

# GENERALIZATION OF LEVICH EQUATION WITH RESPECT TO CONCENTRATION DEPENDENCE OF DIFFUSION COEFFICIENT AND KINEMATIC VISCOSITY

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The theory of the rotating disc method is derived taking into account a linear dependence of the diffusion coefficient and viscosity on concentration. The case of the dissolution of the disc and the ionic transport in the presence of an indifferent electrolyte are considered. The results of the calculation lead to the determination of the correction term in the Levich equation for the diffusion current or for the limiting current density; further the relation between the apparent diffusion coefficient and the heterodiffusion coefficient is determined.

The theory of the rotating disc method involves the hydrodynamics of the liquid (melt) set into motion by the disc and the solution of the diffusion equation. The solution of the first part was derived by Karman<sup>1</sup> and later more exactly by Cochran<sup>2</sup>. The diffusion problem was solved by Levich<sup>3</sup> who assumed independence of the diffusion coefficient of the concentration ( $D = \text{const.}$ ).

The aim of the present work is to generalize the theory of the rotating disc electrode in a laminar regime under the assumption that the diffusion coefficient is a linear function of concentration. In the case of the electrochemical variant of the method, this generalization will apply for ionic melts and solutions of electroactive compounds with an indifferent electrolyte.

The diffusion problem will be described by a more general equation of convective diffusion in liquids<sup>4</sup>:

$$\partial C / \partial t = \text{div} (D \cdot \text{grad } C) - \text{div} (C \cdot \vec{V}), \quad (1)$$

where  $C$  denotes concentration of the diffusing substance and  $\vec{V}$  velocity of flow of the liquid. Since the latter is incompressible ( $\text{div } V = 0$ ) and we shall consider the convective diffusion only in the stationary state ( $\partial C / \partial t = 0$ ), Eq. (1) takes the form

$$(V \cdot \text{grad}) \cdot C = D \cdot \Delta C + \text{grad } D \cdot \text{grad } C. \quad (2)$$

As usual in the rotating disc method, the concentration will be considered indepen-

dent of the coordinates  $r$  and  $\varphi$  and dependent only on the distance  $y$  from the disc surface. Then we obtain from Eq. (2)

$$v(y) \frac{dC(y)}{dy} = D[C(y)] \frac{d^2C(y)}{dy^2} + \frac{dD[C(y)]}{dy} \frac{dC(y)}{dy} . \quad (3)$$

The generalization of the Levich equation is hence reduced to finding the function  $C(y)$ . Let  $C(y)$  obey the convective diffusion equation (3), where  $D[C(y)]$  is expressed as

$$D[C(y)] = D_h[1 + \alpha C(y)] . \quad (4)$$

The boundary conditions are

$$C(y = 0) = C_{\text{sat}} , \quad \lim_{y \rightarrow \infty} C(y) = 0 , \quad (5)$$

or

$$C(y = 0) = 0 , \quad \lim_{y \rightarrow \infty} C(y) = C_0 , \quad (6)$$

where  $D_h$  denotes heterodiffusion coefficient and  $\alpha$  coefficient characterizing the linear concentration dependence of the diffusion coefficient. The boundary conditions (5) characterize the dissolution of the solid phase (the rotating sample) in the liquid (or in the melt) and  $C_{\text{sat}}$  denotes equilibrium concentration (or concentration of the saturated solution). The obvious assumption is that the concentration of the dissolved substance at the end of the experiment is very small, practically zero. The conditions (6) characterize the application of the rotating disc method in electrochemistry, where during current passage through the rotating disc electrode and the melt the concentration at the electrode surface is zero and the concentration of the depolarizer in the melt is  $C_0$ .

To obtain the solution  $C(y)$ , first it is necessary to note the relations for the rotating disc method assuming  $D = D_h = \text{const.}$ ,  $v = v_0 = \text{const.}$ , where  $v$  denotes kinematic viscosity (*i.e.*, the Levich's solution). Eq. (3) then takes the form

$$v(y) [dC_1(y)/dy] = D_h [d^2C_1(y)/dy^2] . \quad (7)$$

The solution of Eq. (7) with boundary conditions (6) is<sup>4</sup>

$$C_1(y) = C_0 [J_1(B \cdot y) / J_1(B \cdot y \rightarrow \infty)] . \quad (8)$$

