

## MASS TRANSPORT TO A VIBRATING ELECTRODE

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The present work is concerned with the concentration field near an electrode located on a vibrating plate. A mathematical model is proposed to describe the hydrodynamic behaviour of a streaming electrolyte solution and of the resulting concentration distribution at the vibrating plate. The results of numerical calculations were used to derive an empirical formula for the concentration gradient as function of dimensionless parameters. The influence of the electrode width and of the width of the vibrating plate on the value of the diffusion flux was also determined.

The title problem has a concrete physical motivation and interpretation; it originates from the study of the diffusion transport of mass at vibrating electrodes<sup>1-3</sup>. We shall discuss its physical aspects in short.

The rate of mass transport plays a significant role in many physical and technical processes. It is, in general, desirable that this rate be as high as possible and controllable. Increasing the diffusion transport of mass by vibration of plates with an electroactive part of their surface has received attention in recent years. We shall, however, not deal with the construction details of vibrating plate electrodes. The aim of the present work was the derivation of a mathematical model describing the physical processes at a vibrating plate electrode.

## THEORETICAL

*Mathematical Model*

We shall consider a vibrating plate in the  $y$ - $z$  plane of a Cartesian coordinate system. The plate is unlimited in both directions of the  $y$  axis, which is also its symmetry axis. The plate width is denoted as  $d$  and the width of the electroactive region is  $a < d$ . The plate is assumed to vibrate along the  $z$  axis (Fig. 1); the coordinate system is fixed with respect to the plate, which is immersed in a liquid of a so large volume that the boundary effects can be neglected. Finally, we assume that the streaming velocity of the liquid at a large distance from the plate is given as  $v_x =$

$= r\omega \sin(\omega t)$ , where  $r$  denotes the amplitude and  $\omega$  the angular frequency of harmonic vibrations.

The mathematical description of the hydrodynamic behaviour is based on the boundary layer theory<sup>4</sup>; the convection-diffusion system is described by the following system of partial differential equations:

$$\partial v_z / \partial t + v_x \partial v_z / \partial x + v_z \partial v_z / \partial z = \nu \partial^2 v_z / \partial x^2 \quad (1)$$

$$\partial v_x / \partial x + \partial v_z / \partial z = 0 \quad (2)$$

$$\partial c / \partial t + v_x \partial c / \partial x + v_z \partial c / \partial z = D(\partial^2 c / \partial x^2 + \partial^2 c / \partial z^2), \quad (3)$$

where  $v_x$  and  $v_z$  are the  $x$ - and  $z$ -components of the velocity,  $\nu$  denotes dynamic viscosity,  $c$  solution concentration at the point considered, and  $D$  diffusion coefficient of the transported solute. The boundary conditions are (since the problem is symmetrical,  $x \geq 0$ )

a) for the  $x$  component of velocity

$$\begin{aligned} v_x(0, z, t) &= 0 \quad \text{for } z \in R, t \in \langle 0, 2\pi/\omega \rangle \\ \lim_{x \rightarrow \infty} v_x(x, z, t) &= 0 \quad \text{for } z \in R, t \in \langle 0, 2\pi/\omega \rangle \\ \lim_{z \rightarrow \pm \infty} v_x(x, z, t) &= 0 \quad \text{for } x \geq 0, t \in \langle 0, 2\pi/\omega \rangle \\ v_x(x, z, 0) &= v_x(x, z, 2\pi/\omega), \end{aligned} \quad (4)$$

b) for the  $z$  component of velocity

$$\begin{aligned} v_z(0, z, t) &= 0 \quad \text{for } |z| \leq d/2, t \in \langle 0, 2\pi/\omega \rangle \\ \partial v_z / \partial x(0, z, t) &= 0 \quad \text{for } |z| > d/2, t \in \langle 0, 2\pi/\omega \rangle \end{aligned} \quad (5)$$

