
**CONCENTRATION BOUNDARY LAYER UNDER GENERAL
STEADY-FLOW CONDITIONS**

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Received March 30, 1990

Accepted April 20, 1990

An explicit formula is given for the overall mass-transfer coefficient between a steady liquid stream and a small active part of a solid surface in the stream. This is a generalization of the well-known Lighthill formula to the form applicable for any velocity field and any shape of the active surface. Its use is demonstrated for the circular electrodiffusion probes under various kinematic conditions.

Leveque¹ developed his formula for estimating heat transfer coefficients on the assumption that the velocity gradient at the transport-active surface, q , does not vary along the surface. Lighthill² generalized this approach for any planar flow by considering longitudinal variation of q . A further generalization was given by Newman³, who has analyzed two-dimensional electrochemical mass-transfer problems including the axisymmetric transport configurations. It is shown in the present paper that an analogous approach can be used for a general three-dimensional velocity field and for any shape of the active part of a solid surface, by considering the surface distribution of the vectorial velocity gradient \mathbf{q} as the starting kinematic information.

GENERALIZED LIGHTHILL TRANSFORMATION

In the present paper, we aim at the calculation of the diffusion flux J of an active component (a depolarizer) from the bulk of a liquid stream to an active part of a solid surface (an electrode), within the approximation of concentration boundary layer (the diffusion-layer approximation). The following three simplifying assumptions are typical for this approximation:

1. The total volume, occupied by the streaming liquid, can be divided into two parts: the bulk and the diffusion layer. The bulk consists of a fresh solution with a constant initial concentration of the depolarizer. The depolarizer concentration changes appreciably only across the diffusion layer close to the electrode surface.

2. The diffusion layer thickness is small enough in comparison with any characteristic length parameter of the velocity field inside the diffusion layer (the curvature radii of the surface itself, the surface streamlines, the velocity profiles – approximation for high Schmidt numbers.)

3. The diffusion fluxes, parallel to the electrode surface, can be neglected in comparison with the corresponding convective fluxes (approximation of high Péclet numbers).

Such conditions generally cannot be achieved in typical units for heat- and mass-transfer operations, like heat exchangers, agitated vessels, or absorption towers, where the interface nearly overall coincides with the transport-active surface. On the other hand, the regime of the concentration boundary layer is typical for all convective probes used in the electrodiffusion diagnostics of flow⁴⁻⁶.

Let us consider now a steady flow along a solid surface with the given surface field of velocity gradients. Assuming a non-permeable surface and no-slip conditions, the surface velocity gradient is fully represented by the vectorial field \mathbf{q} of the velocity gradients at wall which are locally complanar with the surface. It is useful to introduce the notion of surface streamlines. These oriented spacial curves lay in the surface and \mathbf{q} gives their directional field.

The special orthogonal curvilinear coordinate system (z, x, θ) can be introduced for mapping the given surface vectorial field \mathbf{q} into the three-dimensional Euclidean space, see Fig. 1.

The z -coordinate gives the distance to the solid surface E . In particular, $z = 0$

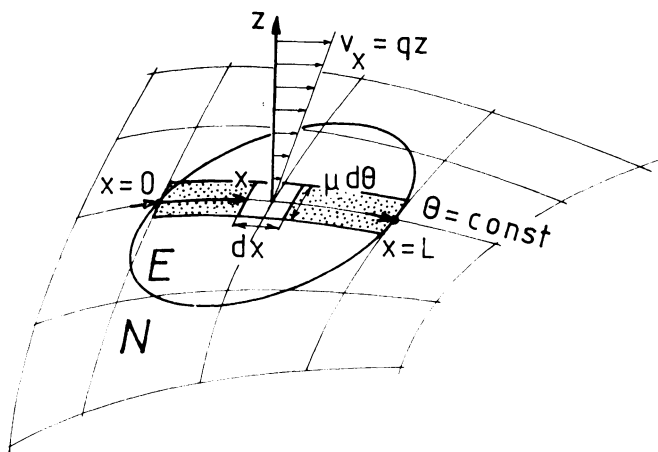


FIG. 1

Surface kinematics and canonical coordinates (z, x, θ)

corresponds to the solid surface whose part is occupied by the electrode under consideration.

