheta = 10°$.

The formed system of linear equations for values of function C in the grid-points of the net was solved by the relaxation method with the relaxation coefficient $r = 0{\cdot}5$. The initial iteration was again estimated according to formula (*D11*). In total 480 iterations have been calculated satisfying the condition

$$\max_{i,k} \left|C_{ik}^{(480)} - C_{ik}^{(479)}\right| < 10^{-5}\,,$$

where the significance of used symbols is obvious. The calculation has been performed

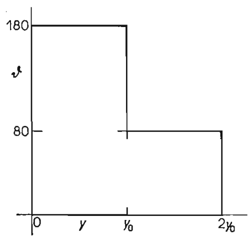

FIG. 2
Region for the Net Method

on the computer ICL 4-72 individually for $\lambda^{1/3}$ = 6,8, 10, 15, 20, 25, 30, 36, 40 and cyclically both for $\lambda^{1/3}$ = 6, 5, 4, 3, 2, 1 and for $\lambda^{1/3}$ = 10, 15, 20, 25, 30, 36, 40. In cyclical evaluation we have made computations for $\lambda^{1/3}$ = 6 or 10 as the source and the last iteration for the given $\lambda^{1/3}$ as the initial iteration for the following value $\lambda^{1/3}$.

For the estimate of the difference between the solutions of the differential and difference equations we have have applied the estimate of errors according to Runge. Thus we have performed the computation for $\lambda^{1/3}$ = 6, 8, 10 with steps twice as long.

We have calculated from the obtained function values C in the grid-points the approximate concentration gradients on the surface of the sphere according to the formula

$$\partial C/\partial y(0, kh_\vartheta) \approx 1/(12h_y)\left(-25C_{0k} + 48C_{1k} - 36C_{2k} + 16C_{3k} - 3C_{4k}\right),$$
$$k = 1, 2, \ldots, 18 .$$

So calculated values of the gradient C were then used for determination of the total diffusion flux toward the sphere

$$Q = 4\pi a^2 c_0 D \,.\, I \,, \tag{33}$$

where

$$I = \frac{1}{2}\int_0^\pi (\partial C/\partial y)_{y=0} \,.\, \sin \vartheta \, d\vartheta \,. \tag{34}$$

The value I was determined by numerical integration. We have found by the Runge estimate of errors that the concentration gradients for individual angles are calculated with the average accuracy 0·2% with the exception of values for $\vartheta \leqq 20°$, the total flux Q with the accuracy 0·025%. Considerable increase in the accuracy in calculation of Q was caused by compensation of errors of the gradient C for individual angles.

In Table III are given values of coefficients $b_1(\vartheta)$, $b_2(\vartheta)$ in the empirical formula

$$\partial C/\partial y(0, \vartheta) \approx b_0(\vartheta)\,\lambda^{1/3} + b_1(\vartheta) + b_2(\vartheta)\,\lambda^{-1/3} \,, \tag{35}$$

where the values of the coefficient $b_0(\vartheta)$ were taken from the analytical solution (Table I) and coefficients $b_1(\vartheta)$, $b_2(\vartheta)$ were calculated by use of relative differences. The choice of formula (*35*) is justified at the end of the first paragraph. From numerical evaluation results that Eq. (*35*) fits well also for angles $\vartheta < 80°$ and $\lambda \geqq 8$. It is suitable to apply it separately in two regions *i.e.* $\lambda^{1/3} \in \langle 2, 10 \rangle$ and $\lambda^{1/3} \in \langle 10, 40 \rangle$. It results from Table III that the values of coefficients $b_1(\vartheta)$, $b_2(\vartheta)$ for $\lambda^{1/3} \in \langle 2, 10 \rangle$ have – contrary to expectation – smoother shape than for the second region as

