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Mathematical modelling of steady-state concentration in immobilized glucose isomerase of packed-bed reactors

V. Margret PonRani · L. Rajendran

Abstract The theoretical model of the steady-state immobilized enzyme electrodes is discussed. This model is based on diffusion equation containing a non-linear term related to Michaelis–Menten kinetics of the enzymatic reaction. Homotopy perturbation method (HPM) is employed to solve the non-linear diffusion equation for the steady-state condition. Simple and approximate polynomial expression of concentration and flux are derived for all small values of parameters ϕ_p (Theiele modulus) and β (kinetic parameter). Furthermore, in this work the numerical solution of the problem is also reported using SCILAB/MATLAB program. The analytical results are compared with the numerical results and found to be in good agreement.

Keywords Glugose isomerase · Mathematical modelling · Diffusion · Homotopy perturbation method · Simulation · Packed bed

List of symbols

G	Reaction of glucose
E	Reaction of enzyme
F	Reaction of fructose
G_0	Initial concentration
D_P	Pore diffusivity
R	Reaction rate
X	Complex intermediate
K_{mf}	Michaelis–Menten constant
v_{mf}	Maximum velocity of the forward reaction

V. M. PonRani · L. Rajendran (✉)
Department of Mathematics, The Madura College, Madurai, 625011, Tamilnadu, India
e-mail: raj_sms@rediffmail.com; dr.rajendran.l@gmail.com

| K_{mr} and v_{mr}	Maximum velocity of the backward reaction
C	Dimensionless concentration
l_p	Pore length
ϕ_p	Pore-level Thiele-modulus
α_1 and α_2	Constants
J_{ij}	Flux
k_1, k_2, k_{-1} and k_{-2}	Kinetic constants

1 Introduction

Pore networks are frequently used in the modelling of transport and reactions in porous materials. This has been reviewed recently Sahimi et al. [1]. By representing the void space with a pore of idealised geometry, usually a cylindrical pore, the equations for diffusion and reaction may be solved. Marshall and Kooi [2] discovered glucose isomerase using enzymatic isomerization (EI). This discovery, which made it possible to produce fructose by EI of glucose isomerase [3–5]. The process was originally carried out in batch reactors with soluble enzymes. It was later extended to one involving immobilized glucose isomerase (IGI); Melkote and Jensen [6] used the percolation properties of a bethe lattice to model deactivation of glucose isomerase. Reyes and Jensen [7] calculate the effective diffusivity of the pore networks. A probabilitic approach was adoped by Forment et al. [8–10] to describe the pore network of the catalyst. Mann et al. [11] simulated fouling within individual catalyst particles using a two-dimensional square pore network. For example, Ching and Chu [12] studied the effect of desperation and flow on the performance of the reactor. Vasic- Racki et al. [13] carried out experimental studies and computer modelling of glucose isomerization. Houng et al. [14] carried out experimental studies of the problem using both a differential and a packed- bed reactor. They also studied the effect of a variety of factors on the reactor's performance. Park et al. [15] and Faqir [16] investigated the effect of the temperature on the performance of the reactor and developed criteria for optimizing the reactor.

Recently, Mitra Dadvar et al. [17] studied deactivation of the microporous particles as a percolation process by developing a network model. However, to the best of our knowledge, no general analytical results for the steady-state substrate concentration and current for all values of parameters ϕ_p, β, α_1 and α_2 have been reported. The purpose of this communication is to derive expression for the steady-state substrate concentration and the current in closed form for small values of parameters using the Homotopy perturbation method.

2 Mathematical formulation of the problem and analysis

The model describes the mechanism by which glugose enzyme moves from the intermediate complex form to the fructose and back to the intermediate to the complex glucose enzyme. This can be written as

$$G + E \underset{k_{-1}}{\overset{k_1}{\rightleftharpoons}} X \underset{k_{-2}}{\overset{k_2}{\rightleftharpoons}} F + \text{E}, \tag{1}$$
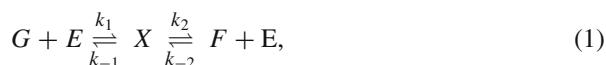
where k_1, k_2 are the kinetic constants for the forward direction and k_{-1}, k_{-2} are the kinetic constants for the backward direction. The non-linear reaction diffusion equation for this model can be represented as follows [17]

