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Research Article

Mathematical Modeling and Analysis of Nonlinear Enzyme Catalyzed Reaction Processes

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A mathematical model for the nonlinear enzymatic reaction process is discussed. An approximate analytical expression of concentrations of substrate, enzyme, and free enzyme-product is obtained using homotopy perturbation method (HPM). The main objective is to propose an analytical solution, which does not require small parameters and avoid linearization and physically unrealistic assumptions. Theoretical results obtained can be used to analyze the effect of different parameters. Satisfactory agreement is obtained in the comparison of approximate analytical solution and numerical simulation.

1. Introduction

The importance of biocatalytic processes and reactions for organic synthesis and the pharmaceutical food and cosmetics industry has been constantly growing during the last few years [1, 2]. From a synthetic point of view, enzymes are highly efficient catalysts for an extremely broad palette of reactions [3]. Enzymes of one type, but from different origins, are specialized for substrates, positions in substrates, and products [4]. Enzyme reactions do not follow the law of mass action directly. The rate of the reaction only increases to a certain extent as the concentration of substrate increases. The maximum reaction rate is reached at high substrate concentration due to enzyme saturation. This is in contrast to the law of mass action that states that the reaction rate increases as the concentration of substrate increases [5]. Various simplified analytical models 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 [6, 7] nonlinear enzyme catalyzed reaction processes.

The simplest model that explains the kinetic behaviour of enzyme reactions is the classic 1913 model of Michaelis and Menten [8] which is widely used in biochemistry for many types of enzymes. The Michaelis-Menten model is based on the assumption that the enzyme binds the substrate to form an intermediate complex which then dissociates to form the final product and release the enzyme in its original form. The schematic representation of this two-step process is given by

$$\mathrm{E} + \mathrm{S} \underset{k_{-1}}{\overset{k_1}{\leftrightarrow}} \mathrm{C} \xrightarrow{k_2} \mathrm{E} + \mathrm{P}, \tag{1}$$

where k_1, k_{-1}, and k_2 are constant parameters associated with the rates of the reaction. Note that it is generally assumed that the second step of the reaction equation (1) is irreversible. In reality, this is not always the case. Typically, reaction rates are measured under the condition that the product is continually removed, which prevents the reverse reaction of the second step from occurring effectively.

In this paper we have derived an expression for concentration of substrate, enzyme-substrate, and free enzyme-product with nonmechanism based enzyme inactivation, in terms of dimensionless reaction diffusion parameters ε, λ_1, λ_2, and λ_3 using homotopy perturbation method (HPM). Comparative study of the same with numerical simulation is presented.

2. Mathematical Formulation of the Problems

If a small amount of enzyme is used and all but one substrate is kept constant, then the rate of the enzymatically catalyzed

reaction depends on the substrate concentration and initial rate as in the equation $v = v_{\max}[S]/(K_M + [S])$, given by [9], where K_M is the Michaelis constant: $K_M = (k_{-1} + k_2)/k_1$. The typical notation of the enzyme catalyzed reaction with one substrate can be given as [10]

$$A + E \underset{k_2}{\overset{k_1}{\leftrightarrow}} X \underset{k_4}{\overset{k_3}{\leftrightarrow}} P + E, \tag{2}$$

where A is substrate, E is enzyme, X is enzyme-substrate complex, and P is free enzyme product. The kinetic equations consist of

$$\frac{d[A]}{dt} = k_2[X] - k_1[A][E], \tag{3a}$$

$$\frac{d[E]}{dt} = (k_2 + k_3)[X] - (k_1[A] + k_4[P])[E], \tag{3b}$$

$$\frac{d[P]}{dt} = k_3[X] - k_4[P][E], \tag{3c}$$

with a conservation relation given in [6]:

$$[E] + [X] = [E]_{\text{total}}. \tag{4}$$

It is obvious that the derivative of a substrate with respect to time gives the rate. Thus, the rate is a function of compounds [V], [A] (intracellular and extracellular), enzyme concentrations E and kinetic parameter k. However, the enzyme concentration is hidden in the kinetic constants in the parameter vector k; herewith, we can write v as a function of [V], [A], and [P]; that is, $v = v([A], [V], k)$ [11].

