

The Steady-State Concentration of the Species in a Reagentless Enzyme-Containing Polymer Modified Electrode Using Akbari-Ganji's method

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A theoretical model is presented for reagentless- conducting polymer modified electrodes. This model is based on nonlinear reaction-diffusion equation with nonlinear term related to Michaels-Menten kinetics. The theoretical representation of species concentration for steady-state conditions for all kinetic model parameters is presented in this report. Akbari-Ganji's method used is to evaluate the analytical expressions of concentration of species in the film and current. The effects of the parameter on the concentration and current are also analyzed. The limiting conditions of catalytic sites (unsaturation and site saturation) are discussed, and corresponding an analytical expression for concentrations and transient current is also derived. Our estimated analytical results are compared with the simulation results. It is observed that a good agreement has been obtained.

Keywords: Mathematical modeling; nonlinear reaction diffusion equation; enzyme, polymer modified electrode; Akbari-Ganji's method.

1. INTRODUCTION

The electrochemical immobilization of enzymes has emerged as a reliable method for the production of enzyme electrodes. The process of immobilization and the kinetic behaviour of the resulting enzyme electrode have been examined [1-3]. Furthermore, because the approach allows for the immobilization of the mediator in the polymer as a dopant, it can be used to make so-called reagentless biosensors [4-6].

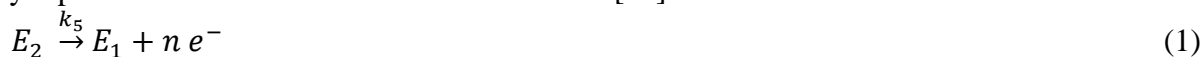
The physical entrapment of enzymes in electrochemically generated polypyrrole has been the focus of the majority of research [7-9]. However, various film geometries, such as immobilization by covalent attachment with functionalized polymers, have been proposed [10,11] or a pyrrole-modified enzyme to copolymerize [12].

Bartlett et al. [13] explored mathematical formulas relating to approximate analytical concentration and current for limiting conditions at enzyme electrodes. Saravanakumar and Rajendran [14] had used the homotopy perturbation method to analyse the enzyme-entrapped conducting polymer modified electrode. Lyons et al. [15] reported the steady-state amperometric current response of a polymer-modified electrode system with Michaelis-Menten kinetics.

For the appropriate limiting conditions alone, Kan and Hui-Huang [16] developed semi-analytical expressions for the concentration of species in the film and the current response of enzyme-entrapped conducting polymer modified electrodes. No analytical equations for the concentration of species in the film and the current have been provided to our knowledge for all values of parameters $\alpha, \beta_s, s_\infty, k_s, K$. The aim of this communication is to use Akbari-Ganji's approach to construct an analytical equation for the concentration and current for all values of the parameters.

2. MATHEMATICAL FORMULATION OF MIXED BOUNDARY VALUE PROBLEM

The reagentless enzyme reactions take place within the polymer film, the regeneration of E_2 is formally represented with an electrochemical reaction [16].



The steady-state mass balance nonlinear differential equation for the concentration of species can be written as follows:

$$D_s \frac{d^2 s(x)}{dx^2} - R(s(x)) = 0 \quad (2)$$

where

$$R(s(x)) = \frac{k_2 e_\epsilon}{1 + K_M/s(x) + k_2/k_5} \quad (3)$$

where $s(x)$ denotes the concentration of species in film, K_m is the Michealis constant, k_2 and k_5 represent the reaction rate constant, e_ϵ is the total concentration of enzyme species, and D_s is the diffusion coefficient of substrate. The following are the corresponding boundary conditions.

$$\text{At } x = 0, \frac{ds}{dx} = 0 \quad (4)$$

$$\text{At } x = L, D_s \left(\frac{ds}{dx} \right) = h_s (s_\infty - k_s s(x)) \quad (5)$$

where s_∞ is the bulk concentration of substrate, L is the inner side of the film, and k_s is the partition coefficient for substrate, and h_s is the mass transport coefficient of substrate. By defining the following dimensionless parameters,

$$\alpha = \left(\frac{k_2 e_\epsilon L^2}{K_M D_s} \right)^{1/2}, \beta_s = \frac{D_s}{h_s k_s L}, \text{ and } K = \frac{k_2 + k_5}{K_M} \quad (6)$$

