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Turbulent mass transfer in the developing diffusion layer at large Schmidt numbers

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Abstract

The aim of this paper is to investigate the turbulent mass transfer in channels, tubes and annuli for fully developed hydrodynamic conditions and for developing diffusional boundary layer at large Schmidt numbers. Three transfer zones are detected: one corresponding to the leading edge of mass transfer surface for which the Lévêque solution is valid, the second far from the entrance where a constant diffusional boundary layer is obtained and the MacAdams equation can be applied, and the third is comprised between the two aforementioned zones. An expression of the mass transfer coefficients is proposed for the intermediate zone. © 1999 Elsevier Science Ltd. All rights reserved.

Nomenclature

$\bar{c}$ mean concentration of the diffusing species
c' concentration pulsations
c_0 surface concentration
c_∞ bulk concentration
C nondimensional concentration $= \bar{c}/(c_\infty - c_0)$
D molecular diffusivity
d_h hydraulic diameter
D_t turbulent diffusivity
j_t turbulent mass flux
j mass flux density
$\hat{j}$ nondimensional mass flux density $= j\delta_\infty/[(c_\infty - c_0)D]$
K average mass transfer coefficient
$\hat{K}$ nondimensional mass transfer coefficient $= K\delta_\infty/D$
k_{yy}, k constants in the equations (2), (23)
l length of the mass transfer active surface
l_* diffusion entrance length, equation (8)
R hydraulic radius
r radial coordinate in polar cylindrical coordinate system
Re Reynolds number $= v \cdot d_h/v$
Sc Schmidt number $= v/D$
T_{corr} correlation time of the normal velocity pulsations
V mean velocity
y coordinate normal to the wall
y_τ dynamic length
y_+ nondimensional normal coordinate $= y/y_\tau$
Y nondimensional normal coordinate with respect to the diffusion layer thickness $= y/\delta_\infty$
z longitudinal coordinate
Z nondimensional longitudinal coordinate $= z/l_*$.

Greek symbols
δ_∞ fully developed diffusion layer thickness, equation (8)
μ dynamic viscosity
ν kinematic viscosity
ω parameter which characterized surface curvature
ρ density
$\bar{\tau}$ mean wall shear stress.

Subscipts
0 corresponds to the Lévêque solution
∞ corresponds to the fully developed diffusion layer.

Superscripts
0 corresponds to the plane geometry
1 corresponds to the correction deals with the surface curvature.

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1. Introduction

Mass transfer to fluids flowing through tubes, channels and annuli is frequently encountered in different industrial processes. The principle of electrodiffusion sensors, which are now widely used for wall shear stress measurements, is based on the study of mass transfer phenomenon also. In this paper, we discuss the mass transfer problem, but all the results can be used for the heat transfer problem if the Prandtl number is rather high. Undoubtedly the turbulent flow regime is the most important for the practical applications. As such, turbulent mass transfer problems have been studied extensively by many workers [1–4].

Nevertheless, even in the simplified case of fully developed hydrodynamics, the previous papers give an incomplete solution of the mass transfer problem. Two types of equations can be found in the literature. The first one is based on the well-known Lévêque solution [5] which predicts the mass transfer coefficient in terms of the mean wall shear stress. The Lévêque solution is justified for the case of short mass transfer length and is usually used in the literature dealing with the electrodiffusion diagnostics of flow.

On the other hand, rather long mass transfer surfaces are usually encountered in chemical engineering processes. For this case, different semi-empirical or pure empirical expressions can be found, for example the well known MacAdams formula [4]. These expressions are based on either experimental data correlations or semi-empirical expressions for turbulent diffusivity.

Undoubtedly it is important to establish the limits of applicability of the equations, and thus it is necessary to predict the diffusion entrance length. Some estimations of the diffusion entrance length were reported [6–8]. For example, Hanratty and Campbell [7] estimated the diffusion entrance length as 600 y_τ, where y_τ is the dynamic length.

In the paper of Kader [6] the cubic law $D_t \alpha y^N$, with $N = 3$, for the near-wall turbulent diffusivity was used, which leads to the conclusion that the diffusion entrance length is independent of the molecular Schmidt number.

On the other hand in [8], by means of statistical method, it was shown that the power factor N in the near-wall turbulent diffusivity law cannot be less than 4 and, as a consequence, the diffusion entrance length is dependent on the molecular Schmidt number. The definition of the diffusion entrance length should be precise, too, as the zones of applicability of the Lévêque and MacAdams formulae are not continuous.