The more general form of (3a)–(3c) can be written in the form of

$$\frac{d[A]}{dt} = \nu_2 - \nu_1, \tag{5a}$$

$$\frac{d[E]}{dt} = \nu_2 + \nu_3 - \nu_1 - \nu_4, \tag{5b}$$

$$\frac{d[X]}{dt} = \nu_1 + \nu_4 - \nu_2 - \nu_3, \tag{5c}$$

$$\frac{d[P]}{dt} = \nu_3 - \nu_4. \tag{5d}$$

The form of rate equations is as follows:

$$\begin{aligned} \nu_1 &= k_1[A][E], \qquad \nu_2 = k_2[X], \\ \nu_3 &= k_3[X], \qquad \nu_4 = k_4[P][E]. \end{aligned} \tag{6}$$

The initial condition at $t = 0$ are as follows:

$$[A] = A_0, \qquad [E] = E_0, \qquad [X] = 0, \qquad [P] = 0. \tag{7}$$

The concentration of the reactants in (5a)–(5d) is denoted by lower case letters

$$s = [A], \qquad e = [E], \qquad c = [X], \qquad p = [P], \tag{8}$$

The law of mass action leads to the system of following nonlinear kinetic equations [12]:

$$\frac{ds}{dt} = -k_1 se + k_2 c, \tag{9a}$$

$$\frac{de}{dt} = -k_1 se + k_2 c + k_3 c - k_4 pe, \tag{9b}$$

$$\frac{dp}{dt} = k_3 c - k_4 pe, \tag{9c}$$

$$\frac{dc}{dt} = k_1 se - k_2 c - k_3 c + k_4 pe, \tag{9d}$$

with the boundary conditions being

$$s(0) = s_0, \qquad e(0) = e_0, \qquad p(0) = 0, \qquad c(0) = 0. \tag{10}$$

Adding (9b) and (9c), we get

$$\frac{de}{dt} + \frac{dc}{dt} = 0. \tag{11}$$

Using the initial conditions (10) we obtain

$$e(t) + c(t) = e_0. \tag{12}$$

With this, the system of ordinary differential equations reduces to the following three differential equations:

$$\frac{ds}{dt} = -k_1 s(e_0 - c) + k_2 c, \tag{13a}$$

$$\frac{dc}{dt} = k_1 s(e_0 - c) - (k_2 + k_3)c + k_4 p(e_0 - c), \tag{13b}$$

$$\frac{dp}{dt} = k_3 c - k_4 p(e_0 - c). \tag{13c}$$

With initial conditions $s(0) = s_0$, $c(0) = 0$, and $p(0) = 0$. By introducing the following set of nondimensional variables and parameters,

$$\tau = \frac{k_1 e_0 t}{\varepsilon}, \qquad u(\tau) = \frac{s(t)}{s_0}, \qquad v(\tau) = \frac{c(t)}{s_0}, \tag{14}$$

$$w(\tau) = \frac{p(t)}{s_0}, \qquad \lambda_1 = \frac{k_2}{k_1 s_0}, \qquad \lambda_2 = \frac{k_3}{k_1 s_0}, \tag{15}$$

$$\lambda_3 = \frac{k_4}{k_1}, \qquad \varepsilon = \frac{e_0}{s_0}, \qquad \eta = \lambda_1 + \lambda_2, \tag{16}$$

the system of (13a)–(13c) and the initial conditions (10) can be represented in dimensionless form as follows:

$$\frac{du}{d\tau} = -\varepsilon u + uv + \lambda_1 v, \tag{17a}$$

$$\frac{dv}{d\tau} = \varepsilon u - \eta v + \lambda_3 \varepsilon w - uv - \lambda_3 v, \tag{17b}$$

$$\frac{dw}{d\tau} = \lambda_2 v - \lambda_3 \varepsilon w + \lambda_3 v w, \tag{17c}$$

with

$$u(0) = 1, \qquad v(0) = 0, \qquad w(0) = 0. \tag{18}$$

3. Implementation of the HPM

We indicate how (33)–(35) in this paper are derived. To find the solution of (14)–(16), we first construct a homotopy as follows:

$$(1-p)\left[\frac{du}{d\tau}+\varepsilon u\right]+p\left[\frac{du}{d\tau}+\varepsilon u-uv-\lambda_1 v\right]=0, \quad (19)$$

$$(1-p)\left(\frac{dv}{d\tau}+\eta v\right) + p\left(\frac{dv}{d\tau}+\eta v-\varepsilon u-\lambda_3\varepsilon w+uv+\lambda_3 vw\right)=0, \quad (20)$$

$$(1-p)\left(\frac{dw}{d\tau}+\lambda_3\varepsilon w\right)+p\left(\frac{dw}{d\tau}+\lambda_3\varepsilon w-\lambda_2 v-\lambda_3 vw\right)=0. \quad (21)$$

