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Simulation of the Dropping Mercury Electrode by Orthogonal Collocation.

By

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Orthogonal collocation techniques are used to simulate the solution to the hydrodynamic partial differential equations describing mass transport to a dropping mercury electrode (DME). Accurate values for the concentration profiles and current are obtained with minimal computational effort.

Simulation of the Dropping Mercury Electrode by Orthogonal Collocation

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The weighted residual method of orthogonal collocation has been used to generate accurate solutions to electrochemical diffusion problems (1-11). The purpose of this work is to demonstrate the utility of the method by application directly to a differential equation containing convective terms. The case considered here is the diffusive and convective transport of an electroactive species to the dropping mercury electrode (DME).

In terms of radial distance r from the center of the mercury drop, the hydrodynamic equation describing the concentration of the electroactive species in time is given (12) by

$$\frac{\delta c}{\delta t} = D\frac{\delta^2 c}{\delta r^2} + \frac{2D}{r}\frac{\delta c}{\delta r} - v_r\frac{\delta c}{\delta r} \qquad [1]$$

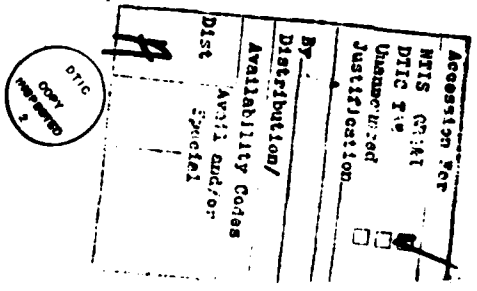
where v_r is the velocity of a point in solution toward the growing mercury drop. The radius r_0 of the drop grows as

$$r_0 = \left(\frac{3mt}{4\pi d}\right)^{1/3} \qquad [2]$$

where m is the flow rate and d the density of mercury. Considering only convection, a point r in solution at t=0 is pushed outwards. As volume must be conserved, its movement is described by

$$r = \left(\frac{3m(t+t')}{4\pi d}\right)^{1/3} \qquad [3]$$

where t' is the time it would take for a sphere of radius r' to be formed at a flow rate m.



The velocity of this point is thus given by

$$V_r = \frac{dr}{dt} = \frac{\left(\frac{3m}{4\pi d}\right)}{3\left(\frac{3m}{4\pi d}t+t'\right)^{2/3}} = \frac{\left(\frac{3m}{4\pi d}\right)}{3r^2} \qquad [4]$$

We wish to normalize the distance coordinate to a [0,1] space where 0 represents the electrode surface, and 1 represents a point in solution where no diffusional processes occur during the experiment. We denote this real point as M. Its movement is given by an equation analogous to [3]:

$$M = \left(\frac{3m(t+t'')}{4\pi d}\right)^{1/3} \qquad [5]$$

where t'' is the time needed to grow a sphere of radius M. The space between the electrode surface and M then is used in normalization.

It is seen that such a space is time dependent, i.e. the space will diminish in time. This is physically observed as the decrease in diffusion layer thickness resulting in an increase in current at constant potential. For our analysis, it is desired to eliminate this time dependency of the distance coordinate. This is done by observing that the difference in the cubes of the quantities M and r_0 is time independent.

$$M^3 - r_0^3 = \frac{3mt''}{4\pi d} \qquad [6]$$

A point in solution will move with respect to the growing mercury drop surface as well. We can refer to a point fixed in the M^3 -

r_0^3 space as R if R is defined as

$$R = \frac{r - r_0}{(M^3 - r_0^3)^{1/3}} \quad [7]$$

where r is the radial distance from the center of the drop to the point in question.

We define our dimensionless time variable T as

$$T = \frac{tD}{(M^3 - r_0^3)^{2/3}} \quad [8]$$

and a constant β as

$$\beta = \frac{a^3}{D(M^3 - r_0^3)^{1/3}} \quad [9]$$

where a is given by

$$a = \left(\frac{3m}{4\pi d}\right)^{1/3} \quad [10]$$

We need now to perform the transformation of the differential equation [1] to dimensionless coordinates. In terms of the new coordinates and constants, we find that

$$t = \frac{a^6 T}{D^3 \beta^2} \quad [11]$$

and

$$r = \frac{Ra^3}{\beta D} + \frac{a^3 T^{1/3}}{\beta^{2/3} D} \quad [12]$$

For a function $w = f(x,y)$ which has continuous real first and second partial differentials, we wish to be able to transform to the new variables u and v where u and v are given by

$$x = \phi(u,v) \quad [13]$$

and

$$y = \phi(u,v) \quad [14]$$

The resulting first partial derivatives are given by

$$\frac{\delta w}{\delta u} = \frac{\delta w}{\delta x}\frac{\delta \phi}{\delta u} + \frac{\delta w}{\delta y}\frac{\delta \phi}{\delta u} \quad [15]$$

$$\frac{\delta w}{\delta v} = \frac{\delta w}{\delta x}\frac{\delta \phi}{\delta v} + \frac{\delta w}{\delta y}\frac{\delta \phi}{\delta v} \quad [16]$$