Eq. (2) can be reduced to the dimensionless form:

$$\frac{d^2s(x)}{dx^2} - \frac{\alpha^2 s(x)}{L^2(1+Ks(x))} = 0 \tag{7}$$

The boundary conditions in dimensionless form in enzyme electrode are as follows:

$$\text{At } x = 0, \frac{ds}{dx} = 0 \tag{8}$$

$$\text{At } x = L, \frac{ds}{dx} = \frac{(s_\infty - k_s s)}{\beta_s k_s L} \Rightarrow \frac{ds}{dx} - \frac{s_\infty}{\beta_s k_s L} = \frac{-s(x)}{\beta_s L} \tag{9}$$

The current response is [16]

$$\frac{i}{nF D_s} = \left(\frac{ds}{dx}\right)_{x=L} \tag{10}$$

3. APPROXIMATE ANALYTICAL EXPRESSION OF THE CONCENTRATION USING AKBARI-GANJI'S METHOD

Recently many asymptotic techniques have been available to solve nonlinear differential equations. The Akbari-Ganji method [17-21], Taylor series method [22,23], homotopy analysis method [24], variational iteration method [25], Adomian decomposition method [26,27], are also examples of such methods.

The Akbari Ganji method (AGM) is a powerful algebraic (semi-analytic) strategy for solving such problems in this regard. In the AGM, a solution function with unknown constant coefficients should satisfy the differential equation and initial conditions initially. Assume that the solution to Eq. (7) is of the following hyperbolic form.

$$s(x) = A_0 \cosh(mx) + B_0 \sinh(mx) \tag{11}$$

where A_0, B_0 and m are constant. The values of A_0, B_0 are found easily from boundary conditions (8) and (9), that is

$$A_0 = \frac{s_\infty}{k_s[\cosh(mL) + mL\beta_s \sinh(mL)]}, B_0 = 0 \tag{12}$$

As a result, Eq. (11) becomes

$$s(x) = A_0 \cosh(mx) = \frac{s_\infty \cosh(mx)}{k_s[\cosh(mL) + mL\beta_s \sinh(mL)]} \tag{13}$$

We use the general form of Eq. (7) to find the constant m in Eq. (13).

$$F(x) = L^2(1 + Ks(x))\frac{d^2s(x)}{dx^2} - \alpha^2 s(x) = 0 \tag{14}$$

We obtain Eq. (14) by substituting it for Eq. (13)

$$F(x = 0) = L^2(1 + K A_0)m^2 A_0 - \alpha^2 A_0 = 0 \tag{15}$$

This gives

$$L^2 m^2 (1 + K A_0) - \alpha^2 = 0 \tag{16}$$

FSubstitute Eq. (16) into Eq. (13) gives the following analytical expression of a substrate concentration in species $s(x)$ for all dimensionless parameters α, β_s and K . The current response can be obtained as follows:

$$\frac{i}{nF D_s} = \left(\frac{ds}{dx}\right)_{x=L} = \left[\frac{m s_\infty \tanh(mL)}{k_s(1+mL\beta_s \tanh(mL))} \right] \tag{17}$$

4. THE ANALYTICAL EXPRESSIONS OF CONCENTRATION AND CURRENT FOR THE LIMITING CASES.

The two limiting conditions are zero-order kinetics and first-order kinetics. The limiting case identities are based on the extent to which the enzyme kinetics have been saturated and hence on the magnitude of the substrate concentration about the enzyme's Michaelis constant.

When the substrate concentration exceeds the Michaelis constant, zero-order kinetics is used. The substrate binds all enzyme active sites, and the reaction rate is unaffected by substrate concentration.

The kinetics are unsaturated when the substrate concentration is less than the Michaelis constant. Instead, they refer to how the substrate has saturated the enzyme catalyst sites; not all active sites have been occupied. As a result, the reaction rate varies linearly as the substrate concentration increases. According to these limiting constraints, the nonlinear kinetic term can be reduced to a linear form. The reaction/diffusion equation is no longer nonlinear in these limiting circumstances, allowing for a more straightforward solution.

4.1 Unsaturated (First-Order) Catalytic Kinetics.

In this case, $KS < 1$ or $(K_M/s) \gg [1 + (k_2/k_5)]$, Eq. (3) reduce to $R = k_2 e_E s/K_M$ and Eq. (7) reduces to the linear reaction-diffusion equations

$$\frac{d^2s(x)}{dx^2} - \frac{\alpha^2 s(x)}{L^2} = 0 \tag{18}$$

The boundary conditions are

$$\text{At } x = 0, \quad \frac{ds}{dx} = 0 \tag{19}$$

$$\text{At } x = L, \quad \frac{ds}{dx} - \frac{s_\infty}{\beta_s k_s L} = \frac{-s(x)}{\beta_s L} \tag{20}$$

