

Application of He's variational iteration method in nonlinear boundary value problems in enzyme–substrate reaction diffusion processes: part 1. The steady-state amperometric response

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Abstract A mathematical model of amperometric biosensors has been developed. In this paper, He's variational iteration method is implemented to give approximate and analytical solutions of non-linear reaction diffusion equations containing a non linear term related to Michaelis–Menten kinetic of the enzymatic reaction. The variational iteration method which produces the solutions in terms of convergent series, requiring no linearization or small perturbation. These analytical results are compared with available limiting case result and are found to be in good agreement.

Keywords Variational iteration method · Reaction diffusion system · Enzyme kinetics · Nonlinear equation

1 Introduction

Considerable advances have been made during the last decade in the development of polymer-based materials for use as electrocatalysis and as chemical and biological sensors operating in the batch amperometric mode [1, 2]. Useful summaries of recent advances in this area has been provided by Hillman [3], Lyons [4–6], Evans [7], Wring and Hart [8], Murray [9] Albery [10–13], Bartlett [14, 15] and Rahamathunissa and Rajendran [16]. Various simplified analytical models describing electrocatalysis at electroactive polymer films have been developed over the last 20 years. In brief, the analysis involves the construction and solution of reaction/diffusion differential equations, resulting in the development of approximate analytical expressions for the amperometric current response. The analysis is not simple since one is concerned with the modeling of reaction/diffusion processes in the films (mathematically,

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this translates to reaction/diffusion with in the finite diffusion space). In many cases, addition of the chemical reaction term to the Fick's diffusion term during formulation of the differential equation results in the generation of a non-linear expression which is not readily solved using standard analytical methods.

The investigation of exact solution of nonlinear equation is interesting and important. In the past several decades, many authors mainly had paid attention to study solution of nonlinear equations by using various methods, such as Backlund transformation [17], Darboux transformation [18], Inverse scattering method [19], Bilinear method [20], the tanh method [21], the sine–cosine method [22], the homogeneous balance method [23] and variational iteration method [24,25] etc.

The variational iteration method [24–28] has been extensively worked out over a number of years by numerous authors. The VIM was first proposed by He [24,29] and was successfully applied to autonomous ordinary differential equations in [30] to nonlinear polycrystalline solids [31] and other fields. This method has been proved by many authors to be a powerful mathematical tool for various kinds of nonlinear problems. It is a promising and evolving method. Besides its mathematical importance and its links to other branches of mathematics, it is widely used in all ramifications modern sciences [32]. In this method the solution procedure is very simple by means of variational theory and only few iterations lead to high accurate solutions which are valid for the whole solution domain.

The purpose of this paper is to derive steady state analytical solution of concentration for at polymer modified electrode for all values α and K using variational iteration method. The chosen configuration is the most used in the design of nowadays enzymatic biosensor realizations such as the use of polymeric matrices as an enzyme support and the mass production of biosensors by the screen printing technique [33].

2 Mathematical formulation of the problem

The enzyme kinetics in biochemical systems have usually been modeled by ordinary differential equations which are based only on reaction without spatial dependence of the various concentrations. Recent attention has been given to the effect of diffusion in the process of interactions [34,35]. When this effect is taken into consideration, the various concentrations in the reaction process are spatially dependent and the equations governing these concentrations become partial differential equations of parabolic type [34]. In an irreversible monoenzyme system the reaction scheme for free enzyme E and substrate concentration S may be expressed by



where ES is the enzyme–substrate complex and P is the product. Suppose that the reaction-diffusion takes place in an arbitrary n -dimensional medium Ω (membrane), where Ω is a bounded domain in R^n ($n = 1, 2, \dots$). Then the rate of change of substrate concentration $S = S(t, \chi)$ at time t , position $\chi \in \Omega$ is equal to the sum of the rate due to reaction and diffusion, and is given by Pao[34]

$$\partial S/\partial t = D_S \nabla \cdot (\nabla S) - v(t, \chi) \tag{2}$$

where D_S is the substrate diffusion coefficient, ∇ is the gradient operation and v is the so-called “initial reaction velocity”. Various models regarding the expression for v are formulated by researchers in this field. In this paper, we discuss some mathematical properties of the solutions for type of such models using Michaelis–Menton hypothesis. Based on the Michaelis hypothesis, the velocity function v for the simple reaction process without competitive inhibition is given by Pao [34] and Baronas et al. [35]

$$v(t, \chi) = k_2 E_0 S / (K_M + S) \tag{3}$$

where E_0 is the total amount of enzyme and K_M is the “Michaelis constant”. In this model, the equation for S becomes

$$\partial S/\partial t - D_S \nabla \cdot (\nabla S) = -k_2 E_0 S / (K_M + S) \quad (t > 0, \chi \in \Omega). \tag{4}$$