If $w(x,y)$ corresponds to $c(r,t)$, and ϕ corresponds to $\frac{a^6 T}{\beta^2 D^3}$ and ϕ corresponds to $\frac{a^3 R}{\beta D} + \frac{a^3 T^{1/3}}{\beta^{2/3} D}$, then we have

$$\frac{\delta c}{\delta R} = \frac{\delta c}{\delta r}\frac{\delta}{\delta R}\left(\frac{a^3 R}{\beta D} + \frac{a^3 T^{1/3}}{\beta^{2/3} D}\right) + \frac{\delta c}{\delta t}\frac{\delta}{\delta R}\left(\frac{a^6 T}{\beta^2 D^3}\right) \quad [17]$$

and

$$\frac{\delta c}{\delta T} = \frac{\delta c}{\delta r}\frac{\delta}{\delta T}\left(\frac{a^3 R}{\beta D} + \frac{a^3 T^{1/3}}{\beta^{2/3} D}\right) + \frac{\delta c}{\delta t}\frac{\delta}{\delta T}\left(\frac{a^6 T}{\beta^2 D^3}\right) \quad [18]$$

Thus

$$\frac{\delta c}{\delta R} = \frac{a^3}{\beta D}\frac{\delta c}{\delta r} \quad ; \quad \frac{\delta c}{\delta r} = \frac{\beta D}{a^3}\frac{\delta c}{\delta R} \qquad [19]$$

$$\frac{\delta c}{\delta t} = \frac{a^3}{3\beta^{2/3}DT^{2/3}}\frac{\delta c}{\delta t} + \frac{a^6}{\beta^2 D^3}\frac{\delta c}{\delta t} \quad ; \quad \frac{\delta c}{\delta t} = \frac{\beta^2 D^3}{a^6}\frac{\delta c}{\delta T} - \frac{\beta^{4/3}D^2}{3a^3T^{2/3}}\frac{\delta c}{\delta r} \qquad [20]$$

Combining [19] and [20], we have

$$\frac{\delta c}{\delta t} = \frac{\beta^2 D^3}{a^6}\frac{\delta c}{\delta T} - \frac{\beta^{7/3}D^3}{3a^6T^{2/3}}\frac{\delta c}{\delta R} \qquad [21]$$

The second partial with respect to the distance coordinate is obtained by recognizing that

$$\frac{\delta^2 c}{\delta r^2} = \frac{\delta}{\delta r}\left(\frac{\delta c}{\delta r}\right) = \frac{\delta}{\delta r}\left(\frac{\beta D}{a^3}\frac{\delta c}{\delta R}\right) \qquad [22]$$

It can be shown that

$$\frac{\delta}{\delta r} = \frac{\beta D}{a^3}\frac{\delta}{\delta R} \qquad [23]$$

so that we now have

$$\frac{\delta^2 c}{\delta r^2} = \frac{\beta^2 D^2}{a^6}\frac{\delta^2 c}{\delta R^2} \qquad [24]$$

Equations [12], [19], [21], and [24] are substituted into the differential equation to get

$$\frac{\beta^2 D^3}{a^6}\frac{\delta c^*}{\delta T} - \frac{\beta^{7/3}D^3}{a^6 T^{2/3}}\frac{\delta c^*}{\delta R} = \frac{\beta^2 D^3}{a^6}\frac{\delta^2 c^*}{\delta R^2} + \left[\frac{2D^3}{\frac{Ra^3}{\beta D} + \frac{a^3 T^{1/3}}{\beta^{2/3}D}} - \frac{a^3}{3\left(\frac{Ra^3}{\beta D} + \frac{a^3 T^{1/3}}{\beta^{2/3}D}\right)^2}\right]\frac{\beta D}{a^3}\frac{\delta c^*}{\delta R} \qquad [25]$$

where

$$c^* = \frac{c}{c^b} \qquad [26]$$

c^b being the bulk concentration of substrate. Equation [25] is readily reduced to

$$\frac{\delta c^*}{\delta T} = \frac{\delta^2 c^*}{\delta R^2} + \left[\frac{2}{R+\beta^{1/3}T^{1/3}} + \frac{\beta^{1/3}}{3T^{2/3}} - \frac{\beta}{3(R+\beta^{1/3}T^{1/3})^2}\right]\frac{\delta c^*}{\delta R} \qquad [27]$$