$$D_P(\lambda)\frac{d^2G}{dx^2} - \frac{2}{ra}R = 0 \tag{2}$$

where $\lambda = R_M/r$ (R_M is the molecular radius and r is the pore radius) and D_P is the pore diffusivity. To proceed further, we make the non-linear differential equation outlined in the Eq. (2) dimensionless by introducing the following parameters:

$$R = \frac{v_m\overline{G}}{K_m + G}\ \overline{G} = G - G_e, \quad v_m = \frac{K_{mr}v_{mr}(1 + K^{-1})}{K_{mr} - K_{mf}},$$

$$K_m = \frac{K_{mf}K_{mr}}{K_{mr} - K_{mf}}\left[1 + \left(K_{mf}^{-1} + \frac{K}{K_{mf}}\right)\frac{G_0}{1 + K}\right] \tag{3}$$

Now the Eq. (2) becomes,

$$D_P\frac{d^2\overline{G}}{dx^2} - \frac{2}{r}\frac{v_m\overline{G}}{K_m + \overline{G}} = 0 \tag{4}$$

By introducing the following dimensionless quantities

$$C = \frac{\overline{G}}{(G_0 - G_e)}, \quad z = \frac{x}{l_p}, \quad \beta = \frac{\overline{C}_0}{K_m}, \quad \phi_p^2 = \frac{2l_p^2v_m}{rD_pK_m} \tag{5}$$

the Eq. (4) becomes, [17]

$$\frac{d^2C}{dz^2} - \phi_p^2\frac{C}{1 + \beta C} = 0 \tag{6}$$

The boundary conditions may be presented as follows:

$$C = \alpha_1 \text{ at } z = 0 \tag{7}$$

$$C = \alpha_2 \text{ at } z = 1 \tag{8}$$

The dimensionless current is given by

$$J_{ij} = \left[\frac{\partial C}{\partial z}\right]_{z=1} \tag{9}$$

3 Analytical solution of the concentration and current using Homotopy perturbation method (HPM)

Recently, various analytical methods are applied to solve non-linear equations. Lesnic [18] used the Adomian decomposition method, He [19], Ozis et al. [20] used Homotopy perturbation method and Zhu [21,22] used exp-function method. The homotopy algorithm might be improved, if Rentoul and Ariel algorithm is adopted [23]. In this paper, the Homotopy perturbation method is applied [24,25]. This method is very effective and simple. Using this method, we obtain the analytical expression of substrate concentration (see Appendix A), as follows:

$$
C = \frac{1}{6A\,(B^2 - 1)}
$$
$$
\times \begin{Bmatrix}
2\beta\alpha_1\alpha_2 AB\,\mathrm{Cosh}(2\phi_p z) + 6\alpha_2\,\mathrm{Sinh}(\phi_p z)\,\mathrm{Cosh}^2(\phi_p z) - 2\beta\alpha_1^2 AB^2\mathrm{Cosh}(2\phi_p z) \\
+ \mathrm{Cosh}(2\phi_p)\mathrm{Sinh}(\phi_p z)\left[2\beta\alpha_1^2 B^2 - 2\beta\alpha_1\alpha_2 B - \beta\alpha_1\right] + \mathrm{Sin}h(2\phi_p)\mathrm{Sinh}(\phi_p z)\left[2\beta\alpha_1\alpha_2 A - 2\beta\alpha_1^2 AB\right] \\
+ \mathrm{Sinh}(2\phi_p z)\left[2\beta\alpha_1\alpha_2 - 2\beta\alpha_1^2 B - 2\beta\alpha_1\alpha_2 B^2 + 2\beta\alpha_1^2 B^3\right] \\
+ \mathrm{Sinh}(\phi_p z)\left[6\alpha_1 B - 4\beta\alpha_1\alpha_2 B + 18\beta\alpha_1\alpha_2 B - 6\beta\alpha_1^2 B^2 + 4\beta\alpha_1^2 B^3 - 2\beta\alpha_1 B - 6\alpha_1 B^3 - 6\alpha_2\right] \\
+ \mathrm{Cosh}(\phi_p z)\left[2\beta\alpha_1 A - 6\alpha_1 A + 12\alpha_1\alpha_2\beta A - 4\beta\alpha_1^2 AB^2 + 4\alpha_1\alpha_2\beta AB + 6\alpha_1 AB^2\right] + 12\alpha_1\alpha_2\beta A \\
-6\beta\alpha_1\alpha_2 AB + 6\beta\alpha_1^2 AB^2 - 3\beta\alpha_1 A
\end{Bmatrix}
$$
$$(10)$$