In the case of dissolution of a rotating disc-shaped sample in a liquid we have the

boundary conditions (5) and the solution  $C_1(y)$  is

$$C_1(y) = C_{\text{sat}}[1 - J_1(By)/J_1(By \rightarrow \infty)], \quad (9)$$

where  $J_1(By)$  is given as<sup>4</sup>

$$J_1(By) = \int_0^{By} \exp(-\xi^3) d\xi, \quad J_1(By \rightarrow \infty) \rightarrow 0.893, \quad (10)$$

$$B = (2/\delta_0)(v_0/D_h)^{1/3} = 1/\delta. \quad (11)$$

Here  $\delta_0$  denotes the thickness of the hydrodynamic boundary layer (Prandtl layer),  $v_0$  kinematic viscosity at  $C_1(y) = 0$ , and  $0.893\delta$  is the thickness of the boundary diffusion layer. The function  $V(y)$  in Eq. (7) is given by<sup>4</sup>

$$V(y) \approx -A_0 y^2, \quad A_0 = 0.51\sqrt{(\omega^3/v_0)}, \quad y \ll \sqrt{(v_0/\omega)}, \quad (12)$$

where  $\omega$  denotes angular velocity of the disc rotation. The quantities  $\delta_0$  and  $\delta$  are defined as<sup>4</sup>

$$\delta_0 = 3.6\sqrt{(v_0/\omega)}, \quad \delta = 0.5(D_h/v_0)^{1/3} \delta_0, \quad \delta \ll \delta_0. \quad (13)$$

To solve the more general equation (3), we shall make use of the following approximation<sup>4</sup>:

$$D[C(y)] = D_h[1 + \alpha C(y)] \approx D_h[1 + \alpha C_1(y)] \quad (14)$$

assuming  $\alpha C_{\text{sat}} \ll 1$ ,  $\alpha C_0 \ll 1$ . The function  $C_1(y)$  is according to the choice of the boundary conditions (5) or (6) given by the solution (9) or (8). If we introduce Eq. (4) into (3), the approximate solution of the latter is advantageously expressed in the form

$$C(y) \approx C_1(y) + \alpha C_2(y) + \alpha^2 C_3(y) + \dots, \quad (15)$$

where the function  $C_1(y)$  is given by Eq. (8) or (9). If we introduce then the solution (15) into Eq. (4) and compare with (14) it is seen that we neglected in Eq. (14) the terms containing powers of  $\alpha$  of higher order than the first.

We now shall solve Eq. (3) with boundary conditions (5) or (6) using the expression (14) for  $D[C(y)]$ :

$$\left[ V(y) - \alpha D_h \frac{dC_1(y)}{dy} \right] f(y) = D_h[1 + \alpha C_1(y)] \frac{df(y)}{dy}, \quad (16)$$

where

$$f(y) = dC(y)/dy. \quad (17)$$

Further we shall set  $[1 + \alpha C_1(y)]^{-1} \approx 1 - \alpha C_1(y)$  and obtain the integral of Eq. (16):

$$\frac{1}{D_h} \int_0^y \left[ V(y') - \alpha C_1(y') V(y') - \alpha D_h \frac{dC_1(y')}{dy'} \right] dy' = \ln f(y) + \text{const.} \quad (18)$$

The functions  $V(y)$  and  $C_1(y)$  are given by Eqs (12) and (9) or (8). We now shall discuss Eq. (12) for the rate of flow, which is dependent on the kinematic viscosity  $\nu$ . Since the Einstein-Stokes relation holds as a first approximation<sup>4</sup>,

$$D \approx \text{const}/\nu, \quad (19)$$

then the dependence of viscosity on concentration according to Eqs (14) and (19) is

$$\nu[C(y)] \approx \text{const}/D[C(y)] \approx \nu_0[1 - \alpha C_1(y)], \quad (20)$$

where  $\nu_0$  denotes kinematic viscosity at zero concentration. This result together with (12) gives the flow velocity in the form

$$V(y) = V[C(y)] \approx -A_0[1 + \frac{1}{2}\alpha C_1(y)] y^2, \quad y \ll \sqrt{(\nu_0/\omega)}, \quad (21)$$

where  $A_0 = 0.51\sqrt{(\omega^3/\nu_0)}$ . If we introduce this result into Eq. (18) we obtain after rearrangement

$$\begin{aligned} D_h^{-1} \int_0^y \left[ -A_0 y'^2 + \frac{1}{2}\alpha A_0 C_1(y') y'^2 - \alpha D_h (dC_1(y')/dy') \right] dy' = \\ = \ln f(y) + \text{const.} \end{aligned} \quad (22)$$

