NATURAL CONVECTIVE DIFFUSION TO A SPHERE

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The problem of natural convective diffusion to a sphere was solved both analytically for the asymptotic case Gr. Sc $\to \infty$ and numerically for values of the product Gr.Sc from about 1 000 to 20 000. Evaluation of numerical results gave basic characteristics of the velocity profile and the form of the diffusion layers. A formula for the total diffusion current to a sphere in the mentioned range of the product Gr.Sc was obtained by combining the analytical solution with the numerical results.

The problems of free convection play a significant role in various physico-chemical and chemical engineering applications. They were solved hitherto especially for systems with a plane boundary, *e.g.*, with a plate-shaped electrode[1-3,5,6], and in the case of axial symmetric systems mostly for heat transfer[7-12].The convective mass transfer is characterized during forced convection[4,13] and according to our results also during free convection by the circumstance that the boundary diffusion layer is by an order of magnitude thinner than the Prandtl boundary layer. For this reason, it was necessary to use rather effective approximation methods during the numerical solution.

THEORETICAL

Mathematical Formulation of the Boundary Value Problem

Spherical coordinates r, φ, and ϑ are best suited for the mathematical description of convective diffusion phenomena in the vicinity of a sphere. If we choose the z axis perpendicular to the earth's surface, then the sought components of the flow velocity, v_r and v_ϑ, and concentration c are independent for symmetry reasons of the polar angle φ and the velocity component $v_\varphi = 0$. The corresponding boundary value problem for stationary convective diffusion at zero concentration on the sphere surface and non-zero concentration c_0 at infinity is then given by the following system of partial differential equations:

$$v_r\, \partial c/\partial r + r^{-1} v_\vartheta\, \partial c/\partial \vartheta = D \Delta_r c\,, \tag{1a}$$

$$v_r\,\partial v_r/\partial r + r^{-1}v_\vartheta\,\partial v_r/\partial\vartheta - r^{-1}v_\vartheta^2 = -k\boldsymbol{g}(c - c_0)\,\varrho^{-1}\cos\vartheta - \varrho^{-1}\,\partial p/\partial r +$$
$$+ \nu(\Delta_r v_r - 2r^{-2}v_r - 2r^{-2}v_\vartheta\operatorname{cotg}\vartheta - 2r^{-2}\,\partial v_\vartheta/\partial\vartheta)\,, \qquad (1b)$$

$$v_r\,\partial v_\vartheta/\partial r + r^{-1}v_\vartheta\,\partial v_\vartheta/\partial\vartheta + r^{-1}v_r v_\vartheta = k\boldsymbol{g}(c - c_0)\,\varrho^{-1}\sin\vartheta -$$
$$- (\varrho r)^{-1}\,\partial p/\partial\vartheta + \nu(\Delta_r v_\vartheta - r^{-2}\sin^{-2}\vartheta v_\vartheta + 2r^{-2}\,\partial v_r/\partial\vartheta)\,, \qquad (1c)$$

$$\partial v_r/\partial r + 2r^{-1}v_r + r^{-1}\,\partial v_\vartheta/\partial\vartheta + r^{-1}v_\vartheta\operatorname{cotg}\vartheta = 0\,, \qquad (1d)$$

where

$$\Delta_r = \partial^2/\partial r^2 + 2r^{-1}\,\partial/\partial r + r^{-2}\,\partial^2/\partial\vartheta^2 + r^{-2}\operatorname{cotg}\vartheta\,\partial/\partial\vartheta\,.$$

The boundary conditions are

$$c(a, \vartheta) = 0\,, \qquad \lim_{r\to\infty} c(r, \vartheta) = c_0 > 0\,,$$
$$\partial c/\partial\vartheta(r, 0) = 0\,, \qquad \partial c/\partial\vartheta(r, \pi) = 0\,, \qquad (2a)$$

$$v_r(a, \vartheta) = 0\,, \qquad \lim_{r\to\infty} v_r(r, \vartheta) = 0\,,$$
$$\partial v_r/\partial\vartheta(r, 0) = 0\,, \qquad \partial v_r/\partial\vartheta(r, \pi) = 0\,, \qquad (2b)$$

$$v_\vartheta(a, \vartheta) = 0\,, \qquad \lim_{r\to\infty} v_\vartheta(r, \vartheta) = 0\,,$$
$$\partial v_\vartheta/\partial\vartheta(r, 0) = 0\,, \qquad \partial v_\vartheta/\partial\vartheta(r, \pi) = 0\,. \qquad (2c)$$

Here D denotes diffusion coefficient, ϱ density of the solution, $k = (\partial\varrho/\partial c)_{c=c_0}$, $\boldsymbol{g}$ denotes acceleration of gravity, p pressure, ν kinematic viscosity, a radius of the sphere.

For the solution of the given problem, it is advantageous to introduce the stream function ψ

$$v_r = r^{-2}\sin^{-1}\vartheta\,.\,\partial\psi/\partial\vartheta\,, \qquad v_\vartheta = -r^{-1}\sin^{-1}\vartheta\,.\,\partial\psi/\partial r\,, \qquad (3)$$

the dimensionless variables

$$y = (r - a)/a\,, \qquad \tilde{C} = c/c_0\,, \qquad \tilde{\Psi} = \psi/\nu a\,,$$