Eq. (*35*) should suit better for larger λ. This fact has two reasons: *1*) Absolute errors of gradient C and thus the values $b_1(\vartheta)$, $b_2(\vartheta)$ as well, are greater for the second region at the same percentage error 0·2%. For example at uniform distribution of errors to the second and third terms in Eq. (*35*) are for $\vartheta = 90°$ possible these absolute errors:

For $\lambda^{1/3} = 5$ is $\Delta b_1 = 0{\cdot}004$, $\Delta b_2 = 0{\cdot}019$; for $\lambda^{1/3} = 25$ is $\Delta b_1 = 0{\cdot}017$ and $\Delta b_2 = 0{\cdot}430$. The results given in Table III demonstrate that the errors are smaller in the average. *2*) Visible fluctuation of values $b_1(\vartheta)$ and $b_2(\vartheta)$ for $\lambda^{1/3} \in \langle 10, 40\rangle$ is probably affected by the coefficient at $\partial C/\partial y$ in Eq. (*4*) whose values in the neighbourhood of 90° suddenly change due to the term $-\lambda \cos \vartheta \, P_r(y)$. This term has less profound effect for smaller values of λ.

Comparison of theoretical values $b_1(\vartheta)$ of Table I with the numerical results (Table III) demonstrates a fair agreement for angles greater than 90°.

In Fig. 3 are graphically plotted theoretical diffusion layers with their thickness d calculated by use of the numerically obtained results according to the formula

$$d^{-1} = (\partial C/\partial y)_{y=0} \,.$$

TABLE III

Values $b_1(\vartheta)$ and $b_2(\vartheta)$

	$2 \leqq \lambda^{1/3} \leqq 10$		$10 \leqq \lambda^{1/3} \leqq 40$	
ϑ^0	$b_1(\vartheta)$	$b_2(\vartheta)$	$b_1(\vartheta)$	$b_2(\vartheta)$
10	0·4643	0·2053	0·2828	1·9221
20	0·3579	0·5592	0·3379	0·4904
30	0·4372	0·3260	0·4790	−0·0616
40	0·4445	0·2899	0·4873	−0·3142
50	0·4750	0·1831	0·5435	−0·5689
60	0·4745	0·1689	0·5186	−0·3361
70	0·4871	0·1139	0·5478	−0·4949
80	0·4896	0·0899	0·5235	−0·3445
90	0·4890	0·0774	0·5458	−0·5570
100	0·4802	0·0950	0·5076	−0·1868
110	0·4850	0·0674	0·5358	−0·5739
120	0·4769	0·0828	0·4898	−0·0613
130	0·4802	0·0653	0·5253	−0·4252
140	0·4746	0·0763	0·4814	−0·0206
150	0·4762	0·0667	0·5161	−0·3951
160	0·4733	0·0744	0·4923	−0·2269
170	0·4716	0·0828	0·4965	−0·2377
180	0·4787	0·0704	0·4887	−0·0501

From the total diffusion fluxes calculated by use of Eqs (*33*), (*34*) for individual values λ we have for Q analogically obtained the next empirical relations

$$Q = 4\pi a^2 c_0 D(0{\cdot}6246\lambda^{1/3} + 0{\cdot}4745 + 0{\cdot}1301\lambda^{-1/3}) \quad \text{pro} \quad \lambda^{1/3} \in \langle 2, 10 \rangle, \qquad (36a)$$

$$Q = 4\pi a^2 c_0 D(0{\cdot}6246\lambda^{1/3} + 0{\cdot}5140 - 0{\cdot}3187\lambda^{-1/3}) \quad \text{pro} \quad \lambda^{1/3} \in \langle 10, 40 \rangle, \qquad (36b)$$

The coefficient $b_0 = 0{\cdot}6246$ has been taken from the analytical solution (see Eq. (*20*)). The use of empirical formula (*36a*) can be extended even to $\lambda^{1/3} \in \langle 1, 2 \rangle$ approximatelly with the 1% error. By direct net method calculation we have obtained for $\lambda = 1$, $Q = 4\pi a^2 c_0 D \,.\, 1{\cdot}2427$, while from Eq. (*36a*) we obtain $Q = 4\pi a^2 c_0 D \,.\, 1{\cdot}2292$.