The Θ -coordinate is chosen in such a way that the curve ($\Theta = \text{const}$, $z = 0$) corresponds to a surface streamline (i.e., the Θ coordinate is in every point of the area normal to the surface streamlines).

The x -coordinate follows the direction of the streamlines: it grows in the local flow direction \mathbf{q} and dx gives the differential length of a surface streamline.

The choice of the starting points, $x = 0$, on the individual streamlines depends both on the flow kinematics and electrode geometry. Surface streamlines can begin either on the electrode territory, E , in the so called critical forward points, $\mathbf{q} = \mathbf{O}$, or somewhere outside the electrode territory, N . In the first case, the starting point, $x = 0$, is placed into the critical point. In other cases, it corresponds to the point of intersection of a surface streamline and the forward boundary to the electrode.

As the common conditions of no permeation and no slip are considered, the velocity field close to the surface, $z \rightarrow 0$, can be represented by the following asymptotic relations:

$$v_x \approx q(x, \Theta) z, \quad v_\theta \approx 0, \quad v_z \approx -A(x, \Theta) z^2. \quad (1)$$

The metrics of the coordinate system (z, x, Θ) is characterized by the expression of a differential area dW in the electrode surface E , see Fig. 1:

$$dW = \mu(x, \Theta) d\Theta dx. \quad (2)$$

Under this assumption, the continuity equation for an incompressible liquid inside the diffusion layer can be written in the following form:

$$\mu \partial_z v_z + \partial_{\mathbf{x}}(\mu v_{\mathbf{x}}) = 0, \quad (3)$$

the Θ -component of velocity being zero by definition. The longitudinal velocity field is characterized completely by the surface field of the magnitude of the velocity gradient $q = q(x, \Theta)$. The normal velocity component then results from the continuity equation, as anticipated in Eqs (1):

$$v_z = -\frac{1}{2} q \partial_{\mathbf{x}} \ln(\mu q) z^2 = -A(x, \Theta) z^2. \quad (4)$$

It should be repeatedly noticed that these expressions hold correctly only within the diffusion-layer approximation, assuming that the diffusion thickness δ is thin enough in comparison with any other characteristic length scale of the flow under consideration. Within the corresponding approximation, the equation of steady convective diffusion can be written in the following form:

$$qz(\hat{c}_x c - \frac{1}{2} \partial_x \ln(\mu q) z \partial_z c) = D \partial_{zz}^2 c \quad (5)$$

and integrated with the following boundary conditions:

$$c|_{z=0} = 0, \quad c|_{z=\infty} = c_0. \quad (6a,b)$$

The essential analytic feature of this three-dimensional parabolic boundary-value problem is the absence of the streamline coordinate Θ in an explicit form. This makes possible to find the explicit similarity solution on a given surface streamline, $\Theta = \text{const}$. The similarity transform with Θ as a constant parameter,

$$C(w) = c(z, x, \Theta)/c_0, \quad w = z/\delta, \quad \delta = \delta(x, \Theta), \quad (7a,b,c)$$

results in separating the original three-dimensional system into two ordinary boundary-value problems, for $C = C(w)$:

$$C'' + 3\sigma w^2 C' = 0, \quad C(0) = 0, \quad C(\infty) = 1, \quad (8a,b,c)$$

and for $\delta = \delta(x, \Theta)$:

$$\frac{q\delta^3}{3D} (\hat{c}_x \ln(\delta) + \frac{1}{2} \hat{c}_x \ln(\mu q)) = \sigma, \quad (9)$$

$$\lim_{x \rightarrow 0} (q\sigma)^{1/2} \delta = 0. \quad (10)$$

The initial condition (10) implicitly includes the assumption of the presence of solution with the initial concentration c_0 outside the diffusion layer. If the entrance point $x = 0$ lies on the electrode boundary and is a regular one, $q > 0$, then it follows $\delta \rightarrow 0$ for $x \rightarrow 0$ from the condition (10). If a forward critical point ($q = 0$, $A > 0$) lies on the electrode territory, the condition (10) is compatible with a finite initial thickness of the diffusion layer, in accordance with the existing analyses for the convective diffusion in a neighbourhood of the forward critical point (or line).