And the initial approximations are as follows:

$$u(0)=1, \qquad v(0)=0, \qquad w(0)=0. \quad (22)$$

Approximate solutions of (33)–(35) are

$$u=u_0+pu_1+p^2u_2+p^3u_3+\cdots \quad (23)$$

$$v=v_0+pv_1+p^2v_2+p^3v_3+\cdots \quad (24)$$

$$w=w_0+pw_1+p^2w_2+p^3w_3+\cdots. \quad (25)$$

Substituting (23)–(25) into (19)–(21), respectively, and comparing the coefficients of like powers of p, we can obtain the following differential equations for the concentration of substrate:

$$p^0: \frac{du_0}{d\tau}+\varepsilon u_0=0,$$
$$p^1: \frac{du_1}{d\tau}+\varepsilon u_1-u_0v_0-\lambda_1 v_0=0, \quad (26)$$
$$p^2: \frac{du_2}{d\tau}+\varepsilon u_2-(u_0v_1+u_1v_0)-\lambda_1 v_1=0.$$

For enzyme substrate concentration v,

$$p^0: \frac{dv_0}{d\tau}+\eta v_0=0,$$
$$p^1: \frac{dv_1}{d\tau}+\eta v_1-\varepsilon u_0-\lambda_3\varepsilon w_0+u_0v_0+\lambda_3 v_0w_0=0, \quad (27)$$
$$p^2: \frac{dv_2}{d\tau}+\eta v_2-\varepsilon u_1-\lambda_3\varepsilon w_1+u_0v_1 + u_1v_0+\lambda_3(v_0w_1+v_1w_0)=0.$$

For product concentration w

$$p^0: \frac{dw_0}{d\tau}+\lambda_3\varepsilon w_0=0,$$
$$p^1: \frac{dw_1}{d\tau}+\lambda_3\varepsilon w_1-\lambda_2 v_0-\lambda_3 v_0w_0=0,$$
$$p^2: \frac{dw_2}{d\tau}+\lambda_3\varepsilon w_2-\lambda_2 v_1-\lambda_3(v_0w_1+v_1w_0)=0,$$
$$p^3: \frac{dw_2}{d\tau}+\lambda_3\varepsilon w_3-\lambda_2 v_2-\lambda_3(v_0w_2+v_1w_1+v_2w_0)=0. \quad (28)$$

Solving (26)–(28), and using the boundary conditions (22), we can find the following results:

$$u_0(\tau)=e^{-\varepsilon\tau},$$
$$u_1(\tau)=0,$$
$$u_2(\tau)=\frac{1}{(\eta-\varepsilon)}\left[-e^{-2\varepsilon\tau}+e^{-\varepsilon\tau}\right]+\frac{\varepsilon}{\eta(\eta-\varepsilon)}\left[e^{-(\eta+\varepsilon)}-e^{-\varepsilon t}\right] + \frac{\lambda_1\varepsilon\tau\, e^{-\varepsilon\tau}}{(\eta-\varepsilon)}+\frac{\lambda_1\varepsilon}{(\eta-\varepsilon)}\left[e^{-\eta\tau}-e^{-\varepsilon\tau}\right],$$
$$v_0(\tau)=0,$$
$$v_1(\tau)=\frac{\varepsilon}{\eta-\varepsilon}\left[e^{-\varepsilon\tau}-e^{-\eta\tau}\right],$$
$$v_2(\tau)=\frac{\varepsilon\left[e^{-\eta\tau}-e^{-2\varepsilon\tau}\right]}{(\eta-\varepsilon)(2\varepsilon-\eta)}+\frac{\left[e^{-(\varepsilon+\eta)\tau}-e^{-\eta\tau}\right]}{(\eta-\varepsilon)},$$
$$w_0(\tau)=0,$$
$$w_1(\tau)=0,$$
$$w_2(\tau)=\frac{\lambda_2\varepsilon}{(\eta-\varepsilon)}\left[\frac{\left(e^{-\varepsilon\tau}-e^{-\lambda_3\varepsilon\tau}\right)}{(\lambda_3-1)\varepsilon}-\frac{\left(e^{-\eta\tau}-e^{-\lambda_3\varepsilon\tau}\right)}{(\lambda_2\varepsilon-\eta)}\right],$$
$$w_3(\tau)=\frac{\lambda_2\left[e^{-2\varepsilon\tau}-e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\eta-2\varepsilon)(\lambda_3-2)}-\frac{\lambda_2\left[e^{-(\varepsilon+\eta)\tau}-e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\lambda_3-\varepsilon-\eta)} - \frac{\lambda_2\varepsilon\left[e^{-\eta\tau}-e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\eta-2\varepsilon)(\lambda_3\varepsilon-\eta)}+\frac{\lambda_2\left[e^{-\eta\tau}-e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\lambda_3\varepsilon-\eta)}. \quad (29)$$