For physical reasons it is obvious that the dependence of quantity $Q/4\pi a^2 c_0 D$ on $\lambda^{1/3}$ will be smooth in the interval $\langle 0, 1 \rangle$. Thus it is possible to complete by graphical extrapolation this dependence with a sufficient accuracy where directly calculated numerical values are not available. The described numerical calculation gives results which are not as good as those obtained by extrapolation because the convective components of diffusion in the extrapolated region are relatively small. Calculation of more accurate values would be very cumbersome which was considered uneconomical with regard to smaller significance of this region.

In Fig. 4 are plotted the dependences of quantities $\partial C/\partial y\ (0, \vartheta)$ for $\vartheta = 20°, 90°, 180°$ and quantites $Y = Q/4\pi a^2 c_0 D$ on $\lambda^{1/3}$ for the region $0 \leqq \lambda \leqq 1000$. In the neighbourhood of zero we have used formula (*27*), for $\lambda \geqq 1$ the numerically calculated values of gradient on the surface or of the over-all diffusion flux toward the sphere.

Finaly it could be concluded that the final equations (*36a,b*) are, when we take into consideration the errors due to empirical evaluation in the considered regions,

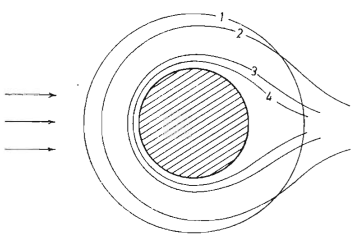

FIG. 3
Diffusion Layers
λ: *1* 0, *2* 1, *3* 125, *4* 1000.
The arrows denote the flow direction.

affected by the maximum error 0·1%, Eq. (*36a*) used in the interval $\lambda^{1/3} \in \langle 1, 2 \rangle$ by the error 1%, which in our opinion is quite satisfactory with regard to experimental possibilities at various applications.

APPENDIX

Computation of the Gradient of First Iteration and Choice of Function f

From Eqs (*14*) and (*13*) results

$$(\partial C_1/\partial y)_{y=0} = f(\vartheta) \Big/ \int_0^\infty \exp\left(-\int_0^w B(t)/A(t)\, dt\right) dw\,. \tag{D1}$$

We express approximately the integral in the denominator of Eq. (*D1*) for large values of Peclet number λ. At first we calculate the integral in the exponent. From comparison of Eqs (*8*) and (*9*) results

$$A(u) = f^2 + (f')^2\, u^2/(f + u)^2\,,$$

$$B(u) = 2f^2/(f + u) - \lambda f \cos \vartheta\, P_r(u/f) + f\,.\,f'' u/(f + u)^2 +$$
$$+ f' u/(f + u)\,.\,\lambda \sin \vartheta\, P_\vartheta(u/f) + f\,.\,f' u/(f + u)^2\,.\,\text{cotg}\, \vartheta\,,$$

where functions P_r and P_ϑ are defined after Eq. (*4*). It is necessary to realize that functions A and B include also the variable ϑ which we have neglected for simplicity also in the argument of function f and in their derivatives. Into the integral

$$\int_0^w B(t)/A(t)\, dt$$

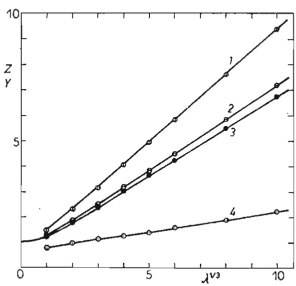

Fig. 4

Graphs of Functions $Z = (\partial C/\partial y)(0, \vartheta)$ and Y

Z for ϑ *4* 20°, *2* 90°, *1* 180°; *3* Y.