and criteria

$$Gr = k\boldsymbol{g}c_0a^3\varrho^{-1}\nu^{-2}\,, \qquad Sc = \nu/D\,.$$

By eliminating the pressure p and rearranging, Eqs ($1a-c$) take the form

$$(1 + y)^{-2} \sin^{-1}\vartheta \, \mathrm{Sc}[(\partial\tilde{C}/\partial y)\,\partial\tilde{\Psi}/\partial\vartheta - (\partial\tilde{C}/\partial\vartheta)\,\partial\tilde{\Psi}/\partial y] = \partial^2\tilde{C}/\partial y^2 + $$
$$+ 2(1 + y)^{-1}\,\partial\tilde{C}/\partial y + (1 + y)^{-2}\,\partial^2\tilde{C}/\partial\vartheta^2 + (1 + y)^{-2}\cot\vartheta \, . \, \partial\tilde{C}/\partial\vartheta\,, \qquad (4a)$$

$$(1 + y)^{-2} \sin^{-1}\vartheta[(\partial\Omega_y\tilde{\Psi}/\partial y)\,\partial\tilde{\Psi}/\partial\vartheta - (\partial\Omega_y\tilde{\Psi}/\partial\vartheta)\,\partial\tilde{\Psi}/\partial y] + 2(1 + y)^{-2}\,.$$
$$. \sin^{-2}\vartheta \, . \, \Omega_y\tilde{\Psi}(\cos\vartheta \, . \, \partial\tilde{\Psi}/\partial y - (1 + y)^{-1}\sin\vartheta \, . \, \partial\tilde{\Psi}/\partial\vartheta) = \Omega_y\Omega_y\tilde{\Psi} -$$
$$- Gr[(1 + y)\sin^2\vartheta \, . \, \partial\tilde{C}/\partial y + \sin\vartheta\cos\vartheta \, . \, \partial\tilde{C}/\partial\vartheta] + kc_0\varrho^{-1}(M\,\partial\tilde{C}/\partial y +$$
$$+ N\,\partial\tilde{C}/\partial\vartheta)\,, \qquad (4b)$$

where

$$\Omega_y = \partial^2/\partial y^2 + (1 + y)^{-2}\,\partial^2/\partial\vartheta^2 - (1 + y)^{-2}\cot\vartheta \, . \, \partial/\partial\vartheta\,. \qquad (5a)$$

Eq. ($1d$) is identically fulfilled by the stream function defined by (3). The last term in Eq. ($4b$) was formed from the terms containing the derivatives $\partial\varrho/\partial r$ and $\partial\varrho/\partial\vartheta$ by using the relations $\partial\varrho/\partial r = (\partial\varrho/\partial c)\,\partial c/\partial r = kc_0a^{-1}\,\partial\tilde{C}/\partial y$, $\partial\varrho/\partial\vartheta = (\partial\varrho/\partial c)\,.\,\partial c/\partial\vartheta = kc_0\,\partial\tilde{C}/\partial\vartheta$. Since the ratio of the terms $kc_0\varrho^{-1}/Gr = \nu^2 g^{-1}a^{-3}$ occurring on the right side of Eq. ($4b$) is in practical situations of the order of 10^{-4} or smaller, we neglect the terms with the multiplicative factor $kc_0\varrho^{-1}$. Thus, we obtain instead of Eq. ($4b$)

$$(1 + y)^{-2}\sin^{-1}\vartheta[(\partial\Omega_y\tilde{\Psi}/\partial y)\,\partial\tilde{\Psi}/\partial\vartheta - (\partial\Omega_y\tilde{\Psi}/\partial\vartheta)\,\partial\tilde{\Psi}/\partial y] +$$
$$+ 2(1 + y)^{-2}\sin^{-2}\vartheta \, . \, \Omega_y\tilde{\Psi}[(\partial\tilde{\Psi}/\partial y)\cos\vartheta - (\partial\tilde{\Psi}/\partial\vartheta)(1 + y)^{-1}\sin\vartheta] =$$
$$= \Omega_y\Omega_y\tilde{\Psi} - Gr((1 + y)\sin^2\vartheta \, . \, \partial\tilde{C}/\partial y + \sin\vartheta\cos\vartheta \, . \, \partial\tilde{C}/\partial\vartheta)\,. \qquad (5b)$$

The boundary conditions for the Eqs ($4a$) and ($5b$) are

$$\tilde{C}(0, \vartheta) = 0\,, \qquad \lim_{y\to\infty}\tilde{C}(y, \vartheta) = 1\,,$$
$$\partial\tilde{C}/\partial\vartheta(y, 0) = 0\,, \qquad \partial\tilde{C}/\partial\vartheta(y, \pi) = 0\,, \qquad (6a)$$

$$\tilde{\Psi}(0, \vartheta) = 0\,, \qquad \lim_{y\to\infty}\tilde{\Psi}(y, \vartheta) = 0\,,$$
$$\tilde{\Psi}(y, 0) = 0\,, \qquad \tilde{\Psi}(y, \pi) = 0\,, \qquad (6b)$$

$$\partial\tilde{\Psi}/\partial y(0, \vartheta) = 0\,, \qquad \lim_{y\to\infty}\partial\tilde{\Psi}/\partial y(y, \vartheta) = 0\,,$$
$$\partial\tilde{\Psi}/\partial\vartheta(y, 0) = 0\,, \qquad \partial\tilde{\Psi}/\partial\vartheta(y, \pi) = 0\,. \qquad (6c)$$

The conditions $(6b, c)$ except for the limits follow from the corresponding conditions $(2b, c)$ and from the fact that the function ψ is given by Eqs (3) regardless of an additive constant. The limit in $(6b)$ follows from the physically plausible assumption that the radial velocity component v_r decreases to zero for $r \to \infty$ faster than r^{-2}; and the limit in $(6c)$ analogously from the assumption that the tangential velocity component v_ϑ diminishes faster than r^{-1}.