According to the HPM, we can conclude that

$$u(\tau)=\lim_{p\to 1}u(\tau)=u_0+u_1+u_2, \quad (30)$$

$$v(\tau)=\lim_{p\to 1}v(\tau)=v_0+v_1+v_2, \quad (31)$$

$$w(\tau)=\lim_{p\to 1}w(\tau)=w_0+w_1+w_2+w_3. \quad (32)$$

Substitute (29) in (30)-(31) we obtain (33)–(35) in the text.

4. Analytical Solution of Substrate, Enzyme, Enzyme-Substrate Complex, and Free Enzyme Product Using HPM

Non-linear phenomena play a crucial role in applied mathematics and chemistry. Construction of particular exact solutions for these equations remains an important problem. Finding exact solutions that have a physical, chemical, or biological interpretation is of fundamental importance. The investigation of exact solution of non-linear equation is

interesting and important. In the past, many authors mainly had paid attention to study solution of non-linear equations by using various methods, variational iteration method [13], and homotopy perturbation method [14–17].

The homotopy perturbation method has been extensively worked out over a number of years by numerous authors. The idea has been used to solve nonlinear boundary value problems [15], integral equations [18–20], Klein-Gordon and Sine-Gordon equations [21], Emden-Flower type equations [22], and many other problems. This wide variety of applications shows the power of the HPM to solve functional equations. The HPM is unique in its applicability, accuracy and efficiency. The HPM [23] uses the imbedding parameter p as a small parameter, and only a few iterations are needed to search for an asymptotic solution. Using this method, we can obtain the following solution to (14)–(16) (see the appendix):

$$u(\tau) = e^{-\varepsilon\tau} + \frac{\left[-e^{-2\varepsilon\tau} + e^{-\varepsilon\tau}\right]}{(\eta-\varepsilon)} + \frac{\varepsilon\left[e^{-(\eta+\varepsilon)} - e^{-\varepsilon\tau}\right]}{\eta(\eta-\varepsilon)} + \frac{\lambda_1\varepsilon\tau\, e^{-\varepsilon\tau}}{(\eta-\varepsilon)} + \frac{\lambda_1\varepsilon\left[e^{-\eta\tau} - e^{-\varepsilon\tau}\right]}{(\eta-\varepsilon)}, \tag{33}$$

$$v(\tau) = \frac{\varepsilon}{\eta-\varepsilon}\left[e^{-\varepsilon\tau} - e^{-\eta\tau}\right] + \frac{\varepsilon\left[e^{-\eta\tau} - e^{-2\varepsilon\tau}\right]}{(\eta-\varepsilon)(2\varepsilon-\eta)} + \frac{\left[e^{-(\varepsilon+\eta)\tau} - e^{-\eta\tau}\right]}{(\eta-\varepsilon)}, \tag{34}$$

$$\begin{aligned} w(\tau) = {} & \frac{\lambda_2\varepsilon}{(\eta-\varepsilon)}\left[\frac{\left(e^{-\varepsilon\tau} - e^{-\lambda_3\varepsilon\tau}\right)}{(\lambda_3-1)\varepsilon} - \frac{\left(e^{-\eta\tau} - e^{-\lambda_3\varepsilon\tau}\right)}{(\lambda_2\varepsilon-\eta)}\right] \\ & + \frac{\lambda_2\left[e^{-2\varepsilon\tau} - e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\eta-2\varepsilon)(\lambda_3-2)} - \frac{\lambda_2\left[e^{-(\varepsilon+\eta)\tau} - e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\lambda_3-\varepsilon-\eta)} \\ & - \frac{\lambda_2\varepsilon\left[e^{-\eta\tau} - e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\eta-2\varepsilon)(\lambda_3\varepsilon-\eta)} + \frac{\lambda_2\left[e^{-\eta\tau} - e^{-\lambda_3\varepsilon\tau}\right]}{(\varepsilon-\eta)(\lambda_3\varepsilon-\eta)}. \end{aligned} \tag{35}$$