The concentration of substrate species

$$s(x) = \frac{s_\infty \cosh\left(\frac{\alpha x}{L}\right)}{k_s(\cosh \alpha + \alpha \beta_s \sinh \alpha)} \tag{21}$$

The current response is

$$\frac{i}{nFD_s} = \frac{s_\infty \alpha \tanh \alpha}{L k_s(1 + \alpha \beta_s \tanh \alpha)} \tag{22}$$

4.2 Saturated (Zero-Order) Catalytic Kinetics.

The significant limiting situation in practice under consideration is when $KS > 1$ or

$(K_M/s) \ll [1 + (k_2/k_5)]$, Eq. (3) reduce to $R = k_2 e_E / (1 + \frac{k_2}{k_5})$ and Eq. (7) reduces to the

linear reaction-diffusion equations

$$\frac{d^2s(x)}{dx^2} - \frac{\alpha^2}{L^2 K} = 0 \tag{23}$$

$$\text{At } x = 0, \quad \frac{ds}{dx} = 0 \tag{24}$$

$$\text{At } x = L, \quad \frac{ds}{dx} - \frac{s_\infty}{\beta_s k_s L} = \frac{-s(x)}{\beta_s L} \tag{25}$$

The concentration of substrate species

$$s(x) = \frac{1}{2} \left[\frac{\alpha^2 x^2}{L^2 K} - \frac{\alpha^2}{2K} - \frac{\beta_s L^2}{K} + \frac{s_\infty}{k_s} \right] \tag{26}$$

The current response is

$$\frac{i}{n F D_s} = \frac{s_\infty \alpha^2}{L K} = \frac{s_\infty \alpha^2 K_M}{L(k_s + k_2)} \tag{27}$$

Our results for the limiting case are identical to the results obtained by Kan and Hui-Huang [16].

5. DISCUSSION.

Equation (13) is the new, general and simple analytical expressions of the concentration profile of the species $s(x)$ in conducting polymer electrodes.

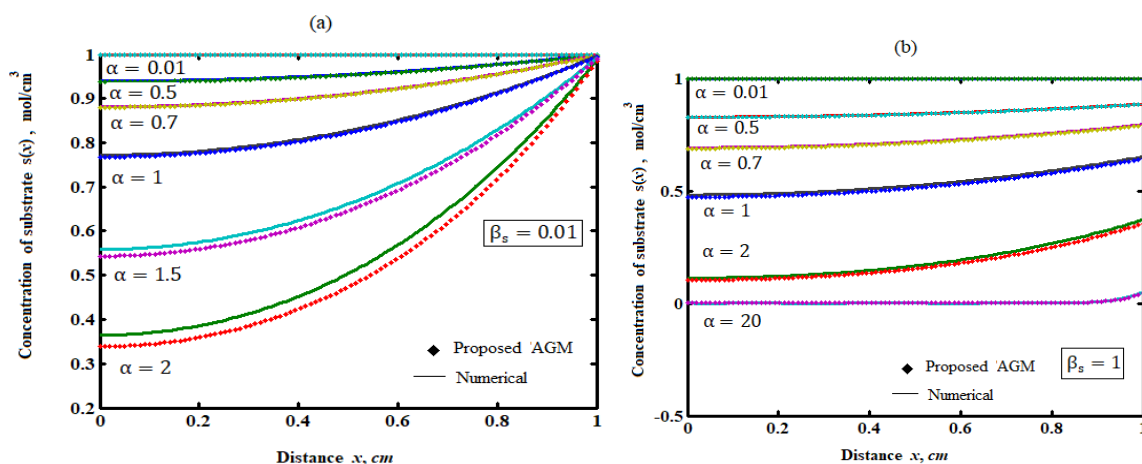


Figure 1. Comparison of analytical expression (Eq. (13)) of substrate concentration of species $s(x)$ with simulation result for various values of parameters α . The other parameter values are $s_\infty = 0.1 \text{ (mol/cm}^3\text{)}, k_s = 0.1, K = 1,$ and $L = 1 \text{ (cm)}$.

