J Math Chem (2012) 50:1277–1288
DOI 10.1007/s10910-011-9968-3

ORIGINAL PAPER

Analytical expression of non steady-state concentration for the CE mechanism at a planar electrode

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Received: 11 October 2011 / Accepted: 22 December 2011 / Published online: 4 January 2012

Abstract The analytical solutions of the non-steady-state concentrations of species at a planar microelectrode are discussed. The analytical expression of the kinetics of CE mechanism under first or pseudo-first order conditions with equal diffusion coefficients at planar electrode under non-steady-state conditions are obtained by using Homotopy perturbation method. These simple new approximate expressions are valid for all values of time and possible values of rate constants. Analytical equations are given to describe the current when the homogeneous equilibrium position lies heavily in favour of the electroinactive species. Working surfaces are presented for the variation of limiting current with a homogeneous kinetic parameter and equilibrium constant. In this work we employ the Homotopy perturbation method to solve the boundary value problem. Furthermore, in this work the numerical simulation of the problem is also reported using Scilab program. The analytical results are found to be in excellent agreement with the numerical results.

Keywords CE mechanism · Planar electrode · Reaction/diffusion equation · Mathematical modelling · Homotopy perturbation method

1 Introduction

The planar electrode is a popular tool in electroanalysis and mechanistic electrochemistry due to its favourable attributes in an electrochemical experiment. Microelectrodes are of great practical interest for quantitative in vivo measurements, e.g. of oxygen tension in living tissues [1–3], because electrodes employed in vivo should be smaller than the unit size of the tissue of interest. Microelectrodes having the geometry of a

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hemisphere resting on an insulating plane are difficult to fabricate, but their behaviour is easily predicted [4]. Planar microelectrode processes involving a follow-up homogenous chemical reaction are very common and largely examined in the literature.

As far back as 1984, Fleischmann et al. [5,6] used planar electrodes to determine the rate constants of coupled homogeneous reactions (CE, EC', ECE, and DISPI mechanism). Also, the measurement of the current at microelectrodes is one of the easiest and yet most powerful electrochemical methods for quantitative mechanistic investigations. In this work, we are interested in finding the mass transport limiting current response for the CE mechanism at a planar microelectrode. For each mechanism, the electroinactive species A is in dynamic equilibrium with the electroinactive species B via a homogeneous chemical step. The decay of species A is described by the first order forward rate constant k_f and the reverse of this process is described by the backward rate constant k_b, which is first order for the CE mechanism. In Ref. [7], CE mechanism was considered when species C is present in great excess. All the species are considered to have a diffusion coefficient D.

In general, the characterization of subsequent homogenous reactions involves the elucidations of the mechanism of reaction, as well as the determination of the rate constants. Values for the limiting current were presented for a range of equilibrium constants and rate constants. Refs. [8] and [9] also used the same simulation technique to CE mechanism investigating the case of very fast homogeneous kinetics but where the three species have unequal diffusion coefficients. However, to the best of our knowledge, there were no rigorous analytical solutions of the kinetics of CE reaction schemes under first or pseudo-first order conditions with equal diffusion coefficients at planar microelectrodes under non-steady-state conditions for all possible values of parameters k_1, k_2, k_3, k_4, k_5 and k_6 have been reported. The purpose of this communication is to derive approximate analytical expressions for the non-steady-state concentrations and current at planar electrode for all possible values of parameters and for short time using Homotopy perturbation method.

2 Mathematical formulation of the boundary value problem

As a representative example of the reaction-diffusion problems considered, the standard pseudo-first-order homogeneous and heterogeneous steps

$$A \rightleftharpoons B + C$$
$$B \pm e^- \rightarrow products$$

has been chosen, with initial and boundary conditions corresponding to the potential step for all planar electrodes. Under steady-state conditions, the local concentrations of the species do not change. The mass transport equations are set equal to zero. We consider the differential equations with diffusion described by the concentration of the three species leads to the following equations [10].

$$\frac{\partial a}{\partial t} = D\frac{\partial^2 a}{\partial x^2} - k_f a + k_b bc = 0 \tag{1}$$

$$\frac{\partial b}{\partial t} = D\frac{\partial^2 b}{\partial x^2} + k_f a - k_b bc = 0 \tag{2}$$