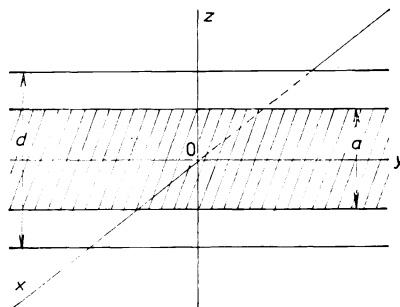


FIG. 1  
The vibrating plate with the coordinate system

$$\begin{aligned} \lim_{x \rightarrow \infty} v_z(x, z, t) &= r\omega \sin(\omega t) \quad \text{for } z \in R, t \in \langle 0, 2\pi/\omega \rangle \\ \lim_{z \rightarrow \pm \infty} v_z(x, z, t) &= r\omega \sin(\omega t) \quad \text{for } x \geq 0, t \in \langle 0, 2\pi/\omega \rangle \\ v_z(x, z, 0) &= v_z(x, z, 2\pi/\omega), \end{aligned}$$

c) for the concentration

$$\begin{aligned} c(0, z, t) &= 0 \quad \text{for } |z| \leq a/2, t \in \langle 0, 2\pi/\omega \rangle \\ \partial c/\partial x(0, z, t) &= 0 \quad \text{for } |z| > a/2, t \in \langle 0, 2\pi/\omega \rangle \\ \lim_{x \rightarrow \infty} c(x, z, t) &= c_0 \quad \text{for } z \in R, t \in \langle 0, 2\pi/\omega \rangle \\ \lim_{z \rightarrow \pm \infty} c(x, z, t) &= c_0 \quad \text{for } x \geq 0, t \in \langle 0, 2\pi/\omega \rangle \\ c(x, z, 0) &= c(x, z, 2\pi/\omega), \end{aligned} \quad (6)$$

where  $c_0$  denotes concentration of the diffusing substance at a large distance from the plate.

The mathematical problem defined by Eqs (1)–(6) can be considerably simplified. The number of unknown functions ( $v_x$ ,  $v_z$ ,  $c$ ) can be lowered by introducing the stream function  $\psi$  as

$$v_x = \partial\psi/\partial z, \quad v_z = -\partial\psi/\partial x. \quad (7)$$

Thus, the continuity equation (2) is automatically satisfied and Eqs (1) and (3) take the form

$$-\partial^2\psi/\partial x\partial t - \partial\psi/\partial z \cdot \partial^2\psi/\partial x^2 + \partial\psi/\partial x \cdot \partial^2\psi/\partial x\partial z = -v\partial^3\psi/\partial x^3, \quad (8)$$

$$\partial c/\partial t + \partial\psi/\partial z \cdot \partial c/\partial x - \partial\psi/\partial x \cdot \partial c/\partial z = D(\partial^2 c/\partial x^2 + \partial^2 c/\partial z^2). \quad (9)$$

It is convenient to introduce the dimensionless variables

$$X = x/r, \quad Z = z/r, \quad T = \omega t/2\pi, \quad C = c/c_0, \quad \Psi = \psi/v \quad (10)$$

and the dimensionless parameters

$$Sc = v/D, \quad B = \omega r^2/2\pi v. \quad (11)$$

Thus, the functions  $\psi$  and  $c$  of the variables  $x$ ,  $z$ , and  $t$  are replaced by the functions  $\Psi$  and  $C$  of the variables  $X$ ,  $Z$ , and  $T$

$$\frac{1}{v} \psi(x, z, t) = \frac{1}{v} \psi \left( rX, rZ, \frac{2\pi}{\omega} T \right) = \Psi(X, Z, T)$$

$$\frac{1}{c_0} c(x, z, t) = \frac{1}{c_0} c \left( rX, rZ, \frac{2\pi}{\omega} T \right) = C(X, Z, T).$$

The original integration domain  $\langle 0, \infty \rangle \times (-\infty, \infty)$  in the  $x$ - $z$  plane is transformed to a finite one,  $\langle 0, X_0 \rangle \times (-1, 1)$ , where  $X_0 > 0$ , by substituting  $Z_1 = (2/\pi) \operatorname{arctg} Z$  and by a suitable definition of the range of the variable  $X$ . The choice of the quantity  $X_0$  follows from physical analysis of the problem and depends on the parameters  $Sc$  and  $B$ . Thus, we arrive at the system of equations for the functions

$$\Psi_1(X, Z_1, T) = \Psi(X, \operatorname{tg}(\pi Z_1/2), T), \quad C_1(X, Z_1, T) = C(X, \operatorname{tg}(\pi Z_1/2), T)$$