Equations (33)–(35) represent the analytical expression of the dimensionless substrate concentration $u(\tau)$, dimensionless enzyme-substrate concentration $v(\tau)$, and dimensionless free enzyme product concentration $w(\tau)$ for all values of parameters ε, λ_1, λ_2 and λ_3. For steady condition, the differential equations (17a)–(17c) become as follows:

$$\begin{aligned} -\varepsilon u + uv + \lambda_1 v &= 0, \\ \varepsilon u - \eta v + \lambda_3\varepsilon w - uv - \lambda_3 v &= 0, \\ \lambda_2 v - \lambda_3\varepsilon w + \lambda_3 v w &= 0. \end{aligned} \tag{36}$$

Solving the above equations, we can obtain the concentrations of substrate u, enzyme substrate complex v, and product free enzyme w as follows: $u = 0$, $v = 0$, $w = 0$. When τ tends to infinity, the analytical expression corresponding to the substrate concentration u, enzyme substrate concentration v and free enzyme product concentration w from (33)–(35) confirm the validity of mathematical analysis.

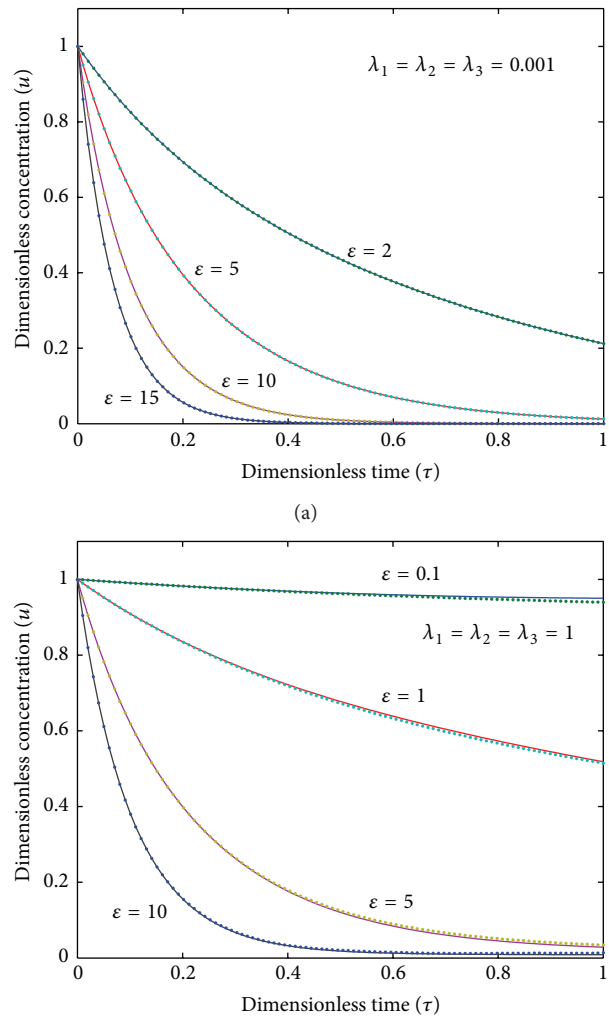


Figure 1: Profile of the normalized concentrations of substrate u is calculated using (33) for various values of dimensionless parameters ε, λ_1, λ_2, and λ_3. Solid line: (33); dotted line: numerical simulation.

5. Results and Discussion

The substrate concentration versus time is plotted in Figures 1 and 2 using (33). From Figures 1(a) and 1(b), it is observed that the dimensionless substrate concentration u decreases. When parameters $\lambda_1 = \lambda_2 = \lambda_3 = 0.001$, the dimensionless substrate concentration u gradually decreases as the value of parameter ε increases and reaches the steady state when $\tau \geq 0.5$. When $\lambda_1 = \lambda_2 = \lambda_3 = 1$, the concentration decreases rapidly as ε increases. $u \approx 1$ when $\varepsilon \leq 0.1$. In Figure 2, it is inferred that the concentration decreases as dimensionless time τ increases and reaches its minimum when $\tau = 1$. The graph is shown for various values of λ_1 when $\lambda_2 = \lambda_3 = 0.001$, $\varepsilon = 1$.