For the first case, we express  $C_1(y)$  from the solution (9) to obtain

$$\begin{aligned} D_h^{-1} \int_0^y \left\{ -A_0 \left( 1 - \frac{1}{2}\alpha C_{\text{sat}} \right) y'^2 + \frac{1}{2}(\alpha C_{\text{sat}}/J_1(By \rightarrow \infty)) \cdot \right. \\ \left. \cdot [-A_0 J_1(By') y'^2 + 2D_h (dJ_1(By')/dy')] \right\} dy' = \ln f(y) + \text{const.} \end{aligned} \quad (23)$$

We now shall calculate the integral

$$I(y) = \int_0^y J_1(By') y'^2 dy', \quad (24)$$

where  $J_1(By)$  is given by Eq. (10) and for  $B$  we obtain from Eqs (11) and (13):

$$B = \sqrt{\omega/1.8 D^{1/3} \nu_0^{1/6}} = 1/\delta, \quad (25)$$

$$I(y) = \frac{1}{3} y^3 J_1(By) - \frac{1}{3} B^{-3} \int_0^{By} \xi^3 \exp(-\xi^3) d\xi, \quad (26)$$

$$J_1(By) = By \cdot \exp(-By)^3 + 3 \int_0^{By} \xi^3 \exp(-\xi^3) d\xi. \quad (27)$$

From the integrals (26) and (27), which can be calculated by the *per partes* method, follows

$$I(y) = \frac{1}{3} y^3 J_1(By) - J_1(By)/9B^3 + (y/9B^2) \exp(-By)^3. \quad (28)$$

With the aid of Eqs (24) and (28) the integral in (23) can be calculated. If we consider Eqs (21) and (25), we have

$$A_0 = 3D_h B^3. \quad (29)$$

Based on Eqs (28) and (29), Eq. (23) can be after integration put into the form

$$F(By, \alpha C_{\text{sat}}) = -(1 - 0.5\alpha C_{\text{sat}})(By)^3 - \frac{\alpha C_{\text{sat}}}{2J_1(By \rightarrow \infty)} \cdot \left[ (By)^3 J_1(By) - \frac{7}{3} J_1(By) + \frac{1}{3} By \cdot \exp(-By)^3 \right] = \ln f(y) + \text{const.} \quad (30)$$

The function  $f(y)$  is expressed from Eq. (17) and Eq. (30) can be integrated to give

$$C(y) = (a_1/B) \int_0^{By} \exp F(u, \alpha C_{\text{sat}}) du + a_2. \quad (31)$$

The constants  $a_1$  and  $a_2$  can be determined from the boundary conditions (5). If we denote the integral in Eq. (31) as  $J(By, \alpha C_{\text{sat}})$ , then

$$a_1 = -BC_{\text{sat}}/J(By \rightarrow \infty, \alpha C_{\text{sat}}), \quad a_2 = C_{\text{sat}}. \quad (32)$$

It follows from this derivation that the course of the concentration of the diffusing particles is in the case of dissolution of the rotating disc given by

$$C(y) = C_{\text{sat}} \left( 1 - \frac{J(By, \alpha C_{\text{sat}})}{J(By \rightarrow \infty, \alpha C_{\text{sat}})} \right), \quad (33)$$

where

$$J(By, \alpha C_{\text{sat}}) = \int_0^{\text{By}} \exp \left\{ -(1 - 0.5\alpha C_{\text{sat}}) u^3 - \frac{\alpha C_{\text{sat}}}{2J_1(\infty)} \cdot \right. \\ \left. \cdot \left[ u^3 J_1(u) - \frac{7}{3} J_1(u) + \frac{1}{3} u \cdot e^{-u^3} \right] \right\} du, \quad (34)$$

$B = 0.555\sqrt{\omega/D^{1/3}v_0^{1/6}}$ ,  $J_1(\infty) = 0.893$  according to Eqs (25) and (10).