In further text, the subscript 1 will be omitted for simplicity. Equations (8) and (9) acquire the form

$$\frac{\partial^3 \Psi}{\partial X^3} - \frac{2}{\pi} \cos^2 \frac{\pi Z}{2} \frac{\partial \Psi}{\partial Z} \frac{\partial^2 \Psi}{\partial X^2} + \frac{2}{\pi} \cos^2 \frac{\pi Z}{2} \frac{\partial \Psi}{\partial X} \frac{\partial^2 \Psi}{\partial X \partial Z} = B \frac{\partial^2 \Psi}{\partial X \partial T}, \quad (12)$$

$$\begin{aligned} \frac{\partial^2 C}{\partial X^2} + \frac{4}{\pi^2} \cos^3 \frac{\pi Z}{2} \left( \cos \frac{\pi Z}{2} \frac{\partial^2 C}{\partial Z^2} - \pi \sin \frac{\pi Z}{2} \frac{\partial C}{\partial Z} \right) + \\ + \frac{2}{\pi} Sc \cos^2 \frac{\pi Z}{2} \left( \frac{\partial C}{\partial Z} \frac{\partial \Psi}{\partial X} - \frac{\partial C}{\partial X} \frac{\partial \Psi}{\partial Z} \right) = BSc \frac{\partial C}{\partial T}. \end{aligned} \quad (13)$$

The boundary conditions for the function  $\Psi$  must be in accord with Eqs (4), (5), and (7). According to Eqs (7) and (10), the dimensionless stream function  $\Psi$  is defined except for an additive quantity which depends, in general, on the variable  $T$ . The function  $\Psi$  can be chosen so that  $\Psi(0, 0, T) = 0$ . Then

$$\Psi(0, Z, T) = 0 \quad \text{for} \quad |Z| \leq (2/\pi) \operatorname{arctg}(d/2r), \quad T \in \langle 0, 1 \rangle$$

$$\frac{\partial \Psi}{\partial X}(0, Z, T) = 0 \quad \text{for} \quad |Z| \leq (2/\pi) \operatorname{arctg}(d/2r), \quad T \in \langle 0, 1 \rangle \quad (14)$$

$$\Psi(0, Z, T) = 0 \quad \text{for} \quad (2/\pi) \operatorname{arctg}(d/2r) < |Z| \leq 1, \quad T \in \langle 0, 1 \rangle$$

$$\frac{\partial^2 \Psi}{\partial X^2}(0, Z, T) = 0 \quad \text{for} \quad (2/\pi) \operatorname{arctg}(d/2r) < |Z| \leq 1, \quad T \in \langle 0, 1 \rangle,$$

$$\frac{\partial \Psi}{\partial X}(X_0, Z, T) = -2\pi B \sin(2\pi T) \quad \text{for} \quad |Z| \leq 1, \quad T \in \langle 0, 1 \rangle \quad (15a)$$

$$\Psi(X, \pm 1, T) = -2\pi BX \sin(2\pi T) \quad \text{for } 0 \leq X \leq X_0, T \in \langle 0, 1 \rangle \quad (15b)$$

$$\Psi(X, Z, 0) = \Psi(X, Z, 1). \quad (15c)$$

The boundary conditions for the dimensionless concentration are

$$C(0, Z, T) = 0 \quad \text{for } |Z| \leq (2/\pi) \operatorname{arctg}(a/2r), T \in \langle 0, 1 \rangle \quad (16a)$$

$$\frac{\partial C}{\partial X}(0, Z, T) = 0 \quad \text{for } (2/\pi) \operatorname{arctg}(a/2r) < |Z| \leq 1, T \in \langle 0, 1 \rangle \quad (16b)$$

$$C(X_0, Z, T) = 1 \quad \text{for } |Z| \leq 1, T \in \langle 0, 1 \rangle \quad (16c)$$

$$C(X, \pm 1, T) = 1 \quad \text{for } 0 \leq X \leq X_0, T \in \langle 0, 1 \rangle \quad (16d)$$

$$C(X, Z, 0) = C(X, Z, 1). \quad (16e)$$

### Numerical Solution

Our task is to calculate the mass transfer by diffusion to the vibrating electrode after attainment of a dynamic equilibrium. The diffusion flux is given by the concentration gradient at the electrode surface. To this purpose, the concentration field at the vibrating electrode must be known, hence the boundary value problem (12)–(16) must be solved. This was done by the finite difference method.