There is a free numerical coefficient σ which joints the both ordinary differential equations (8, 9). If the choice of σ is made implicitly by introducing the additional normalizing condition

$$C'(0) = 1, \quad (8d)$$

then δ becomes identical with the Nernst diffusion thickness:

$$\partial_z c|_{z=0} = c_0/\delta(x, \Theta). \quad (11)$$

Both the ordinary boundary-value problems can be solved in quadratures. The

resulting general expression for similarity concentration profiles is

$$C(w) = \int_0^w \exp(-\sigma s^3) ds / \int_0^\infty \exp(-\sigma s^3) ds. \quad (12)$$

The value of σ follows from the normalizing condition (8d):

$$1 = \int_0^\infty \exp(-\sigma s^3) ds = \sigma^{-1/3} \Gamma(4/3). \quad (13)$$

The Eq. (9) can be rearranged by using the identities

$$\begin{aligned} q\delta^3[\partial_x \ln(\delta) + \frac{1}{2}\partial_x \ln(\mu q)] &= \\ &= q^{1/2}\mu^{-1/2}\delta^2\partial_x(q^{1/2}\mu^{1/2}\delta) = (q\mu^3)^{-1/2} \partial_x((q^{1/2}\mu^{1/2}\delta)^3) \end{aligned} \quad (14)$$

and then integrated with the initial condition (10) to give:

$$\delta(x, \Theta) = \left[\frac{9\sigma D Q}{q^{3/2}\mu^{3/2}} \right]^{1/3}, \quad (15)$$

where

$$Q(x, \Theta) = \int_0^x q^{1/2}\mu^{3/2} dx. \quad (16)$$

The last but essential step in the analysis is to find an expression for the macroscopic diffusion flux J_W over a given part (a segment) W of the electrode E . From the common definition of the diffusion fluxes across a solid boundary it follows:

$$\begin{aligned} J_W/(c_0 D) &= \int_W \delta^{-1} dW = \\ &= \int_{\Theta \in W} \left[\int_{x_1(\Theta)}^{x_0(\Theta)} \delta^{-1}(x, \Theta) \mu(x, \Theta) dx \right] d\Theta, \end{aligned} \quad (17)$$

with x_i, x_o corresponding to the input and output points on the surface streamlines, respectively.

By applying the obvious identities,

$$\delta^{-1}\mu = (9\sigma D)^{-1/3} q^{1/2}\mu^{3/2} = \frac{3}{2} (9\mu D)^{-1/3} \partial_x Q, \quad (18)$$

it is possible to simplify the general surface integral (17) to the form of a double quadrature:

$$J_W/(c_0 D^{2/3}) \equiv \alpha_W, \quad (19)$$

$$\alpha_W = \frac{3^{1/3}}{2\Gamma(4/3)} \int_{\Theta \in W} [Q^{2/3}(x_o(\Theta), \Theta) - Q^{2/3}(x_i(\Theta), \Theta)] d\Theta. \quad (20)$$

The newly introduced transport coefficient κ_W does not depend on the depolarizer concentration c_0 nor diffusivity D , and therefore it is a purely hydrodynamical quantity. For the total flux to the electrode, $W = E$, the formula (20) can be simplified slightly, as it is $x_0 = 0$, and $x_i = L(\theta)$ corresponds to the total lengths of surface streamlines over the electrode territory:

$$J_E/(c_0 D^{2/3}) \equiv \kappa_E = \frac{3^{1/3}}{2\Gamma(4/3)} \int_{\theta \in E} Q^{2/3}[L(\theta), \theta] d\theta. \quad (21)$$

The suggested general algorithm is illustrated in the following text by giving several simple examples.

USE FOR SPECIAL CONFIGURATIONS

Strip Electrode Placed Aslant in Simple Shear Flow

This idealized flow configuration is shown in Fig. 2. The velocity field in the rectangular coordinates (x_1, x_2, z) is given by

$$\begin{aligned} v_1 &= q_1 z, & v_2 &= q_2 z, & v_z &= 0, \\ q_1 &= q \cos(\beta), & q_2 &= q \sin(\beta). \end{aligned} \quad (22)$$

The territory of an infinite strip electrode is bounded by two parallel straight lines $x_1 = 0$ and $x_1 = h_1$. The longitudinal coordinate x is given unambiguously, $x =$