where

$$
A = \mathrm{Sinh}(\phi_p) \text{ and } B = \mathrm{Cosh}(\phi_p) \tag{11}
$$

Equation (10) is the new analytical expressions for the dimensionless substrate concentration in terms dimensionless kinetic parameter β and Thiele modulus ϕ_p. Using Eq. (9), the current density is

$$
J_{ij} = \left[\frac{0.1666}{A\,(B^2 - 1)}\right]
$$
$$
\times \begin{Bmatrix}
+6\alpha_2 B^3\phi_p - 6\alpha_2 B - 6\alpha_1 A^2\phi_p + 6\alpha_1 B^2\phi_p - 4\alpha_1\alpha_2\beta B^3\phi_p + 18\alpha_1\alpha_2\beta B^2\phi_p + 2\alpha_1\beta A^2\phi_p \\
- 12\alpha_1\alpha_2\beta A^2\phi_p - 6\alpha_1^2\beta B^3\phi_p + 4\alpha_1^2\beta B^4\phi_p - 4\alpha_1^2\beta A^2 B^2\phi_p + 4\alpha_1\alpha_2\beta A^2 B\phi_p \\
-2\alpha_1\beta B^2\phi_p + 3\alpha_1\beta B\phi_p - 6\alpha_1 B^4\phi_p + 6\alpha_1 A^2 B^2\phi_p - 12\alpha_1\alpha_2\beta B\phi_p \\
+ \mathrm{Cosh}(2\phi_p)\left[6\alpha_1^2\beta B^3\phi_p - \alpha_1\beta B\phi_p + 4\alpha_1\alpha_2\beta\phi_p - 4\alpha_1^2\beta B\phi_p\right] \\
+ \mathrm{Sinh}(2\phi_p)\left[2A\alpha_1\beta\phi_p + 6\alpha_1^2\beta AB^2\phi_p\right]
\end{Bmatrix}
$$
$$(12)$$

4 Numerical Simulation

The diffusion equation, Eq. (6) for the boundary conditions Eqs. (7–8) is solved by numerical methods. The function pdex4 in Scilab software which is a function of solving the initial-boundary value problems for partial differential equation is used to solve these equations. Its numerical solution is compared with our analytical results in Tables 1, 2, 3, 4 and it gives good agreement between them. The SCILAB/MATLAB program is also given in Appendix B.

Table 1 Comparison of the analytical result Eq. (10) with the numerical result of concentration C when $\phi_P = 2$, $\alpha_1 = 0.5$, $\alpha_2 = 1$ and for various values of β

X	$\beta = 0.01$			$\beta = 1$			$\beta = 2.5$			$\beta = 5$		
	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)
0	0.5000	0.5000	0.00	0.4999	0.5000	0.02	0.5000	0.5000	0.00	0.5000	0.5000	0.00
0.2	0.4411	0.4383	0.63	0.4799	0.4851	1.08	0.5231	0.5387	2.98	0.6367	0.5514	13.40
0.4	0.4534	0.4492	0.93	0.5020	0.5234	4.26	0.5827	0.5756	1.22	0.6982	0.6263	10.30
0.6	0.5390	0.5347	0.80	0.5780	0.6175	6.83	0.6805	0.6370	6.39	0.7355	0.7255	1.36
0.8	0.7118	0.7087	0.44	0.7298	0.7735	5.99	0.8187	0.7573	7.50	0.8098	0.8498	5.84
1	1.0000	1.0000	0.00	1.0000	1.0000	0.00	1.0000	1.0000	0.00	1.0000	1.0000	0.00
	Average deviation		0.47	Average deviation		3.03	Average deviation		3.02	Average deviation		5.15

Table 2 Comparison of the analytical result Eq. (10) with the numerical result of concentration C when $\beta = 0.01$, $\alpha_1 = 0.1$, $\alpha_2 = 1$ and for various values of ϕ_p