It should be noted for completeness that for the dissolution of the rotating disc the boundary conditions for Eq. (3) can have in certain cases the form

$$C(y = 0) = C_{\text{sat}}, \quad \lim_{y \rightarrow \infty} C(y) = C_1 < C_{\text{sat}}. \quad (35)$$

The solution  $C(y)$  will then have the form

$$C(y) = C_{\text{sat}} - (C_{\text{sat}} - C_1) \frac{J(By, \alpha C_{\text{sat}}, \alpha C_1)}{J(By \rightarrow \infty, \alpha C_{\text{sat}}, \alpha C_1)}, \quad (36)$$

where

$$J(By, \alpha C_{\text{sat}}, \alpha C_1) = \int_0^{\text{By}} \exp \left\{ -(1 - 0.5\alpha C_{\text{sat}}) u^3 - \frac{\alpha(C_{\text{sat}} - C_1)}{2J_1(\infty)} \cdot \right. \\ \left. \cdot \left[ u^3 J_1(u) - \frac{7}{3} J_1(u) + \frac{1}{3} u \cdot \exp(-u^3) \right] \right\} du. \quad (37)$$

It should again be pointed out that the solutions (33) and (36) apply if the total quantity of the dissolved substance from the rotating disc at the end of the experiment is very small and can be hence considered equal to zero, so that the boundary conditions (5) and (35) are fulfilled.

We now shall calculate analogously the solution  $C(y)$  for the boundary conditions (6). This is the case of the well known rotating disc electrode:

$$C(y) = C_0 J(By, \alpha C_0) / J(By \rightarrow \infty, \alpha C_0), \quad (38)$$

where

$$J(By, \alpha C_0) = \int_0^{\text{By}} \exp \left\{ -u^3 + \frac{\alpha C_0}{2J_1(u \rightarrow \infty)} \cdot \right. \\ \left. \cdot \left[ J_1(u) u^3 - \frac{7}{3} J_1(u) + \frac{1}{3} u \cdot \exp(-u^3) \right] \right\} du. \quad (39)$$

The meaning of other symbols is the same as with Eq. (34). It is apparent that for  $\alpha \rightarrow 0$ , i.e.,  $D \rightarrow D_h$  the solutions (33), (36) and (38) take the form of (9), (8) etc. derived by Levich.

The integration of Eq. (18) requires some comments. From Eqs (12) and (13) it follows that the expression for the velocity  $C(y)$  applies for  $y \ll \sqrt{(v_0/\omega_0)} < \delta_0$ , as a first approximation. For other  $y$  values, we have<sup>4</sup>

$$V(y) = -0.89\sqrt{(v_0\omega)}. \quad (40)$$

Accordingly, Eq. (18) should be integrated in two steps: from zero to  $y \approx \delta \ll \delta_0$  with the use of Eq. (12) or (21) for  $V(y)$ , and from  $y \approx \delta$  to infinity with the use of (40). However, the value of the second integral is negligible against that of the first one and therefore it is possible to use the procedure given in the present paper. (For more details, see ref.<sup>4</sup>).

To calculate the flux density,  $j$ , from the disc in the case of dissolution or to the disc in the case of the disc electrode, we shall make use of the definition

$$|j| = -D[C(y=0)](\partial C(y)/\partial y)_{y=0}. \quad (41)$$

In the case of the dissolution of the rotating disc we obtain from the solution (33):

$$j = [(1 + \alpha C_{\text{sat}})/J(By \rightarrow \infty, \alpha C_{\text{sat}})] \cdot 0.55 D_h^{2/3} v_0^{-1/6} \omega^{1/2} C_{\text{sat}}. \quad (42)$$

For the rotating disc electrode, the limiting current density  $i$  (A/cm<sup>2</sup>) rather than the flux density  $j$  is used;  $i = nFj$ , where  $n$  denotes the number of exchanged electrons and  $F$  Faraday's constant. From the solution (38) and Eq. (41) we obtain

$$i = [0.55nF/J(By \rightarrow \infty, \alpha C_0)] D_h^{2/3} v_0^{-1/6} \omega^{1/2} C_0. \quad (43)$$

The values of the integrals  $J(By, \alpha C_{\text{sat}})$  and  $J(By, \alpha C_0)$  for  $y \rightarrow \infty$  were calculated by a computer and are shown graphically in Figs 1–3. The last of them suggests the following relations:

$$J(By \rightarrow \infty, \alpha C_{\text{sat}}) \approx 0.893(1 + K_{11}\alpha C_{\text{sat}}), \quad (44)$$

where  $i = 1$  or  $2$ ,  $|\alpha| C_{\text{sat}} \leq 0.3$ ,  $K_{11} = 0.60$  for  $\alpha > 0$ , and  $K_{12} = 0.51$  for  $\alpha < 0$ .

$$J(By \rightarrow \infty, \alpha C_0) \approx 0.893(1 - K_{21}\alpha C_0), \quad (45)$$

where  $i = 1$  or  $2$ ,  $|\alpha| C_0 \leq 0.3$ ,  $K_{21} = 0.35$  for  $\alpha > 0$ , and  $K_{22} = 0.41$  for  $\alpha < 0$ . The right sides in these equations approximate the function  $J$  in the mentioned interval with an accuracy of  $10^{-3}$ ; the error in the coefficients  $K_{ji}$  is smaller than 2%.

