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The Short-Time Response of a Disk Electrode

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September, 1973

Abstract

An asymptotic treatment of the transient response of a disk electrode is given for short times. Results are obtained by expressing the potential in terms of an integral equation at the surface. A similar formulation is used also to calculate the steady-state current distribution for large exchange current densities.

Key Words: Singular perturbation, galvanostatic control, potentiostatic control, exchange-current density, current distribution

Introduction

The authors have recently reported[1,2] a theoretical study of the transient behavior of a disk electrode in the presence of faradaic and capacitive effects. Series expressions were obtained for the potential and the current under galvanostatic and potentiostatic control, respectively. Those results are readily applicable to describe the long-time response of the disk and determine the relaxation time of the overpotential after a step change in the applied current or the current decay after a step change in the potential. However, a large number of terms need to be included in the series to express the short time behavior accurately. This can be accomplished more efficiently by deriving an asymptotic solution to the problem valid at small times.

Shortly after the cell current is switched on, the current distribution on the surface is given by the primary distribution[3,4] everywhere except at a small region near the edge of the disk. Since the primary current density is infinite at the edge, the double-layer capacity is charged more rapidly in this region than at other parts of the disk, so that the current density is reduced to a finite value.

From a mathematical standpoint, we have a singular-perturbation problem at hand. A similar problem at high frequencies for the alternating-current distribution on a disk electrode has been treated earlier.[5] We follow here the same guidelines in the mathematical formulation as developed in that paper. Another problem of the same type is encountered for large values of the exchange-current density.[6]

The singular nature of this problem has been recognized;[5] however its consequences were not of immediate interest. It nevertheless conforms with the context of the present paper, and an analysis is outlined in Appendix II, thereby rendering more complete the overall treatment of the secondary current distribution at a disk electrode.

Mathematical Model

The potential in the solution satisfies Laplace's equation,

$$\nabla^2\Phi = 0 \; . \tag{1}$$

The current density vanishes on the insulating plane of the disk, which implies

$$\frac{\partial\Phi}{\partial z} = 0 \quad \text{at} \quad z = 0 \; , \; r > r_o \; . \tag{2}$$

Away from the edge region, the potential in the solution is given by the primary distribution, Φ^p . Furthermore, at the surface, the passage of current is primarily due to the charging of the electric double layer,

$$i = -\kappa \frac{\partial\Phi}{\partial z} = C \frac{\partial(V - \Phi)}{\partial t} \quad \text{at} \quad z = 0 \; , \; r \leq r_o \; , \tag{3}$$

where V is the potential of the electrode, κ is the conductivity of the solution, and C is the double-layer capacity. Charge transfer may also occur by means of a faradaic reaction. This effect is small at short times as long as the exchange-current density is not

too large, and it is neglected here. The other extreme, where the faradaic reaction dominates over the double-layer charging, is discussed in Appendix II. Diffusion is also neglected. The transient response of a rotating-disk electrode under mass-transfer-controlled situations has been treated elsewhere.[7,8]

Analysis and Results

In terms of the rotational elliptic coordinates, the potential distribution outside the edge region with the counter electrode placed at infinity is given by[3]

$$\Phi^p/\Phi_o^p = 1 - \frac{2}{\pi}\tan^{-1}\xi \ , \tag{4}$$

where Φ_o^p is the potential in the solution adjacent to the electrode surface corresponding to the primary distribution.[1] If the electrode potential is kept at a constant value V, then $\Phi_o^p = V$. If, however, the electrode is under galvanostatic control, $\Phi_o^p = I/4r_o\kappa$. With this difference in mind, the present analysis applies to both galvanostatic and potentiostatic cases unless stated otherwise. For small ξ, equation 4 can be approximated by

$$\Phi/\Phi_o^p = 1 - 2\xi/\pi \ , \tag{5}$$

which may be regarded as the outer solution for the potential, with which the potential in the edge region must match. For instance, in the inner region, the potential approaches equation 5 asymptotically

as $\xi \to \infty$ and also as $t \to 0$. The rotational elliptic coordinate system reduces to parabolic coordinates in the edge region,[5] and Laplace's equation can be expressed as

$$\frac{\partial^2 \Phi}{\partial \eta^2} + \frac{\partial^2 \Phi}{\partial \xi^2} = 0 \ . \tag{6}$$