X	$\phi_p = 0.01$			$\phi_p = 0.1$			$\phi_p = 1$			$\phi_p = 3$		
	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)
0	0.1000	0.1000	0.00	0.1000	0.1000	0.00	0.1000	0.1000	0.00	0.1000	0.1000	0.00
0.2	0.2799	0.2800	0.04	0.2789	0.2796	0.25	0.2390	0.2468	3.26	0.1181	0.1136	3.81
0.4	0.4589	0.4600	0.24	0.4584	0.4594	0.22	0.3917	0.4036	0.04	0.1800	0.1721	4.39
0.6	0.6398	0.6400	0.03	0.6383	0.6393	0.16	0.5640	0.5767	2.25	0.3087	0.2986	3.27
0.8	0.8199	0.8200	0.01	0.8188	0.8195	0.09	0.7635	0.7728	1.22	0.5519	0.5427	1.67
1	1.0000	1.0000	0.00	1.0000	1.0000	0.00	1.0000	1.0000	0.00	1.0000	1.0000	0.00
	Average deviation		0.05	Average deviation		0.12	Average deviation		1.13	Average deviation		2.19

Table 3 Comparison of the analytical result Eq. (10) with the numerical result of concentration C when $\beta = 0.01$, $\phi_p = 2$, $\alpha_2 = 1$ and for various values of α_1

X	$\alpha_1 = 0.01$			$\alpha_1 = 0.1$			$\alpha_1 = 0.5$			$\alpha_1 = 1$		
	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)
0	0.0099	0.0100	1.01	0.9999	0.1000	0.10	0.5000	0.5000	0.00	0.9999	1.0000	0.01
0.2	0.1197	0.1184	1.09	0.1786	0.1772	0.78	0.4411	0.4383	0.63	0.7707	0.7650	0.74
0.4	0.2490	0.2466	0.96	0.2863	0.2838	0.07	0.4534	0.4492	0.93	0.6645	0.6563	1.23
0.6	0.4186	0.4157	0.69	0.4405	0.4375	0.68	0.5390	0.5347	0.80	0.6643	0.6563	1.20
0.8	0.6561	0.6538	0.36	0.6662	0.6639	0.35	0.7118	0.7087	0.44	0.7703	0.7650	0.69
1	0.9999	1.0000	0.01	1.0000	1.0000	0.00	1.0000	1.0000	0.00	1.0000	1.0000	0.00
	Average deviation		0.69	Average deviation		0.33	Average deviation		0.47	Average deviation		0.65

Table 4 Comparison of the analytical result Eq. (10) with the numerical result of concentration C when $\beta = 0.01$, $\phi_P = 2$, $\alpha_1 = 1$ and for various values of α_2