In one dimensional form Eq. 4 can be written as

$$\frac{\partial S}{\partial t} = D_S \frac{\partial^2 S}{\partial \chi^2} - \frac{k_2 E_0 S}{K_M + S} \tag{5}$$

Introducing a pseudo-first order rate constant $K = k_2 E_0 / K_M$, we can write the above equation as

$$\frac{\partial S}{\partial t} = D_S \frac{\partial^2 S}{\partial \chi^2} - \frac{K S}{1 + S/K_M} \tag{6}$$

Here we consider, an initial condition is given in the usual form,

$$S(0, \chi) = s_0(\chi) \quad (\chi \in \Omega) \tag{7}$$

The system governs the substrate concentration S when there is no competitive inhibition in the reaction. We make the non-linear PDE (Eq. 6) dimensionless by defining the following parameters:

$$u = s/k_s^\infty, \quad x = \chi/L, \quad \tau = D_S t/L^2, \quad K = kL^2/D_S = \phi^2, \quad \alpha = ks^\infty/K_M \tag{8}$$

where u , x and τ represent dimensionless concentrations, distance and time, respectively. Here α denotes a saturation parameter and K denotes reaction diffusion parameter. Now the Eq. 6 reduces to the following dimensionless form:

$$\frac{\partial u}{\partial \tau} = \frac{\partial^2 u}{\partial x^2} - \frac{Ku}{1 + \alpha u} \quad 0 < u \leq 1 \tag{9}$$

whereas the initial condition reduces to

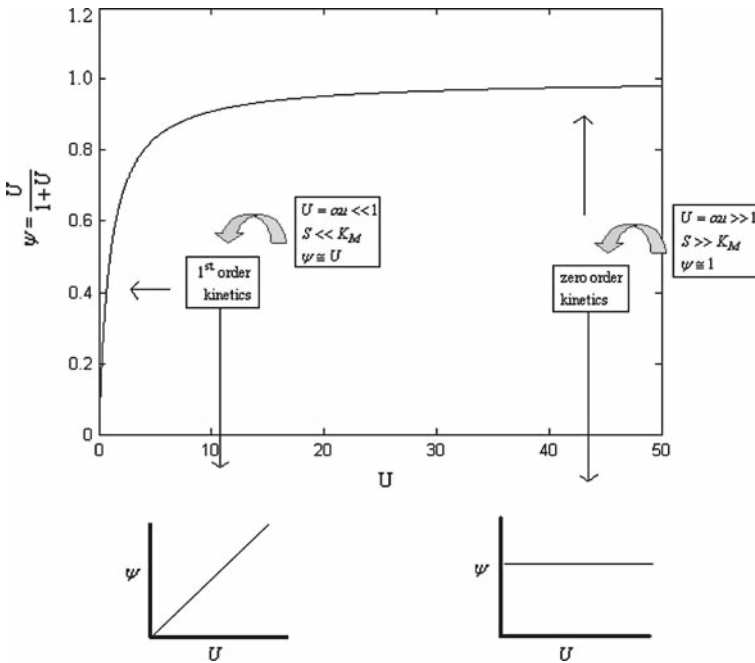


Fig. 1 Diagrammatic representation of saturated (zero order kinetics) and unsaturated (1st order kinetics) catalytic kinetics

$$u(x = 0) = a \text{ (constant)} \tag{10}$$

Lyons and co-workers [1] solved the above equations only for the limiting cases ($\alpha u \ll 1$ and $\alpha u \gg 1$ (refer Fig. 1) using Dirichlet and Neumann boundary conditions. But we wish to obtain an analytical expression for the concentration profile $u(x)$ of substrate for all values of α . In steady state, $\frac{\partial u}{\partial \tau} = 0$. In this case the steady state diffusion Eq. 9 takes the form

$$\frac{\partial^2 u}{\partial x^2} - \frac{Ku}{1 + \alpha u} = 0 \tag{11}$$

Again, this is a non-linear differential equation. Now the boundary condition (Eq. 10) is [1]

$$u(0) = a = \text{Sech}(\sqrt{K}) \text{ for } \alpha \ll 1 \tag{12a}$$

$$= 1 - K/2\alpha \text{ for } \alpha \gg 1 \tag{12b}$$

The non-linear Eq. 11 is solved for the above boundary conditions using variational iteration method. The variational iteration method proposed by He [24,29] has been successfully applied to finding the solution of differential equation in closed form. The basic concept of variational iteration method is summarized briefly here for completeness.