Approximate Analytical Solution for Gr . Sc → ∞

The functions $\tilde{C}$ and $\tilde{\Psi}$ can close to the sphere surface be expressed by series in powers of y, where it is advantageous to separate the multiplicative factor Gr in the series for $\tilde{\Psi}$:

$$\tilde{C}(y, \vartheta) = a_0(\vartheta) + a_1(\vartheta)\, y + a_2(\vartheta)\, y^2 + \ldots, \tag{7a}$$

$$Gr^{-1}\tilde{\Psi}(y, \vartheta) = A_0(\vartheta) + A_1(\vartheta)\, y + A_2(\vartheta)\, y^2 + A_3(\vartheta)\, y^3 + \ldots. \tag{7b}$$

The boundary conditions on the sphere surface give

$$a_0(\vartheta) = 0\,, \quad A_0(\vartheta) = 0\,, \quad A_1(\vartheta) = 0\,.$$

The coefficient $a_1(\vartheta)$ has the meaning of the concentration gradient on the sphere surface. We introduce a new variable u and functions C, Ψ by the relations

$$u = a_1(\vartheta)\, y\,, \tag{8}$$

$$C(u, \vartheta) = \tilde{C}(a_1^{-1}(\vartheta)\, u, \vartheta)\,, \quad \Psi(u, \vartheta) = \tilde{\Psi}(a_1^{-1}(\vartheta)\, u, \vartheta)\,. \tag{9}$$

As shown in the Appendix, the diffusion equation $(4a)$ can be after substitution of (8) approximated as

$$a_1\, \partial^2 C/\partial u^2 + (2 - Sc \sin^{-1} \vartheta\, \partial\Psi/\partial\vartheta)\, \partial C/\partial u = 0\,. \tag{10}$$

This equation can be considered as an ordinary differential one for the function C with parameter ϑ. Its solution is obtained in an elementary way with regard to the conditions $(6a)$ in the form

$$C(u, \vartheta) = J^{-1} \int_0^u \exp\left[a_1^{-1} \int_0^w (Sc \sin^{-1} \vartheta\, \partial\Psi/\partial\vartheta - 2)\, dt\right] dw\,, \tag{11}$$

where

$$J = \int_0^\infty \exp\left[a_1^{-1} \int_0^w (Sc \sin^{-1} \vartheta \; \partial\Psi/\partial\vartheta - 2) \, dt\right] dw \,. \tag{12}$$

The formal solution (*11*) contains unknown functions a_1 and $\partial\Psi/\partial\vartheta$. From Eqs (*7a*), (*8*) and (*11*) we obtain

$$\partial C/\partial u(0, \vartheta) = 1 = J^{-1}$$

hence

$$J = 1 \,. \tag{13}$$

This equation serves to calculate $a_1(\vartheta)$. A differential equation for the function a_1 is derived from it in the second part of the Appendix in the form

$$3a_1^{-1}a_1' \sin \vartheta - 2 \cos \vartheta = 120 \, Gr^{-1}Sc^{-1}H^5a_1^4 \,, \tag{14}$$

where $H = \int_0^\infty \exp(-x^5) \, dx \approx 0{\cdot}91817$. Its solution is

$$a_1(\vartheta) = \tfrac{1}{2} \sqrt[4]{(Gr \,.\, Sc)} / \sqrt[4]{(10H^5)} \; h(\vartheta) \,, \tag{15}$$

where

$$h(\vartheta) = \sin^{2/3} \vartheta / \sqrt[4]{\int_\vartheta^\pi \sin^{5/3} \tau \, d\tau} \,. \tag{16}$$

Now it is possible to express the function C in (*11*) for the asymptotic case $Gr \,.\, Sc \rightarrow \rightarrow \infty$. We obtain in an analogous way as in rearranging the integral (*12*) (see Appendix)

$$C(u, \vartheta) = H^{-1} \int_0^{Hu} \exp(-x^5) \, dx \,. \tag{17}$$

We used this asymptotic formula in calculating the starting iteration of C during numerical solution of the given boundary problem.

The formula (*15*) for the concentration gradient on the sphere surface was derived by solving Eq. (*D7*), which is an asymptotic approximation of (*D6*). If we express the integral on the left side of Eq. (*D6*) with the use of the Taylor expansion for the function $\exp(2\omega^{-1}a_1^{-1}x)$ and introduce the parameter

$$\eta = (Gr \,.\, Sc)^{-1/4} \,, \tag{18}$$

we obtain the following asymptotic expansion of the gradient $a_1(\vartheta)$

$$a_1(\vartheta) = \eta^{-1}(a_1^{(0)}(\vartheta) + a_1^{(1)}(\vartheta)\,\eta + a_1^{(2)}(\vartheta)\,\eta^2 + \ldots)\,, \qquad (19)$$

where $a_1^{(0)}(\vartheta) = \frac{1}{2}h(\vartheta)/\sqrt[4]{(10H^5)} \approx 0{\cdot}31284h(\vartheta)$, as follows from the approximation (*15*). The values of the angle function h and function $a_1^{(0)}$ are given in Table I. The analytical calculation of the coefficients $a_1^{(1)}(\vartheta)$, $a_1^{(2)}(\vartheta)$, ... is very difficult, hence we shall not deal with it. We use the expansion (*19*) with several terms as an empirical formula in evaluating numerical results.