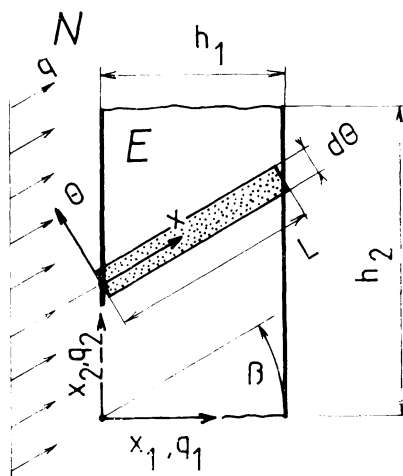


FIG. 2
Strip electrode in simple shear flow

$= x_1/\cos(\beta)$, but the streamlines can be identified in various ways, $\Theta = x_2/b$, with a free choice of the positive constant b . It follows from the identities $dW = \mu \cdot dx$. $d\Theta = dx_1 dx_2$ that the metric coefficient μ is constant over the whole electrode surface, $\mu = b \cos(\beta)$. The lengths of all the streamlines on the electrode territory are the same, $x_0 = h_1/\cos(\beta)$, and the result $Q(x_0) = q^{1/2}(b \cos(\beta))^{3/2} (h_1/\cos(\beta))$ is obvious. The mean flux on the electrode can be now calculated according to Eq. (21), with $\Delta\Theta = h_2/b$:

$$\frac{2\Gamma(4/3)}{3^{1/3}} \frac{J_E}{c_0 D^{2/3} h_1 h_2} = \frac{\Delta\Theta}{h_1 h_2} [Q(x_0)]^{2/3} = \frac{h_2/b}{h_1 h_2} [b q^{1/3} \cos^{1/3}(\beta) h_1^{2/3}] = (q_1/h_1)^{1/3}. \quad (23)$$

The final result is identical with the well-known Leveque's formula: the resulting flux on the strip electrode depends only on the strip width, h_1 , and on the q_1 -component of vectorial velocity gradient which is perpendicular to the strip edges. The alternative way of obtaining the result (23) consists in neglecting the convective fluxes due to the v_2 -velocity component. This is substantiated by the assumed planar symmetry of the concentration field at the infinitely long strip electrode, which results in the formal assumption $dc/dx_2 = 0$.

Circular Electrode in Simple Shear Flow

The velocity field and rectilinear coordinates x_1, x_2 are the same as in the previous example, assuming $q_2 = 0$, see Fig. 3. By choosing $dx = dx_1$ and $\Theta = x_2$, it is $\mu = 1$ and hence $Q(x) = q^{1/2}x$. For completing the calculation, it is necessary to know the

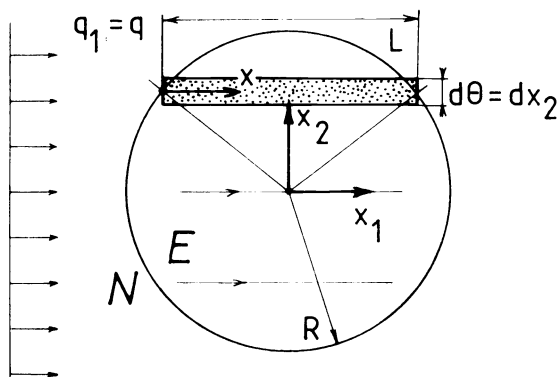


FIG. 3

Circular electrode in simple shear flow

lengths of the individual streamlines on the electrode territory, $L(\theta) = 2\sqrt{(R^2 - \theta^2)}$. Then the well-known result⁴⁻⁶ is obtained in a straightforward way by applying the general formula (21):

$$\begin{aligned} \kappa_E/\pi R^2 &= \frac{1}{\pi R^2} \frac{3^{1/3}}{2\Gamma(4/3)} \int_{-R}^R [2q^{1/2}(R^2 - \theta^2)^{1/2}]^{2/3} d\theta = \\ &= \frac{12^{1/3}}{\pi\Gamma(4/3)} \int_0^1 (1 - s^2)^{1/3} ds (q/R)^{1/3} = 0.68660 (q/R)^{1/3}. \end{aligned} \quad (24)$$

Analogous calculations for the various radial segments of a circular electrode are given both for the simple shear flow⁷ and a class of two dimensional flows⁸.