With respect to the limited possibilities of the available computers, the following method was used for the numerical solution. The periodicity conditions (15c) and (16e) were replaced by the initial conditions

$$\Psi(X, Z, 0) = 0 \quad \text{for } 0 \leq X \leq X_0, |Z| \leq 1 \quad (17a)$$

$$C(0, Z, 0) = 0 \quad \text{for } |Z| \leq (2/\pi) \operatorname{arctg}(a/2r) \quad (17b)$$

$$C(0, Z, 0) = 1 \quad \text{for } (2/\pi) \operatorname{arctg}(a/2r) < |Z| \leq 1 \quad (17c)$$

$$C(X, Z, 0) = 1 \quad \text{for } 0 < X \leq X_0, |Z| \leq 1. \quad (17d)$$

The problem thus modified was solved with  $T$  as the parameter, beginning from  $T = 0$  and ending at  $T = T_k$ , when the functions  $\Psi$  and  $C$  varied during one period by less than the allowed error. The accuracy was tested both for the stream function  $\Psi$  and for the concentration  $C$ . It turned out that the stream function is already after two periods constant. Therefore, in further subroutines, only the concentration  $C$  was tested, and this in the first three columns of the chosen grid, adjacent to the "surface" column  $X = 0$ , since the values of  $\operatorname{grad} C$  were approximated as

$$(\operatorname{grad} C)_j = (-11C_{0j} + 18C_{1j} - 9C_{2j} + 2C_{3j})/(6h_x) + \mathcal{O}(h_x^3), \quad (18)$$

where  $h_x$  denotes the step of the grid along the  $X$  axis.

It is known that diffusion processes are slow. Therefore, after calculations during a number of periods (40–50), the essential functions,  $\Psi$  and  $C$ , were extrapolated for  $T \rightarrow \infty$ , the values obtained were used as new input data and further changes of  $\Psi$  and  $C$  were followed. When the changes were smaller than the allowed error, the calculations were stopped. In the opposite case, the described cycle was repeated.

The numerical calculations started at  $T = 0$  and continued with a time step equal to  $h_T$ . When passing from the  $k$ -th time level corresponding to the time  $T = kh_T$  to the  $(k + 1)$ -st time level corresponding to  $T = (k + 1)h_T$ , we used the finite-difference method with the implicit difference scheme in the integration domain  $\langle 0, X_0 \rangle \times \langle -1, 1 \rangle$  in the  $X$ - $Z$  plane. In this way, Eqs. (12) for the stream function was solved first. Since it is non-linear, the corresponding system of difference equations was solved by the Newton method. In calculating values at the  $(k + 1)$ -st time level, values at the  $k$ -th time level were used as initial approximations. After obtaining the stream function values at the grid points with the required accuracy, which was tested, the  $\Psi$  value were introduced into the diffusion equation (13), which was solved in an analogous manner. Since this equation is linear, the corresponding system of difference equations was solved by using the algorithm described earlier<sup>5</sup>.

## RESULTS AND DISCUSSION

After solving Eq. (12), the solution of Eq. (13) yielded the concentration field and gradients at the vibrating electrode. Physical reasons allow us to expect that the second and third terms on the left side of Eq. (13) have a much lesser influence on the concentration  $C$ ; for reasons of symmetry,  $\partial C/\partial Z(X, 0, T) = 0$  in the second term, and hence negligible for  $Z$  values close to zero. The term  $\partial^2 C/\partial Z^2(X, Z, T)$  in the neighbourhood of  $Z = 0$  plays the greater role the narrower is the active zone. Accordingly, the term  $(\partial C/\partial Z) \partial \Psi/\partial X$  in the third term can be neglected, whereas the value of  $(\partial C/\partial X) \partial \Psi/\partial Z$ , though small, is not negligible.