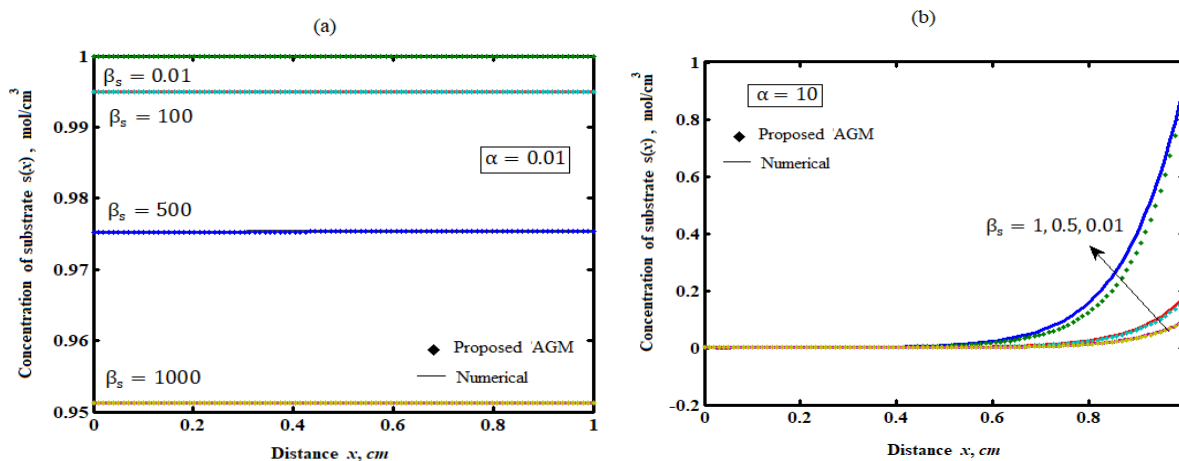


Figure 2. Comparison of analytical expression (Eq. (13)) of substrate concentration of species $s(x)$ with simulation result for various values of parameters β_s . The other parameter values are $s_\infty = 0.1 \text{ (mol/cm}^3\text{)}, k_s = 0.1, K = 1,$ and $L = 1 \text{ (cm)}$.

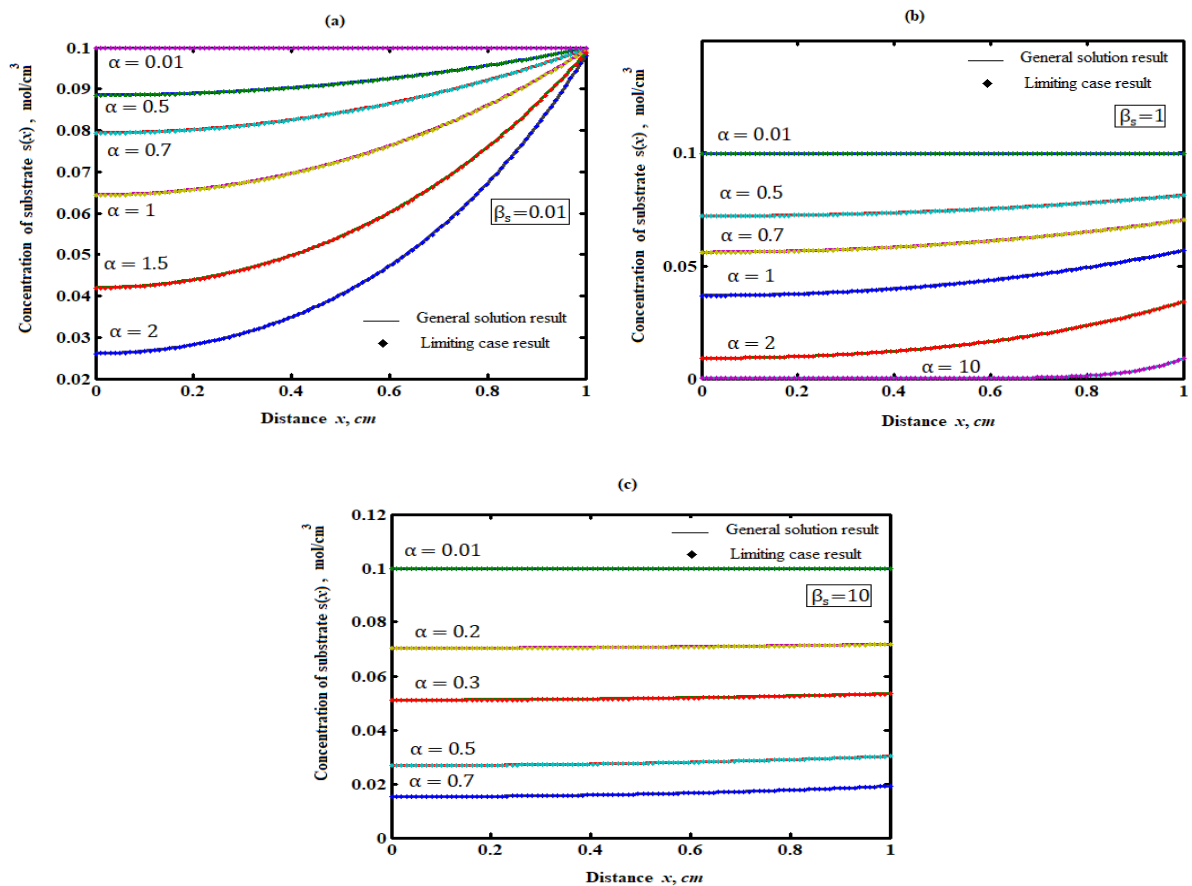
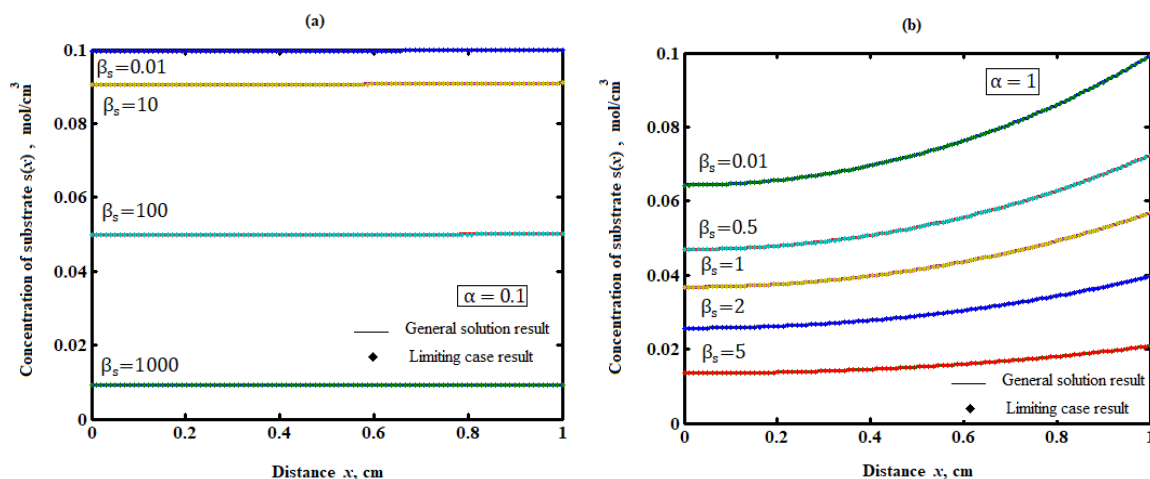