Let us now introduce the stretched variables appropriate to the edge region:

$$\bar{\phi} = \frac{\pi}{2\sqrt{\theta}} \left(\frac{V - \Phi}{\Phi_o^p} \right) , \tag{7}$$

$$\bar{\eta} = \eta/\sqrt{\theta} \ , \ \bar{\xi} = \xi/\sqrt{\theta} \ , \tag{8}$$

where $\theta = \kappa t/r_o C$ is the dimensionless time. Equation 7 represents a separation of variables, such that $\bar{\phi}$ is a function of $\bar{\eta}$ and $\bar{\xi}$ only. When the basic equations are transformed into the stretched coordinates, the problem reads

$$\frac{\partial^2 \bar{\phi}}{\partial \bar{\eta}^2} + \frac{\partial^2 \bar{\phi}}{\partial \bar{\xi}^2} = 0 \ , \tag{9}$$

$$\frac{\partial \bar{\phi}}{\partial \bar{\xi}} = \frac{1}{2} \bar{\eta}\bar{\phi} - \frac{\bar{\eta}^2}{2} \frac{\partial \bar{\phi}}{\partial \bar{\eta}} \quad \text{at} \quad \bar{\xi} = 0 \ , \tag{10}$$

$$\frac{\partial \bar{\phi}}{\partial \bar{\eta}} = 0 \quad \text{at} \quad \bar{\eta} = 0 \ , \tag{11}$$

$$\bar{\phi} \to \bar{\xi} \quad \text{as} \quad \bar{\eta}^2 + \bar{\xi}^2 \to \infty \ . \tag{12}$$

These results indicate that the inner region is of order $r_o\theta$ in the original, cylindrical coordinate system. Moreover, the dimensionless potential is of order $\sqrt{\theta}$, and the dimensionless current density, given by

$$\frac{ir_o}{\kappa\Phi_o^p} = -\frac{V - \Phi_o}{2\Phi_o^p\theta}, \tag{13}$$

is of order $1/\sqrt{\theta}$.

One possible way of solving the system of equations 9 to 12 is by finite-difference methods.[9] This scheme did not prove to be straightforward in converging to a stable solution due to the complex nature of the surface boundary condition, equation 10. The problem for the potential at the surface can also be expressed (see Appendix I) in terms of the integral equation,

$$\bar{\phi}_o(\bar{\eta}) = \frac{1}{\pi}\int_0^\infty \ln\left|\bar{\eta}_*^2 - \bar{\eta}^2\right| \left[\frac{\bar{\eta}_*}{2}\,\bar{\phi}_o(\bar{\eta}_*) - \frac{\bar{\eta}_*^2}{2}\left.\frac{\partial\bar{\phi}}{\partial\bar{\eta}}\right|_{\substack{\bar{\xi}=0\\ \bar{\eta}=\bar{\eta}_*}} - 1\right] d\bar{\eta}_* . \tag{14}$$

In order to bring the problem into a finite domain, the integral can be written in the form

$$\begin{aligned}\bar{\phi}_o = \frac{1}{\pi}\int_0^{\eta_s} \ln\left|\bar{\eta}_*^2 - \bar{\eta}_*^2\right| \left(\frac{\bar{\eta}_*}{2}\,\bar{\phi}_o - \frac{\bar{\eta}_*^2}{2}\frac{\partial\bar{\phi}_o}{\partial\bar{\eta}_*} - 1\right) d\bar{\eta}_* \\ + \frac{1}{2\pi}\int_0^{1/\eta_s^4} \ln\left|\frac{1}{\sqrt{x_*}} - \frac{1}{\sqrt{x}}\right| \frac{1}{x_*^{3/4}}\frac{\partial\bar{\phi}_o'}{\partial x_*}\, dx_*,\end{aligned} \tag{15}$$

where

$$\bar{\phi}_o' = (\bar{\phi}_o - 1/\bar{\eta})/\bar{\eta} \ , \ x = 1/\bar{\eta}^4 \ , \tag{16}$$

and η_s is a convenient breaking point. Equation 15 can now be integrated for the entire range of $\bar{\eta}$, whereas equation 14 has to be truncated at some point with the possibility of neglecting an important contribution to the integral. Numerical solution of this integral equation [10] turned out to be more efficient than the finite difference technique in obtaining results for the present problem.

The results are shown in figure 1. The shape of the curves is also characteristic of the current distribution near the edge since the dimensionless potential $\bar{\phi}_o$ is proportional to the current density as indicated by equation 13. The current distribution approaches the primary distribution toward the center of the disk. However, it remains finite and much more uniform than the primary distribution in the edge region; the finite capacity of the electric double layer does not allow it to become infinite.

The electrode potential for the galvanostatic case or the applied current for the potentiostatic case is given by

$$\left.\begin{array}{l} \text{(galvanostatic) } V/\Phi_o^p - 1 \\ \\ \text{(potentiostatic) } 1 - I/4r_o\kappa\Phi_o^p \end{array}\right\} = \int_0^1 \left(\frac{V - \Phi_o}{\Phi_o^p}\right) d\eta \ . \tag{17}$$