X	$\alpha_2 = 0.01$			$\alpha_2 = 0.1$			$\alpha_2 = 0.5$			$\alpha_2 = 1$		
	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)	Eq. (10)	Numerical	% deviation of % Eq. (10)
0	0.9999	1.0000	0.01	0.9999	1.0000	0.01	0.9999	1.0000	0.01	0.9999	1.0000	0.01
0.2	0.6562	0.6538	0.82	0.6693	0.6639	0.81	0.7144	0.7080	0.90	0.7707	0.7650	0.74
0.4	0.4499	0.4157	3.30	0.4448	0.4375	0.64	0.5425	0.5347	1.44	0.6645	0.6563	1.23
0.6	0.2531	0.2466	2.57	0.2904	0.2838	2.27	0.4566	0.4492	1.62	0.6643	0.6563	1.20
0.8	0.1225	0.1184	3.35	0.1814	0.1772	2.32	0.4431	0.4383	1.08	0.7703	0.7650	0.69
1	0.0999	0.0100	1.01	0.1000	0.1000	0.00	0.5000	0.5000	0.00	1.0000	1.0000	0.00
	Average deviation		1.84	Average deviation		1.01	Average deviation		0.84	Average deviation		0.65
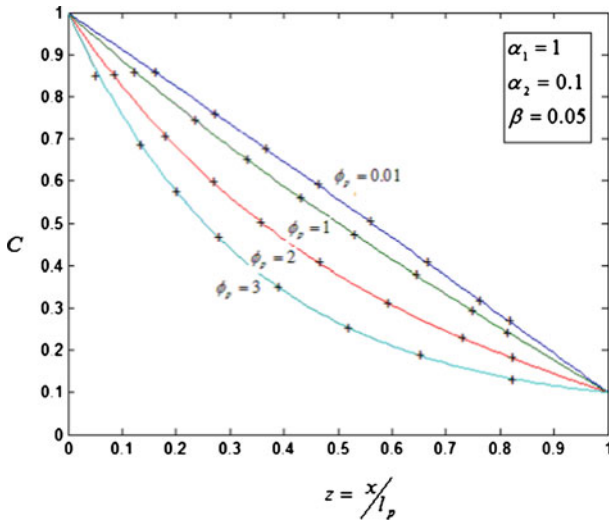
Fig. 1 Plot of the concentration C versus the normalised distance z for various values of ϕ_p and some fixed values of α_1, α_2 and β. The concentrations were computed using Eq. (10). The key to the graph: (*dottedline*) represents the Eq. (10) and (+) represents the numerical simulation
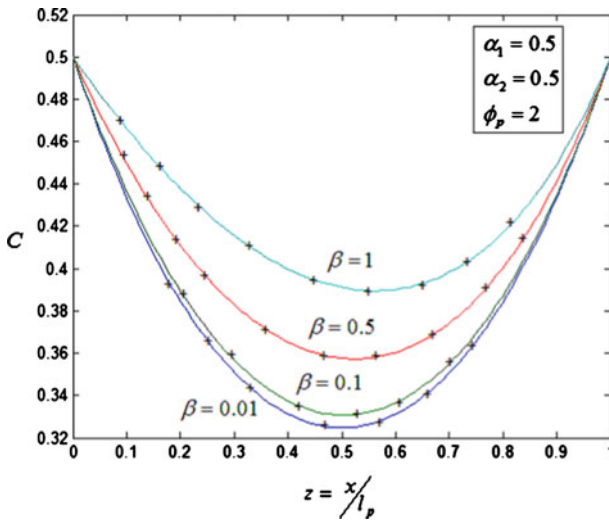

Fig. 2 Plot of the concentration C versus the normalised distance z for various values of β and some fixed value of α_1, α_2 and ϕ_p. The concentrations were computed using Eq. (10). The key to the graph: (*dottedline*) represents the Eq. (10) and (+) represents the numerical simulation

5 Result and discussion

Equation (10) represents the closed and simple approximate analytical expression of the concentration of substrate for small values of parameters ϕ_p, β, α_1 and α_2. Equation (12) represents new simple analytical expression of flux. The concentration of
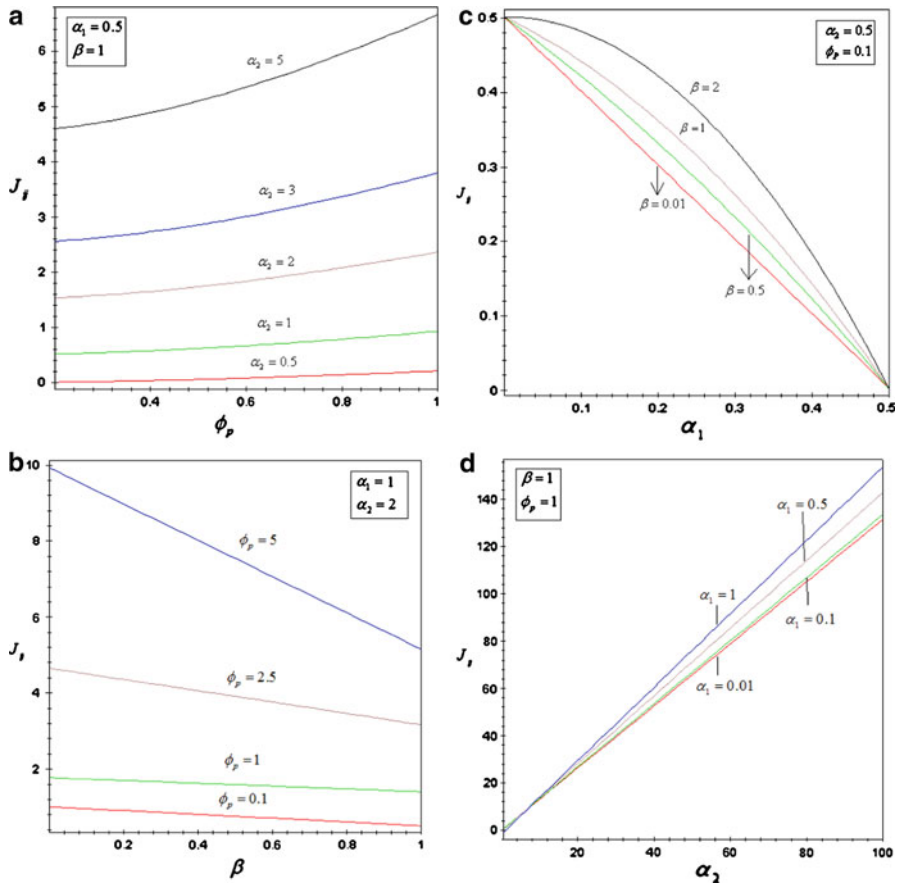
Fig. 3 **a** Variation of flux J_{ij} versus dimensionless parameter ϕ_p. The current is computed using Eq. (12). **b** Variation of flux J_{ij} versus dimensionless parameter β. The current is computed using Eq. (12). **c** Variation of flux J_{ij} versus dimensionless parameter α_1. The current is computed using Eq. (12). **d** Variation of flux J_{ij} versus dimensionless parameter α_2. The current is computed using Eq. (12)