we introduce the variable z by substitution

$$z = (t/f)/(1 + (t/f)) \quad \text{or} \quad t/f = z/(1 - z) \tag{D2}$$

and denote

$$(w/f)/(1 + (w/f)) = x\,. \tag{D3}$$

So functions P_r and P_ϑ take the form

$$P_r(z/(1-z)) = \tfrac{3}{2}z^2 - \tfrac{1}{2}z^3\,, \quad P_\vartheta(z/(1-z)) = \tfrac{3}{2}z - \tfrac{3}{4}z^2 + \tfrac{1}{4}z^3\,.$$

$$\int_0^w B(t)/A(t)\,\mathrm{d}t = \int_0^x [2(1-z) - \lambda\cos\vartheta(\tfrac{3}{2}z^2 - \tfrac{1}{2}z^3) + f^{-1}f''(z - z^2) +$$
$$+ f^{-1}f'\lambda\sin\vartheta(\tfrac{3}{2}z^2 - \tfrac{3}{4}z^3 + \tfrac{1}{4}z^4) + f^{-1}f'(z - z^2)\,.\,\mathrm{cotg}\,\vartheta]/$$
$$/(1 + (f^{-1}f')^2 z^2)(1 - z)^2\,.\,\mathrm{d}z\,.$$

From substitution $(D2)$ it is obvious that $0 \leqq z < 1$ and thus we can use the binomic expansion

$$(1 - z)^{-2}(1 + (f^{-1}f')^2 z^2)^{-1} = (1 + 2z + 3z^2 + \ldots)(1 - (f^{-1}f')^2 z^2 + \ldots) =$$
$$= 1 + 2z + (3 - (f^{-1}f')^2 z)^2 + \ldots\,.$$

By substitution, arrangement and integration we obtain

$$\int_0^w B(t)/A(t)\,\mathrm{d}t \approx 2x + (1 + f''/(2f) + f'/(2f)\,.\,\mathrm{cotg}\,\vartheta)\,x^2 +$$
$$\lambda/2\,.\,(f'/f\,.\,\sin\vartheta - \cos\vartheta)\,x^3 + \lambda/4\,.\,(\tfrac{9}{4}f'/f\,.\,\sin\vartheta - \tfrac{5}{2}\cos\vartheta)\,x^4 +$$
$$+\lambda/5\,.\,(13/4\,.\,f'/f\,.\,\sin\vartheta - \tfrac{7}{2}\cos\vartheta + \tfrac{3}{2}(f^{-1}f')^2\cos\vartheta - \tfrac{3}{2}(f^{-1}f')^3\sin\vartheta)\,x^5\,. \tag{D4}$$

Now we start with the computation of integral in the denominator of Eq. $(D1)$. We introduce a new variable s by substitution (see $D3$))

$$s = \gamma x = \gamma\,.\,(w/f)/(1 + (w/f))\,, \tag{D5}$$

where

$$\gamma^3 = \lambda/2\,.\,(f'/f\,.\,\sin\vartheta - \cos\vartheta)\,. \tag{D6}$$

After arrangement with the use of the binomic expansion and expansion of the exponential function we obtain the approximation including the terms up to the order $\lambda^{-2/3}$ after the integral sign

$$\int_0^\infty \exp\left(-\int_0^w B(t)/A(t)\,\mathrm{d}t\right)\mathrm{d}w \approx f/\gamma \int_0^\gamma [1 - \tfrac{9}{8}s^4/\gamma - (f''/(2f) +$$
$$+ f'/(2f)\,.\,\mathrm{cotg}\,\vartheta)\,s^2/\gamma^2 - (13/10 - \tfrac{3}{5}(f^{-1}f')^2)\,s^5/\gamma^2 + 81/128\,.\,s^8/\gamma^2 +$$
$$+ \lambda/16\,.\,\cos\vartheta\,.\,s^4/\gamma^4 + \lambda/20\,.\,\cos\vartheta\,.\,s^5/\gamma^5 - 9/128\,.\,\lambda\cos\vartheta\,.\,s^8/\gamma^5 +$$
$$+ \lambda^2/512\,.\,\cos^2\vartheta\,.\,s^8/\gamma^8]\exp(-s^3)\,\mathrm{d}s\,. \qquad (D7)$$