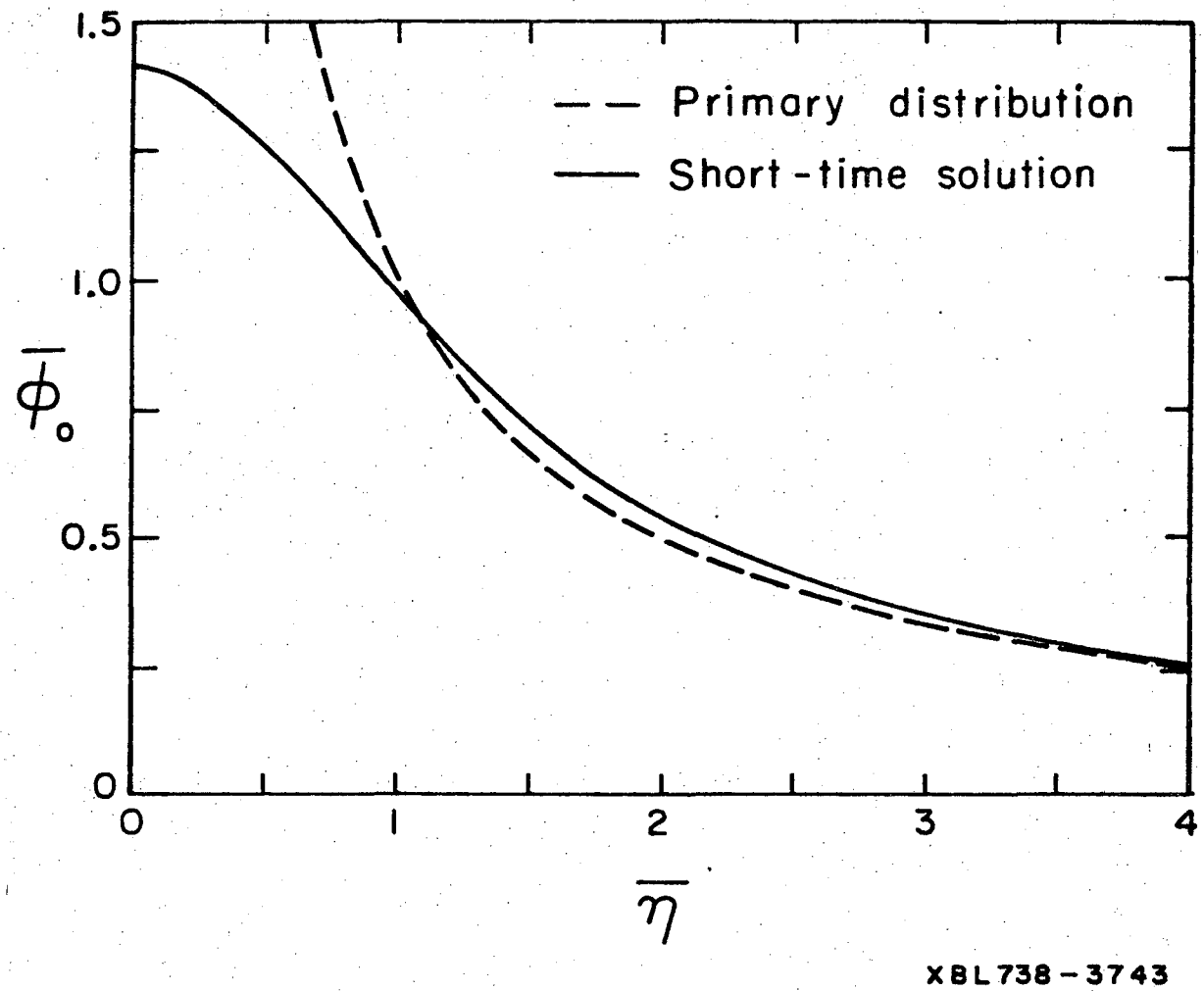

Figure 1. The surface potential distribution at short times near the edge of a disk electrode

Substitution of the stretched variables and integration yields

$$\int_0^1 \left(\frac{V - \Phi_o}{\Phi_o^p} \right) d\eta = - \frac{1}{\pi} \theta \, \ell n \, \theta + A\theta \, , \tag{18}$$

where[5]

$$A = \frac{2}{\pi} \left\{ \int_0^b \bar{\Phi}_o d\bar{\eta} + \int_b^\infty \left(\bar{\Phi}_o - \frac{1}{\bar{\eta}} \right) d\bar{\eta} - \ell n \, b \right\} . \tag{19}$$

The integral is broken up this way as a convenience in numerical calculation since the first integral does not converge as $b \to \infty$ and the second integral as $b \to 0$. Equation 19 was integrated[10] for the whole range of $\bar{\eta}$ using the numerical solution obtained from equation 15, and the value of A was thus found to be 0.841 . Equation 18 is plotted in figure 2 using this value and compared to previous calculations.[1,2] The long-time series approach the present short-time solution as an asymptote for small values of θ .