The generalized Levich equation will hence have the form:

a) for the dissolution of the disc – Eqs (42) and (44) –

$$\begin{aligned} |j| &= 0.62 D_h^{2/3} v_0^{-1/6} \omega^{1/2} C_{\text{sat}} (1 + \alpha C_{\text{sat}}) / (1 + K_{11}\alpha C_{\text{sat}}) \approx \\ &\approx 0.62 (1 + K_{31}\alpha C_{\text{sat}}) D_h^{2/3} v_0^{-1/6} \omega^{1/2} C_{\text{sat}}, \end{aligned} \quad (46)$$

where  $i = 1$  or  $2$ ,  $K_{31} = 0.40$  for  $\alpha > 0$ ,  $K_{32} = 0.48$  for  $\alpha < 0$ , and  $|\alpha| C_{\text{sat}} \leq 0.3$ ; b) for the rotating disc electrode – Eqs (43) and (45) –

$$i \approx 0.62(1 + K_{2i}\alpha C_0) nFD_h^{2/3} v_0^{-1/6} \omega^{1/2} C_0 \quad (47)$$

for  $|\alpha| C_0 \leq 0.3$ .

According to the original theory of the rotating disc, the diffusion coefficient was calculated from Eq. (42) or (43) for  $\alpha = 0$ . In this way the “effective” diffusion coefficient,  $\bar{D}$ , was found:

$$\bar{D} = [ |j| (0.62 v_0^{-1/6} \omega^{1/2} C_{\text{sat}}) ]^{-3/2}. \quad (48)$$

If we consider Eqs (42)–(48), we can simply deduce the relation between the value of  $\bar{D}$  and the heterodiffusion coefficient  $D_h$ . As a first approximation, we have for the case of the disc dissolution

$$\bar{D} \approx D_h(1 + K_{4i}\alpha C_{\text{sat}}), \quad i = 1, 2, \quad (49)$$

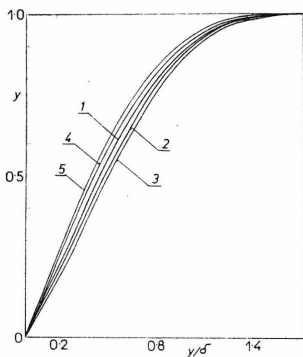


FIG. 1

Dependence of  $J(y/\delta, \alpha C_{\text{sat}})/J(y/\delta \rightarrow \infty, \alpha C_{\text{sat}})$ , ( $y$ -axis) from Eq. (34) on  $y/\delta = By$  ( $x$ -axis) for Various Values of  $\alpha C_{\text{sat}}$

Values of  $\alpha C_{\text{sat}}$ : 1 0; 2 0.2; 3 0.4; 4 -0.2; 5 -0.4. The curves give the courses of the function  $1 - C(y)/C_{\text{sat}}$ , compare Eq. (33), where  $C(y)$  denotes concentration of diffusing substance in the case of dissolution of the rotating disc.

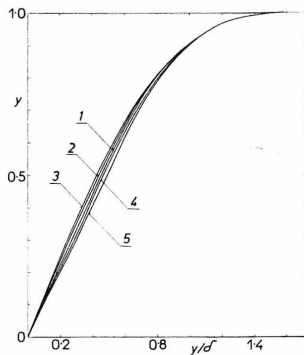


FIG. 2

Dependence of  $J(y/\delta, \alpha C_0)/J(y/\delta \rightarrow \infty, \alpha C_0)$ , ( $y$ -axis) from Eq. (39) on  $y/\delta = By$  ( $x$ -axis) for Various Values of  $\alpha C_0$

Values of  $\alpha C_0$ : 1 0; 2 0.2; 3 0.4; 4 -0.2; 5 -0.4. The curves give the courses of the concentration of the diffusing substance,  $C(y)/C_0$  – compare Eq. (38), in the case of the rotating disc electrode.