DISCRETIZATION OF THE DIFFERENTIAL EQUATION

Accurate solutions of the differential equation [27] are obtained at the roots of shifted Jacobi polynomials on the (0,1) interval as a function of time by orthogonal collocation (1,3,11). In terms of the collocation coefficients A_{ij} and B_{ij} (functions of the roots of the polynomial only as previously defined) we have that, at the roots R_i of the polynomial,

$$\left.\frac{dc^*}{dR}\right|_{R_i} = \sum_{j=1}^{N+2} A_{ij}\, c^*(R_j,T) \qquad [28]$$

and

$$\left.\frac{d^2c^*}{dR^2}\right|_{R_i} = \sum_{j=1}^{N+2} B_{ij}\, c^*(R_j,T) \qquad [29]$$

where N is the order of the polynomial. N+2 terms are taken in order to include the electrode surface (R=0) and the point N in solution where no diffusion occurs (R=1).

We take as an experiment the imposition of a constant potential to the electrode surface such that $c^*(0,T)$ is equal to zero during the growth of the drop. The mechanism is simple single electron transfer. We refer to the concentration of the substrate only.

We thus recognize that

$$c^*(0,T) = 0 \qquad [30]$$

and

$$c^*(1,T) = 1 \qquad [31]$$

such that

$$\left.\frac{dc^*}{dR}\right|_{R_i} = A_{i,N+2} + \sum_{j=2}^{N+1} A_{ij}\, c^*(R_j,T) \qquad [32]$$

and

$$\left.\frac{d^2c^*}{dR^2}\right|_{R_i} = B_{i,N+2} + \sum_{j=2}^{N+1} B_{ij}\, c^*(R_j,T) \qquad [33]$$

These differentials, now referring to explicit points in simulation space, reduce the partial differential equation to the following set of N first order ordinary differential equation in N unknowns which may be solved simultaneously for the $c(R_j,T)$ (1,3,11)

$$\left.\frac{dc^*}{dT}\right|_{R_i} = B_{i,N+2} + \sum_{j=2}^{N+1} B_{ij}\, c^*(R_j,T) + \left[\frac{2}{R+\beta^{1/3}\tau^{1/3}} + \frac{\beta^{1/3}}{3\tau^{2/3}} - \frac{\beta}{3(R+\beta^{1/3}\tau^{1/3})^2}\right]\left[A_{i,N+2} + \sum_{j=2}^{N+1} A_{ij}\, c^*(R_j,T)\right] \qquad [34]$$

THE SIMULATED CURRENT

The current is given by Fick's first law:

$$i = nFAD\left(\frac{\delta c}{\delta r}\right)_{r_0} \qquad [35]$$

where A is the area of the DME

$$A = 4\pi r_0^2 \qquad [36]$$

Substitution of [4] and [19] into [35] and simplifying, results, in terms of the dimensionless parameters,

$$i = 4\pi nFc^b a^3 \beta^{-1/3}\tau^{2/3}\left(\frac{\delta c^*}{\delta R}\right)_{R=0} \qquad [37]$$

The differential is immediately discretized, considering the

boundary conditions $c^*(0,T) = 0$ and $c^*(1,f) = 1$, to

$$i = 4\pi nFc^b m^{2/3}\beta^{-1/3}T^{2/3}\left[A_{i,N+2} + \sum_{j=2}^{N+1} A_{ij}\, c^*(R_j,T)\right] \quad [38]$$

$$= 21.39\ nc^b m\beta^{-1/3}T^{2/3}\left[A_{i,N+2} + \sum_{j=2}^{N+1} A_{ij}\, c^*(R_j,T)\right] \quad [39]$$

The current is calculated in the output subroutine of the program.