In order to substantiate these considerations numerically, it is convenient to replace the variable  $X$  by a new one,  $U$ :

$$U = \sqrt{(B \cdot Sc)} X.$$

Equation (13) then takes the form

$$B Sc \frac{\partial^2 C}{\partial U^2} + \frac{4}{\pi^2} \cos^3 \frac{\pi Z}{2} \left( \cos \frac{\pi Z}{2} \frac{\partial^2 C}{\partial Z^2} - \pi \sin \frac{\pi Z}{2} \frac{\partial C}{\partial Z} \right) + \\ + Sc \sqrt{(B \cdot Sc)} \frac{2}{\pi} \cos^2 \frac{\pi Z}{2} \left( \frac{\partial C}{\partial Z} \frac{\partial \Psi}{\partial U} - \frac{\partial C}{\partial U} \frac{\partial \Psi}{\partial Z} \right) = B Sc \frac{\partial C}{\partial T}$$

and after dividing with  $B \cdot Sc$

$$\begin{aligned} \frac{\partial^2 C}{\partial U^2} + \frac{1}{B \cdot Sc} \frac{4}{\pi^2} \cos^3 \frac{\pi Z}{2} \left( \cos \frac{\pi Z}{2} \frac{\partial^2 C}{\partial Z^2} - \pi \sin \frac{\pi Z}{2} \frac{\partial C}{\partial Z} \right) + \\ + \sqrt{\left( \frac{Sc}{B} \right)} \frac{2}{\pi} \cos^2 \frac{\pi Z}{2} \left( \frac{\partial C}{\partial Z} \frac{\partial \Psi}{\partial U} - \frac{\partial C}{\partial U} \frac{\partial \Psi}{\partial Z} \right) = \frac{\partial C}{\partial T}. \end{aligned} \quad (19)$$

If the second and third terms on the left-hand side of Eq. (19) are neglected, then

$$\frac{\partial^2 C}{\partial U^2} = \frac{\partial C}{\partial T}. \quad (20)$$

Here, the parameters  $B$  and  $Sc$  do not occur, and the solution is  $C = C(U, Z, T)$ , whence

$$\frac{\partial C}{\partial U}(0, Z, T) = k_1(Z, T)$$

and

$$\text{grad } C(0, Z, T) = \frac{\partial C}{\partial U}(0, Z, T) \sqrt{(B \cdot Sc)} = k_1(Z, T) \sqrt{(B \cdot Sc)} \quad (21)$$

With respect to Eq. (19), it can be shown by a thorough analysis that

$$\frac{\partial C}{\partial U}(0, Z, T) = k_1(Z, T) + \frac{k_2(Z, T)}{B \cdot Sc} + k_3(Z, T) \sqrt{\left( \frac{Sc}{B} \right)}$$

and hence

$$\text{grad } C(0, Z, T) = k_1(Z, T) \sqrt{(B \cdot Sc)} + \frac{k_2(Z, T)}{\sqrt{(B \cdot Sc)}} + k_3(Z, T) Sc. \quad (22)$$

The results of numerical solution show that the dependence on  $T$  is in the dynamic equilibrium not marked. Therefore, it is possible to replace the time average of the concentration gradient during one period by the value of the gradient at the end of the period. Since the concentration gradient at the centre of the active area (at the point  $Z = 0$ ) is important, the following semiempirical equation can be proposed for the mean value of  $\text{grad } C$  by analogy to Eq. (22):

$$\text{grad } C = K_1 \sqrt{(B \cdot Sc)} + \frac{K_2}{\sqrt{(B \cdot Sc)}} + K_3 Sc, \quad (23)$$

TABLE I  
Dependence of grad  $C$  on the width of vibrating plate ( $Sc = 1\ 000$ )

$B = 100$			$B = 1\ 000$		
$A$	$E$	grad $C$	$A$	$E$	grad $C$
1	2	38·3005	1	2	127·661
1	4	38·3024	1	4	127·683
1·5	2	37·4567	1·5	2	123·867
1·5	4	37·3705	1·5	4	123·498

TABLE II  
Dependence of grad  $C$  on the electrode width ( $Sc = 1\ 000$ )