Conclusions

The present work demonstrates once more the efficiency and convenience of the singular-perturbation method in obtaining asymptotic solutions to problems which would otherwise be laborious to solve numerically. In electroanalytical applications, one usually focuses attention to the conditions at the electrode surface, and the conditions in the bulk can be accounted for adequately by using the expression

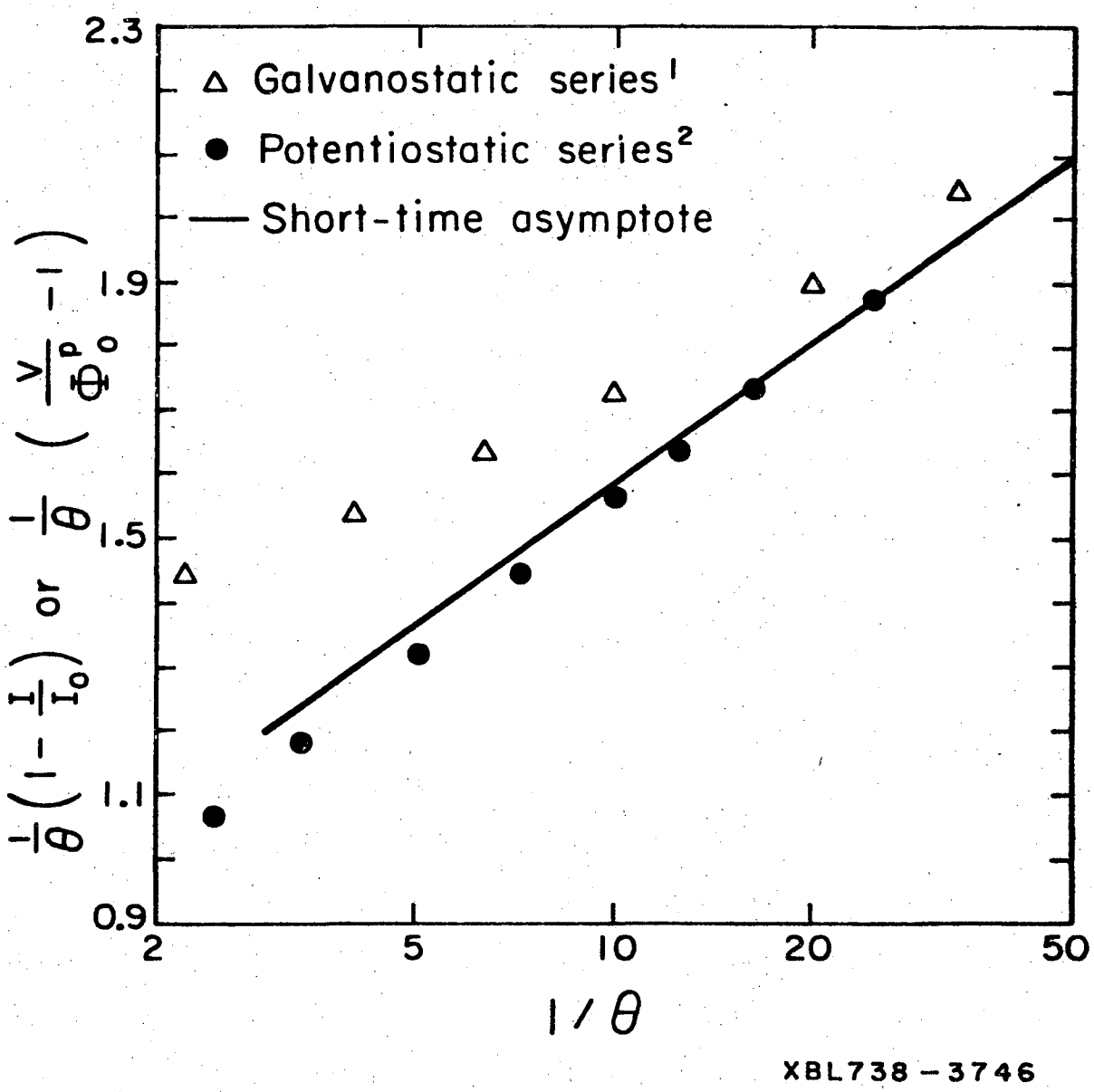

Figure 2. Comparison of the short-time solution and the longtime series[1,2] for the transient response of a disk

for the primary distribution, equation 4. Therefore, expressing the surface potential in terms of an integral equation provides additional economy in the numerical work since the bulk of the solution does not enter the calculations explicitly.

The results reported above are universally applicable since no parameters appear in the problem, and numerical calculations need not be repeated. Furthermore, the results are independent of the disk geometry because the formulation is confined to a small region near the edge. Hence, they can also be made to apply under similar conditions to any electrode geometry embedded in an insulating plane.

APPENDIX I

Integral Representation for the Potential at Short Times

It is possible to solve Laplace's equation by superposing solutions due to current sources on an electrode. For a disk, this is given by[11]

$$\Phi(r,z) = \frac{2}{\pi\kappa} \int_0^{r_o} \frac{i(r_*)K(m)r_* dr_*}{\sqrt{z^2 + (r + r_*)^2}} \tag{20}$$

where $K(m)$ is the complete elliptic integral of the first kind,[12]

$$K(m) = \int_0^{\pi/2} \frac{d\alpha}{\sqrt{1 - m \sin^2 \alpha}} , \tag{21}$$

and

$$m = \frac{4rr_*}{z^2 + (r + r_*)^2} . \tag{22}$$

After converting into the rotational elliptic coordinates[3] and substituting equation 3, one obtains

$$\frac{\Phi_o}{\Phi_o^p} = \frac{2}{\pi} \int_0^1 \frac{K(m)}{1 + \sqrt{1 - \eta_*^2}} \frac{\partial}{\partial\theta} \left(\frac{V - \Phi_o}{\Phi_o^p} \right) \eta_* d\eta_* , \tag{23}$$

where the assumption $\eta << 1$ has been introduced for the edge region. The assumption cannot be applied to the dummy variable η_* because the integration has to be carried over the entire surface of the disk.