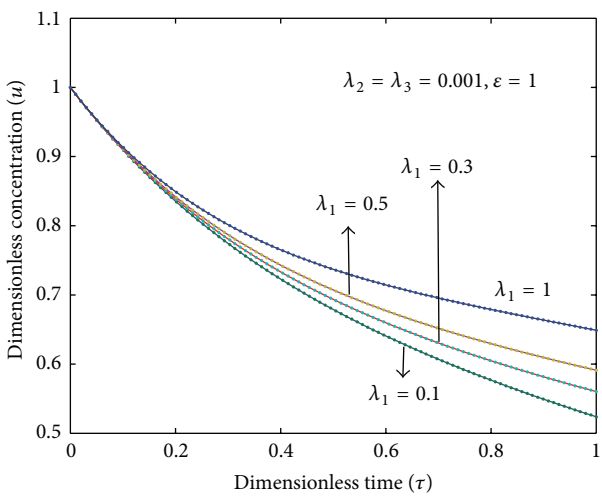


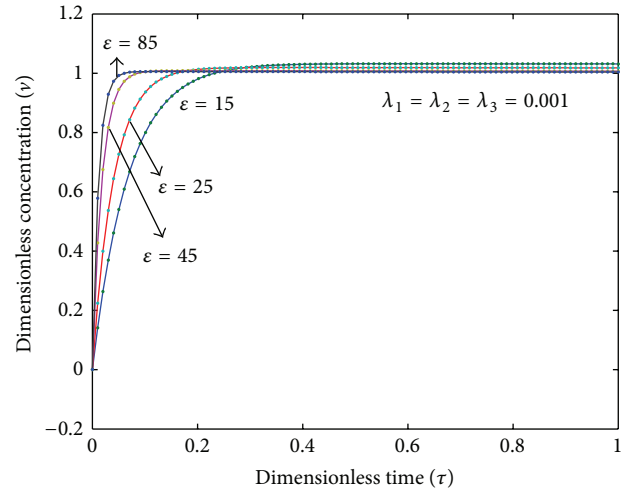
FIGURE 2: Profile of the normalized concentrations of substrate u is calculated using (33) for various values of dimensionless parameter λ_1. Solid line: (33); dotted line: numerical simulation.

Figures 3 and 4 are plotted using (34), dimensionless time τ as abscissa, and enzyme concentration v as ordinate. From Figures 3(a) and 3(b), it is observed that the enzyme concentration increases when $\tau \leq 0.2$ and reaches the steady state value when $\tau > 0.2$. The graph is shown for various values of the parameter ε, when $\lambda_1 = \lambda_2 = \lambda_3 = 0.001$ and 0.01. From Figure 4, it is inferred that enzyme concentration v reaches its maximum between the time 0.03–0.09, and reaches the steady state when $\tau \geq 0.5$.

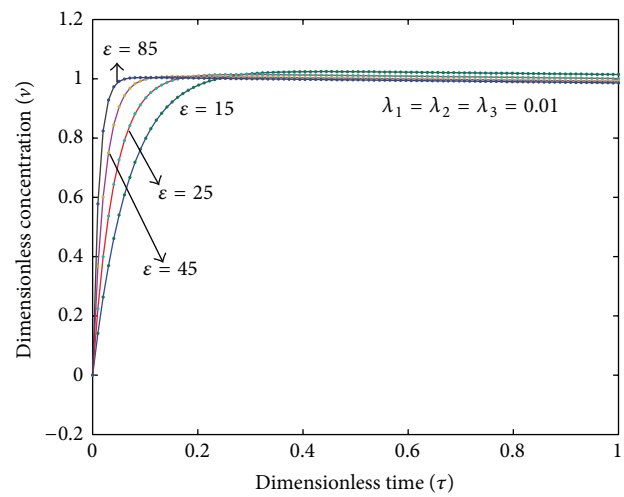
Figures 5 and 6 shows the free enzyme product concentration versus time for various values of parameter λ using (35). From Figures 5(a)-5(b), it is inferred that product concentration w increases very slowly as the time increases. The graphs are shown for various values of the parameter ε, when $\lambda_1 = \lambda_2 = \lambda_3 = 0.001$ and 0.01. In Figure 6, it is noted that the product concentration increases slowly and reaches the steady state at $\tau \geq 0.3$. Profile of dimensionless concentrations u, v, and w versus the dimensionless time τ using (33), (34), and (35) for the fixed values of the parameters is plotted in Figure 7. From this figure, it is inferred that the concentration of substrate decreases, whereas the concentration of enzyme increases. But for all time the concentration of free enzyme-product have at most constant value.