$B = 100$			$B = 1\ 000$		
$A$	$E$	grad $C$	$A$	$E$	grad $C$
0·5	2	45·3584	0·5	2	154·100
1	2	38·3005	1	2	127·661
1·5	4	37·3705	1·5	4	123·498
2	4	37·1653	2	4	122·448
5	15	33·8117	5	15	106·659
10	30	33·8117	10	30	106·659

TABLE III  
Values of grad  $C$  as function of parameters  $B$  and  $Sc$  for  $A = 10$ ,  $E = 30$

$B$	$Sc$	$\sqrt{(B \cdot Sc)}$	grad $C$
$10^3$	$10^3$	$10^3$	106·51
$10^2$	$10^4$	$10^3$	99·83
10	$10^5$	$10^3$	99·09
$10^2$	$10^2$	$10^2$	11·238
10	$10^3$	$10^2$	10·661
1	$10^4$	$10^2$	9·935
10	10	10	0·9877
0·1	$10^3$	10	1·0679



where the coefficients  $K_1$ ,  $K_2$  and  $K_3$  depend on the width of the active area,  $a$ , and on the width of the vibrating plate,  $d$ , as can be seen from Tables I and II. We denote, for convenience,

$$A = \frac{a}{2r}, \quad E = \frac{d}{2r}. \quad (24)$$

It follows from Table I that the concentration gradient is practically independent of  $E$  at constant  $A$ , which is in accord with expectation. Changes of the width of the vibrating plate,  $d$ , i.e. of the parameter  $E$  influence only the boundary conditions for the streaming velocity of the electrolyte (Eqs (14)), and this has a negligible effect on the concentration field in the vicinity of the electrode. It can be expected for physical reasons that the dependence of  $\text{grad } C$  on the electrode width will be much stronger than on the width of the vibrating plate, since the quantity  $a$  (and hence  $A$ ) is involved in the boundary conditions (16a, b). This is substantiated by the results in Table II, according to which the values of the concentration gradient decrease with increasing ratio of  $a/r$ , hence the influence of the boundaries diminishes. The last two couples of the mentioned values suggest that the boundary effect becomes negligible for sufficiently high values of  $a/r$ . Therefore, further calculations were made for  $A = 10$  and  $E = 30$ .

The results of calculations are given in Table III, from which the coefficients  $K_1$ ,  $K_2$ , and  $K_3$  in Eq. (23) were determined by the least squares method. Thus,

$$\text{grad } C = 0.104 \sqrt{(B \cdot Sc)} + \frac{2.01}{\sqrt{(B \cdot Sc)}} - 4.85 \cdot 10^{-5} Sc. \quad (25)$$

Here, the first term on the right-hand side is dominant, the second is negligible in common cases, i.e. for  $B \in \langle 10, 100 \rangle$  and  $Sc \in \langle 100, 10\,000 \rangle$ , and the last one comes into play only at very large values of  $Sc$ . Therefore, it can be concluded that the concentration gradient,  $\text{grad } C$ , is under suitably chosen and often encountered physical conditions directly proportional to  $\sqrt{(B \cdot Sc)}$ .

#### SYMBOLS

$a$	width of active area
$A$	dimensionless parameter
$B$	dimensionless parameter
$c$	concentration
$c_0$	concentration in the bulk of solution
$C(X, Z, T)$	dimensionless concentration
$C_1(X, Z_1, T)$	dimensionless concentration
$C_{ij}$	value of dimensionless concentration at the grid point
$d$	width of vibrating plate

$D$	diffusion coefficient
$E$	dimensionless parameter
$h_x, h_z, h_T$	grid steps
$k_i(Z, T)$	empirical coefficient, $i = 1, 2, 3$
$r$	amplitude of harmonic vibrations
$Sc$	Schmidt number
$t$	time
$T$	dimensionless time
$U$	dimensionless coordinate
$v_x, v_y, v_z$	velocity components
$x, y, z$	Cartesian coordinates
$X, Z$	dimensionless coordinates
$X_0$	limit of the integration domain
$Z_1$	auxiliary space coordinate
$\nu$	dynamic viscosity of solution
$\psi$	stream function
$\Psi(X, Z, T)$	dimensionless stream function
$\Psi_1(X, Z_1, T)$	dimensionless stream function
$\omega$	angular frequency of harmonic vibrations

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