Therefore, the numerical solution of the mass transfer problem with respect to physically well-founded turbulent diffusivity law $N = 4$ is important both from the practical and fundamental points of view. The other goal of this paper is to study both plane and cylindrical flow geometries, which will allow us to use the results of the calculations to predict the mass transfer rate in channels, tubes and annuli.

2. Statement of the problem

In this section, we shall consider turbulent flow of an incompressible liquid along the flat solid boundary, $y = 0$. The cylindrical geometry will be considered in Section 4. At the core of the flow, $y \to \infty$, the concentration of the diffusing substance is c_∞. On the mass transfer active part of the solid boundary ($y = 0$, $z > 0$), the concentration is equal to c_0. The Oz-axis is along the surface, the Oy-axis is perpendicular to the boundary. In the transversal direction the problem is homogeneous.

We shall restrict the problem to the case where the molecular diffusion in the longitudinal direction is negligible and only the normal component of the turbulent diffusion flux caused by the normal concentration gradient is important. Then for the fully developed hydrodynamic conditions the turbulent diffusion equation can be written as [6, 8]:

$$\bar{\tau}/\mu y \partial\bar{c}/\partial z = \partial/\partial y[D + D_t(y)]\partial\bar{c}/\partial y \tag{1}$$

where $\bar{\tau}$ and $\bar{c}$ are the mean wall shear stress and concentration. D and μ are the molecular diffusivity and dynamic viscosity, respectively.

Equation (1) is of a parabolic type and boundary conditions which should be used are the following:

$$c \to c_\infty \quad \text{as} \quad y \to \infty \quad \text{or} \quad z \to 0$$

$$c \to c_0 \quad \text{as} \quad y \to 0 \quad \text{or} \quad z > 0.$$

The turbulent diffusion coefficient D_t is a function of the normal coordinate y

$$D_t(y) = k_{yy}\left(\frac{y}{y_\tau}\right)^N \tag{2}$$

where $y_\tau = v(\rho/\bar{\tau})^{1/2}$ is the dynamic length.

Two different power factors, $N = 3$ and $N = 4$, were proposed in the literature [1–3, 6–10] in the near-wall turbulent diffusivity law, equation (2), mainly in relation to the problem of fully developed diffusion layer. the only paper [6], where the problem of the developing diffusion layer was studied numerically, was founded on the cubic ($N = 3$) turbulent diffusivity law. In our opinion, the cubic law has no physical meaning due to results of the treatment of this problem by means of statistical method [8]. By multiplying the equation for the concentration fluctuations with the fluctuating velocity in another space-time point, $v\ (t', r')$, an equation is obtained [8] for the velocity and concentration fluctuations correlator:

$$\frac{\partial S}{\partial t} + u\frac{\partial c'}{\partial z} - D\Delta S = -W_Z\frac{\partial \bar{c}}{\partial z} - W_y\frac{\partial \bar{c}}{\partial y}$$

$$S(t, r; t', r') = \langle c'(t, r)v'(t', r')]$$

$$W_{z,y}(t, r; t', r') = \langle v'_{z,y}(t, r)v'(t', r')]. \tag{3}$$

Here u is the mean flow velocity and $\langle\rangle$ denotes statistical averaging. The estimation of the different terms in equation (3) with respect to available information about characteristics of the near-wall turbulence have shown [8] that, in the main part of the diffusion layer, the term $\partial/\partial t$ dominates in the left-hand of this equation. If we keep only the term $\partial S/\partial t$ in the left-hand side of equation (3), the following expression for the turbulent mass flux can be obtained:

$$j_t(z,y) = \langle c'(t,r)v'(t,r)\rangle = -D_Z(y)\frac{\partial\bar{c}}{\partial z} - D_y(y)\frac{\partial\bar{c}}{\partial y} \quad (4)$$

$$D_{z,y}(y) = -\int_{-\infty}^{0} W_{z,y}(t-t'; r'=r)\,\mathrm{d}(t-t'). \quad (5)$$