Numerical Solution

It is necessary to solve the boundary value problem (*4a*), (*5b*), (*6a*–*c*). At first, we shall study the influence of the quadratic terms in (*5b*). We introduce a new function $\hat{\Psi}$

$$\tilde{\Psi} = Gr\hat{\Psi}\,. \qquad (20)$$

The system (*4a*) and (*5b*) takes then the form

$$Gr\,Sc(1 + y)^{-2}\sin^{-1}\vartheta[(\partial\tilde{C}/\partial y)\,\partial\hat{\Psi}/\partial\vartheta - (\partial\tilde{C}/\partial\vartheta)\,\partial\hat{\Psi}/\partial y] = \partial^2\tilde{C}/\partial y^2 +$$
$$+ 2(1 + y)^{-1}\,\partial\tilde{C}/\partial y + (1 + y)^{-2}\,\partial^2\tilde{C}/\partial\vartheta^2 + (1 + y)^{-2}\operatorname{cotg}\vartheta\,\partial\tilde{C}/\partial\vartheta\,, \qquad (21a)$$

TABLE I
Values of functions h and $a_1^{(0)}$

ϑ^0	$h(\vartheta)$	$a_1^{(0)}(\vartheta)$	ϑ^0	$h(\vartheta)$	$a_1^{(0)}(\vartheta)$
10	0·27343	0·08554	100	1·09479	0·34249
20	0·43083	0·13478	110	1·13872	0·35623
30	0·55850	0·17472	120	1·17625	0·36797
40	0·66756	0·20884	130	1·20764	0·37779
50	0·76273	0·23861	140	1·23310	0·38576
60	0·84657	0·26484	150	1·25277	0·39191
70	0·92073	0·28804	160	1·26674	0·39628
80	0·98632	0·30856	170	1·27510	0·39890
90	1·04414	0·32665	180	1·27789	0·39977

$$Gr\{(1+y)^{-2}\sin^{-1}\vartheta[(\partial\Omega_y\hat{\Psi}/\partial y)\,\partial\hat{\Psi}/\partial\vartheta - (\partial\Omega_y\hat{\Psi}/\partial\vartheta)\,\partial\hat{\Psi}/\partial y] +$$
$$+ 2\Omega_y\hat{\Psi}(1+y)^{-2}\sin^{-2}\vartheta[(\partial\hat{\Psi}/\partial y)\cos\vartheta - (\partial\hat{\Psi}/\partial\vartheta)\sin\vartheta(1+y)^{-1}]\} =$$
$$= \Omega_y\Omega_y\hat{\Psi} - [(1+y)\sin^2\vartheta\,\partial\tilde{C}/\partial y + \sin\vartheta\cos\vartheta\,\partial\tilde{C}/\partial\vartheta]\,. \qquad (21b)$$

If we choose $Gr\,.\,Sc = \eta^{-4}$ = const., then it follows from Eq. (*21b*) that the quadratic terms in it are negligible for sufficiently small Gr values. To gain some idea about their influence, we solved the system (*21a*, *b*) in the finite region $(0, b) \times (0, \pi)$ by the finite difference method; these calculations were repeated after omitting the quadratic terms in Eq. (*21b*). We chose in turn $Gr = 1, 10, 100, 500$, and $b = 2, 2{\cdot}4, 2{\cdot}8$ at constant $Gr\,.\,Sc = 10^4$. It is apparent from the numerical results that the quadratic terms in Eq. (*21b*) play no role at $Gr \leqq 100$ within the limits of the computational accuracy, hence we neglected them in further calculations. Thus, Eq. (*21b*) is linearized and the solution $\tilde{C}$, $\tilde{\Psi}$ of the mentioned system will depend on a single parameter, η, defined by Eq. (*18*). The stream function $\tilde{\Psi}$ and the radial and tangential components of the velocity derived from it will be directly proportional to the criterion Gr at constant η as follows from Eq. (*20*).

For the numerical solution of the simplified system, it is advantageous to introduce an auxiliary function $\tilde{\Phi}$:

$$\Omega_y\tilde{\Psi} = Gr\tilde{\Phi}\,. \qquad (22)$$

Thus, Eq. (*5b*) takes after neglecting the mentioned terms the form of a second-order equation

$$\Omega_y\tilde{\Phi} = (1+y)\sin^2\vartheta\,\partial\tilde{C}/\partial y + \sin\vartheta\cos\vartheta\,\partial\tilde{C}/\partial\vartheta\,. \qquad (23)$$

To pass from the infinite region $(0, \infty) \times (0, \pi)$ to a finite one, we introduce a new variable z instead of y defined as $z = y/(1+y)$, *i.e.*, $y = z/(1-z)$, and functions C, Φ, Ψ defined as

$$C(z,\vartheta) = \tilde{C}(z/(1-z),\vartheta)\,,\quad \Phi(z,\vartheta) = \tilde{\Phi}(z/(1-z),\vartheta)\,,$$
$$\Psi(z,\vartheta) = \tilde{\Psi}(z/(1-z),\vartheta)\,. \qquad (24)$$

Thus, the original region $(0, \infty) \times (0, \pi)$ is transformed to $(0, 1) \times (0, \pi)$ and the system of equations (*4a*), (*22*), (*23*) to the following one for the functions C, Φ, Ψ:

$$(1-z)^2\,\partial^2 C/\partial z^2 + \partial^2 C/\partial\vartheta^2 + \operatorname{cotg}\vartheta\,\partial C/\partial\vartheta -$$
$$-(1-z)^2\,Sc\,.\,\sin^{-1}\vartheta[(\partial C/\partial z)\,\partial\Psi/\partial\vartheta - (\partial C/\partial\vartheta)\,\partial\Psi/\partial z] = 0\,, \qquad (25a)$$

$$(1 - z)^2 \Omega_z \Phi - (1 - z) \sin^2 \vartheta \, \partial C/\partial z - \sin \vartheta \cos \vartheta \, \partial C/\partial \vartheta = 0 \,, \qquad (25b)$$

$$(1 - z)^2 \Omega_z \Psi - Gr\Phi = 0 \,, \qquad (25c)$$

where

$$\Omega_z = (1 - z)^2 \, \partial^2/\partial z^2 + \partial^2/\partial \vartheta^2 - 2(1 - z) \, \partial/\partial z - \operatorname{cotg} \vartheta \, \partial/\partial \vartheta \,.$$