In integration in $(D7)$ there appears the sum of several first terms of the series in powers of $\lambda^{-1/3}$. The integrals of the form

$$E_n(u) = \int_0^u s^n\,.\,\exp(-s^3)\,\mathrm{d}s\,, \qquad (D8)$$

will appear in their coefficients while in the limiting case $E_n(\infty) = E_n$ (see (19)). By substitution into $(D1)$and repeated use of the binomic expansion there originates

$$(\partial C_1/\partial y)_{y=0} = \lambda^{1/3}(a_0(\vartheta) + a_1(\vartheta)\,\lambda^{-1/3} + a_2(\vartheta)\,\lambda^{-2/3} + \ldots) \qquad (D9)$$

(see (15)).

Now we determine the function f. We start with the requirement that the second iteration G_2 should differ from the first iteration G_1 as little as possible. This happens obviously in the case when F_1 introduced generally after Eq. (11) will be in the absolute value as small as possible. As we consider the case of rather large values of λ we can approximate

$$F_1(u, \vartheta) \approx -\lambda f \sin\vartheta/(f + u)\,.\,P_\vartheta(u/f)\,.\,\partial G_1/\partial\vartheta\,. \qquad (D10)$$

To be able to express F_1 it is necessary to know the iteration G_1 given in Eq. (13). If the integral in the numerator of the quoted formula is calculated by the method described in the above given part of this study with the first two terms of the corresponding expansions used, we obtain

$$G_1(u, \vartheta) \approx \lambda^{-1/3}\{E(\xi) + \gamma^{-1}(-9/8 + \lambda\cos\vartheta/(16\gamma^3))\,E_4(\xi)\}/$$
$$/\{E(\gamma) + \gamma^{-1}(-9/8 + \lambda\cos\vartheta/(16\gamma^3))\,E_4(\gamma)\}\,, \qquad (D11)$$

where

$$\xi = \gamma(u/f)/(1 + (u/f))\,. \qquad (D12)$$

For large value of λ the values of γ are also large according to $(D6)$ and thus it holds

$$E - E(\gamma) = \int_\gamma^\infty \exp(-s^3)\,ds = \exp(-\gamma^3)/(3\gamma^2) -$$
$$- \frac{2}{3}\int_\gamma^\infty s^{-3}\exp(-s^3)\,ds < \gamma^{-2}\,.\,\tfrac{1}{3}\exp(-\gamma^3)\,,$$

which is the quantity proportional to $\lambda^{-2/3}$. Similar result can be derived also for difference $E_4 - E_4(\gamma)$, so that in the denominator of the formula $(D11)$ can be at the given approximation order the numbers $E(\gamma)$, $E_4(\gamma)$ substituted by values E, E_4. If we now calculate $\partial G_1/\partial\vartheta$, we obtain after arrangements with the accuracy λ^{-1}

$$\partial G_1/\partial\vartheta \approx \lambda^{-1/3}/E^2\,.\,[E\exp(-\xi^3)\,\xi' + (EE_4(\xi) - E_4E(\xi))\,.$$
$$.\,((9\gamma')/(8\gamma^2) - \lambda/16\,.\,(\gamma\sin\vartheta + 4\gamma'\cos\vartheta)/\gamma^5) +$$
$$+ \exp(-\xi^3)\,\xi'/\gamma\,.\,(\xi^4E - E_4)(\lambda\cos\vartheta/(16\gamma^3) - 9/8)]\,, \qquad (D13)$$

