Int. J. Electrochem. Sci., 17**(2022)** Article Number: 221064, doi: 10.20964/2022.10.47

International Journal of
ELECTROCHEMICAL
SCIENCE
www.electrochemsci.org

Mathematical Modelling of Amperometric Glucose Biosensor Based on Immobilized Enzymes: New Approach of Taylors Series Method

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Received: 10 July 2022 / *Accepted:* 21 August 2022 / *Published:* 10 September 2022

An amperometric glucose biosensor's theoretical model is discussed. The glucose oxidase enzyme in this model is immobilized in conducting polypyrrole.This model includes a nonlinear term that corresponds with the kinetics of enzyme reactions. The solution of coupled nonlinear reaction diffusion equations is obtained using new approach of Taylor method . Additionally, a comparison of numerical simulation and analytical approximation is provided. There is an agreement between numerical results and analytical expressions.

Keywords: Nonlinear equations; Mathematical modelling; an amperometric glucose biosensor; immobilized enzymes.

1. INTRODUCTION

The development of insoluble immobilized enzymes has received much attention since the second half of the 20th century [1]. These applications, such as reusable heterogeneous biocatalysts [2], stable and reusable devices for analytical and medical applications [3-7], selective adsorbents for purifying proteins and enzymes [8], fundamental tools for solid-phase protein chemistry [9,10], and microbial sensors, can benefit from using immobilised enzymes instead of their soluble counterparts.

It has been observed that the fabrication of biosensors [11-14] is well suited to the immobilization of enzymes in conducting polymer during the electro-polymerization step [15]. The process is simple to control. A theoretical model for an amperometric polypyrrole+glucose oxidase (PPY+GOD) electrode has already been presented by Bartlett and Whitaker [19]. Another significant advantage of this technique is the ability to entrap the mediator in the polymer as a dopant anion [20-24] or by covalent fixation on the pyrrole monomer [19].For low substrate and high benzoquinone concentrations compared to the corresponding Michaelis constants, Marchesiello and Genesis [20] developed analytical expressions of the concentrations of the substrate and benzoquinone. Analytical solutions have been derived for the steady state nonlinear reaction/diffusion equations in an amperometric glucose sensor [21].

To the best of my knowledge, no rigorous analytical expressions of substrate concentrations and mediators of amperometric glucose biosensor under non-steady-state conditions have been reported for all value parameters.Using the new approach of Taylor series method, this communication aims to derive approximate analytical expressions for non-steady-state concentrations and current.

2. MATHEMATICAL FORMULATION OF THE PROBLEM

The catalytic reaction scheme between benzoquinone and glucose is given by[20]
Glucose + GOD(FAD) ® GOD(FADH2) + gluconolactone
Q + GOD($FADH_2$) → GOD (FAD) + H2Q.
The oxidation of hydroquinone: - H_2Q → Q + 2H + 2e +.
Schematic representation of the above reaction scheme is given in Fig-1.

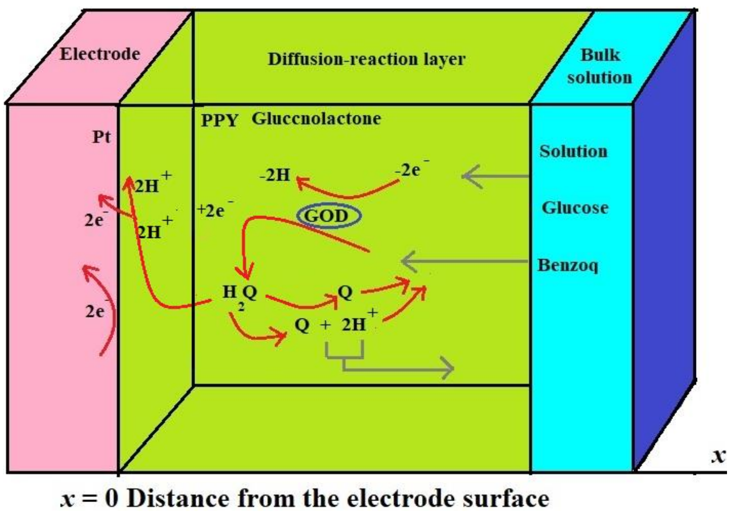

Figure 1. Schematic diagram of reaction scheme

The derivation and concise description of mass transport nonlinear equations in glucose biosensors by Marchesiello and Genesis [20] are summarised below. The enzymatic reaction rate between glucose and benzoquinone proceeds at a ping-pong mechanism [20]. The system of nonlinear differential equations describing the concentrations of S andH_2Q at steady- state are as follows [20]:

$$\mathrm{D}_s \frac{d^2[s]}{dX^2} - \frac{k_{cat}[E]}{1+\frac{K_Q}{[Q]}+\frac{K_S}{[S]}} = 0 \tag{1}$$

$$\mathrm{D}_{\mathrm{H}_2Q} \frac{d^2[\mathrm{H}_2Q]}{dX^2} - \frac{k_{cat}[E]}{1+\frac{K_Q}{[Q]}+\frac{K_S}{[S]}} = K\Sigma[\mathrm{H}_2Q] \tag{2}$$

The boundary conditions are as follows [28]:

$[s] = [s]_0, [\mathrm{H}_2Q] = 0$ at $X = L$ (polymer/solution interface) (3)

$\frac{d[\mathrm{S}]}{dx} = 0, [\mathrm{H}_2Q] = 0$ at $X = 0$ (platinum/polymer interface) (4)

where [S], $[\mathrm{H}_2Q]$ and [Q] are glucose, benzoquinone and enzyme concentrations in the film, $k_{cat}$is the turnover number for GOD, $K_s$and K_Q are the Michaelis constants for glucose and benzoquinone, respectively. The third term is the oxidation term.The charge-transfer constant for oxidation in the conducting polymer is$K$, so the electrochemical reaction rate in the conducting polymer is $v = K\Sigma[\mathrm{H}_2Q]$. $Also$ $[s]_0$ is bulk concentration of glucose. The total electro-catalytic current I_T is the sum of two currents. The current

$$I_S = 2FAD_{\mathrm{H}_2Q}\left(\frac{d[\mathrm{H}_2Q]}{dX}\right)_{X=0} \quad (5)$$

For H_2Q oxidation on the platinum electrode, and

$$I_P = \int_0^L dI = \int_0^L 2FAK\Sigma[\mathrm{H}_2Q]dx \quad (6)$$

where L is the film thickness of the reaction layer. The partition coefficients are assumed to equal one, and there is no concentration polarisation of S and H_2Q in the solution.. We make the non-linear PDE outlined in Eqs (1) and (2) dimensionless by introducing the following parameters:

$$x = \frac{X}{L}, u = \frac{[S]}{[S]_0}, v = \frac{[\mathrm{H}_2Q]}{[S]_0}, \alpha = L/\Lambda, \alpha' = L/\Lambda',$$

$$\Lambda = \left(\frac{\mathrm{D}_s\mathrm{K}_s}{[E]\mathrm{K}_{cat}}\right)^{1/2}, \Lambda' = \left(\frac{D_{\mathrm{H}_2Q}}{K\Sigma}\right)^{1/2}, M = \frac{[S]_0}{\mathrm{K}_s} + \frac{[S]_0\,\mathrm{K}_S}{\mathrm{K}_S[Q]} \quad (7)$$

The system of reaction-diffusion equations (1) and (2) are normalised as follows:

$$\frac{d^2u}{dx^2} - \frac{\alpha^2 u}{Mu+1} = 0 \quad (8)$$

$$\frac{d^2v}{dx^2} + \frac{\alpha^2 u}{Mu+1} = \alpha'^2 v \quad (9)$$

where u and v are the dimensionless concentration of S and H_2Q. Dimensionless boundary conditions are

$$\frac{du}{dx} = 0, v = 0 \text{ when } x = 0 \quad (10)$$

$$u = 1, v = 0 \text{ when } x = 1 \quad (11)$$

The dimensionless parameters α compare the enzymatic reaction rate to the substrate diffusion in the polymer. The dimensionless parameter compares α' the rate of $[\mathrm{H}_2Q]$oxidation in the conducting polymer with the rate of $[\mathrm{H}_2Q]$diffusion in the polymer. For electrochemically inert polymer, α' has low value and for electrochemically active polymer, α' has high value. The parameter M is a dimensionless constant. The dimensionless form of the current is given by

$$\psi_S = \frac{I_S}{2FAD_{\mathrm{H}_2Q}} = \left(\frac{dv}{dx}\right)_{x=0} \quad (12)$$

$$\psi_P = \frac{I_P}{2FAK\Sigma} = \int_0^1 v\,dx \quad (13)$$

3. RESULTS AND DISCUSSION

Exact solutions to nonlinear equations are extremely difficult to find.Asymptotic methods are developed for solving nonlinear differential equations approximately. The homotopy perturbation method [22-26],Taylor series method [27-36],and Akbari-Ganji method [37-40] Rajendran-Joy method

[41]. In this paper, the Taylor series method and Akbari-Ganji method is applied to solve the nonlinear model (equations (1)-(3)) for the immobilized enzyme reactions as per our assumptions considered for our enzyme kinetics .