The boundary conditions $(6a-c)$ take the form

$$C(0, \vartheta) = 0 \,, \quad C(1, \vartheta) = 1 \,, \quad \partial C/\partial \vartheta(z, 0) = \partial C/\partial \vartheta(z, \pi) = 0 \,, \qquad (26a)$$

$$\Psi(0, \vartheta) = \Psi(1, \vartheta) = \Psi(z, 0) = \Psi(z, \pi) = 0 \,, \qquad (26b)$$

$$\partial \Psi/\partial z(0, \vartheta) = \partial \Psi/\partial z(1, \vartheta) = \partial \Psi/\partial \vartheta(z, 0) = \partial \Psi/\partial \vartheta(z, \pi) = 0 \,. \qquad (26c)$$

The boundary value problem modified in this way was solved approximately by the method of finite differences. The partial derivatives in *(25a—c)* were approximated by symmetrical relative differences with an order of accuracy of h^2, where h denotes the grid spacing, and in the boundary conditions *(26a,c)* by nonsymmetrical relative differences with the same order of accuracy.

We shall denote: m number of divisions in the interval $\langle 0, 1 \rangle$, n number of divisions in the interval $\langle 0, \pi \rangle$, $h_z = m^{-1}$ step of the variable z, $h_\vartheta = \pi/n$ step of the variable ϑ, $(z_i, \vartheta_j) = (ih_z, jh_\vartheta)$ coordinates of a general grid point of the chosen network, $C_{ij} = C(z_i, \vartheta_j)$ and analogously Φ_{ij} and Ψ_{ij}.

Further we denote Resc_{ij}, Resf_{ij}, Resp_{ij} the values of the difference approximations of the left sides of Eqs *(25a—c)* and *(26a,c)* in a general grid point (z_i, ϑ_j); Eq. *(25a)* was moreover divided by Sc in order that the numbers Resc_{ij} be comparable with Resf_{ij} and Resp_{ij} with regard to their order of magnitude.

The nonlinear system of difference equations for the vector of unknowns $\mathbf{x} = (C_{ij}, \Phi_{ij}, \Psi_{ij})$ formed by the mentioned discretization was solved by a modified gradient method according to the iteration scheme

$$\mathbf{x}^{(k+1)} = \mathbf{x}^{(k)} - \lambda_k \boldsymbol{H} \operatorname{grad} G \,, \quad k = 0, 1, 2, \ldots \qquad (27)$$

where

$$G = \sum_{i,j} \left(\mathrm{Resc}_{ij}^2 + \mathrm{Resf}_{ij}^2 + \mathrm{Resp}_{ij}^2 \right) ,$$

λ_k is a suitably chosen parameter, $\boldsymbol{H}$ a suitably chosen diagonal matrix. When we chose the latter equal to a unit matrix $\boldsymbol{E}$ (gradient method proper), it was necessary for the convergence to be achieved to choose a very small value of λ_k so that the convergence of the series $(\mathbf{x}^{(k)})$ was very slow. We therefore chose the matrix

$$H = \begin{bmatrix} r_c & & & & & & & \\ & \ddots & & & & & & \\ & & r_c & & & & & \\ & & & r_f & & & & \\ & & & & \ddots & & & \\ & & & & & r_f & & \\ & & & & & & r_p & \\ & & & & & & & \ddots \\ & & & & & & & & r_p \end{bmatrix} . \qquad (28)$$

The constants r_c, r_f, r_p modify in turn the parameter λ_k for the components of grad G corresponding to the unknowns C_{ij}, Φ_{ij}, Ψ_{ij}. The numbers r_c, r_f, r_p were estimated separately by solving the difference system for $r_f = r_p = 0$ according to the scheme (*27*) and finding a suitable value of r_c, and proceeding analogously to find r_f and r_p. These three numbers were after a minor used to form the matrix $\boldsymbol{H}$, whereby the convergence of the sequence $(\boldsymbol{x}^{(k)})$ was considerably accelerated. This can be seen from the used values of $\lambda_0 = 1$, $r_c = 6$, $r_f = 0{\cdot}15$, $r_p = 3 \, . \, 10^{-5}$ as compared with $\lambda_0 = 3 \, . \, 10^{-5}$ corresponding to the choice of $\boldsymbol{H} = \boldsymbol{E}$.

It is known that the sequence of iterations in the gradient method converges at a suitable choice of λ_k. The criterion of the convergence is the diminishing of the function G with increasing k. We therefore checked the values of G after every iteration. To speed up the convergence, we started from the initial value of λ_0 which we multiplied after every 25 iterations by a suitably chosen factor of $r_1 > 1$ if the values of G decreased. As soon as they began to increase, we divided in turn the obtained factor λ_k by a factor of $r_2 > r_1$ after every iteration until the values of G began again to decrease.

The most important physical quantity in our problem is the concentration and its gradient on the sphere surface. Therefore, the following quantities were calculated in addition:

$$d_1^{(k)} = \max_{i,j} \left| C_{ij}^{(k+1)} - C_{ij}^{(k)} \right| ,$$

$$d_2^{(k)} = \left[\sum_{i,j} \left(C_{ij}^{(k+1)} - C_{ij}^{(k)} \right)^2 \right]^{1/2} .$$

The problem under study was solved numerically on an ICL 4—72 type computer. The grid parameters were $m = 20$, $n = 18$. The input iteration for C was obtained from Eq. (*17*), the input values of Φ and Ψ were set initially equal to zero and then their values were derived from preliminary calculations by the relaxation method.