6. Conclusion

Approximate analytical solutions to the system of non-linear reaction equations in enzyme reaction mechanism are presented using homotopy perturbation method. A simple, straight forward, and a new method of estimating the concentrations of substrate, enzyme-substrate, and product are derived. This solution procedure can be easily extended to all kinds of system of coupled non-linear equations with various complex boundary conditions in enzyme-substrate nonlinear reaction diffusion processes.



(a)



(b)

FIGURE 3: Profile of dimensionless enzyme concentration v is calculated using (34) for various values of dimensionless parameters ε, λ_1, λ_2 and λ_3. Solid line: (34); dotted line: numerical simulation.

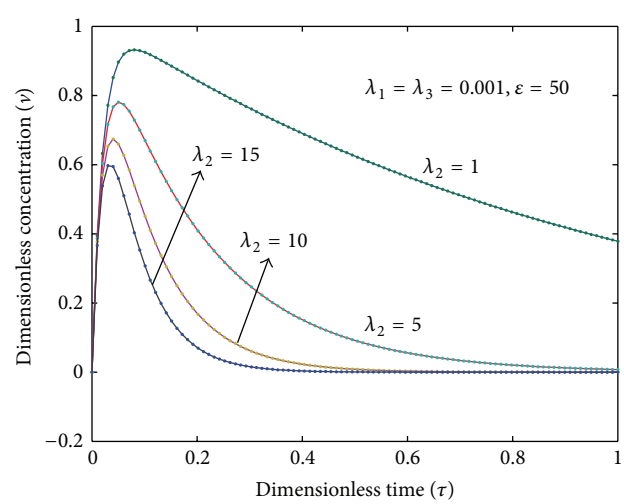
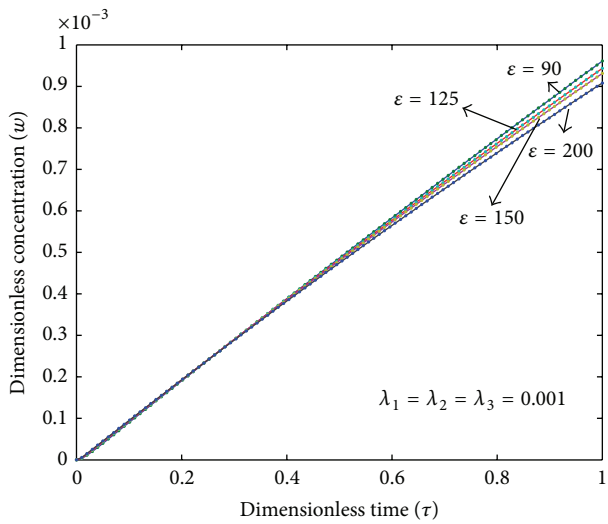
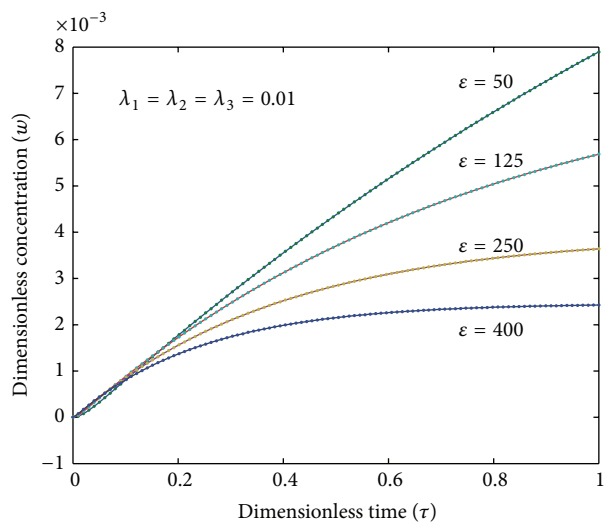


FIGURE 4: Profile of dimensionless enzyme concentration v is calculated using (34) for various values of dimensionless parameter λ_2. Solid line: (34); dotted line: numerical simulation.



(a)



(b)

Figure 5: Profile of dimensionless free enzyme product concentration w is calculated using (35) for various values of dimensionless parameters ε, λ_1, λ_2 and λ_3. Solid line: (35); dotted line: numerical simulation.