Define

$$F(\eta,\eta') = \frac{2}{\pi} \int_0^{\eta'} \frac{K(m)}{1 + \sqrt{1 - \eta_*^2}} \frac{\partial}{\partial\theta} \left(\frac{V - \Phi_o}{\Phi_o^p} \right) \eta_* d\eta_* . \tag{24}$$

The potential distribution in the outer region is approximated by equation 5. Substitution into equation 3 and integration gives

$$\frac{V - \Phi_o}{\Phi_o^p} = \frac{2}{\pi} \frac{\theta}{\eta} . \tag{25}$$

Hence, for the outer region, equation 24 can be written as

$$\tilde{F}(\eta,\eta') = F(\eta,1) - \frac{4}{\pi^2}\left[\int_0^1 \frac{K(m)}{1+\sqrt{1-\eta_*^2}}\,d\eta_* - \int_0^{\eta'} \frac{K(m)}{1+\sqrt{1-\eta_*^2}}\,d\eta_*\right] . \quad (26)$$

The first integral in the brackets corresponds to the primary distribution. Proof:

Let us write equation 20 for the primary distribution by replacing Φ with Φ^p and i with the primary current density:[3]

$$\Phi_o^p = I/4\kappa r_o = \frac{2}{\pi\kappa}\int_0^{r_o} \frac{I}{\pi r_o^2}\,\frac{0.5}{\sqrt{1-(r_*/r_o)^2}}\,\frac{K(m)r_*dr_*}{r+r_*}$$

$$= \frac{4\Phi_o^p}{\pi^2}\int_0^1 \frac{K(m)}{\sqrt{1-\eta^2}+\sqrt{1-\eta_*^2}}\,d\eta_* \quad . \quad (27)$$

Thus, independent of the value of η, we have

$$\frac{4}{\pi^2}\int_0^1 \frac{K(m)}{\sqrt{1-\eta^2}+\sqrt{1-\eta_*^2}}\,d\eta_* = 1 \ . \quad (28)$$

For small values of η', the second integral in equation 26 can be approximated by

$$\int_0^{\eta'} \frac{K(m)}{1+\sqrt{1-\eta_*^2}}\,d\eta_* \approx \int_0^{\eta'} \ln\frac{4}{\eta}\,d\eta = \eta'\left(\ln\frac{4}{\eta'}+1\right) \ . \quad (29)$$

Equation 26 for the outer region therefore reduces to

$$\tilde{F}(\eta,\eta') = F(\eta,1) - 1 + \frac{4\eta'}{\pi^2}\left(\ln\frac{4}{\eta'} + 1\right) . \tag{30}$$

In the inner region, equation 24 can be expressed in the form

$$\bar{F}(\eta,\bar{\eta}') = \frac{\sqrt{\theta}}{\pi^2}\int_0^{\bar{\eta}'} K(m)\left[\bar{\phi}_o(\bar{\eta}_*) - \bar{\eta}_*\left.\frac{\partial\bar{\phi}_o}{\partial\bar{\eta}}\right|_{\bar{\eta}=\bar{\eta}_*}\right]\bar{\eta}_* d\bar{\eta}_* . \tag{31}$$

Both η and η_* are small in the inner region. Therefore,

$$\lim_{\eta,\eta_*\to 0} K(m) = \lim_{\eta,\eta_*\to 0}\frac{1}{2}\ln\frac{16}{1-m} = \ln\frac{16/\theta}{|\bar{\eta}^2 - \bar{\eta}_*^2|} . \tag{32}$$

Equation 31 can now be broken up into several parts:

$$\bar{F}(\bar{\eta},\bar{\eta}') = \frac{2\sqrt{\theta}}{\pi^2}\left\{\int_0^\infty \ln\,(16/\theta)G(\bar{\eta}_*)d\bar{\eta}_* - \int_0^\infty \ln\,|\bar{\eta}_*^2 - \bar{\eta}^2|\;G(\bar{\eta}_*)d\bar{\eta}_* - \int_{\bar{\eta}'}^\infty \ln\frac{16/\theta}{|\bar{\eta}_*^2 - \bar{\eta}^2|}\,G(\bar{\eta}_*)d\bar{\eta}_* + \int_0^{\bar{\eta}'} \ln\frac{16/\theta}{|\bar{\eta}_*^2 - \bar{\eta}^2|}\,d\bar{\eta}_*\right\} , \tag{33}$$

where

$$G(\eta_*) = \frac{\bar{\eta}_*}{2}\,\bar{\phi}_o(\bar{\eta}_*) - \frac{\bar{\eta}_*^2}{2}\left.\frac{\partial\bar{\phi}_o}{\partial\bar{\eta}}\right|_{\bar{\eta}=\bar{\eta}_*} - 1 \,. \tag{34}$$