3 Basic concepts in the variational iteration method

To illustrate the basic concepts of variational iteration method (VIM), we consider the following nonlinear partial differential equation:

$$L [u(x)] + N [u(x)] = g(x) \tag{13}$$

where L is a linear operator, N is a nonlinear operator, and $g(x)$ is a given continuous function [24,29]. According to the variational iteration method, we can construct a correct functional as follows:

$$u_{n+1}(x) = u_n(x) + \int_0^x \lambda [L [u_n(\tau)] + N[\tilde{u}_n(\tau)] - g(\tau)]d\tau \tag{14}$$

where λ is a general Lagrange multiplier [24,29] which can be identified optimally via variational theory, u_n is the n th approximate solution, and \tilde{u}_n denotes a restricted variation, i.e., $\delta\tilde{u}_n = 0$.

4 Solution of boundary value problem

Using above variation iteration method we can write the correction functional of Eq. 11 as follows

$$u_{n+1}(x) = u_n(x) + \int_0^x \lambda \left[u_n''(s) - \frac{\hbar\hbar\hbar\hbar}{Ku_n(s)} \right] ds \tag{15}$$

Taking variation with respect to the independent variable u_n , noticing that $\delta u_n(0) = 0$

$$\delta u_{n+1}(x) = \delta u_n(x) + \delta \int_0^x \lambda \left[u_n''(s) - \frac{\hbar\hbar\hbar\hbar}{Ku_n(s)} \right] ds \tag{16}$$

For all variational δu_n and $\delta u'_n$, implying the following stationary conditions

$$\delta u_n : 1 - \lambda'(s)|_{s=x} = 0 \tag{17a}$$

$$\delta u'_n : \lambda(s)|_{s=x} = 0 \tag{17b}$$

$$\delta u_n : \lambda''(s)|_{s=x} = 0 \tag{17c}$$

The Lagrange multiplier can be identified as

$$\lambda(s) = s - x \tag{18}$$

Substituting the Lagrange multiplier in the iteration formula (Eq. 15) we get the following approximation

$$u_1(x) = a + \frac{Ka}{(1 + \alpha a)}x^2 \quad (19)$$

$$u_2(x) = a + \frac{Kx^2}{2\alpha} + \frac{(1 + \alpha a)}{a\alpha^2} \ln \left[\frac{Ka\alpha}{2(1 + \alpha a)^2}x^2 + 1 \right] - \frac{\sqrt{2K}}{\alpha^{3/2}\sqrt{a}}x \tan^{-1} \left[\frac{\sqrt{Ka\alpha}}{\sqrt{2}(1 + \alpha a)}x \right] \quad (20)$$

$$u_3(x) = u_2(x) + \frac{K}{\alpha} \left[-\frac{x^2}{2} - \frac{(1 + \alpha a)}{2Ka\alpha} \left\{ -\frac{2(1 + \alpha a)^2}{(Ka\alpha x^2 + 2(1 + \alpha a)^2)} + \frac{3}{2} - \frac{1}{2} \ln \left(\frac{Ka\alpha x^2}{2(1 + \alpha a)^2} + 1 \right) \right\} + \frac{x^2(1 + \alpha a)}{(Ka\alpha x^2 + 2(1 + \alpha a)^2)} \right] - \int_0^x (s - x) \frac{Ku_2(s)}{1 + \alpha u_2(s)} ds \quad (21)$$

Last term in the Eq. 21 is not integrated. Hence $u_3(x)$ is not expressed in the closed form. Therefore we are taking $u_2(x) = u(x)$. Equation 20 represents the most general approximate new analytical expression for the substrate concentration profiles for all values of α and K . The time independent concentration $u(x)$ using Eq. 20 are represented in Figs. 2 and 3 for various values of α and K .