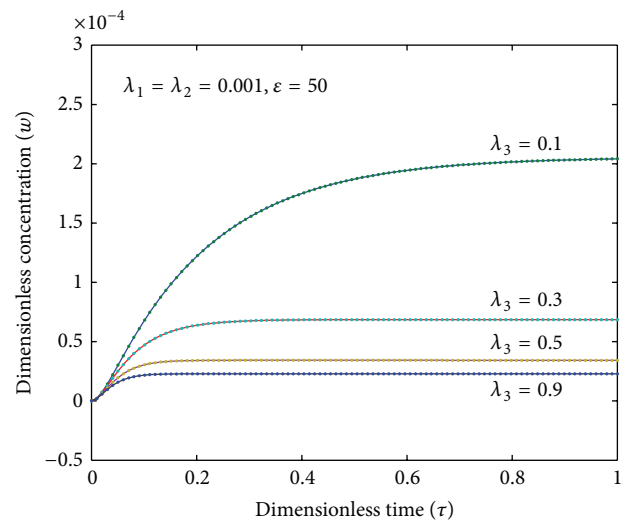


Figure 6: Profile of dimensionless free enzyme product concentration v is calculated using (35) for various values of dimensionless parameter λ_3. Solid line: (35); dotted line: numerical simulation.

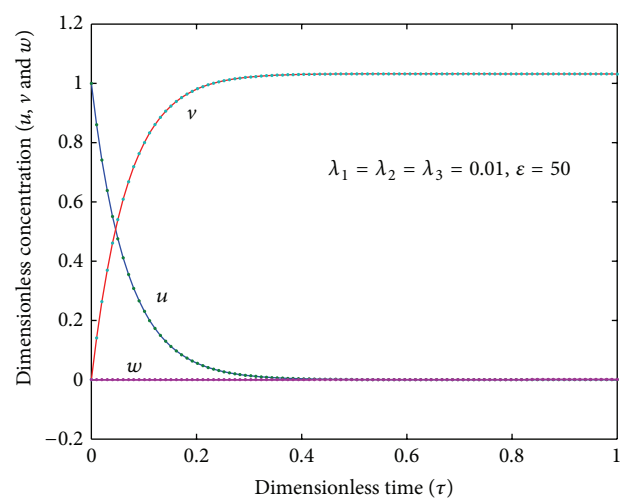


Figure 7: Profile of dimensionless concentrations u, v, and w versus the dimensionless time τ using (33), (34), and (35) for the fixed values of the parameters λ_1, λ_2, λ_3, and ε. Solid line: (33), (34), and (35); dotted line: numerical simulation.

Appendix

Numerical Simulation Program for (14)–(16)

```
function graphmain3
options = odeset ("RelTol", 1e − 6, "Stats", "on");
% initial conditions
X0 = [1; 0; 0];
t span = [0, 5];
tic
[t, X] = ode45 (@TestFunction, t span, X0, options);
toc
figure
hold on
plot (t, X(:, 1), "−" )
plot (t, X(:, 2),"−")
plot (t, X(:, 3),"∗")
legend ("x1","x2","x3")
y label ("x")
x label ("t")
return
function [dx_dt] = TestFunction (t, x)
e = 1; c1 = 0.1; c2 = 0.001; c3 = 0.1;
dx_dt(1) = −e ∗ x(1) + x(1) ∗ x(2) + c1 ∗ x(2);
```

$dx\_dt(2) = e * x(1) - c1 * x(2) - c2 * x(2) + c3 * e * x(3) - x(1) * x(2) - c3 * x(2) * x(3);$

$dx\_dt(3) = c2 * x(2) - c3 * e * x(3) + c3 * x(2) * x(3);$

$dx\_dt = dx\_dt,$

return

Nomenclature and Units

[E]:	Enzyme concentration (μM)
[C]:	Enzyme-substrate complex (μM)
[S]:	Substrate concentration (μM)
$[E_0]$:	Initial enzyme concentration (μM)
K_M:	Michaelis-Menten constant
$[S_0]$:	Initial substrate concentration (μM)
k_1, k_2, k_3:	Positive rate constants (none)
$\lambda_1, \lambda_2, \lambda_3, \varepsilon$:	Reaction diffusion parameter (none)
u:	Dimensionless substrate concentration (none)
v:	Dimensionless enzyme substrate concentration (none)
w:	Dimensionless product concentration (none)
t:	Time (sec)
τ:	Dimensionless time (none).

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