RESULTS AND DISCUSSION

The solution of the boundary value problem for stationary free convection as formulated in the theoretical part gives the values of the functions Ψ and C. The first one characterizes the hydrodynamic conditions in the vicinity of the sphere due to the proceeding diffusion. The stationary state is the result of interaction of two counteracting effects, namely gravitation, which causes a motion of solution layers of different concentration and hence of different density, and viscosity, which hinders this motion. Since the concentration gradient is the proper reason of the motion, it is obvious that the motion will be most pronounced near to the sphere surface while it will extinguish with increasing distance from the sphere. Thus, the streaming velocity will increase with distance from zero on the surface to a certain maximum and then it will slowly decrease to zero, theoretically at an infinite distance. Its magnitude and direction will depend, in addition, at constant *Gr* and *Sc* values also on the spherical angle ϑ. We shall assume that the z axis of the coordinate system is perpendicular and oriented oppositely to the gravitational force and that $Gr > 0$. This means that the solution density increases with increasing concentration, hence the solution layer at the sphere surface is lighter than the more distant one, so that a motion takes place in the upward direction. For values of $\vartheta \approx \pi$, *i.e.*, under the sphere (with respect to the Earth), the streaming starts to form and is hence relatively weak, whereas on the opposite side (above the sphere), for $\vartheta \approx 0$, it is strongest.

To express this situation quantitatively, we introduce two dimensionless quantities analogous to the Reynolds criterion, characterizing the streaming in both radial and tangential directions:

$$(Re)_r = av^{-1}v_r\,, \quad (Re)_\vartheta = av^{-1}v_\vartheta\,. \tag{29a, b}$$

Both these quantities depend on the coordinates r, ϑ, eventually y, ϑ, and can be expressed by means of the stream function $\Psi(z, \vartheta)$ as follows:

$$(Re)_r = (1 - z)^2 \sin^{-1} \vartheta \, \partial\Psi/\partial\vartheta\,, \tag{30a}$$

$$(Re)_\vartheta = -(1 - z)^3 \sin^{-1} \vartheta \, \partial\Psi/\partial z\,. \tag{30b}$$

Vectors with radial components $(Re)_r$ and tangential components $(Re)_\vartheta$ for the case $Gr = 1$, $Sc = 10^4$ are shown in Fig. 1, representing in essence the velocity field in the vicinity of the sphere and illustrating our qualitative considerations.

The absolute value of the velocity vector,

$$V = [(Re)_r^2 + (Re)_\vartheta^2]^{1/2} = av^{-1}(v_r^2 + v_\vartheta^2)^{1/2} \tag{31}$$

as a function of the relative distance, y, from the sphere surface is shown in Fig. 2 for four different values of ϑ and for $Gr = 1$, $Sc = 10^4$. This figure also substantiates our qualitative considerations and shows that the maximum velocity is attained in a relatively small distance from the sphere surface, namely $y \approx 0{\cdot}2$ at the mentioned values of *Gr* and *Sc*, and this is only little dependent on the angle ϑ.

Further we calculated the dependence of the character of streaming on the Schmidt criterion *Sc* at constant *Gr*; the results are shown in Fig. 3, whence it is seen that the dependence of V on y for $\vartheta = 90°$ and $Gr = 1$ is for all four selected values of *Sc* qualitatively the same: the position of the maximum does not change much but its height diminishes appreciably with increasing *Sc*. This effect can be physically easily explained. Increasing *Sc* means either increasing viscosity or smaller diffusion coefficient. This means in the first case a larger hindrance of motion, in the second a thinner diffusion layer bringing about hindering by the sphere surface where the streaming velocity is zero. The form of the differential equations (*4a*), (*5b*) suggests that both these factors have the same effect, which can be determined from Fig. 3.

The dependence of the streaming on the value of *Gr* at constant product $Gr \,.\, Sc$ was discussed in the section Numerical Solution, where it was stated that the stream function Ψ and hence the velocities derived from it are directly proportional to *Gr* up to rather high values of *Gr*.

The second calculated function, the concentration *C*, enables to determine the most important physical quantity, the diffusion flux to the sphere, which is directly

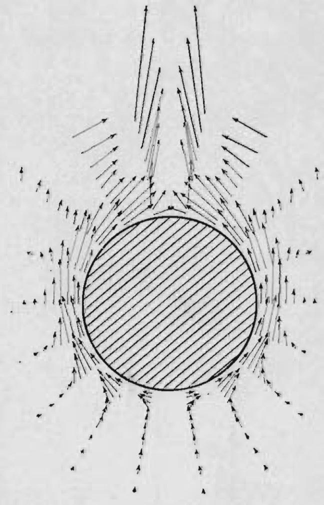

FIG. 1

Velocity field in the vicinity of a sphere

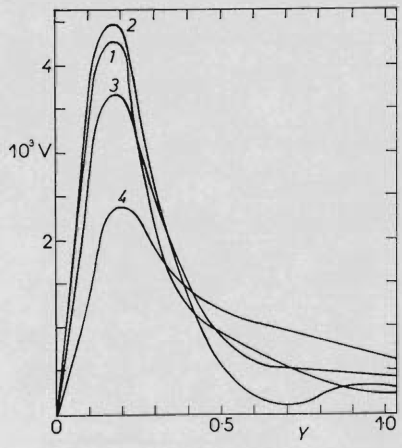

FIG. 2

Diagrams of the function V (formula (*31*)). *1* V for $\vartheta = 60°$; *2* V for $\vartheta = 90°$; *3* V for $\vartheta = 120°$; *4* V for $\vartheta = 150°$

proportional to the concentration gradient on the sphere surface. The obtained values of this function in the grid points served to calculate approximate values of the concentration gradient on the sphere surface according to the difference formula

$$\partial\tilde{C}/\partial y(0, kh) \approx (12h_z)^{-1}(-25C_{0k} + 48C_{1k} - 36C_{2k} + 16C_{3k} - 3C_{4k}), \quad (32)$$

for $k = 1, 2, \ldots 18$. Fig. 4 shows the theoretical diffusion layers, whose thickness d was calculated from the numerical results according to