Circular Electrode in Cone-and-Plate Viscometric Flow

The cone-and-plate viscometric configuration, see Fig. 4a, is popular among rheologists⁶, as it produces the homogeneous field of shear rates, $q \approx \Omega/\alpha$. It can be useful for electrodiffusion calibration measurements, as well.

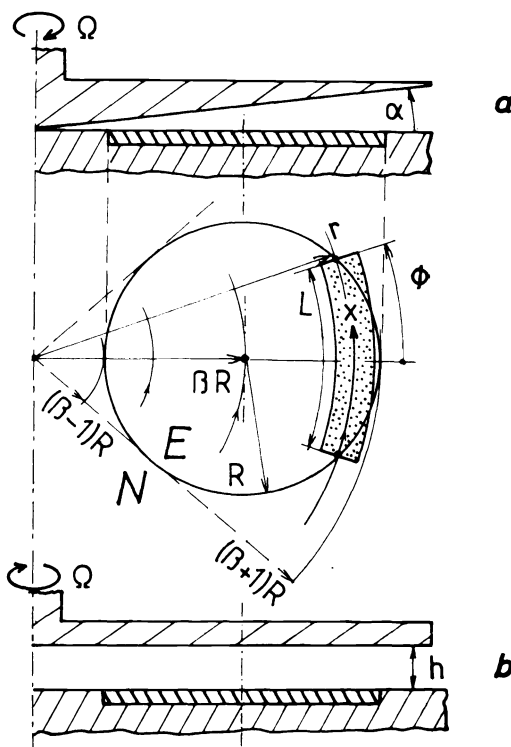


FIG. 4
Circular electrode in cone-and-plate (a) or torsional (b) flow

The surface streamlines are concentric circles. In the polar coordinates (z, r, Φ) it can be written $dx = r d\Phi$, $d\Theta = dr$, i.e. $\mu = 1$. For the circular electrode of the radius R and distance βR between the centre of the electrode and the axis of flow symmetry, the lengths of individual streamlines on the electrode territory can be found from simple trigonometry considerations:

$$L = 2r\Phi = 2Rt \arccos \left(\frac{t^2 + \beta^2 - 1}{2\beta t} \right), \quad (25)$$

where $t = r/R$. By substituting this result into the starting formulas (16) and (21) it follows $Q = q^{1/2}L(t)$ and

$$\kappa_E/\pi R^2 = F(\beta)(q/R)^{1/3}, \quad q = \Omega/\alpha, \quad (26)$$

with

$$F(\beta) = \frac{12^{1/3}}{\pi\Gamma(4/3)} \frac{1}{2} \int_{\beta-1}^{\beta+1} \left[t \arccos \left(\frac{t^2 + \beta^2 - 1}{2\beta t} \right) \right]^{2/3} dt. \quad (27)$$

For $\beta \rightarrow \infty$, the integral in Eq. (27) reduces to $2 \int_0^1 (1-w^2)^{1/3} dw$ and the formula (26) gives the well-known result for the circular electrode in simple shear flow, $F(\infty) = 0.68660$. For any $\beta \geq 1$, the correction factor $F(\beta)/F(\infty)$ can be represented with an acceptable accuracy by the empirical formula

$$\frac{F(\beta)}{F(\infty)} = 1 - 0.0145\beta^{-2}[1 - 0.4(1 - 1/\beta)^{1/2}]. \quad (28)$$

In particular, it is obvious from Eq. (28), that the position of the electrode in the cone-and-plate viscosimeter has nearly negligible effect on the total diffusion flux.

Circular Electrode in Torsional Viscometric Flow

The disk-disk rotational viscometer, see Fig. 4b, produces the viscometric torsional flow field with the shear rate q proportional to the radial distance r to the axis of rotation,

$$q = \Omega r/h = (\Omega R/h) t. \quad (29)$$

With this difference, the situation is analogous to that considered as the previous example. By using the same notation, it is $Q(t) = q^{1/2}(t) L(t)$, and the final result can be read as follows

$$\kappa_E/\pi R^2 = G(\beta)(q/R)^{1/3}, \quad q = \beta\Omega R/h \quad (30)$$

with

$$G(\beta) = \frac{12^{1/3}}{\pi\Gamma(4/3)} \frac{\beta^{-1/3}}{2} \int_{\beta-1}^{\beta+1} t \left[\arccos \left(\frac{t^2 + \beta^2 - 1}{2\beta t} \right) \right]^{2/3} dt. \quad (31)$$

In the asymptote $\beta \rightarrow \infty$ it is $G(\infty) = 0.68660$ and the result (30) becomes identical with the formulas for the circular electrode in simple shear flow, the local shear rate q being evaluated at the electrode center.