substrate depends upon the values parameters ϕ_p, β, α_1 and α_2. The parameter Thiele modulus ϕ_p can be varied by changing either the thickness of the enzyme layer or the amount of enzyme immobilized in the matrix. This parameter describes the relative importance of diffusion and reaction in the enzyme layer. Figures 1 and 2 represent the dimensionless steady state concentration C for different values of the Thiele modulus ϕ_p and dimensionless kinetic parameter β. From these figures, it is obvious that the substrate concentration C reaches the maximum value 1, when $x = 1$. Figure 3a–d represent the dimensionless flux J_{ij} versus dimensionless kinetic parameter β for various values of Thiele modulus ϕ_p. From this figure, it is evident that the flux value increases as the dimensionless kinetic parameter β and Thiele modulus ϕ_p increases. In Tables 1, 2, 3 and 4 our result is compared with numerical result for small values of parameters. The substrate concentration C is compared numerically for various values of the dimensionless kinetic parameter β (Table 1), Thiele modulus ϕ_p (Table 2), α_1
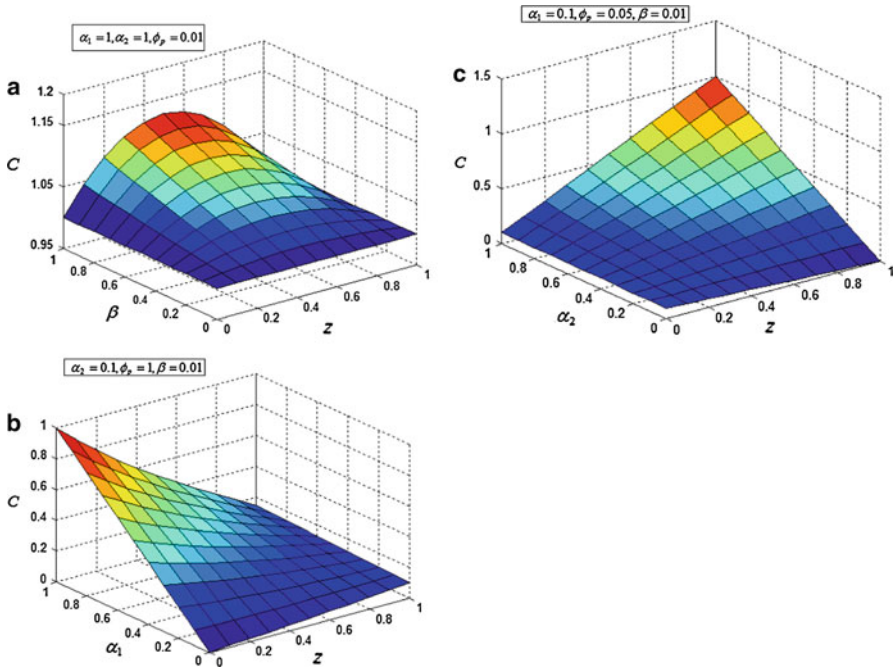
Fig. 4 **a** The normalized three-dimensional normalized concentration profile Eq. (10) when $0 \leq \beta \leq 1$, $\phi_p = 0.01$, $\alpha_1 = 1$ and $\alpha_2 = 1$. **b** The normalized three-dimensional normalized concentration profile Eq. (10) when $0 \leq \alpha_1 \leq 1$, $\beta = 0.01$, $\phi_p = 1$ and $\alpha_2 = 0.1$. **c** The normalized three-dimensional normalized concentration profile Eq. (10) when $0 \leq \alpha_2 \leq 1$, $\beta = 0.01$, $\phi_p = 1$ and $\alpha_1 = 1$