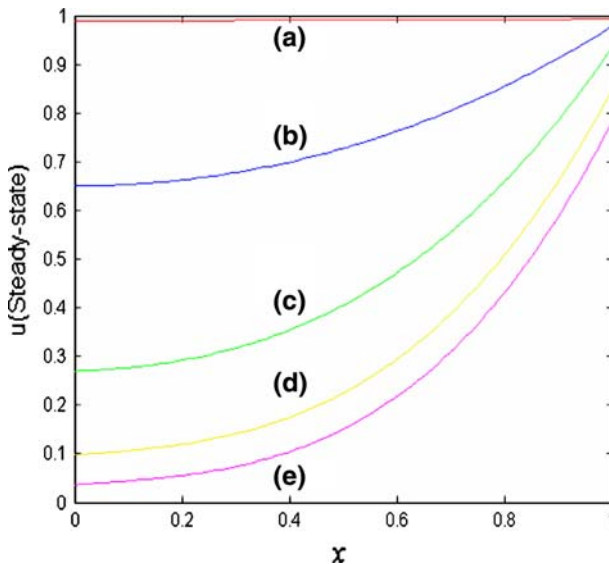


Fig. 2 Plot of normalized steady state substrate concentration u versus normalized distance x when $\alpha u \ll 1$ (Here $\alpha = 0.1$) for various values of K using Eq. 20. (a) $K = 0.01$; (b) $K = 1$; (c) $K = 4$; (d) $K = 9$; (e) $K = 16$

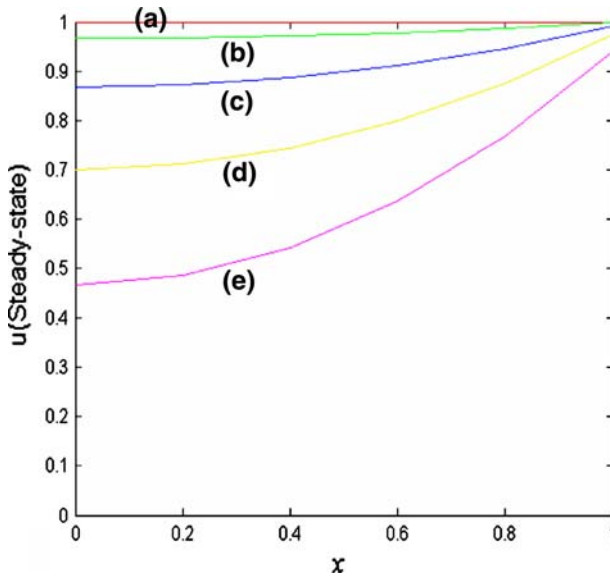


Fig. 3 Plot of normalized steady state substrate concentration u versus normalized distance x when $\alpha u \gg 1$ (Here $\alpha = 15$) various values of K using Eq. 20. (a) $K = 0.01$; (b) $K = 1$; (c) $K = 4$; (d) $K = 9$; (e) $K = 16$

5 Limiting cases

(1) *unsaturated (first order) catalytic kinetics*

In this cases, the substrate concentration in the film $S(\chi)$ is less than the Michaelis constant K_M . This is explained in Fig. 1. When $\alpha u \ll 1$, the Eq. 11 reduces to

$$\frac{\partial^2 u}{\partial x^2} - Ku = 0 \tag{22}$$

Using the He’s variational iteration method and boundary condition (12a), its correction functional can be written in the form

$$u_{n+1}(x) = u_n(x) + \int_0^x \lambda \left[u_n''(s) - \frac{hu_n(s)}{K} \right] ds \tag{23}$$

By the same manipulation, the multiplier ($\lambda(s) = s - x$) can be identified and the following iteration formula can be obtained as

$$u_{n+1}(x) = u_n(x) + \int_0^x (s - x) \left[u_n''(s) - \frac{hu_n(s)}{K} \right] ds \tag{24}$$

Now $u_1(x)$, $u_2(x)$, $u_3(x)$ becomes,

$$u_1(x) = a \left[1 + \frac{Kx^2}{2} \right] \quad (25a)$$

$$u_2(x) = a \left[1 + \frac{Kx^2}{2!} + \frac{(Kx^2)^2}{4!} \right] \quad (25b)$$

$$u_3(x) = a \left[1 + \frac{Kx^2}{2!} + \frac{(Kx^2)^2}{4!} + \frac{(Kx^2)^3}{6!} \right] \quad (25c)$$

$$u_n(x) = a \sum_{m=0}^n \frac{(Kx^2)^m}{2m!} \quad (25d)$$

The solution of $u(x)$ in a closed form is

$$u(x) = a \operatorname{Cosh}(\sqrt{K}x) \quad (25)$$

The Eq. 25 derived by us is identical with Eq. 10 in Ref. [1].