Figure 3. Comparison of substrate concentration of species $s(x)$ for limiting case result species $s(x)$ (Zhu Kan and Hui-huang) (Eq. (21)) and general solution result using Eq. (13) for various values of parameters α . The other parameter values are $s_\infty = 0.1 \text{ (mol/cm}^3\text{)}$, $k_s = 1$, $K = 0.1$, and $L = 1 \text{ (cm)}$.

This concentration depends on parameters $\alpha, \beta_s, s_\infty, k_s, K$, and L . The steady-state substrate concentration profile of the substrate $s(x)$ is compared with simulation results (Figs 1,2) and limiting case results (Figs 3,4) [16] respectively.



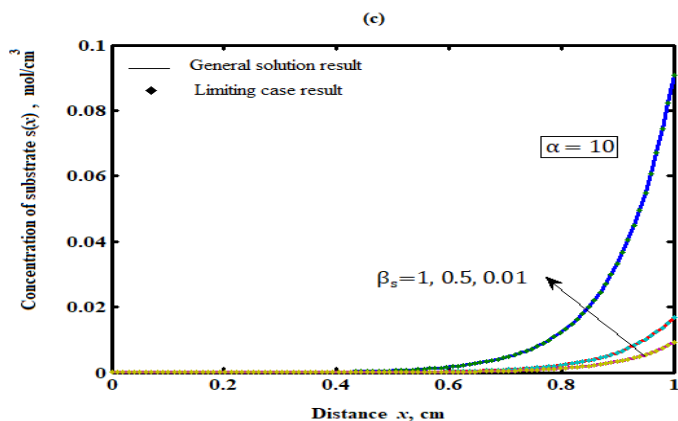


Figure 4. Comparison of substrate concentration of species $s(x)$ for limiting case result (Zhu Kan and Hui-huang) (Eq. (21)) and general solution result using Eq. (13) for various values of parameters β_s . The other parameter values are $s_\infty = 0.1 \text{ (mol/cm}^3\text{)}$, $k_s = 1$, $K = 0.01$, and $L = 1 \text{ (cm)}$.