In the original unstretched coordinate system, the first integral can be expressed as

$$\begin{aligned}
\ln\,(16/\theta)\int_0^\infty G(\bar{\eta}_*)d\bar{\eta}_* &= -\frac{\ln\,(16/\theta)}{\sqrt{\theta}}\int_0^1\left(\frac{\pi}{2\Phi_o^p}\left.\frac{\partial\Phi}{\partial\xi}\right|_{\xi=0} + 1\right)d\eta \\
&= \frac{\ln\,(16/\theta)}{\sqrt{\theta}}\left(\frac{1}{4\kappa r_o\Phi_o^p}\int_0^{r_o} 2\pi i r dr - 1\right) \\
&= \frac{\ln\,(16/\theta)}{\sqrt{\theta}}\left(\frac{I}{4\kappa r_o\Phi_o^p} - 1\right) .
\end{aligned} \tag{35}$$

Under galvanostatic control, $I = 4\kappa r_o\Phi_o^p$, and the integral is identically zero. For potentiostatic control, the current is given by equation 18, and the integral is therefore of order $\sqrt{\theta}$ and still small compared to the second integral, which is of order unity. For large $\bar{\eta}'$, the last integral in equation 33 can be approximated by

$$\int_0^{\bar{\eta}'} \ln\frac{16/\theta}{|\bar{\eta}_*^2 - \bar{\eta}^2|}\,d\bar{\eta}_* \approx \bar{\eta}'\,\ln\frac{16}{\theta} - \bar{\eta}'\,\ln\bar{\eta}'^2 + 2\bar{\eta}' \,. \tag{36}$$

Equation 33 for the inner region thus becomes at large $\bar{\eta}'$

$$\bar{F}(\bar{\eta},\bar{\eta}') = -\frac{2\sqrt{\theta}}{\pi^2}\left\{\int_0^\infty \ln\,|\bar{\eta}_*^2 - \bar{\eta}^2|\; G(\bar{\eta}_*)d\bar{\eta}_*\right.$$

$$+\int_{\bar{\eta}'}^\infty \ln\frac{16/\theta}{|\bar{\eta}_*^2 - \bar{\eta}^2|}\, G(\bar{\eta}_*)d\bar{\eta}_* \tag{37}$$

$$\left. - \bar{\eta}'\,\ln\frac{16}{\theta} + \bar{\eta}'\,\ln\bar{\eta}'^2 - 2\bar{\eta}'\right\}.$$

Finally, the matching condition,

$$\lim_{\eta'\to 0}\tilde{F}(\eta,\eta') = \lim_{\bar{\eta}'\to 0}\bar{F}(\bar{\eta},\bar{\eta}') \tag{38}$$

has to be satisfied. Substitution of equations 30 and 37 gives, after cancelling the matching terms,

$$\bar{\phi}_o(\bar{\eta}) = \frac{1}{\pi}\int_0^\infty \ln\,|\bar{\eta}_*^2 - \bar{\eta}^2|\; G(\bar{\eta}_*)d\bar{\eta}_*\,, \tag{39}$$

which is identical to equation 14.

APPENDIX II

Current Distribution on a Disk Electrode at Large Exchange-Current Densities

Previous numerical calculations for the rotating disk[6] have shown that the secondary current distribution approaches the primary distribution as the exchange-current density goes to infinity. For

large but finite exchange current densities, the current distribution approximates closely the primary distribution on the disk except near the edge, where the faradaic impedance is large enough to force the current density to remain finite.

When the current density is small compared to the exchange-current density $(i \ll |i_o|)$, one may assume linear kinetics.[6] The dimensionless, exchange-current density can then be defined as

$$J = (\alpha_a + \alpha_c) \frac{i_o r_o F}{RT\kappa} , \tag{40}$$

where α_a and α_c are the kinetic parameters of the general kinetic expression for an electrode reaction.[11] The condition at the electrode surface can be expressed by

$$i = -\kappa \left.\frac{\partial\Phi}{\partial z}\right|_{z=0} = J(V - \Phi_o) \qquad (r \leq r_o) . \tag{41}$$

The stretched variables appropriate for this problem are

$$\bar{\phi} = \frac{\pi}{2}\left(1 - \frac{\Phi}{V}\right)\sqrt{J} , \tag{42}$$

$$\bar{\eta} = \eta\sqrt{J} , \quad \bar{\xi} = \xi\sqrt{J} . \tag{43}$$

Substitution into Laplace's equation and the boundary conditions gives a system identical to equations 9, 11, and 12, and the condition at the surface becomes

$$\left.\frac{\partial\bar{\phi}}{\partial\bar{\xi}}\right|_{\bar{\xi}=0} = \bar{\eta}\bar{\phi} \ . \tag{44}$$

The finite-difference method by successive overrelaxations was successful in this case in obtaining a stable solution. The results are plotted in figure 3.

