

Latest updates on Mathematical Research

Chapter 1

Mathematical Modeling and Reaction-Diffusion Processes at Ultra Microelectrodes

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Abstract

The inimitable belongings of ultramicroelectrodes can be used for shaping thermodynamic and kinetic constraints of electrode progressions. By means of ultramicroelectrode through diverse radii, preceding (CE) and supplement responses of the ECE and EC' nature can be quantitatively investigated. In this chapter, mathematical models for a ultramicroelectrode for the steady and transient states are discussed. Ultramicroelectrode can be modeled with linear and non-linear convection differential equations of EC', EC, and ECE reactions mechanism. The chapter focuses on the behavior of ultramicroelectrodes. We discuss the recent modeling developments (analytical solution) of the concentration produced and current in a ultramicroelectrode from all the electrochemical reactions are reviewed.

Keywords: Ultramicroelectrode; Boundary value problems; Michaelis-Menten kinetics; Analytical Solution.

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1. Introduction

Ultramicroelectrodes (UMEs) are electrodes for which as a minimum one dimension is in the array among $0.1 \mu\text{m}$ and $50 \mu\text{m}$. Ultramicroelectrode are lesser than minor electrodes (microelectrodes) in addition intended for electroanalytical chimsts, convectional electrode is of millimetric dimensions hence span microelectrode is suitable the technique to describe electrodes of micrometric dimensions. In wide-ranging, it remained imagined that conductive polymer film accepts geometry of the hemisphere if electrodeposited scheduled an ultramicrodisc surface. Succeeding this, the microelectrodes stand well-thought-out to deposit on an electrode polymeric hemisphere coated on a disk electrode.

Figure 1 denotes a hemispherical polymer on topmost of a plane, through a line inside insulating plane, in place of conducting wire that arises at the origin of the hemisphere. Following, a short-term response mechanistic model is given in **Figure 2**. Newly, there has been abundant curiosity in the growth of ultramicroelectrodes offer quite a lot of rewards in electrochemical measurements [1] such as lesser interfacial capacitance, slighter time constants, reduced ohmic drop, improved current density, etc. The outcomes of ultramicroelectrodes of several geometries to potential step experiments have been studied through an experiment [2, 3]. Ultramicroelectrodes have been functional in the studies of electrode kinetics, exclusively together with homogeneous kinetic processes [4-6]. An UME is a working electrode used in a three electrode system. The small size of UME gives them comparatively huge diffusion layers and slight total currents. These structures permit UME towards realize suitable steady-state conditions and very high scan rates (V/s) with restricted distortion. UME was established independently by Wightmann and Fleischmann around 1980 [7]. The rewards depending upon electrode geometry may be summed up as follows [8]: (a) A steady state (Disc, Ring, Hemisphere) or quasi-steady state (band, cylinder) current is attained even in a quiescent solution. (b) The steady state current permits to produce chemical and electrochemical kinetic measurements. (c) The current-potential curve with little deformation renders rapid measurements are likely in non-polar solvents or resistive solution without deliberately adding supportive electrolytes. Several kinds of microelectrode arrays have been studied for the design of sensors, micro-discs, bands, squares, inter digitated electrodes. The electrochemical response rest on the figure of the electrodes and shape of the array. Quite a lot of models originated on analytical and numerical solutions also have been delivered to investigate the diffusion process happening at a regular ensemble of micro-disc electrodes [9-18].

Boundary value problems connected through systems of linear and nonlinear ordinary differential equations happen with various divisions of science and engineering. In this problems boundary conditions are mentioned at the end points of the problem domain, and a solution of the differential equation on this area is sought that satisfies the given boundary conditions. Initial boundary value problems in two space dimensions (IBVP2Ds) show an vital

role in electrochemical kinetic studies by means of a variety of transient electroanalytical techniques [19-21] via microelectrodes [22-25].

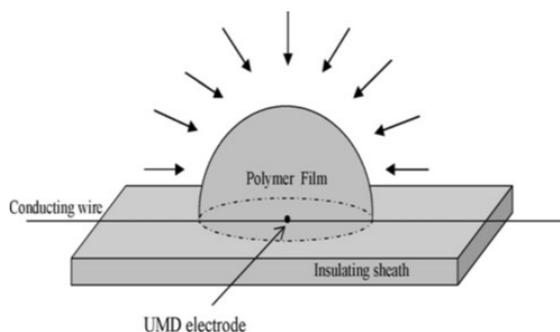


Figure 1: Schematic representations of the geometry adopted through the polymer coated microelectrode adopted by the analytes.