Circular Electrode in the Forward Critical Flow Region

The axisymmetric velocity field in a proximity of the forward critical point can be expressed by the formulas³

$$v_r = Arz, \quad v_z = -\frac{1}{2}Az^2, \quad (32)$$

containing the single kinematic constant A . Here, the angular coordinate Φ can be identified with the streamline coordinate, $\Theta = \Phi$, the radial coordinate r with the longitudinal one, $dx = dr$. The metrics is then identical with that for the polar cylindrical system, $\mu(x, \Theta) = r$.

Two different transport regimes should be distinguished. In the case $\beta < 1$, the critical point $r = 0$ lies inside the electrode territory. All the surface streamlines begin in the critical point and therefore $x = r$. Then it holds $q = Ax$, $Q = (A/9)^{1/2} x^3$, and the following expression can be developed in an obvious way:

$$\kappa_E/\pi R^2 = \frac{3^{1/3}}{\Gamma(4/3)} \left(\frac{A}{9}\right)^{1/3} \frac{1}{2\pi R^2} \int_0^{2\pi} L^2(\Theta) d\Theta \quad (33)$$

If the electrode is placed axisymmetrically with the velocity field, it is $L(\Theta) = R$, and hence $\int_0^{2\pi} L^2(\Theta) d\Theta = 2\pi R^2$. The same relation holds even for eccentric position of the electrode, if the critical point lies inside the electrode territory. The final result

$$\kappa_E/\pi R^2 = \frac{1}{\Gamma(4/3)} \left(\frac{A}{3}\right)^{1/3} \quad (34)$$

is identical with the well-known Levich formula for the rotating disk electrode^{3,5,6}, where $A = 0.510 \sqrt{(\Omega^3/\nu)}$ and ν stands for the kinematic viscosity.

In the opposite case $\beta > 1$, the critical point $r = 0$ lies outside the electrode. All the surface streamlines begin outside the electrode territory and it is necessary to know the coordinates of their intersections with the electrode perimeter. The radii r_o , r_i of these points, and the corresponding lengths L of the surface streamlines

follow from simple considerations, see Fig. 5,

$$r_{i,o}/R = \beta \cos(\Phi) \pm \sqrt{(1 - \beta^2 \sin^2(\Phi))}, \quad (35)$$

$$L = r_o(\Phi) - r_i(\Phi) = 2 \sqrt{(1 - \beta^2 \sin^2(\Phi))}. \quad (36)$$

The values $\Phi = \pm \Phi_0$, for which it is $\beta^2 \sin^2(\Phi) = 1$, correspond to the limit streamlines which only touch the electrode perimeter.

By successively using the outlined systematic approach, the calculation proceeds in the following steps:

$$\begin{aligned} \Theta = \Phi, \quad x = r - r_i(\Phi), \quad q = Ar, \quad \mu = r, \\ Q(\Theta) = \int_0^{L(\Theta)} q^{1/2}(x) \mu^{3/2}(x) dx = \frac{1}{3} A^{1/2} [r_o^3(\Theta) - r_i^3(\Theta)], \end{aligned} \quad (37)$$

and finally:

$$\kappa_E/\pi R^2 = \frac{1}{\Gamma(4/3)} \left(\frac{A}{3}\right)^{1/3} H(\beta) \quad (38)$$