In particular, for the normal component, j_y, of the turbulent mass flux caused by the normal concentration gradient, we obtain the traditional relation:

$$j_y = -D_t(y)\frac{\partial\bar{c}}{\partial y} \quad (6)$$

where the turbulent diffusivity D_t has the form [8]:

$$D_t(y) = \int_0^{\infty} \langle v'_y(t,r)v'_y(0,r)\rangle\,\mathrm{d}t = \overline{v'^2_y}T_{\mathrm{corr}} \quad (7)$$

where r is the space vector and T_{corr} the correlation time of the hydrodynamical pulsations within the viscous sublayer. So, the turbulent diffusivity near the wall is equal to the product of the normal velocity fluctuations intensity and their correlation time. The normal velocity near the wall is proportional to the square of the normal coordinate. So, the power factor in the near-wall turbulent diffusivity law should be equal to at least 4 and the cubic law, which is frequently used, has no physical meaning. So, below we shall use the power factor $N = 4$ in the near-wall turbulent diffusivity law.

3. Numerical solution of the problem

Let us introduce the dimensionless variables:

$$Z = z/l_*; \quad Y = y/\delta_\infty; \quad C = (c-c_0)/(c_\infty - c_0) \quad (8)$$

with

$$\delta_\infty = \left(\frac{D}{k_{yy}}\right)^{1/4} y_\tau; \quad l_* = \delta_\infty^3 \frac{\bar{\tau}}{(D\mu)},$$

where δ_∞ is the fully developed diffusion layer thickness and l_* is the diffusion entrance length. Then, the diffusion-convection equation and the boundary conditions take the form:

$$Y\partial C/\partial Z = \partial/\partial Y[(1+Y^4)\partial C/\partial Y] \quad (9)$$

$$C \to 1 \quad \text{as } Y \to \infty \quad \text{or} \quad Z \to 0$$

$$C \to 0 \quad \text{as } Y \to 0 \quad \text{and} \quad Z > 0. \quad (10)$$

Equations (9) and (10) were solved numerically by a finite different method. The numerical procedure is realized in the range $0 \leqslant Z \leqslant Z_{\max}$, $0 \leqslant Y \leqslant Y_{\max}$. The limiting values $Y_{\max}$ and $Z_{\max}$ are determined in order to provide the necessary accuracy for the mass flux with respect to the known analytical solutions for beginning region layer and for the fully developed diffusion layer.

The boundary condition for the concentration $Z = 0$, $Y = 0$, equation (10), is not continuous, hence it is more convenient to perform the numerical procedure in the range $\varepsilon < Z < Z_{\max}$, $\varepsilon \ll 1$, where the concentration distribution is continuous. The boundary condition at $Z = 0$ can be changed.

$$C \to C_0(\varepsilon, Y) \quad \text{as} \quad Z \to \varepsilon, \quad Y > 0, \quad (11)$$

where $C_0(Z, Y)$ is the well-known Lévêque solution for the entrance zone of the diffusion layer.

The inertial term in the equation (9) tends to zero as $Y \to 0$. As a result, the system of the finite difference equations in orthogonal uniform grid becomes unstable, i.e. a small deviation of coefficients causes a large variation of the numerical solution.

The correct system of the finite difference equations can be obtained if we use a nonuniform grid with respect to the Y-variable in order to avoid a sharp variation of the term $1/[Y\Delta Y^2(Y)]$ when Y tends to zero. Here ΔY corresponds to the grid step. The finite difference approximation of equation (9) leads to a system of linear algebraical equations, which was solved by a Gauss–Seidel algorithm.

The results of the calculations of the mass transfer coefficient

$$\hat{K}(Z) = 1/Z\int_0^Z \hat{j}(\tilde{Z})\,\mathrm{d}\tilde{Z}; \quad \hat{J}(Z) = \partial C(Z, Y=0)/\partial Y \quad (12)$$

are presented in Fig. 1. For the small values of Z, the mass transfer coefficient can be approximated by means of the Lévêque solution at $Z = 0.025$ with 1% accuracy and at $Z = 0.09$ with 5% accuracy respectively. The Lévêque solution approximates the mass flux density, $\hat{J}(Z)$ at $Z = 0.06$ and at $Z = 0.22$ for 1 and 5% accuracy, respectively.

For the large values of Z the density of the mass flux becomes constant and is equal to:

$$\hat{J}_\infty = \left(\int_0^\infty \frac{1}{1+y^4}\,\mathrm{d}y\right)^{-1} \quad (13)$$

The numerical values of the longitudinal coordinate which characterize the zone of applicability of the fully developed mass flux density solution are $Z = 0.6$ and $Z = 0.35$ for 1 and 5% accuracy, respectively.