(2) *saturated (zero order) catalytic kinetics*

In this cases, the substrate concentration in the film $S(\chi)$ is greater than the Michaelis constant K_M (Refer Fig. 1). Hence $\alpha u \gg 1$ reduces the Eq. 11 to

$$\frac{\partial^2 u}{\partial x^2} - \frac{K}{\alpha} = 0 \quad (26)$$

Using He's variational iteration method and using the boundary condition (12b) its correction functional can be written in the form

$$u_{n+1}(x) = u_n(x) + \int_0^x \lambda [u_n''(s) - K/\alpha] ds \quad (27)$$

By the same manipulation, the multiplier ($\lambda(s) = s - x$) can be identified and the following iteration formula can be obtained as

$$u_{n+1}(x) = u_n(x) + \int_0^x (s - x) [u_n''(s) - K/\alpha] ds \quad (28)$$

We start with initial approximation given by Eq. (12b) and by the above iteration formula, we can obtain the Eq. (29) in a closed form

$$u(x) = a + \frac{K}{2\alpha} x^2 \quad (29)$$

Table 1 Numerical values of steady state concentration calculated using Eqs. 20 and 25 for various values x and K when $\alpha = 0.1$ (Case 1)

x	$K = 0.01$			$K = 1$			$K = 4$			$K = 9$			$K = 16$		
	Eq. 20	Eq. 25	% deviation of Eq. 20	Eq. 20	Eq. 25	% deviation of Eq. 20	Eq. 20	Eq. 25	% deviation of Eq. 20	Eq. 20	Eq. 25	% deviation of Eq. 20	Eq. 20	Eq. 25	% deviation of Eq. 20
0	0.995	0.9950	0.00	0.6481	0.6481	0.00	0.2658	0.2658	0.00	0.0993	0.0993	0.00	0.0366	0.0366	0.00
0.2	0.9951	0.9952	-0.01	0.6603	0.6611	-0.12	0.2861	0.2874	-0.45	0.1172	0.1177	-0.43	0.0521	0.0490	5.95
0.4	0.9955	0.9958	-0.03	0.6975	0.7006	-0.44	0.3501	0.3555	-1.54	0.1770	0.1798	-1.58	0.1059	0.0944	10.86
0.6	0.9961	0.9968	-0.07	0.7608	0.7682	-0.97	0.4671	0.4813	-3.04	0.2974	0.3087	-3.80	0.2201	0.2035	7.54
0.8	0.9970	0.9982	-0.12	0.8525	0.8667	-1.67	0.6526	0.6851	-4.98	0.5088	0.5520	-8.49	0.4308	0.4499	-4.43
1	0.9982	1	-0.18	0.9754	1	-2.52	0.9276	1	-7.81	0.8531	1	-17.22	0.7882	1	-26.87
Average			-0.09	Average		-1.26	Average		-3.90	Average		-8.61	Average		-13.44

Table 2 Numerical values of steady state concentration calculated using Eqs. 20 and 29 for various values x and K when $\alpha = 15$. (Case 2)

x	$K = 0.01$			$K = 1$			$K = 4$			$K = 9$			$K = 16$		
	Eq. 20	Eq. 29	% deviation of Eq. 20	Eq. 20	Eq. 29	% deviation of Eq. 20	Eq. 20	Eq. 29	% deviation of Eq. 20	Eq. 20	Eq. 29	% deviation of Eq. 20	Eq. 20	Eq. 29	% deviation of Eq. 20
0	0.9997	0.9997	0	0.9667	0.9667	0.00	0.8667	0.8667	0.00	0.7	0.7	0.00	0.4667	0.4667	0.00
0.2	0.9997	0.9997	0	0.9679	0.968	-0.01	0.8717	0.872	-0.03	0.7120	0.712	0.00	0.4854	0.488	-0.54
0.4	0.9997	0.9997	0	0.9717	0.972	-0.03	0.8865	0.888	-0.17	0.7439	0.748	-0.55	0.5416	0.552	-1.92
0.6	0.9998	0.9998	0	0.9779	0.9787	-0.08	0.9113	0.9147	-0.37	0.7988	0.808	-1.15	0.6358	0.6587	-3.60
0.8	0.9999	0.9999	0	0.9866	0.988	-0.14	0.9460	0.952	-0.63	0.8759	0.892	-1.84	0.7687	0.808	-5.11
1	1	1	0	0.9979	1	-0.21	0.9907	1	-0.94	0.9753	1	-2.53	0.9407	1	-6.30
Average			0	Average		-0.11	Average		-0.47	Average		-1.27	Average		-3.15