3.1 Analytical expression of concentrations using Taylor series method

One of the earliest analytic-numerical algorithms for approximating initial value problems for ordinary linear/nonlinear differential equations is the Taylor series method [27-32]. This method also can be applied to solve the nonlinear differential equation with complex functions with mixed boundary conditions defined on finite intervals. The concentration of glucose using Taylor's series method is obtained as follows(Appendix-A):

$$u(x) = u(0) + \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{x^2}{2!} + \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3}\frac{x^4}{4!} + \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5}\frac{x^6}{6!} + \frac{(\alpha^2)^4 u(0)\left(90\,(\alpha^2)^2(u(0))^2 - 36(\alpha^2)u(0)+1\right)}{(Mu(0)+1)^7}\frac{x^8}{8!}. \tag{14}$$

where $u(0)$is obtained by solving the eqn.(15) for the given values of the parameters.

$$u(0) + \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3}\frac{1}{4!} + \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5}\frac{1}{6!} = 1 \tag{15}$$

The concentration of benzoquinone using Taylor's series is

$$v(x) = v'(0)\frac{x}{1!} - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{x^2}{2!} + \alpha'^2 v'(0)\frac{x^3}{3!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{x^4}{4!} + (\alpha'^2)^2\, v'(0)\frac{x^5}{5!} \tag{16}$$

The eqn. (17) is solved for the given specified parameter values to get the value of unknown parameter$v'(0)$

$$0 = v'(0) - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \alpha'^2 v'(0)\frac{1}{3!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{1}{4!} + (\alpha'^2)^2\, v'(\tag{17}$$

Now current becomes

$$\psi_S = \frac{I_S}{2FAD_{\mathrm{H_2}Q}} = \left(\frac{dv}{dx}\right)_{x=0} = v'(0) \tag{18}$$

$$\psi_P = \frac{I_P}{2FAK\Sigma} = \int_0^1 v\,dx = \int_0^1 v\,dx = v'(0)\frac{1}{2!} - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{3!} + \alpha'^2 v'(0)\frac{1}{4!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{1}{5!} + (\alpha'^2)^2\, v'(0)\frac{1}{6!} \tag{19}$$

3.2 Analytical expression of concentration using new approach of Taylor series method

The nonlinear differential equations governing the mentioned system has been investigated using a simple and novel method known as the new approach of Taylor series method.Using this method, we obtain the concentrations as follows (Appendix-B)

$$u(x) = \frac{\cosh mx}{\cosh m} \tag{20}$$

$$v(x) = \sinh n\,(x^2 - x) \tag{21}$$

Here the $u(0) = 1/\cosh m$ can be obtained from the below equation.

$$u(0) + \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3}\frac{1}{4!} + \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5}\frac{1}{6!} = 1 \tag{22}$$

The following equation (23) can be used to determine unknown constant $v'(0) = -$

$$v'(0) - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \alpha'^2 v'(0)\frac{1}{3!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{1}{4!} + (\alpha'^2)^2 v'(0)\frac{1}{5!} = 0 \tag{23}$$

Now current can obtained as follows:

$$\psi_S = \frac{I_S}{2FAD_{H_2Q}} = \left(\frac{dv}{dx}\right)_{x=0} = -n \tag{24}$$

$$\psi_P = \frac{I_P}{2FAK\Sigma} == \int_0^1 v\,dx = \frac{\sqrt{\pi}e^{-n/4}\left\{erfi\left(\sqrt{\frac{n}{4}}\right) - e^{n/2}\left(erf\left(\sqrt{\frac{n}{4}}\right)\right)\right\}}{2\sqrt{n}} \tag{25}$$

3.3 Validation of analytical results with numerical simulation

The numerical method offers an approximate solution to a mathematical problem. It is also useful to validate the analytical method. We solved the initial-boundary value problems for nonlinear differential equations (8) and (9)numerically using the MATLAB function pdex1.The simulation results solution is compared with our analytical results using new approach of Taylor series methodin Figures 2-3. Upon comparison, it gives a satisfactory agreement for all values of the parametersγ (dimensionless activation energy), β (dimensionless heat ofreaction), and Φ (Thiele modulus).