The mass transfer coefficients for large values of Z tends to its limiting value

$$\hat{K}_\infty = \hat{J}_\infty + A/Z; \quad A = 0.13 \quad (14)$$

where the constant A was obtained by means of the

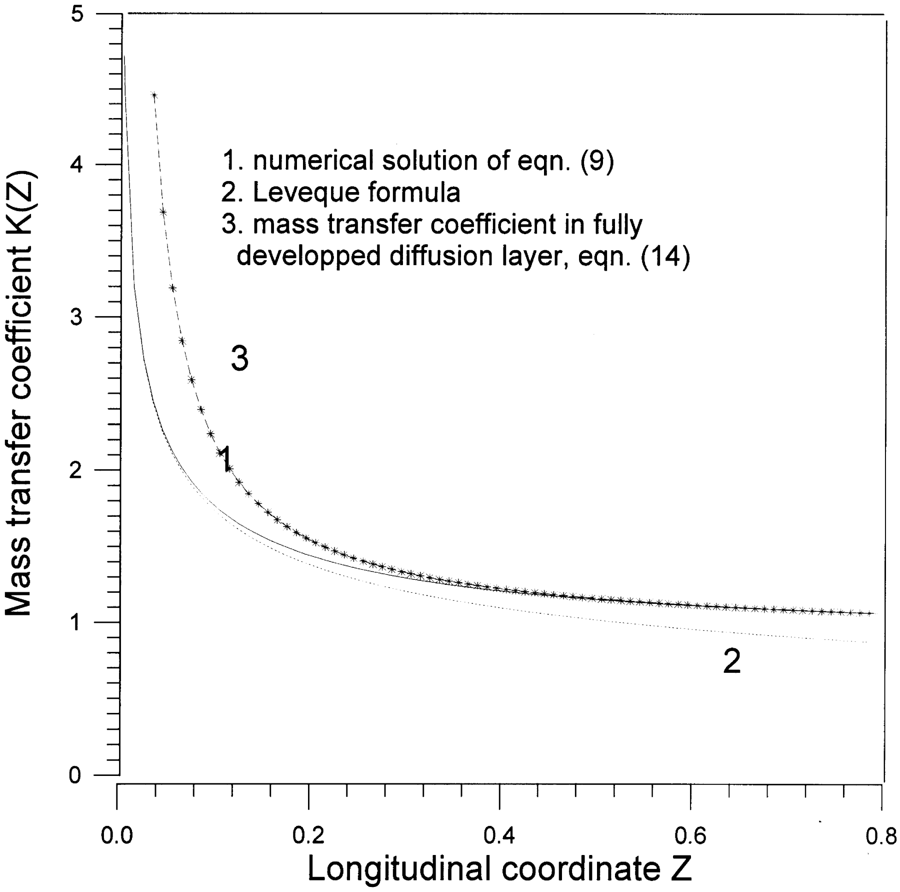

Fig. 1. Mean transfer coefficient as a function of longitudinal coordinate in nondimensional variables.

results of the numerical calculations. The parameter A characterizes the influence of the entrance section on the value of the mass transfer coefficient in the fully developed diffusion layer. Equation (14) can be used for the prediction of the mass transfer coefficient with 1% accuracy at $Z = 0.5$ and with 5% accuracy at $Z = 0.25$. In the zone $0 < Z < 0.7$ the following formulae present the numerical results with 99% accuracy:

$$\hat{j}(Z) = \hat{j}_0(Z) = P(Z); \quad P(Z) = \sum_{n=0}^{3} p_n Z^n;$$

$$p_0 = 9.6\times10^{-4}; \quad p_1 = 0.68;$$
$$p_2 = -0.50; \quad p_3 = 0.21 \tag{15}$$

$$\hat{K}(Z) + \hat{K}_0(Z) + Q(Z); \quad Q(Z) = \sum_{n=0}^{2} q_n Z^n;$$

$$q_0 = 1.7\times10^{-3}; \quad q_1 = 0.32; \quad q_2 = -0.10 \tag{16}$$

where the functions

$$\hat{j}_o(Z) = \alpha Z^{-1/3}; \quad \hat{K}_o(Z) = \frac{3\alpha}{2} Z^{-1/3};$$

$$\alpha = \frac{3^{1/3}}{\Gamma(1/3)} \approx 0.54 \tag{17}$$

correspond to the Lévêque solution. Equations (15) and (16) are only a satisfactory approximation, obtained by linear regression, for the numerical solution in the transition zone. It means that we have not considered equation (12) for the determination of p_n and q_n.

