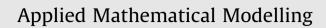
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Mathematical analysis of an enzyme-entrapped conducting polymer modified electrode



K. Saravanakumar, L. Rajendran*

Department of Mathematics, The Madura College, Madurai 625011, Tamil Nadu, India

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1. Introduction

ABSTRACT

A mathematical model for the enzyme-entrapped conducting polymer modified electrode is discussed. Approximate expressions for the steady state concentration of the various species in the film and the current response of the enzyme electrode are derived. In this work we employ the Homotopy analysis method (HAM) to solve the nonlinear equations. These approximate analytical results are found to be in good agreement with the previously reported limiting case results. The obtained results are valid for the whole solution domain. Also, the redox enzyme in conducting polymer electrode is the most fundamental requirement for the development of amperometric biosensor and biofuel cell elements. © 2015 Elsevier Inc. All rights reserved.

Developments in science and technology have revealed new application possibilities for conducting homopolymers and their derivatives [1]. Conducting polymers have been used widely in many areas such as rechargeable batteries, condensators, diodes, LED's, biofuel cells and sensors [2]. Of this class of polymers, polyaniline and polypyrrole have been intensively studied due to their favorable processibility and relative stability. Immobilization of an enzyme onto a solid support increases the rigidity of the enzyme molecule and, as a result, frequently improves the stability. Covalent attachment of biomolecules to the surface of polymers ensures immobilization without leaching of the biomolecule from the substrate surface [3]. The precise mechanism of the reactions within the conducting-polymer film and the effect of the immobilization on the enzyme kinetics are of interest. The behavior of these entrapped enzyme films is sufficiently stable and reproducible from film to film that we are able to carry out kinetic studies of the glucose oxidase catalyzed reaction within the film [4]. An ideal immobilization method should employ mild chemical conditions, allow for large quantities of enzyme to be immobilized, provide a large surface area for enzyme-substrate contact within a small total volume, minimize barriers to mass transport of substrate and product, and provide a chemically and mechanically robust system. Electrochemical-based enzyme immobilization methods are a convenient way of immobilizing enzymes on microelectrodes. The general features of amperometric response were analyzed by Mell and Maloy [5].

Earlier, Bartlett et al. [6] evaluated the mathematical expressions pertaining to approximate analytical concentration and current for limiting cases at enzyme electrodes. Furthermore mass transport and partition of both the reactants and the products within the film can also play an important role. Consequently, the entrapment of enzymes within electropolymerized films has become an increasingly important technique for electronically addressing biological molecules. The technique

* Corresponding author. Tel.: +91 9442228951. E-mail address: raj_sms@rediffmail.com (L. Rajendran).

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Nomenclature

Symbols

S	concentration of the substrate (mol/cm ²)
b	concentration of the reduced mediator (mol/cm ³)
а	concentration of the oxidized mediator (mol/cm ³)
e_{Σ}	total concentration of the enzyme species (mol/cm ³)
S_{∞}	substrate concentration in bulk solution (mol/cm ³)
b_∞	reduced mediator concentration in bulk solution (mol/cm ³)
a_{∞}	oxidized mediator concentration in bulk solution (mol/cm ³)
D_S	diffusion coefficient of substrate $(cm^2 s^{-1})$
D_B	diffusion coefficient of reduced mediator $(cm^2 s^{-1})$
D_A	diffusion coefficient of oxidized mediator $(cm^2 s^{-1})$
K_M	Michealis–Menten constant (mol/cm ³)
k _s	partition coefficient for substrate (none)
k_B	partition coefficient for reduced mediator (none)
<i>k</i> _A	partition coefficient for oxidized mediator (none)
k ₁ , k ₃	rate constants (L mol ^{-1} s ^{-1})
k_{-1}, k_2	rate constants (s ⁻¹)
h _s	mass transport coefficient of substrate (m/s)
h_B	mass transport coefficient of reduced mediator (m/s)
h_A	mass transport coefficient of oxidized mediator (m/s)
L	inner side of the film (cm)
η	dimension variable (cm ³ /mol)
Dimensionless parameters	
$\beta_i = D_i / h_i$	<i>kL</i> ratio of the mass-transport coefficient in the polymer film to that in the solution
$\theta = (k_4 L^2)$	$(D_B)^{1/2}$ ratio of the reduction/oxidation rate of mediator to its mass-transport rate in the
1/2	

concentration of the substrate (mol/cm^3)

 $\theta = (k_4 L^2 / D_B)^{1/2}$ ratio of the reduction/oxidation rate of mediator to its mass-transport rate in the film $\alpha = (k_2 e_{\Sigma} L^2 / K_M D_S)^{1/2}$ ratio of the rate of enzyme catalytic reaction to the rate of diffusion in the polymer film

Subscripts

- S substrate
- *B* reduced mediator
- A mediator
- ∞ bulk of the solution

has found a wide range of applications in the fields of both biosensors and molecular electronics [7]. The stability of the enzyme-entrapped electrodes was compared to electrodes prepared by covalent grafting [8]. A mediator based fuel cell has been reported by Pizzariello et al. [9]. Several biofuel cell configurations based on immobilization of enzyme and mediator in polypyrrole was introduced [10,11].

Recently, Kan and Hui-huang [12] obtained approximate analytical expressions for the concentrations and the current response of an enzyme-entrapped conducting polymer modified electrodes for the suitable limiting cases only. However, to the best of our knowledge, no analytical expressions corresponding to the substrate concentration and the current for all values of parameters α (ratio of the rate of enzyme catalytic reaction to the rate of diffusion in the polymer film) and θ (ratio of the reduction/oxidation rate of mediator to its mass-transport rate in the film) at polymer modified electrode have been reported. However, in general, analytical solutions of nonlinear differential equations are more interesting, as they are used for various kinds of data analysis. The aim of this paper is to derive an analytical expressions for the concentrations and the current response for all values of the parameters by using Homotopy analysis method.

2. Mathematical formulation of the boundary value problem

Fig. 1 represents the general kinetics scheme for an amperometric enzyme electrode. We assume the diffusion of substrate and oxidized mediator from the bulk of the solution to the polymer/solution interface with the respective diffusion coefficient. Also the enzyme reactions take place within the polymer film. We can consider that the diffusion of the product *P* and *B* in film, towards the polymer/solution interface and then into the bulk of the solution. We can assume that the enzyme containing polymer film of thickness *L* is coated on the surface of a metal electrode (at x = 0). The reactions occurring within the film are: Download English Version:

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