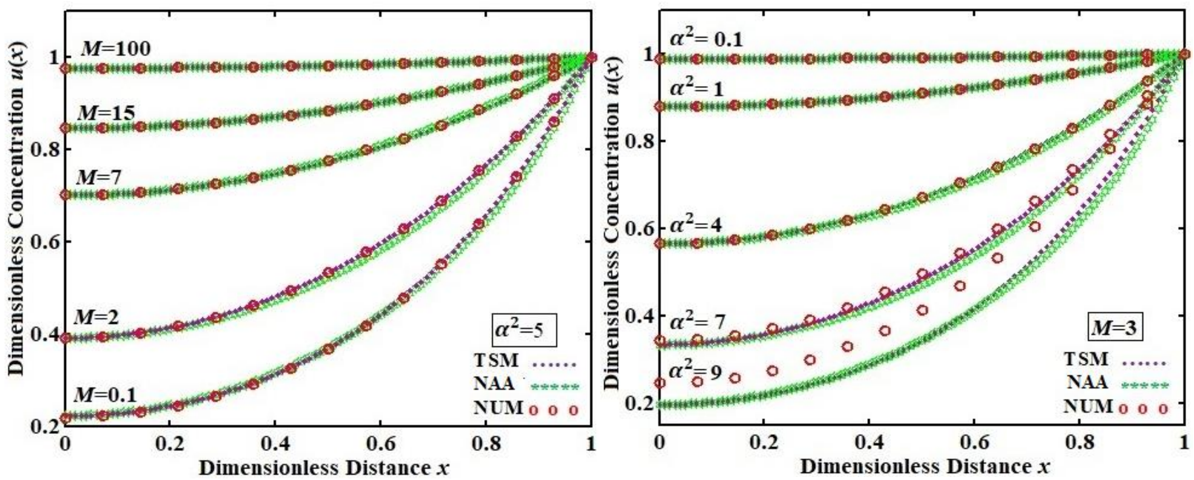

Figure 2. Comparison of analytical expression concentration of glucose with numerical simulation results for different values of dimensionless reaction diffusion parameter α and M.

Figures 2(a)-2(c) show the normalised non-steady state glucose concentration versus dimensionless distance X for various values of the dimensionless reaction diffusion parameter αandα' The concentration of substrate increases whenαdecreases and M increases, as shown in the graph. From this Figures 1(a)–1(c), it is evident that the value of concentration is uniform when α< 0.5. 0r M>100.

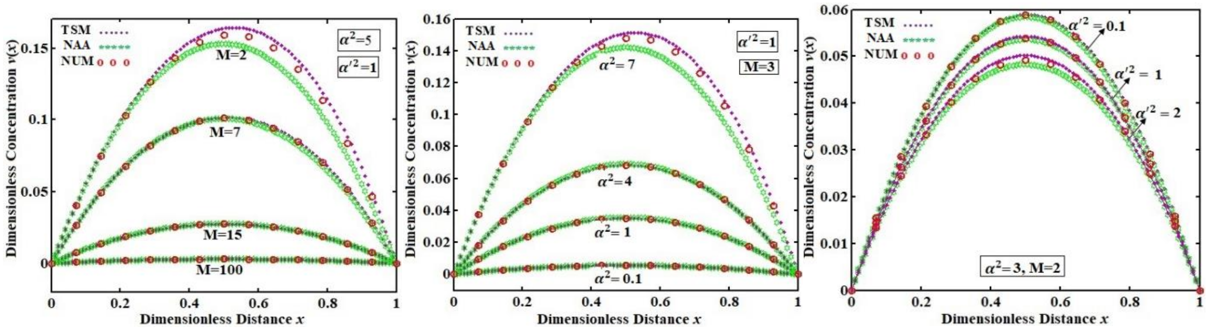

Figure 3. Comparison of analytical expression concentration of benzoquinone V with numerical simulation results for different values of dimensionless reaction diffusion parameter α and M.