4. Mass transfer on the cylindrical surface

The influence of the surface curvature on the mass transfer is determined by the ratio of the diffusion layer thickness δ_∞ to the radius of the mass transfer active surface R, $|\omega| = \delta_\infty/R$. For large values of the molecular

Schmidt number, $Sc \approx 10^3$, the order of magnitude of the diffusion boundary layer thickness does not exceed the dynamic length: $\delta_\infty \leqslant y_\tau$. By means of the well-known Blasius law it is possible to present the dynamic length in terms of the equivalent hydraulic diameter (or radius R) and the Reynolds number, $|\omega| \approx 10/Re^{7/8}$. For $Re = 10^3$, the magnitude of $|\omega|$ is about 2.4×10^{-2}. So for turbulent flows, $|\omega|$ can be considered as a small parameter.

Under the same conditions as for the plane surface (molecular and turbulent mass transfer in the longitudinal direction are negligible) the convective diffusion equation in the polar cylindrical coordinates takes the form:

$$Y\partial C/\partial Z = \partial/\partial Y[(1+Y^4)\partial C/\partial Y] - \omega(1+Y^4)\partial^2 C/\partial Y^2, \tag{18}$$

where $Y = (R \pm r)/\delta_\infty$, the sign $(+)$ corresponds to the case when the active mass transfer surface is the internal surface of a tube or the outer annular cylinder ('inner flow') and the sign $(-)$ corresponds to the case when the active mass transfer surface is the inner annular cylinder ('outer flow'), and $\omega = |\omega|$ for the case of the 'inner flow' and $\omega = -|\omega|$ for the case of the 'outer flow'.

The solution can be presented as a series with respect to the parameter ω:

$$C(Z, Y, \omega) = C^0(Z, Y) + \omega C^1(Z, Y) + o(\omega^2) \tag{19}$$

where $C^0(Z, Y)$ corresponds to the concentration profile for the plane surface and $C^1(Z, Y)$ gives the correction with respect to the curvature factor. In the same manner the mass flux density and the mass transfer coefficient can be expressed by:

$$\hat{j}(Z, \omega) = \hat{j}^0(Z, 0) + \omega \hat{j}^1(Z) + o(\omega^2) \tag{20}$$

$$\hat{K}(Z, \omega) = \hat{K}^0(Z, 0) + \omega \hat{K}^1(Z) + o(\omega^2) \tag{21}$$

where J^0 and K^0 correspond to the plane case solution. The equation for the correction C^1 has the form:

$$Y\partial C^1/\partial Z - \partial/\partial Y[(1+Y^4)\partial C^1/\partial Y] = (1-Y^4)\partial^2 C^0/\partial Y^2. \tag{22}$$

In order to determine the correction for mass flux density for all the values of Z, the numerical solution of equation (18) was done for the value $\omega = \pm 0.01$. It can be concluded that the ratio of the 'curvature correction' to the plane case solution is less than $5Re^{-7/8}$ and is negligible for the turbulent flows at least with 99% accuracy. For a given value of the wall shear stress, the curvature increases the mass flux on the surface of the 'inner flow' and decreases it for the 'outer flow'.

5. Discussion

As shown in the previous section, the influence of the surface curvature on the mass transfer in turbulent flows is negligible at least with 1% accuracy. So, mass transfer in tubes and annuli does not need any additional treatment and we can restrict the discussion to the case of the plane geometry.

The numerical solution of the problem (Section 3) gives the complete description of the mass transfer phenomena in dimensionless variables. Three zones can be distinguished. In the beginning of the active mass transfer surface, the development of the diffusion boundary layer is due to the mean longitudinal velocity and the molecular diffusion. In this zone only one hydrodynamical parameter, namely, the mean wall shear stress is important to predict the mass transfer coefficient and the well-known Lévêque solution can be used.

Relatively far from the beginning of the mass transfer active surface, the fully developed diffusion layer of a constant thickness is formed. In this zone the mass transport is due to the turbulent and molecular diffusion in the normal direction only, and the other hydrodynamical parameter k_{yy} is important to predict the mass transport. The constant k_{yy} determines the magnitude of the turbulent diffusivity, see equation (2).