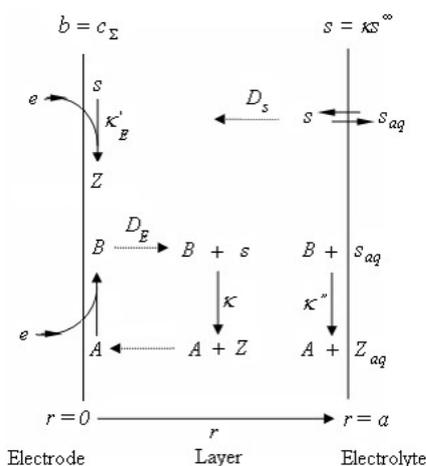


Figure 2: Schematic model and a basic notation of a modified electrode expected diffusion profile.

2. Boundary Value Problems in Ultramicroelectrodes:

Recently Rajendran et. al [26,27] calculated the two point Padé approximation designed for unsteady state chronoamperometric current at ultramicrodisc electrodes and showed that the estimated multidimensional analytical appearance meant for the transient chronoamperometric current for a catalytic electrode reaction (EC' reaction) at a hemispheroidal ultramicroelectrode. Further newly, Rajendran et. al [28] extended this work by calculating the transient chronoamperometric current response at hemispheroidal ultramicroelectrodes.

Rajendran et al. [29-32] (i) proposed two point Pade' approximants for chronoamperometric current at ultramicroelectrodes refer to reversible electron transfer reactions reacting to potential step experiments, moreover reviewed the analytical expressions, then numerical solutions of transient chronoamperometric current at a microring electrode underneath diffusion control, (ii) provided the stable and unstable current for microring electrode for an EC' response, (iii) indicated the analogy of steady state EC' reactions with non-steady state diffusion versions and (iv) obtained transient chronoamperometric current response at hemispheroidal ultramicroelectrodes.

Rajendran et al [33- 35] derived accurate analytical appearance designed for the current by cylindrical electrodes for an EC' reactions for minimum time and slow reaction rate, using Danckwert's presence and reported the transient current for an EC' reaction at a cylindrical electrodes for all that period and overall reaction rate, and also derived an analytical expression of current for steady state in addition non-steady state at polymer-modified electrodes for entire values of saturation parameter α within the polymer film using Danckwerts' expression and compared the steady and non-steady-state currents for entire ultramicroelectrodes(circular disc, circular ring, elliptical disc, elliptical ring, band, hemisphere, hemicylinder, hemi-oblate and hemi-prolate electrodes) for EC -reactions.

Hence, it may be understood that in earlier period, substantial progress was achieved in the understanding of the electrode diffusion process at microring electrodes. This work examined the theory of mass transport at microring electrodes under restricted diffusion reaction and electrochemical reaction. Also non-steady-state current at microring electrodes has been investigated with distinct topics, for example, short-time current expression, long-time current expression and all-time current expression. Moreover, steady state and unsteady state current at microring electrodes for EC' reactions were examined in detail. This work is relevant to the applications of microring electrodes in analytical and molecular electrochemistry. This review could benefit experimental researchers and few chemists with expertise in electrochemical analysis.

3. Analytical Solutions of Ultramicroelectrodes:

The concentration/current at ultramicroelectrodes is controlled by diffusion, convection, and migration. Non-linear phenomena show a crucial part in electrical chemistry and biology (heat and mass transfer, filtration of liquids, diffusion in chemical reactions, etc.). Formerly quite a few decades, several authors mostly give their attention towards the resolution of non-linear equations by using several analytical approaches, for instance Pade approximation method [29-32], the Adomian decomposition method (ADM) [36-38], variational iteration method (VIM) [39], homotopy analysis method (HAM) [40-42], homotopy perturbation method (HPM) [43], reduction of order method [44] and complex inversion formula [45].

4. Analytical Expressions of Current:

The recent contributions to the analytical terminologies of current designed for ultramicroelectrodes for various mechanisms are given in the **Table**.