The steady-state current or potential can be calculated from

$$1 - \frac{\Phi_o^p}{V} = 1 - \frac{I}{4r_o\kappa V} = \int_0^1 \left(1 - \frac{\Phi_o}{V}\right) d\eta = \frac{1}{\pi J} \ln J + \frac{A}{J} \ , \tag{45}$$

where A has the same form as equation 19 and the numerical value of 0.708. The quantity V/Φ_o^p can be characterized as a dimensionless, effective direct-current resistance[5] $4r_o\kappa R_{eff}$ and is fixed once J is specified. A few values were reported earlier,[1,5] and these are reproduced in table 1 along with some additional results. These results are compared with the present asymptotic solution in figure 4.

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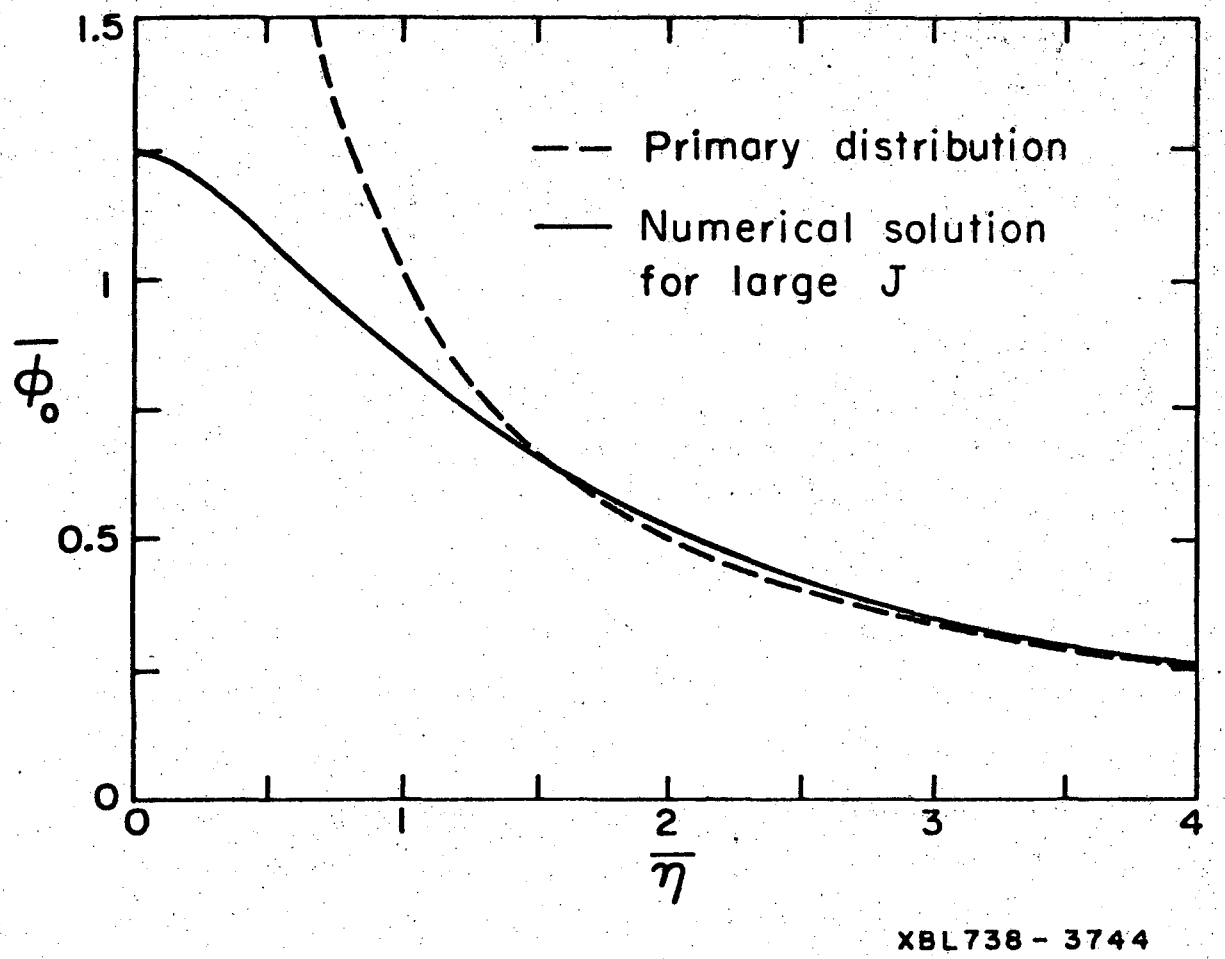

Figure 3. The surface potential distribution for large values of the kinetic parameter J near the edge of a disk electrode