$K_{41} = 0.60$  for  $\alpha > 0$ ,  $K_{42} = 0.73$  for  $\alpha < 0$ ,  $|\alpha| C_{\text{sat}} \leq 0.3$ . For the rotating disc electrode, we have

$$\tilde{D} \approx D_h(1 + K_{5i}\alpha C_0), \quad i = 1, 2, \quad (50)$$

$K_{51} = 0.53$  for  $\alpha > 0$ ,  $K_{52} = 0.62$  for  $\alpha < 0$ ,  $|\alpha| C_0 \leq 0.3$ .

FIG. 3  
Functions 1  $J(B\gamma \rightarrow \infty, \alpha C_{\text{sat}} > 0)$ , 2  $J(B\gamma \rightarrow \infty, \alpha C_0 < 0)$ , 3  $J(B\gamma \rightarrow \infty, \alpha C_0 > 0)$ , 4  $J(B\gamma \rightarrow \infty, \alpha, C_{\text{sat}} < 0)$  Calculated from Eqs (34) and (39) for  $B\gamma \rightarrow \infty$ .

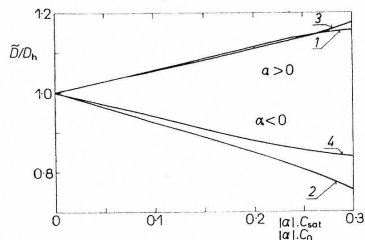
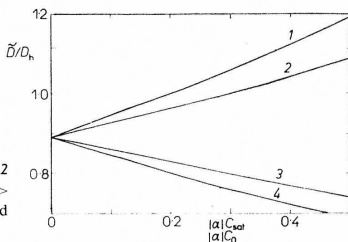
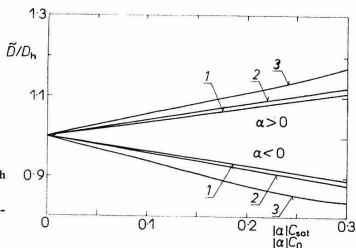


FIG. 4  
Dependences of  $\tilde{D}/D_h$  on  $\alpha C_{\text{sat}}$  or  $\alpha C_0$  Calculated from Eqs (53)–(56)

1 Dissolution of the rotating disc for  $\alpha > 0$ , 2 same for  $\alpha < 0$ , 3 rotating disc electrode for  $\alpha > 0$ , 4 same for  $\alpha < 0$ .

FIG. 5  
Comparison of the Dependences of  $\tilde{D}/D_h$  on  $\alpha C_0$  for Electrochemical Methods

1 Chronoamperometry, 2 chronopotentiometry, 3 rotating disc method.



The dependences of  $\bar{D}/D_h$  on  $|\alpha| C_0$  or  $|\alpha| C_{\text{sat}}$  calculated from Eqs (42)–(45) and (48) are shown graphically in Fig. 4.

It is apparent from the above theory that the influence of the concentration dependence of the diffusion coefficient on the flux at the disc surface or on the  $\bar{D}$  values is somewhat different for the disc dissolution and for the electrochemical variant of the method. This difference is due also to the fact that the flux during dissolution is proportional to  $D(C_{\text{sat}}) \approx D_h(1 + \alpha C_{\text{sat}})$ , whereas in the other case it is proportional to  $D(C \rightarrow 0) = D_h$  – compare Eqs (42), (23), and (4).

Eqs (49) and (50) are criterions which enable us to decide – the values of  $\alpha C_{\text{sat}}$  and  $\alpha C_0$  being known – if the coefficient  $D$  can be in practical cases set equal to the heterodiffusion coefficient  $D_h$ . The values of  $D_h$  and  $\alpha$  can be determined from Eq. (50) if measurements with electrolytes of different concentration are carried out. The value of  $\alpha$  can be substantially different from  $1 \text{ cm}^3/\text{mol}$ .

In the derivation of the generalized Levich equation for the rotating disc electrode, we have neglected the migration current of ions. The case where the migration current is taken into consideration is solved in another paper<sup>5</sup>.

The dissolution of the rotating disc on the assumption of an exponential dependence of the diffusion coefficient on the concentration was solved earlier<sup>6</sup>, however the integration of the equation of convective diffusion was performed numerically by the Runge-Kutta method on a computer, and the dependence of viscosity on concentration was not considered.

Also with other electrochemical methods, namely chronoamperometry and chronopotentiometry, we obtain only “effective” diffusion coefficients  $\bar{D}$  if the nongeneralized theory is used<sup>7,8</sup>. The corresponding dependences of  $\bar{D}$  on  $\alpha C_0$  are, together with Eq. (50) for the rotating disc method, shown in Fig. 5. In the latter case the concentration dependence of  $F$  is most pronounced.

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