<p>[44]</p>	<p>polymer-modified ltramicroelectrodes(non- steady-state current)</p>	$\frac{d^2u}{d\rho^2} = \frac{2}{\rho} \frac{du}{d\rho} - \gamma_S uv = \frac{du}{d\tau}$ $\frac{d^2v}{d\rho^2} + \frac{2}{\rho} \frac{dv}{d\rho} - \gamma_E uv = \frac{dv}{d\tau}$ <p>$\tau = 0; u = 0; v = 1$</p> <p>$\rho = 0; v = 0; \frac{du}{d\rho} = 0$</p> <p>$\rho = 1; \frac{dv}{d\rho} = 0; u = 1$</p>	$\Psi = \frac{ia}{nFAD_S k_S^\infty} = \left(\frac{du}{d\rho} \right)_{\rho=1}$ $\Psi = \frac{ia}{nFAD_E C_S} = \left(\frac{dv}{d\rho} \right)_{\rho=0}$ $\psi = \sqrt{\gamma_S \coth(\sqrt{\gamma_S}) - 1 + 2\pi^2 \sum_{n=1}^{\infty} \frac{n^2}{(n^2\pi^2 + \gamma_S)} \exp[-(n^2\pi^2 + \gamma_S)\tau]}$ $\psi = - \left(\frac{dv}{d\rho} \right)_{\rho=0}$ $= - \frac{\gamma_E}{2} \frac{\sqrt{\gamma_E}(1 - \sqrt{\gamma_E}) \exp(\gamma_E)}{2(\sqrt{\gamma_E} \sinh(\sqrt{\gamma_E}) - \cosh(\sqrt{\gamma_E}))}$ $- \gamma_E \pi^4 \sum_{n=1}^{\infty} \frac{(n-1)^4}{((n-1)^2\pi^2 + \gamma_E)} \exp(-n-1^2\pi^2 + \gamma_E)\tau$
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[33]	<p>Transient Chronoamperometric Cylindrical ultramicroelectrode</p>	<p>$A \pm e^- \rightarrow B$ $B + Z \xrightarrow{k} A + \text{products}$</p>	<p>$\frac{\partial C_B}{\partial \tau} = \frac{\partial^2 C_B}{\partial r^2} + \frac{1}{r} \frac{\partial C_B}{\partial r} - K C_B$ Here $K = \frac{k a^2}{D_B}$</p>	<p>$\phi(\tau) = K \int_0^\tau e^{-ku} \phi^0(u) du + e^{-k\tau} \phi^0(\tau) =$ $\left[K \int_0^\tau e^{-ku} \left(\frac{e^{-\sqrt{\pi u}/10}}{\sqrt{\pi u}} + \frac{1}{\ln[(4e^{-\tau u})^{1/2} + e^{5/3}]} \right) du + e^{-k\tau} \left(\frac{e^{-\sqrt{\pi \tau}/10}}{\sqrt{\pi \tau}} + \frac{1}{\ln[(4e^{-\tau})^{1/2} + e^{5/3}]} \right) \right]$ $\phi =$ $= \left[K \int_0^\tau e^{-ku} \left(0.5642u^{-1/2} \exp(-0.1772u^{1/2}) + \frac{1}{\ln[1.4986u^{1/2} + 5.2945]} \right) du \right]$ $+ e^{-k\tau} \left[0.5642\tau^{-1/2} \exp(-0.1772\tau^{1/2}) + \frac{1}{\ln[1.4986\tau^{1/2} + 5.2945]} \right]$</p>
[34]	<p>Thin electrocatalytically active polymer film</p>	<p>$S + C \xrightarrow{K_M} [SC] \rightarrow [PC] \xrightarrow{k_c} P + C'$ $C' \xrightarrow{k'_c} C$</p>	<p>$\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial \chi^2} - \frac{Ku}{1 + \alpha u}, 0 \leq \chi \leq 1$ $u(x, 0) = 0, \left[\frac{\partial u}{\partial \chi} \right]_{\chi=0} = 0, u(1, \tau) = 1$</p>	<p>$y_{ss}(\alpha, K) = (2K[\alpha - \ln(1 + \alpha)])^{1/2} \tan h \left[K^{1/2} \alpha / (1 + \alpha)(2[\alpha - \ln(1 + \alpha)])^{1/2} \right]$ $y_{ss}(\alpha, K) = \frac{\alpha \sqrt{K} \tan h(\sqrt{K})}{1 + \sqrt{\alpha/2} \tan h(\sqrt{K})}$</p>
[35]	<p>Steady and nonsteady state diffusion currents aimed at EC's reactions for entirely ultramicroelectrodes</p>	<p>$A + e \rightarrow B$ $B + Z \xrightarrow{k} A + \text{products}$</p>	<p>$\nabla^2 c(r, z) - K(r, z) = 0$</p>	<p>$I_{ss}(K) = \frac{i(K) a}{4\pi F D C_0 A} = \frac{a^2}{A} [A_1 + B_1 \sqrt{K} + C_1 \exp(D_1 \sqrt{K})]$ $I_T = \frac{i(t) a}{4\pi F D C_0 A} = I_{ss}(K) + \frac{1}{4} K^{1/2} g(K, t)$ Here A means area of electrode, $I_{ss}(k)$ means steady state current designed for the EC reaction $g(K, t) = (\pi K t)^{-1/2} e^{-Kt} - \text{erfc}(Kt)^{1/2}$ Here A means area of electrode, $I_{ss}(k)$ means steady state current meant for the EC reaction</p>
[43]	<p>polymer microelectrodes</p>	<p>$A + e \rightarrow B$ $B + Z \xrightarrow{k} A + \text{products}$</p>	<p>$\frac{d^2 u}{d\rho^2} + \frac{2 du}{\rho d\rho} - \frac{\gamma u}{1 + \alpha u} = 0$ $\rho = 0; \frac{du}{d\rho} = 0$ $\rho = 1; u = 1$</p>	<p>$\psi = \frac{ia}{nFA D_s \omega} = \left(\frac{du}{d\rho} \right)_{\rho=1}$ $\psi(\gamma, \alpha) = \sqrt{\gamma} \cot h(\sqrt{\gamma}) - 1 + \alpha \gamma \operatorname{cosec} h^2(\sqrt{\gamma}) [\sqrt{\gamma} \coth(\sqrt{\gamma}) - 1]$ $\psi(\gamma, \alpha, \nu) = \frac{\sqrt{\gamma} \cosh(\sqrt{\gamma}) - \sinh(\sqrt{\gamma})}{\left\{ \sinh(\sqrt{\gamma}) + \frac{1}{\nu} [\sqrt{\gamma} \cosh(\sqrt{\gamma}) - \sinh(\sqrt{\gamma})] \right\}} - \frac{\alpha \gamma \sqrt{\gamma} \cosh(\sqrt{\gamma}) - \sinh(\sqrt{\gamma})}{\left\{ \sinh(\sqrt{\gamma}) + \frac{1}{\nu} [\sqrt{\gamma} \cosh(\sqrt{\gamma}) - \sinh(\sqrt{\gamma})] \right\}^3}$</p>