The concentration and current are depends upon the parameter α , β_s , s_∞ , k_s , K , and L . The rate of enzyme catalytic reaction divided by the rate of diffusion in the polymer film yields α^2 . The parameter β_s is the ratio of mass-transport coefficient in the polymer film to the solution. The remaining parameters s_∞ , k_s , K , and L are all ready defined in the section 2.

Table 1. Comparison between analytical and previous results for current $\frac{i}{nfD_s}$ for various values of parameter β_s when $s_\infty = 0.1 \text{ (mol/cm}^3\text{)}$, $k_s = 0.1$, $K = 0.01$ and $L = 0.1 \text{ (cm)}$

| α | $\beta_s = 0.01$ | | | $\beta_s = 10$ | | |
|----------|------------------------|-----------------------------------|-----------|------------------------|-----------------------------------|-----------|
| | This work AGM Eq. (17) | Zhu Kan & Hui-huang [16] Eq. (22) | abs.error | This work AGM Eq. (17) | Zhu Kan & Hui-huang [16] Eq. (22) | abs.error |
| 0.01 | 0.0009900 | 0.0010000 | 0.0000100 | 0.0009891 | 0.0009970 | 0.0000079 |
| 0.5 | 2.2865572 | 2.3052592 | 0.0187020 | 0.6974151 | 0.6979386 | 0.0005235 |
| 1 | 7.5210523 | 7.5583773 | 0.0373250 | 0.8838762 | 0.8839361 | 0.0000599 |
| 5 | 47.612024 | 47.614930 | 0.0029060 | 0.9803904 | 0.9803904 | 0 |
| 10 | 90.909090 | 90.909090 | 0 | 0.9900990 | 0.9900990 | 0 |
| 50 | 333.33333 | 333.33333 | 0 | 0.9980040 | 0.9980040 | 0 |
| 100 | 500.00000 | 500.00000 | 0 | 0.9990010 | 0.9990010 | 0 |
| 500 | 833.33335 | 833.33335 | 0 | 0.9998000 | 0.9998000 | 0 |
| 1000 | 909.09091 | 909.09091 | 0 | 0.9999000 | 0.9999000 | 0 |

Table 2. Comparison between analytical and previous results for current $\frac{i}{nfD_s}$ for various values of parameter β_s when $s_\infty = 0.1$ (mol/cm³), $k_s = 0.1$, $K = 0.01$ and $L = 0.1$ (cm)

| β_s | $\alpha = 0.01$ | | | $\alpha = 0.1$ | | |
|-----------|------------------------|-----------------------------------|-----------|------------------------|-----------------------------------|-----------|
| | This work AGM Eq. (17) | Zhu Kan & Hui-huang [16] Eq. (22) | abs.error | This work AGM Eq. (17) | Zhu Kan & Hui-huang [16] Eq. (22) | abs.error |
| 0.01 | 0.0009900 | 0.0010000 | 0.0000100 | 0.0986796 | 0.0996580 | 0.0009784 |
| 0.5 | 0.0049999 | 0.0099990 | 0.0049991 | 0.4991665 | 0.9917380 | 0.4925715 |
| 1 | 0.0049998 | 0.0099990 | 0.0049992 | 0.4985401 | 0.9868440 | 0.4893039 |
| 5 | 0.0049993 | 0.0099950 | 0.0049957 | 0.4935381 | 0.9493691 | 0.4558310 |
| 10 | 0.0049987 | 0.0099900 | 0.0049913 | 0.4872899 | 0.9063462 | 0.4190563 |
| 50 | 0.0049937 | 0.0099500 | 0.0049563 | 0.4382045 | 0.6651894 | 0.2269849 |
| 100 | 0.0049870 | 0.0099010 | 0.0049140 | 0.3817228 | 0.4991686 | 0.1174458 |
| 500 | 0.0049370 | 0.0095250 | 0.0045880 | 0.1614078 | 0.1665742 | 0.0051664 |
| 1000 | 0.0048750 | 0.0090910 | 0.0042160 | 0.0900726 | 0.0908816 | 0.0008090 |

Fig. 3(a-c), shows the effect of parameter α on the concentration of substrate in conducting polymer electrodes. From the Figures it is inferred that the concentration decreased by increasing the parameter α . Fig. 4(a-c), it is inferred that concentration decreases when β_s decreases for different values of α . The concentration of substrate reaches its maximum when $\alpha < 0.01$. Tables 1 and 2 show that the analytical and limiting case result for the current are in strong agreement.