Figures 3(a)-3(c) show the concentration of mediator V versus dimensionless distance X for different values of the dimensionless diffusion parameters α' and M. According to this graph, the value of the concentration of mediator decreases abruptly when M and α' increase. For all other parameters, the concentration of mediator reaches a maximum at the middle of the membrane (x = 0.5).

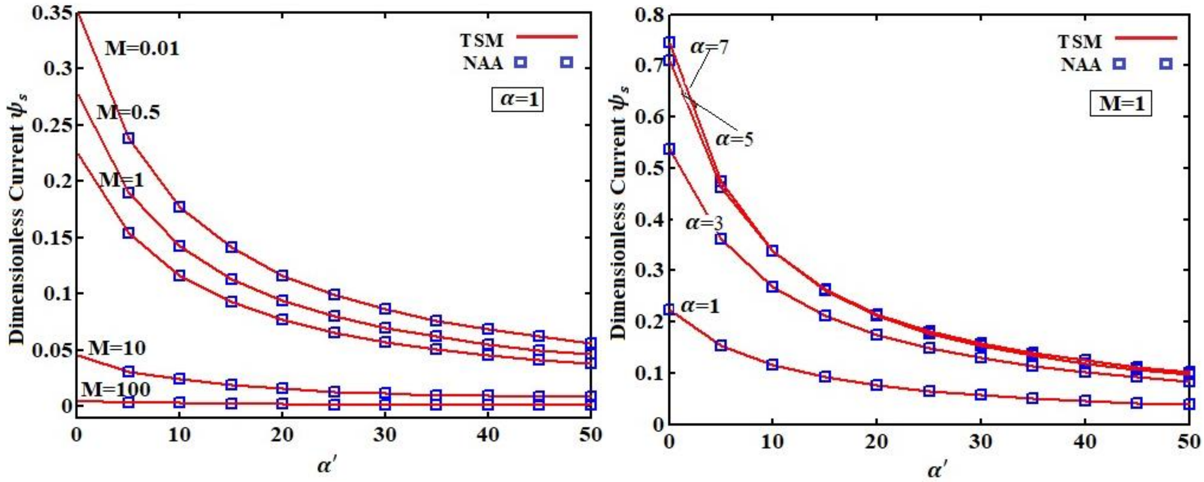

Figure 4. Comparison of normalized steady-state current ψ_S versus the diffusion dimensionless parameter α'

The normalized steady-state current $\psi_S(\alpha, \alpha', M)$ as the function of the dimensionless parameter α, α', and M' is given in Figures 4(a)–4(b). It is clear from these figures that the current values decrease slowly and reach a constant value when $\alpha' \geq 40$. When Mincreases, the current values also decrease.

4. CONCLUSIONS

We have derived approximate analytical expression concentration profiles and current of amperometric glucose biosensor based on immobilized enzymes for steady-state conditions over a wide range of parameters. A nonlinear time-independent differential equation has been solved using the Taylors series and Akbari-Ganji method. The effects of the reaction and diffusion parameters on concentration and current are discussed. The numerical results from the Matlab software are used to validate these analytical results. A satisfactory agreement is noted. In summary, the obtained analytical expressions for the concentration and current are reliable. Therefore, they can be applied to other enzyme-based electrochemical tissue-based biosensors with complex boundary conditions. This method can be used for solving differential in different fields of study such as solid mechanics, fluid mechanics, heat transfer, etc

DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

ACKNOWLEDGEMENT

The authors express their gratitude to Shri J. Ramachandran, Chancellor, Col. Dr. G. Thiruvasagam, Vice-Chancellor and Dr. M. Jayaprakashvel, Registrar, Academy of Maritime Education and Training (AMET), Deemed university, Chennai, Tamil Nadu for their continuous encouragement.