References	Experimental Techniques	Enzymatic scheme and Electrodes	Nonlinear Diff. Eqns. with initial/boundary conditions	Expressions for current
[27]	Hemispheroidal electrodes (Non steady state)	$A + n\bar{e} \rightarrow B, B + Z \xrightarrow{k} A + products$	$\frac{\partial c(r,t)}{\partial t} = \nabla^2 c(r,t) - K(r,t) = 0$	<p>$\frac{i(t)}{nFD_0} = - \int_A \frac{\partial c}{\partial z} ds$</p> <p>$\frac{i(t)a}{nFD_0 C_0 A} = i_\infty(k) + k^{1/2} g(K,t)$</p> <p>Here A means area of electrode, $i_\infty(k)$ means dimensionless steady state current.</p> <p>$\frac{i(t)a}{nFD_0 C_0 A} = i_\infty(k) + k^{1/2} g(K,t)$</p> <p>$g(K,t) = \pi K t^{-1/2} e^{-Kt} - \text{erfc}(Kt)^{1/2}$</p> <p>(non steady state current)</p> <p>Here A means area of electrode, $i_\infty(k)$ means dimensionless non-steady state current</p>
[28]	Transient Chronoamperometric current at prolate hemispheroidal ultra micro electrodes (Non steady state)	$O + ne^- \rightleftharpoons R$	$\frac{\partial c}{\partial t} = \nabla^2 c$ <p>Where c is dimensionless concentration is given by $c = \frac{c^*}{c_0^b}$, c_0^b is bulk concentration $c=0$ at $t=0$</p>	<p>$\frac{i}{4nFD_0 c_0^b a} = \frac{p_0 + p_1 y + p_2 y^2 + p_3 y^3}{1 + q_1 y + q_2 y^2}$</p> <p>Where $y = t \left(\frac{-1}{2}\right)$</p>
[32]	Spheroidal electrodes (Non steady state)		$\frac{\partial c}{\partial t} = \nabla^2 c$ <p>$c = 0 \text{ at } t = 0$</p>	<p>$\frac{i}{4nFD_0 c_0^b a} = 2\pi \int_{electrodes} r q(r,t) dr$</p> <p>$\frac{i}{4nFD_0 c_0^b a} = A + By + C \exp(-Dy)$</p> <p>where $y = t \left(\frac{-1}{2}\right)$</p> <p>data of A, B, C & D are specified in Ref [32]</p>

5. Conclusion

Most mathematical and theoretical models of ultramicroelectrode are constructed on nonlinear reaction-diffusion differential equations. Various novel and advanced analytical systems for instance the homotopy perturbation technique, the Taylor's series method, the Pade approximation technique, reduction of order method, hyperbolic function method, complex inversion formula etc. have been employed to obtain approximate analytical solutions under steady and non-steady state conditions. Reliable analytical results are very useful for the analysis of various parameters like the thickness of electrode, loading of the different species, steady-state current, flux, diffusion rate, rate constant, reaction rate, the permeability of the porous medium, diffusion coefficients, kinematic viscosity, and voltammetry current. In conclusion, ultramicroelectrodes have made significant progress in power efficiency and stability since their conception. However, there is still a need for further theoretical and simulation research to make them a more technically and commercially feasible solution for wearable, implantable, and portable devices powering.

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