5.1 Biosensor Sensitivity

Sensitivity is the ratio of change in stationary current to change in concentration. A low value indicates that it is less sensitive to change in substrate concentration. Variations in current and substrate concentration must be studied to produce high accuracy sensors. Biosensor sensitivity can be obtained as follows:

$$B_s(s_\infty) = \frac{\partial I(s_\infty)}{\partial s_\infty} \times \frac{s_\infty}{I(s_\infty)} \tag{28}$$

where B_s is biosensor sensitivity. $I_{CCE}(s_\infty)$ is steady-state current density measured at the substrate concentration s_∞ . When the thickness of the enzyme L is very small, $A_0 = \frac{s_\infty}{k_s}$. Now from the

eq. (16) we get

$$L^2 m^2 \left(1 + K \frac{s_\infty}{k_s} \right) - \alpha^2 = 0 \tag{29}$$

Simplifying the above equation, we obtain

$$m = \frac{\alpha}{L} \sqrt{\frac{k_s}{k_s + s_\infty}} \tag{30}$$

Sensitivity becomes

$$B_s(s_\infty) = \frac{L k_s B}{\alpha \eta A} \left[\frac{\alpha \eta A}{L k_s B} - \frac{\alpha s_\infty A K}{2 L \eta} - \frac{\alpha^2 s_\infty K (1-A^2)}{L B (k_s + K s_\infty)^2} + \frac{\alpha^2 \beta_s s_\infty K A^2}{2 L B^2 (k_s + K s_\infty)^2} + \frac{\alpha^3 \beta_s s_\infty K \eta A (1-A^2)}{2 L B^2 (k_s + K s_\infty)^2} \right] \tag{31}$$

$$\text{where } \eta = \sqrt{\frac{k_s}{k_s + s_\infty}}, A = \tanh(\alpha \eta), B = 1 + \alpha \beta_s \eta A \tag{32}$$