RESULTS

The simulated current is compared to the solution offered by Koutecky (14) which is corrected for spherical diffusion

$$i_d = 708\ n\ c^b m^{2/3} D^{1/2} t^{1/6}(1+34.7\ D^{1/2}t^{1/6}m^{-1/3}+100\ Dt^{1/3}m^{-2/3}) \quad [40]$$

Again, equation [11] is used to convert to dimensionless time

$$i = 708\ nc^b m\ T^{1/6}\beta^{-1/3}\left(\frac{3}{4\pi d}\right)^{1/3}\left(1 + 34.7\left(\frac{3}{4\pi d}\right)^{1/3}T^{1/6}\beta^{-1/3} + 100\left(\frac{3}{4\pi d}\right)^{2/3}T^{1/3}\beta^{-2/3}\right) \quad [41]$$

$$= 18.43\,nc^b m\ T^{1/6}\beta^{-1/3}(1+0.902\ T^{1/6}\beta^{-1/3}+0.0678\ T^{1/3}\beta^{-2/3}) \quad [42]$$

We may also compare the current to the Ilkovic equation

$$i = 708\ nD^{1/2}c^b m^{2/3}t^{1/6} \quad [43]$$

which is converted by the same substitutions to

$$i = 708\ n\ c^b m\left(\frac{3}{4\pi d}\right)^{1/3}T^{1/6}\beta^{-1/3} \quad [44]$$

$$= 18.43\,nc^b m^{2/3}T^{1/6}\beta^{-1/3} \quad [45]$$

Values for the parameters for a typical Teal case were chosen and the results are given in Figure 2.

COMPUTATIONAL ASPECTS

Results accurate to 0.4 % of Koutecky's calculated values are obtained with minimal computational effort. Using 6 solution points (collocation points; polynomial roots) 1000 current values and concentration profiles are generated by the program in less than 0.5 seconds using an Amdahl 470-V6 computer. The program, which has been described (4,13), is modular in design. Switching from the problem described herein to simple linear diffusion requires the changing of only 2 program statements. Such minimal changing of the program is retained with all other electrochemical modeling problems.

CONCLUSIONS

Orthogonal collocation may be adapted in a straightforward manner to provide accurate solutions to hydrodynamic problems in electrochemistry. The problem herein discussed describes only

the first drop in an actual experiment and useful data may only be obtained from subsequent drops which apparently encounter reproducible concentration perturbations of the reaction layer. These perturbations are due to convective transport initiated by the first drop. We are preparing the simulation treatment for this case, as well as those describing the current obtained when following chemical reactions and adsorption are involved in the overall reaction mechanism.

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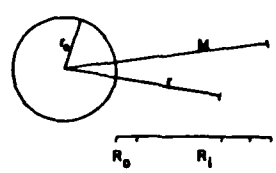

FIGURE LEGENDS

(1) The geometry of the growing spherical mercury drop.

(2) Comparison of (a) simulated results with (b) the Koutecky model and (c) the Ilkovic equation. Parameters: $\beta = 0.35$, $C^b = 10^{-5}$ mol/cm^3, $m = 2$ mg/s.

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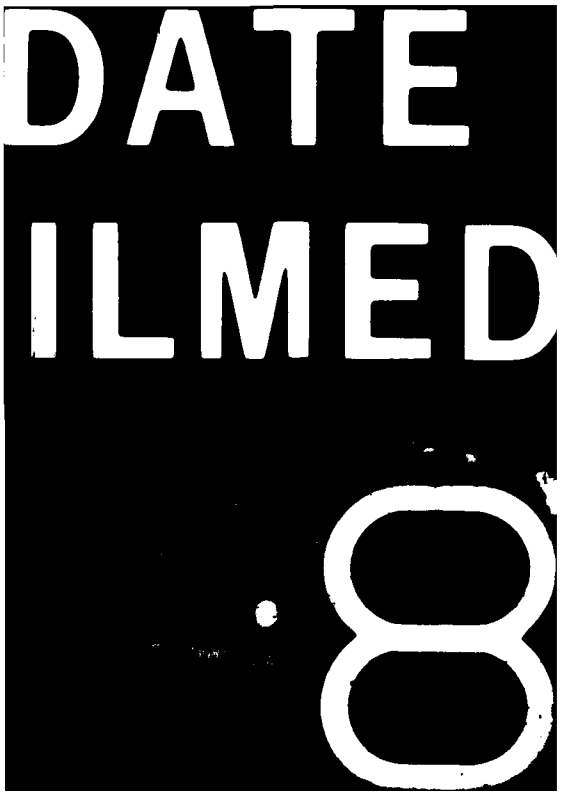
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