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

The values of the gradient from Eq. (*32*) were used to calculate the total diffusion flow to the sphere

$$Q = 4\pi a^2 c_0 DK , \quad (33)$$

where

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

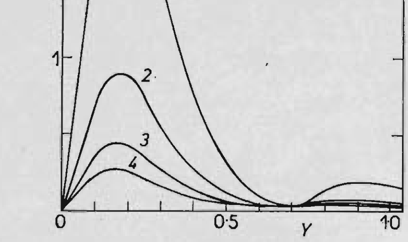

FIG. 3

Diagrams of the function V (formula (*31*)) for $\vartheta = 90°$. *1* V for $Sc = 6^4$; *2* V for $Sc = 8^4$; *3* V for $Sc = 10^4$; *4* V for $Sc = 12^4$

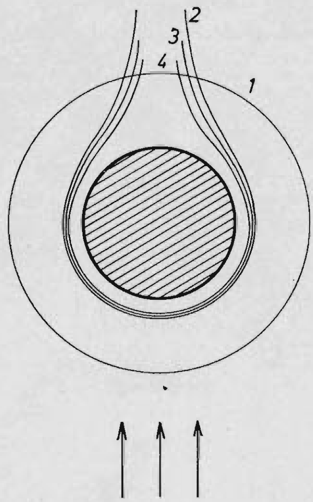

FIG. 4

Diffusion layers. *1* $Gr.Sc = 0$; *2* $Gr.Sc = 6^4$; *3* $Gr.Sc = 8^4$; *4* $Gr.Sc = 12^4$

If we introduce an approximation for the gradient $(\partial\tilde{C}/\partial y)_{y=0} = a_1(\vartheta)$ according to Eq. (*19*)

$$(\partial\tilde{C}/\partial y)_{y=0} \approx \eta^{-1} a_1^{(0)}(\vartheta) + a_1^{(1)}(\vartheta) + a_1^{(2)}(\vartheta)\, \eta\,,$$

we obtain

$$K = \frac{1}{2}\left(\eta^{-1}\int_0^\pi a_1^{(0)}(\vartheta)\sin\vartheta\,\mathrm{d}\vartheta + \int_0^\pi a_1^{(1)}(\vartheta)\sin\vartheta\,\mathrm{d}\vartheta + \eta\int_0^\pi a_1^{(2)}(\vartheta)\sin\vartheta\,\mathrm{d}\vartheta\right) =$$

$$= l_0\eta^{-1} + l_1 + l_2\eta\,. \qquad (35)$$

The value of $l_0 \approx 0{\cdot}30812$ was calculated by numerical integration based on the values of $a_1^{(0)}(\vartheta)$ given in Table I. The other constants l_1 and l_2 were determined by the least squares method from the values of K for $\eta^{-1} = 6, 7, 8, 9$, and 10. Thus, we obtained an approximate formula for the whole diffusion flow

$$Q = 4\pi a^2 c_0 D(0{\cdot}308\eta^{-1} + 0{\cdot}568 + 8{\cdot}525\eta)\,. \qquad (36)$$

Table II summarizes the values, K_n, calculated by numerical integration using Eqs (*34*) and (*32*), and the values, K_e, calculated from Eq. (*35*) for different values of η^{-1}. The deviations, in percent, given in the last column show that the approximate formula (*35*) fits very well the values of K_n and thus the diffusion flow Q. However, it should be pointed out that the values of K_n obtained from the numerical solution may be subject to larger errors than the given deviations with respect to the complexity of the system of difference equations under study.

TABLE II
Values of K_n and K_e

η^{-1}	K_n	K_e	$(K_e - K_n)/K_n \,.\, 100$
6	3·85912	3·837	—0·57
7	3·93651	3·942	0·14
8	4·09667	4·098	0·03
9	4·27028	4·287	0·39
10	4·51347	4·501	—0·28

APPENDIX

Approximation of the Diffusion Equation

We start from Eq. (*4a*) and introduce the variable u and functions C and Ψ according to Eqs (*8*) and (*9*). Thus, we obtain

$$(1 + u/a_1)^{-2} \sin^{-1}\vartheta\, a_1 Sc(\partial C/\partial u\, \partial\Psi/\partial\vartheta - \partial C/\partial\vartheta\, \partial\Psi/\partial u) = a_1^2\, \partial^2 C/\partial u^2 + $$
$$+ 2a_1(1 + u/a_1)^{-1}\, \partial C/\partial u + (1 + u/a_1)^{-2} \operatorname{cotg}\vartheta(ua_1'/a_1\, \partial C/\partial u + \partial C/\partial\vartheta) +$$
$$+ (1 + u/a_1)^{-2}[(ua_1'/a_1)^2\, \partial^2 C/\partial u^2 + u(a_1'/a_1)^2\, \partial C/\partial u + u(a_1'/a_1)'\, \partial C/\partial u +$$
$$+ 2ua_1'/a_1\, \partial^2 C/\partial u\, \partial\vartheta + \partial^2 C/\partial\vartheta^2]\,. \qquad (D1)$$

After introducing Eq. (*8*), the series (*7a,b*) take the form

$$C(u, \vartheta) = u + a_2(\vartheta)/a_1^2(\vartheta)\, u^2 + a_3(\vartheta)/a_1^3(\vartheta)\, u^3 + \ldots, \qquad (D2a)$$

$$Gr^{-1}\Psi(u, \vartheta) = A_2(\vartheta)/a_1^2(\vartheta)\, u^2 + A_3(\vartheta)/a_1^3(\vartheta)\, u^3 + A_4(\vartheta)/a_1^4(\vartheta)\, u^4 + \ldots. \qquad (D2b)$$