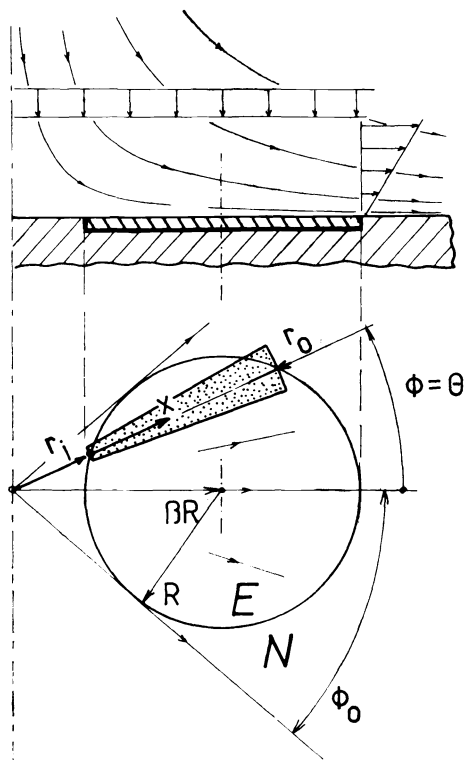


FIG. 5
Circular electrode in axisymmetric forward flow region

with

$$\begin{aligned} \pi H(\beta) &= \int_0^{\Theta_0} [(\beta c + \sqrt{(1 - \beta^2 s)})^3 - (\beta c - \sqrt{(1 - \beta^2 s)})^3]^{2/3} d\Theta = \\ &= 2^{2/3} \int_0^{\Theta_0} (1 - \beta^2 s)^{1/3} (3\beta^2 + 1 - 4\beta^2 s^2)^{2/3} d\Theta = \\ &(36\beta)^{1/3} \int_0^1 (1 - t^2)^{1/3} (1 - t^2/\beta^2)^{-1/2} \left[1 + \frac{1 - 4t^2}{3\beta^2} \right]^{2/3} dt. \end{aligned} \quad (39)$$

In these integrals, s and c stand for $\sin(\Theta)$ and $\cos(\Theta)$, respectively. It is apparent from the last expression that, for $\beta \rightarrow \infty$, the result becomes identical to the formula for the circular electrode in simple shear flow, with the local shear rate q specified at the centre of the electrode, i.e. at $r = \beta R$, where $q(r)/R = A\beta$. For the other limiting case, $\beta = 1$, the expected result $H(1) = 1$ can be easily checked.

The following empirical formula for the correcting factor H ,

$$H(\beta) = \begin{cases} 1; & \beta < 1.08 \\ 0.884\beta^{1/3}(1 + 0.12\beta^{-2}); & \beta > 1.08 \end{cases} \quad (40)$$

can be safely used within the accuracy of three decimal digits.

CONCLUSIONS

The simplifying assumptions, commonly applied in the theory of two-dimensional concentration boundary layer, allow us to develop rather general three-dimensional theory which is based on the knowledge of the surface field of velocity gradients in the Euclidean space. As a result, the analytic predictions are given both for the local and total diffusion fluxes.

The theory is demonstrated by calculating the total fluxes to a circular electrodiffusion electrode under various flow conditions. Less trivial case of a circular electrode, placed eccentrically in the rotating disk body, is analyzed in another paper⁹.

SYMBOLS

A	normal flow coefficient
c	concentration of depolarizer
c_0	initial concentration of depolarizer
D	diffusivity
E	total electrode surface
h	distance between coaxial disks, Fig. 4
h_1, h_2	length and width of a strip electrode
J_E, J_W	macroscopic flux to an electrode, E , or its segment, W
L	geodesic length of surface streamline
N	inert neighbourhood of electrode

q, q	velocity gradient at solid surface and its magnitude
q_1, q_2	components of velocity gradient
Q	kernel of the generalized Lighthill transformation
r	radial coordinate
r_i, r_o	radial coordinates of the input and output points on surface streamlines
R	radius of a circular electrode
v_x, v_z	longitudinal and normal velocity components
x_i, x_o	x -coordinates of the input and output points on surface streamlines
W	surface of an electrode segment
z	normal coordinate
α	angle of the cone in cone-and-plate configuration, Fig. 4
β	geometric simplex
δ	diffusion thickness
θ	streamline coordinate
κ_E, κ_W	modified mass-transfer coefficients
μ	metric coefficient for a coordinate system (z, x, θ) , Eq. (2)
Φ	angular polar coordinate
Φ_0	Φ -coordinates of the central tangents to a circle, Fig. 5
Ω	angular speed of rotation

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Translated by the author.