where

$$\xi' = \xi(\gamma'/\gamma - f'/f) + f'/f\,.\,\xi^2/\gamma\,, \qquad (D14)$$

as results from $(D12)$. The expression for $\partial G_1/\partial\vartheta$ in $(D13)$ includes a single term of the order $\lambda^{-1/3}$ which is $\lambda^{-1/3}/E\,.\,\exp(-\xi^3)\,\xi(\gamma'/\gamma - f'/f)$. If this term equals zero which can occur only under the condition

$$\gamma'/\gamma - f'/f = f/\gamma\,.\,(\gamma/f)' = 0\,, \qquad (D15)$$

the function F_1 will satisfy the above given requirement. Thus the ratio γ/f must be constant. For simplification of other computations we choose

$$\gamma/f = k/\sqrt[3]{2} \triangleq \text{konst.}\,. \qquad (D16)$$

By substitution into $(D6)$ we obtain the differential equation for the function f

$$\lambda(f'/f\,.\,\sin\vartheta - \cos\vartheta) = k^3 f^3\,, \qquad (D17)$$

which is solved[2] by introduction of a new function h by substitution

$$f(\vartheta) = \sin\vartheta/h(\vartheta)\,.$$

At the condition $f(\pi) \neq 0$ from which results $h(\pi) = 0$, we obtain the formulae

(*17a,b*). If we substitute into (*D16*) for f the term from formula (*17a*) then

$$\gamma = \sqrt[3]{(\lambda/6)} \,.\, \bar{f}(\vartheta) \,. \qquad (D18)$$

With the use of relations (*D1*), (*D7*), (*D9*), (*17a*) and (*D18*) we obtain for coefficients in the series (*D9*) the relations

$$a_0(\vartheta) = \bar{f}/(E \,.\, \sqrt[3]{6})\,, \quad a_1(\vartheta) = E_1/(4E^2) \,.\, (3 - \cos \vartheta/\bar{f}^3)\,,$$

$$a_2(\vartheta) = \sqrt[3]{6}/(\bar{f}E^2) \,.\, [11/960 - \tfrac{1}{5}(f'/f)^2 + \tfrac{1}{6}f''/f + \tfrac{1}{6}f'/f \,.\, \mathrm{cotg}\, \vartheta + \qquad (D19)$$
$$+ 29/160 \,.\, \cos \vartheta/\bar{f}^3 - 3/64 \,.\, \cos^2 \vartheta/\bar{f}^6 + E_1^2/(16E) \,.\, (3 - \cos \vartheta/\bar{f}^3)^2]\,.$$

CALCULATION OF THE GRADIENT OF SECOND ITERATION

For second iteration G_2 it holds according to Eq. (*11*)

$$A \,.\, \partial^2 G_2/\partial u^2 + B \,.\, \partial G_2/\partial u = F_1\,, \qquad (D20)$$

where F_1 can be approximated by the relation (*D10*). It is useful to introduce the difference

$$H(u, \vartheta) = G_2(u, \vartheta) - G_1(u, \vartheta)\,,$$

for which according to (*12*) and (*D20*) holds

$$A \,.\, \partial^2 H/\partial u^2 + B \,.\, \partial H/\partial u = F_1 \qquad (D21)$$

with the boundary conditions

$$H(0, \vartheta) = \lim_{u \to \infty} H(u, \vartheta) = 0\,. \qquad (D22)$$

By solution we obtain

$$H(u, \vartheta) = L \int_0^u \exp\left(- \int_0^w B/A \, \mathrm{d}t\right) \mathrm{d}w +$$
$$+ \int_0^u \left[\exp\left(- \int_0^w B/A \, \mathrm{d}t\right) \int_0^w F_1/A \,.\, \exp\left(\int_0^t B/A \, \mathrm{d}\tau\right) \mathrm{d}t\right] \mathrm{d}w\,, \qquad (D23)$$

where

$$L = - \int_0^\infty \left[\exp\left(- \int_0^w B/A \, \mathrm{d}t\right) \int_0^w F_1/A \,.$$

$$. \exp\left(\int_0^t B/A \, d\tau\right) dt \Big] dw \Big/ \int_0^\infty \exp\left(-\int_0^w B/A \, dt\right) dw . \qquad (D24)$$