Thus, we obtain

$$\partial C/\partial u = 1 + 2a_2/a_1^2 u + 3a_3/a_1^3 u^2 + \ldots,$$
$$\partial^2 C/\partial u^2 = 2a_2/a_1^2 + 6a_3/a_1^3 u + \ldots,$$
$$\partial C/\partial\vartheta = (a_2/a_1^2)'\, u^2 + (a_3/a_1^3)'\, u^3 + \ldots,$$
$$\partial^2 C/\partial\vartheta^2 = (a_2/a_1^2)''\, u^2 + (a_3/a_1^3)''\, u^3 + \ldots,$$
$$\partial^2 C/\partial u\, \partial\vartheta = 2(a_2/a_1^2)'\, u + 3(a_3/a_1^3)'\, u^2 + \ldots,$$
$$Gr^{-1}\, \partial\Psi/\partial u = 2A_2/a_1^2 u + 3A_3/a_1^3 u^2 + \ldots,$$
$$Gr^{-1}\, \partial\Psi/\partial\vartheta = (A_2/a_1^2)'\, u^2 + (A_3/a_1^3)'\, u^3 + \ldots.$$

When we substitute these expansions into Eq. (*D1*) and restrict them to the lowest powers of u, we obtain

$$\sin^{-1}\vartheta\, a_1 Sc\, \partial C/\partial u\, \partial\Psi/\partial\vartheta = a_1^2\, \partial^2 C/\partial u^2 + 2a_1\, \partial C/\partial u\,.$$

This simplification is justified because decisive changes of the sought functions C and Ψ occur at the sphere surface, *i.e.*, for small values of u. The simplified equation can be written as (*10*).

Calculation of the Function a_1

Eq. (*13*), which serves to calculate the function a_1, involves also the unknown function $\partial\Psi/\partial\vartheta$. Therefore, we need some information about the coefficients A_2, A_3, A_4 in the expansion (*7b*). When we substitute the expansions (*7a,b*) into Eq. (*5b*), we obtain by comparing the coefficients of the linear terms the condition

$$4!\, A_4 - 4 \operatorname{cotg}\vartheta A_2' + 4A_2'' - a_1 \sin^2\vartheta = 0\,,$$

whence

$$A_4 = (1/4!)\,(a_1 \sin^2 \vartheta + 4A_2' \operatorname{cotg} \vartheta - 4A_2'') \,. \tag{D3}$$

Preliminary numerical results showed that the coefficients A_2, A_3, eventually their derivatives are at least by 2—3 orders of magnitude smaller than a_1. Therefore, we can use the following approximations:

$$A_2(\vartheta) = 0\,, \quad A_3(\vartheta) = 0\,, \quad A_4(\vartheta) = (1/4!) \sin^2 \vartheta a_1(\vartheta)\,.$$

The expansion (*D2b*) then gives

$$\partial \Psi / \partial \vartheta = (Gr/4!)\, \mathrm{d}/\mathrm{d}\vartheta(\sin^2 \vartheta a_1^{-3}(\vartheta))\, u^4 + \ldots\,.$$

After substituting into Eqs (*12*) and (*13*) we obtain the equation

$$\int_0^\infty \exp\,\{a_1^{-1} \int_0^w [Gr\,.\,Sc(24 \sin \vartheta)^{-1}\, \mathrm{d}/\mathrm{d}\vartheta[\sin^2 \vartheta a_1^{-3}(\vartheta)]\, t^4 - 2]\, \mathrm{d}t\}\, \mathrm{d}w = 1$$

whence

$$\int_0^\infty \exp\,\{a_1^{-1}\, [Gr\,.\,Sc\,.\,120^{-1} a_1^{-4}(2a_1 \cos \vartheta - 3a_1' \sin \vartheta)\, w^5 - 2w]\}\, \mathrm{d}w = 1\,. \tag{D4}$$

If we denote

$$\omega^5 = Gr\,.\,Sc\,.\,120^{-1} a_1^{-5}(3a_1' \sin \vartheta - 2a_1 \cos \vartheta)\,, \tag{D5}$$

then it follows from Eq. (*D4*) that $\omega > 0$. After setting $\omega w = x$, Eq. (*D4*) takes the form

$$\omega^{-1} \int_0^\infty \exp\,(-x^5 + 2\omega^{-1} a_1^{-1} x)\, \mathrm{d}x = 1\,. \tag{D6}$$

Since for large values of ω we have $\exp\,(2\omega^{-1} a_1^{-1} x) \approx 1$, we obtain in the asymptotic case $Gr.Sc \to \infty$ the equation

$$H/\omega = 1\,. \tag{D7}$$

By combining Eqs (*D5*) and (*D7*), we obtain the differential equation (*14*) for the function a_1. If we introduce a new function f

$$f(\vartheta) = \sin^{2/3} \vartheta a_1^{-1}(\vartheta)\,,$$

we obtain from Eq. (*14*) after some rearrangement

$$-f^3\, \mathrm{d}f/\mathrm{d}\vartheta = 40 H^5 Gr^{-1} Sc^{-1} \sin^{5/3} \vartheta\,. \tag{D8}$$

Since $a_1(\pi) \neq 0$, we have $f(\pi) = 0$. The solution of Eq. (*D8*) has therefore the form

$$f(\vartheta) = \left(160H^5Gr^{-1}Sc^{-1}\int_\vartheta^\pi \sin^{5/3}\vartheta \, d\vartheta\right)^{1/4},$$

whence we obtain the formula (*15*).

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