(Table 3) and α_2 (Table 4). In all the cases average relative error is less than 3%. From these tables we conclude that there is no significant difference found in the substrate concentration C when the value of all parameters is less than 0.1. The normalized three-dimensional substrate concentration profile C is plotted in Fig 4a–c where the data given by Figs. 1 and 2 are verified.

6 Conclusion

In this work, system of coupled, steady-state non-linear reaction/diffusion equation in pore network model has been solved analytically. Approximate analytical expressions for the concentration and flux in packed-bed reactor for kinetic reactions with diffusion coefficients at a planar microelectrode under steady-state conditions are obtained using the Homotopy perturbation method. The primary result of this work is simple approximate calculation of concentration profiles and flux for small values of the fundamental parameters. The small variation in Thiele modulus caused a significant change in both the magnitude of the current response and the general behavior of the system. The Homotopy perturbation method is extremely simple and promising to solve other non-linear equations. This method can be easily extended to find the solution of all other non-linear reaction diffusion equations in kinetic reactions for all microelectrodes for various complex boundary conditions.

Appendix A

Solution of the Eq. (6) using Homotopy perturbation method

In this Appendix, we indicate how Eq. (10) in this paper are derived. To find the solution of Eq. (10) we first construct a Homotopy as follows:

$$(1-p)\left[\frac{d^2C}{dz^2} - \phi_p^2 C\right] + p\left[\frac{d^2C}{dz^2} - \phi_p^2 C + \beta C \frac{d^2C}{dz^2}\right] = 0 \tag{A1}$$

The boundary conditions are

$$C = \alpha_1 \text{ at } z = 0 \tag{A2}$$
$$C = \alpha_2 \text{ at } z = 1 \tag{A3}$$

The approximate solutions of Eq. (A1) is

$$C = C_0 + pC_1 + p^2 C_2 + p^3 C_3 + \cdots\cdots. \tag{A4}$$

Substituting Eq. (A4) into (A1) and comparing the coefficients of like powers of p

$$p^0 : \frac{d^2C_0}{dz^2} - \phi_p^2 C_0 = 0 \tag{A5}$$

The boundary conditions are

$$C_0 = \alpha_1 \text{ at } z = 0 \tag{A6}$$
$$C_0 = \alpha_2 \text{ at } z = 1 \tag{A7}$$
$$p^1 : \frac{d^2C_1}{dz^2} - \phi_p^2 C_1 + \beta C_0 \frac{d^2C_0}{dz^2} = 0 \tag{A8}$$

The boundary conditions are

$$C_1 = 0 \text{ at } z = 0 \tag{A9}$$
$$C_1 = 0 \text{ at } z = 1 \tag{A10}$$

According to the HPM, we can conclude that

$$C = C_0 + C_1 + \cdots\cdots. \tag{A11}$$

Solving the Eqs. (A5) and (A8), using the boundary conditions Eqs. (A6, A7, A9, A10) and substituting into Eq. (A11), and we can obtain Eq. (10) in the text.

Appendix B

SCILAB/MATLAB program to find the numerical solutions for Eq. (6):

```
function pdex4
m = 0;
x = linspace(0,1);
t = linspace(0,100000);
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,2)')
%————————————————————————

function [c,f,s] = pdex4pde(x,t,u,DuDx)
c = 1;
f = DuDx;
q = 0.1;
B = 10;
F = −q^2*u(1)/(1+B*u(1));
s = F;
% ————————————————————————

function u0 = pdex4ic(x);        %create a initial conditions
u0 = 1;
% ————————————————————————

function [pl,ql,pr,qr] = pdex4bc(xl,ul,xr,ur,t)        %create a boundary conditions
pl = 0;
ql = 1;
pr = ur−1;
qr = 0;
```

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