Appendix A: *Approximate analytical solution of nonlinear Eqns. (7) and (8) using Taylor's series method.*

The Taylor's **s**eries solution of Eqns.(7) and (8) are

$$u(x) = u(0) + u'(0)\frac{x}{1!} + u''(0)\frac{x^2}{2!} + u'''(0)\frac{x^3}{3!} + \cdots \tag{A1}$$

$$v(x) = v(0) + v'(0)\frac{x}{1!} + v''(0)\frac{x^2}{2!} + v'''(0)\frac{x^3}{3!} + \cdots \tag{A2}$$

Let us assume that $u(0)$is constant. From the boundary condition (9) we get

$$u'(0) = 0 \tag{A3}$$

Now Eqn.(7) can be rewritten at x=0 as follows:

$$u''(0) = \frac{\alpha^2 u(0)}{Mu(0)+1} \tag{A4}$$

Differentiate the equation (7) successively with respect to *x and* substituting $x = 0$ *we get*

$$u'''(0) = 0 \tag{A5}$$

$$u^{(iv)}(0) = \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} \tag{A6}$$

$$u^{(v)}(0) = 0 \tag{A7}$$

$$u^{(vi)}(0) = \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5} \tag{A8}$$

Putting these in Taylors series, we get the concentration of glucose as follows:

$$u(x) = u(0) + \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{x^2}{2!} + \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3}\frac{x^4}{4!} + \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5}\frac{x^6}{6!} \quad \text{(A9)}$$

Substitute the initial condition($u(1) = 1$) we get

$$1 = u(0) + \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3}\frac{1}{4!} + \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5}\frac{1}{6!} \quad \text{(A10)}$$

Similarly we can assume that $v'(0)$is constant.

Now Eq.(9) can be rewritten as follows:

$$v''(0) = -\frac{\alpha^2 u(0)}{Mu(0)+1} \quad \text{(A11)}$$

Differentiating the equation (9) successively and substituting ,we get

$$v'''(0) = \alpha'^2 v'(0) \quad \text{(A12)}$$

$$v^{(iv)}(0) = -\left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right) \quad \text{(A13)}$$

$$v^{(v)}(0) = (\alpha'^2)^2\, v'(0) \quad \text{(A14)}$$

The concentration of product using Taylor's series is

$$v(x) = v'(0)\frac{x}{1!} - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{x^2}{2!} + \alpha'^2 v'(0)\frac{x^3}{3!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{x^4}{4!} + (\alpha'^2)^2\, v'(0)\frac{x^5}{5!}$$

(A15)

Substitute the initial condition $v(1) = 0$ we get

$$0 = v'(0) - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \alpha'^2 v'(0)\frac{1}{3!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{1}{4!} + (\alpha'^2)^2\, v'(0)\frac{1}{5!}$$

(A16)

Appendix B: *Approximate analytical solution of nonlinear eqns. (8) and (9 using new approach of Taylors series method.*

Equations (8) and (9) can be rewritten as follows:

$$u''(x) - \frac{\alpha^2 u(x)}{Mu(x)+1} = 0 \quad \text{(B1)}$$

$$v''(x) + \frac{\alpha^2 u(x)}{Mu(x)+1} = \alpha'^2 v(x) \quad \text{(B2)}$$

where u and v are the dimensionless concentration of S and H_2Q . The transformed boundary conditions are

$$\frac{du}{dx} = 0, v = 0 \text{ when } x = 0 \quad \text{(B3)}$$

$$u = 1, v = 0 \text{ when } x = 1 \tag{B4}$$

Assume that the solution of eqn. (B1) is

$$u(x) = A\cosh mx + B\sinh mx \tag{B5}$$

Using the boundary conditions (B3) and (B4) we get

$$u(x) = \frac{\cosh mx}{\cosh m} \tag{B6}$$

Where $u(0) = \frac{1}{\cosh m}$ which can be from obtained by solving the below equation .

$$1 = u(0) + \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3}\frac{1}{4!} + \frac{(\alpha^2)^3 u(0)(1-6(\alpha^2)u(0))}{(Mu(0)+1)^5}\frac{1}{6!} \tag{B11}$$

Similarly we assume the solution of the equation (B2) as

$$v(x) = \sinh n\,(x^2 - x) \tag{B12}$$

where $v'^{(0)} = -n$ can be from obtained by solving the below equation.

$$0 = v'(0) - \frac{\alpha^2 u(0)}{Mu(0)+1}\frac{1}{2!} + \alpha'^2 v'(0)\frac{1}{3!} - \left(\frac{(\alpha^2)^2 u(0)}{(Mu(0)+1)^3} + \frac{(\alpha^2\alpha'^2 u(0))}{(M\,u(0)+1)}\right)\frac{1}{4!} + (\alpha'^2)^2\, v'(0)\frac{1}{5!} \tag{B13}$$

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