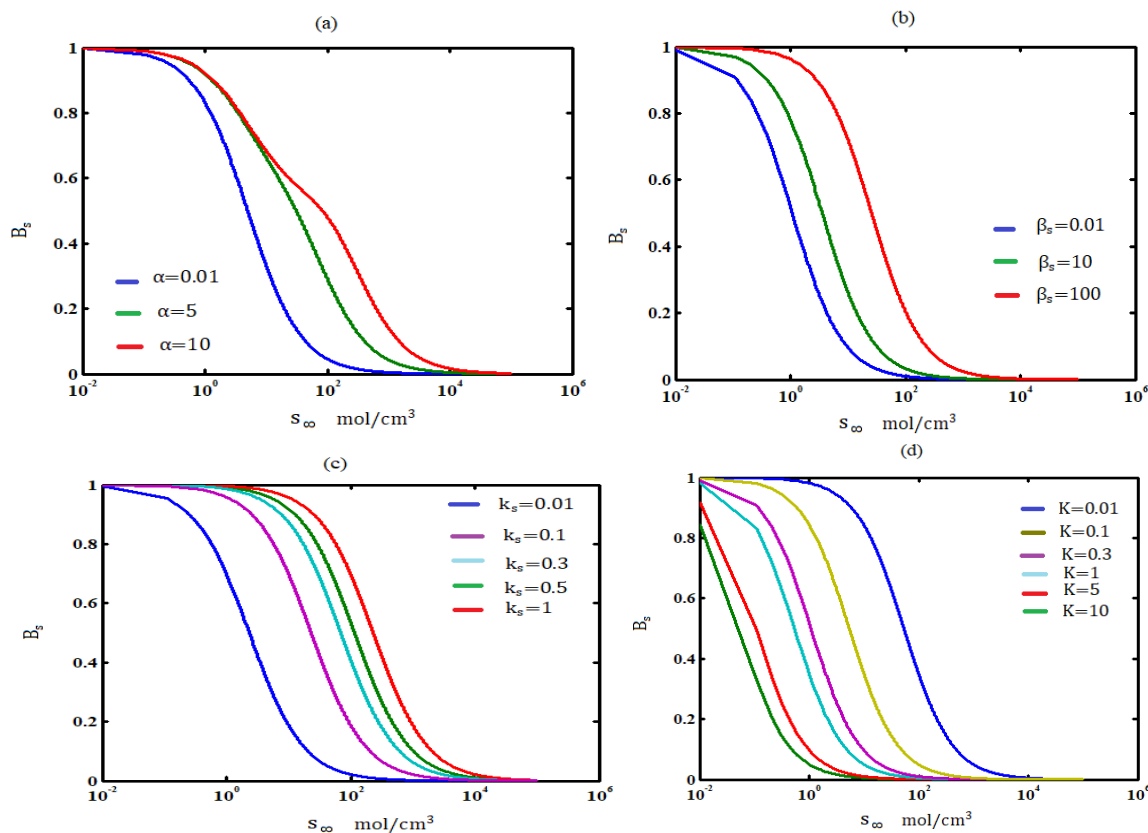


Figure 5. Sensitivity using Eq. (31) for fixed values of parameters (a) $\beta_s=0.01, L = 0.5 \text{ cm}, K = 1, k_s = 5$. (b) $\alpha=0.5, L = 0.5 \text{ cm}, K = 5, k_s = 5$. (c) $\alpha = 0.5, \beta_s=5, L = 0.5 \text{ cm}, K = 0.01$. (d) $\alpha = 0.5, \beta_s=0.01, L = 0.5 \text{ cm}, k_s = 0.5$.

Sensitivity of the biosensor versus substrate concentration is plotted in Fig.5 for various values parameters. From the figure it is inferred that the parameter α , s_∞ increases or β_s, k_s, K decreases the sensitivity B_s decreases. It is possible to observe from the smooth curves of figure 5 (a) to (d) that $0 \leq B_s \leq 1$ for all values of parameters.

5.2 Biosensor Resistance

The resistance B_R is described as the gradient of steady-state biosensor current as a function of membrane thickness L .

$$B_R(L) = \frac{\partial I(L)}{\partial L} \times \frac{L}{I(L)} = \left(\frac{-\alpha \eta s_\infty A}{L^2 k_s B} \right) \left(\frac{L^2 k_s B}{\alpha \eta s_\infty A} \right) = -1 \tag{33}$$

6. CONCLUSIONS

In this paper, the approximate analytical expression of concentration of substrate for all experimental values of parameters is derived using Akbari-Ganji's method. The derived analytical results and simulation results are in good agreement. The effects of various parameter on the concentration, current and sensitivity are discussed. These results can be used to improve and construct enzyme electrodes for amperometric biosensors and biofuel cells.

NOMENCLATURE:

| Symbols | Description | Units |
|---|---|--------------------|
| D_s | Diffusion coefficient of substrate | $cm^2 s^{-1}$ |
| h_s | Mass transport coefficient of substrate | m/s |
| i/nF | current density at an enzyme electrode | $\mu A/cm^2 C$ |
| K_M | Michealis-Menten constant | mol/cm^3 |
| k_s | Partition coefficient of substrate | None |
| K | Dimensionless parameter | None |
| k_2, k_5 | Rate constants | $mole^{-1}/s^{-1}$ |
| L | Inner side of the film | cm |
| $s(x)$ | Concentration of the substrate | mol/cm^3 |
| s_∞ | Substrate concentration in bulk solution | mol/cm^3 |
| e_E | Total concentration of the enzyme species | mol/cm^3 |
| x | Distance | cm |
| $\alpha = \left(\frac{k_2 e_E L^2}{K_M D_s} \right)^{1/2}$ | The ratio of the rate of enzyme catalysis to the rate of diffusion in a polymer film | None |
| $\beta_s = \frac{D_s}{h_s k_s L}$ | The mass-transport coefficient in the polymer film compare to a mass-transport coefficient in the solution. | None |

APPENDIX A:

MATLAB Code for Numerical Solution of the Non-Linear Eq. (7)

```
function pdex4
```

```
m = 0;
```

```
x = linspace(0, 1);
```

```
t = linspace(0, 1000000);
```

```
sol = pdepe(m, @pdex4pde, @pdex4ic, @pdex4bc, x, t);
```

```
u1 = sol(:, :, 1);
```

```
% _____
```

```

figure
plot(x, u1(end,:))
title('u1(x, t)')
xlabel('Distance x')
ylabel('u1(x, 1)')
function [c, f, s] = pdex4pde (x, t, u, DuDx)
c =1;
f =1. * DuDx;
b=1; alpha=10; k=1;
F =-(alpha^2*u (1))/(b^2*(1+k*(u (1))));
s =F;
% -----
function u0 = pdex4ic(x)
u0 = [0];
% -----
function [pl, ql, pr, qr] = pdex4bc (xl, ul, xr, ur, t)
a=0.1; g=0.1; beta=1; n=1;
pl = [0];
ql = [1];
pr = -(a-g*ur(1))/(beta*g*n);
qr = [n];

```

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