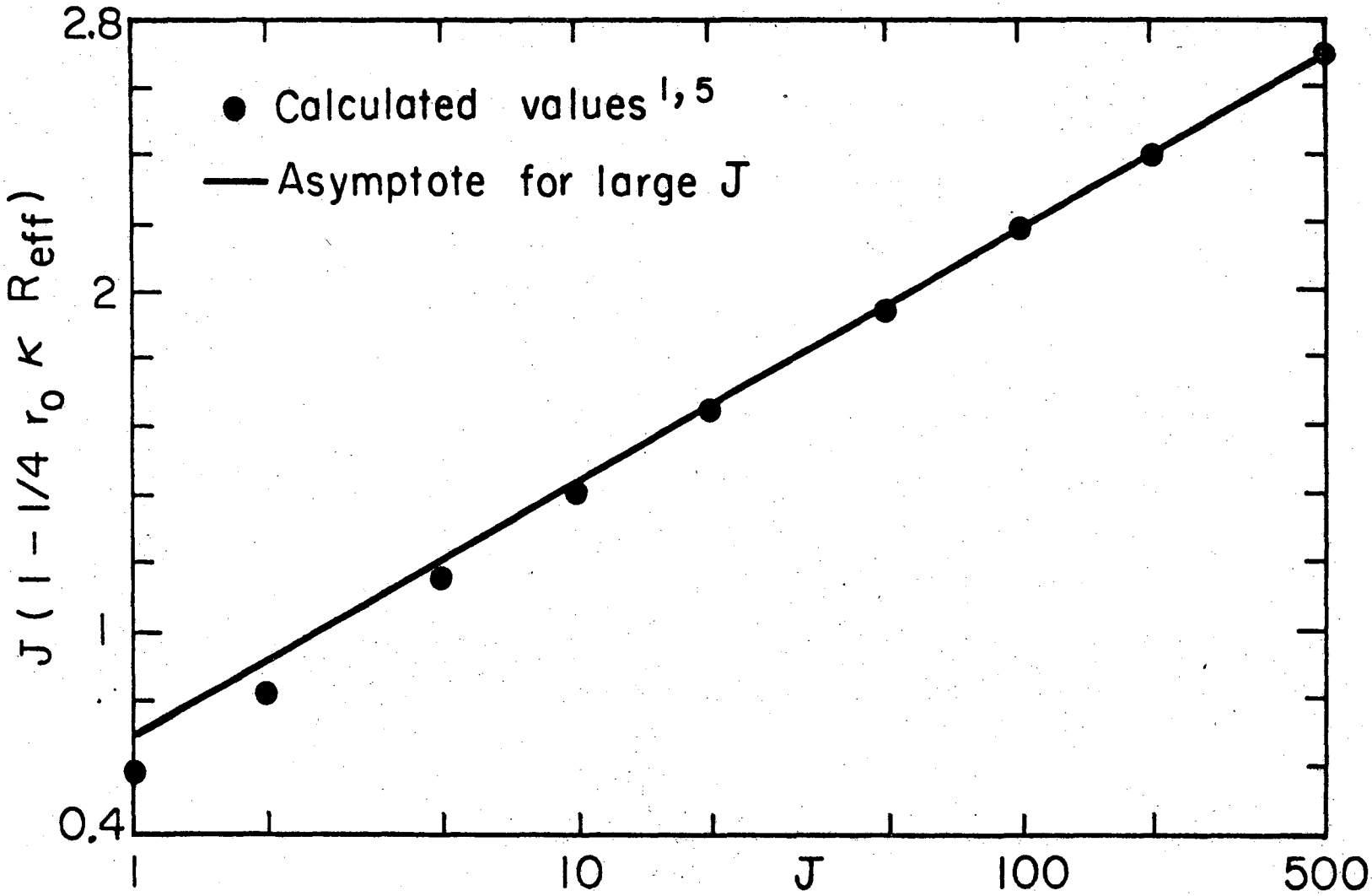

Figure 4. The dependence of the effective direct-current resistance on the kinetic parameter J

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Table 1. The effective cell resistance at different values of the parameter J

J	$4r_o \kappa R_{eff}$
0.1	13.81194
0.2	7.44458
0.5	3.62161
1	2.34368
2	1.69962
5	1.30375
10	1.16459
20	1.09002
50	1.04072
100	1.02231
200	1.01217
500	1.00543

Nomenclature

A	constant defined in equation 18
C	double-layer capacity, f/cm^2
F	Faraday's constant, coulomb/equivalent
$F(\eta,\eta')$	function defined by equation 23
$G(\bar{\eta}_*)$	function defined by equation 33
i	current density, A/cm^2
I	total current, A
J	dimensionless parameter for linear electrode kinetics
$K(m)$	complete elliptic integral of the first kind
r	radial position from axis of disk, cm
r_o	radius of disk, cm
R_{eff}	effective direct-current resistance, ohm
t	time, sec
V	electrode potential, V
z	distance from plane of disk, cm
κ	conductivity of the solution, $ohm^{-1} - cm^{-1}$
η	rotational elliptic coordinate
$\bar{\phi}$	dimensionless potential in the solution
Φ	potential in the solution, V
Φ_o	potential in the solution adjacent to the electrode, V
Φ^p	potential in the solution corresponding to the primary distribution, V
ξ	rotational elliptic coordinate
θ	dimensionless time

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