In the transitional zone all the three mechanisms of the mass transfer are important, namely, convective transport by the mean longitudinal velocity, molecular diffusion and turbulent mass transfer in the normal direction.

The zones of applicability of the Lévêque and fully developed solutions were numerically determined. The mass flux density and mass transfer coefficient are calculated with 99% accuracy by equations (15) and (16) for $0 < Z < 0.7$. For large $Z > 0.7$, the stabilized solution is valid, equations (13) and (14). To return to the starting variables, equation (8) can be used. Here one delicate problem arises. In the definition of the longitudinal l_* and the normal δ_∞ space scales, equation (8), two hydrodynamical parameters are presented. The mean wall shear stress $\bar{\tau}$ can be connected with the Reynolds number by means of the well-established Blasius or Nikuradze laws. Some additional information should be used in order to determine the parameter k_{yy}. For example [9, 10] this parameter is given in terms of the molecular viscosity and involves a fitting empirical constant k:

$$k_{yy} = k\nu; \quad k \approx 2.4 \times 10^{-4}. \tag{23}$$

Applying equation (23) and the Blasius law for the wall shear stress as well, we obtain by means of equation (8):

$$\delta_\infty = k^{1/4} Sc^{-1/4} y_\tau; \quad 1_* = k^{-3/4} Sc^{1/4} y_\tau; \quad y_\tau = 5 d_{\mathrm{h}} Re^{-7/8}, \tag{24}$$

where d_{h} is the equivalent hydraulic diameter. For the value $k = 2.4 \times 10^{-4}$, equation (24) gives:

$$1_*/d_{\mathrm{h}} \approx 2.6 \times 10^3 Sc^{1/4} Re^{-7/8}. \tag{25}$$

Equation (25) can be used for the estimation of the zones of applicability of the Lévêque and the fully developed solutions in terms of hydraulic diameter.

For example, for the case $Re = 10^4$ and $Sc = 10^3$, a constant mass flux density can be obtained with 99% accuracy at $z = 2.7\ d_h$ and with 95% accuracy at $z = 1.6\ d_h$. But for the calculation of mass transfer coefficient, the assumption of the fully developed diffusion layer ($K = j_\infty$) can be used only if the mass transfer active surface is larger than 60 d_h and 12 d_h, for 99 and 95% accuracy, respectively. In the opposite case, the correction with respect to equation (14) should be done. The Lévêque solution can be used for the calculation of the mass flux density with 99 and 95% accuracy up to $z = 0.1\ d_h$ and $z = 0.4\ d_h$, respectively, for the case $Re = 10^4$ and $Sc = 10^3$.

The Sherwood number in fully developed diffusion layer is given by:

$$Sh_\infty = j_\infty d_h/[(c_\infty - c_0)D], \quad (26)$$

where j_∞ was given by [3]:

$$j_\infty = 0.90\, D(C_o - C_\infty)/\delta_\infty. \quad (27)$$

From equations (8), (23), (26) and (27), the Sherwood number is equal to:

$$Sh_\infty = \alpha Sc^{1/4} Re^{7/8}. \quad (28)$$

The exponent $\frac{1}{4}$ for the Schmidt number is due to the power factor 4 in equation (2) for the turbulent diffusion coefficient. The constant α is connected with the constant k:

$$\alpha = 0.180\, k^{1/4} \approx 0.022. \quad (29)$$

6. Conclusion

Both qualitative and quantitative treatments of the mass transfer problem in turbulent flows at large Schmidt numbers Sc are presented in this paper. The conclusions which can be useful for practical application are the following:

(1) The assumption of the fully developed boundary layer can be used for the prediction of the mass transfer coefficient if the length l of the active surface is larger than: $l > (1/\varepsilon)3.34 \times 10^4 Sc^{1/4} Re^{-7/8}\ d_h$, where ε is an acceptable error in percentages. In the opposite case, the influence of the diffusion entrance length should be taken into account.

(2) The mass flux density on the microelectrode surface can be calculated by means of the Lévêque formula if its length l is less than $l < a \times 10^2 Sc^{1/4} Re^{-7/8}\ d_h$, where $a = 0.65$ and $a = 2.34$ for 99 and 95% accuracy, respectively.

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