This equation is identical with Eq. 11 of Ref. [1]. This limiting case results, Eq. 25 (for $\alpha \ll 1$) and Eq. 29 (for $\alpha \gg 1$) are compared with our main result Eq. 20. Tables 1 and 2 indicates the dimensionless substrate concentration evaluated using Eq.20 with the limiting case results. The average relative difference between our results (Eq.20) and limiting case results are $-0.09, -1.26, -3.90, -8.61, -13.44\%$ (for $\alpha \ll 1$) and $0, -0.11, -0.47, -1.27, -3.15\%$ (for $\alpha \gg 1$) when $K = 0.01, 1, 4, 9$ and 16 , respectively.

6 Analysis of moving boundary

Considering the limiting situations for totally unsaturated kinetics when $\alpha u \ll 1$ and saturated kinetics when $\alpha u \gg 1$. The outer part of the region is saturated (region RII) and the inner region (region RI) remains unsaturated. This is illustrated in Fig. 4. Considering a situation where the reaction kinetics are fast compared with substrate diffusion. A moving normalized distance parameter x^* is defined the boundary between regions RI and RII. The substrate S diffuses into the film a reaction front is established at $x = x^*$. When $x^* = 0$, the entire region is saturated and when $x^* = 1$, the entire region is unsaturated. In RI, $\alpha u \ll 1$ and in RII $\alpha u \gg 1$. When $x = x^*$, $u = 1/\alpha$. Now the Eq. 20 becomes,

$$\frac{1}{\alpha} = a + \frac{Kx^{*2}}{2\alpha} + \frac{(1 + \alpha a)}{\alpha\alpha^2} \ln \left[\frac{K\alpha\alpha}{2(1 + \alpha a)^2} x^{*2} + 1 \right] - \frac{\sqrt{2K}}{\alpha^{3/2}\sqrt{a}} x^* \tan^{-1} \left[\frac{\sqrt{K\alpha\alpha}}{\sqrt{2}(1 + \alpha a)} x^* \right] \tag{30}$$

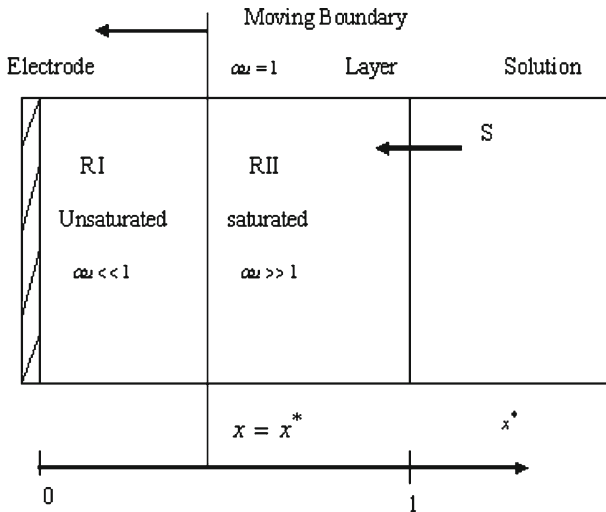


Fig. 4 The description of two region approach used to obtain Eq.32 which corresponds the situation of moving boundary. The inner region RI is unsaturated whereas the outer region RII is saturated. The line between these two regions is set at some value x^* defined in Eq.32. Complete saturation occurs when $x^* = 0$

When $\frac{K\alpha\alpha}{2(1+\alpha a)^2}$ is small, we obtain,

$$\frac{K^2 a \alpha}{12(1 + \alpha a)^3} x^{*4} - \frac{K a \alpha}{2(1 + \alpha a)} x^{*2} - \alpha a + 1 = 0 \quad (31)$$

Solving the above equation to obtain x^*

$$x^* = \left[\frac{3(1 + \alpha a)^2}{K} \left[1 \pm \frac{1}{\sqrt{\alpha a}} \sqrt{\frac{3\alpha^2 a^2 + 7\alpha a - 4}{3(1 + \alpha a)}} \right] \right]^{1/2} \quad (32)$$

Equation 32 describes the position of the boundary as it moves through the film.

7 Conclusion

A nonlinear time independent partial differential equation has been formulated and solved using He's variational iteration method. The primary result of this work is first accurate calculation of substrate concentration for all values of α and K . It gives good agreement with previous published limiting case results. The extension of the procedure to other two dimension and three dimension geometries with various complex boundary conditions seems possible.

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