According to Eq. (*14*) it holds for the gradient of second iteration

$$(\partial C_2/\partial y)_{y=0} = \lambda^{1/3}(\partial G_2/\partial u)_{u=0} \cdot f(\vartheta) = $$
$$= \lambda^{1/3} f(\vartheta) . (\partial G_1/\partial u + \partial H/\partial u)_{u=0} . \qquad (D25)$$

From (*D23*) it results

$$(\partial H/\partial u)_{u=0} = L .$$

After calculation of $(\partial G_1/\partial \vartheta)_u$ from the approximation (*D11*) and substitution into (*D10*) we obtain (variable ξ introduced in (*D12*))

$$F_1(u, \vartheta) \approx -\tfrac{3}{2}\lambda^{2/3} \sin \vartheta . \xi/(\gamma E)^2 . [\tfrac{5}{8} f'/f . E\xi^2 \exp(-\xi^3) +$$
$$+ \lambda/48 . E\gamma^{-3}\xi^2 \exp(-\xi^3)(\sin \vartheta + 4\gamma'/\gamma . \cos \vartheta) +$$
$$+ (\tfrac{3}{4}\gamma'/\gamma - \lambda/(24\gamma^3)(\sin \vartheta + 4\gamma'/\gamma . \cos \vartheta))(E . E_1(\xi) - E_1 E(\xi))] .$$

By the analogous procedure as in derivation of the gradient of the first iteration we obtain

$$(\partial H/\partial u)_{u=0} = L \approx \tfrac{9}{4}\lambda^{-1/3} f^{-1}(E . \bar{f})^{-3}[(\bar{f}^3 - (1 + 3\cos^2 \vartheta)/\bar{f}^3 + \tfrac{5}{3}\cos \vartheta) . J +$$
$$+ E . E_1/36 . (5\bar{f}^3 + 3(1 + 3\cos^2 \vartheta)/\bar{f}^3 + 19 \cos \vartheta)] = \lambda^{-1/3} f^{-1}(\vartheta) . b(\vartheta) , \qquad (D26)$$

where

$$J = \int_0^\infty \exp(-s^3) \int_0^s \xi(EE_1(\xi) - E_1 E(\xi)) . \exp(\xi^3) \, d\xi \, ds . \qquad (D27)$$

If we substitute the first two terms of expansions (*16*) and (*15*) and (*D26*) into (*D25*) we obtain

$$\lambda^{1/3} b_0(\vartheta) + b_1(\vartheta) \approx \lambda^{1/3} a_0(\vartheta) + a_1(\vartheta) + b(\vartheta)$$

and from there

$$b_0(\vartheta) = a_0(\vartheta) , \quad b_1(\vartheta) = a_1(\vartheta) + b(\vartheta) .$$

So we have proved the statements following Eq. (*16*) in the main part of this paper and formulae (*18*) are verified where for the appearing integrals are substituted numerical values. Moreover it holds

$$\lim_{\lambda \to \infty} (\partial G_2/\partial y)_{y=0} = \lim_{\lambda \to \infty} (b_0(\vartheta) + b_1(\vartheta) \,.\, \lambda^{-1/3}) = b_0(\vartheta)\,,$$

which means that for sufficiently large values λ the concentration gradient for the given ϑ is proportional to the quantity $f(\vartheta)$:

$$(\partial C/\partial y)_{y=0} \approx \lambda^{1/3}(\partial G_2/\partial y)_{y=0} \approx \lambda^{1/3}\, b_0(\vartheta) = \text{konst.}\, f(\vartheta)\,.$$

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