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} + k_f a - k_b bc = 0 \tag{3}$$

where a, b and c denote the concentration of the species A, B and C. The variables x and t stand for space and time respectively. D is the diffusion coefficient of the species A, B and C. The constants κ_f and κ_b are described the forward and backward rate constants respectively. The boundary conditions reduce to

$$t = 0,\ a = a_0,\ b = b_0,\ c = c_0 \tag{4}$$

$$x = l,\ \frac{\partial a}{\partial x} = 0,\ b = 0,\ \frac{\partial c}{\partial x} = 0 \tag{5}$$

$$x \to \infty,\ a \to a_0,\ b \to b_0,\ c \to c_0 \tag{6}$$

where a_0, b_0 and c_0 are the bulk concentrations of the species A, B and C. The current density is defined as:

$$i = nFAD\left.\frac{\partial b}{\partial x}\right|_{x=l} \tag{7}$$

where n is the number of electrons and F is the Faraday constant. Using the following dimensionless parameters

$$u = \frac{a}{a_0};\ v = \frac{b}{b_0};\ w = \frac{c}{c_0};\ X = \frac{x}{l};\ T = \frac{Dt}{l^2};\ k_1 = \frac{k_f l^2}{D};\ k_2 = \frac{k_b l^2 b_0 c_0}{D a_0};$$
$$k_3 = \frac{k_f l^2 a_0}{D b_0};\ k_4 = \frac{k_b l^2 c_0}{D};\ k_5 = \frac{k_f l^2 a_0}{D c_0};\ k_6 = \frac{k_b l^2 b_0}{D} \tag{8}$$

we get the dimensionless non-linear reaction diffusion equations for planar electrode as follows:

$$\frac{\partial u}{\partial T} = \frac{\partial^2 u}{\partial X^2} - k_1 u + k_2 vw = 0 \tag{9}$$

$$\frac{\partial v}{\partial T} = \frac{\partial^2 v}{\partial X^2} + k_3 u - k_4 vw = 0 \tag{10}$$

$$\frac{\partial w}{\partial T} = \frac{\partial^2 w}{\partial X^2} + k_5 u - k_6 vw = 0 \tag{11}$$

where k_1, k_2, k_3, k_4, k_5 and k_6 are the dimensionless rate constants. The initial and boundary conditions are represented as follows:

$$T = 0,\ u = 1,\ v = 1,\ w = 1 \tag{12}$$

$$X = 1,\ \frac{\partial u}{\partial X} = 0,\ v = 0,\ \frac{\partial w}{\partial X} = 0 \tag{13}$$

$$X \to \infty,\ u = 1,\ v = 1,\ w = 1 \tag{14}$$

The dimensionless current is as follows:

$$\psi = \frac{il}{nFADb_0} = (\partial v/\partial X)_{X=1} \tag{15}$$

3 Analytical solution of the concentrations of the species and current using Homotopy perturbation method

Recently, many authors have applied the HPM to various problems and demonstrated the efficiency of the HPM for handling non-linear structures and solving various physics and engineering problems [11–14]. This method is a combination of homotopy in topology and classic perturbation techniques. The set of expressions presented in Eqs. (9)–(14) defines the initial and boundary value problem. He [15] used the HPM to solve the Lighthill equation, the Duffing equation [16] and the Blasius equation [17]. The idea has been used to solve non-linear boundary value problems [18], integral equations [19–21], Klein–Gordon and Sine–Gordon equations [22], Emden –Flower type equations [23] and many other problems. This wide variety of applications shows the power of the HPM to solve functional equations. This method is unique in its applicability, accuracy and efficiency. The HPM [24–27] uses the imbedding parameter p as a small parameter, and only a few iterations are needed to search for an asymptotic solution. The dimensionless rate constants k_1, k_2, k_3, k_4, k_5 and k_6 are related to one another, since the bulk solution is at equilibrium in the non-steady state. Using HPM (see Appendix A and B), we can obtain the following solutions to the Eqs. (9–11).

$$\begin{aligned} u(X,T) = 1 &- \frac{k_1}{2}\left\{ T erfc\left(\frac{X-1}{2\sqrt{T}}\right) + \sqrt{T}\,(X-1)\left[\left(\frac{X-1}{2\sqrt{T}}\right) erfc\left(\frac{X-1}{2\sqrt{T}}\right)\right.\right. \\ &\left.\left. - \frac{1}{\sqrt{\pi}} e^{-\frac{(X-1)^2}{4T}}\right]\right\} \\ &- \frac{k_1}{2}\left[2\sqrt{\frac{T}{\pi}} e^{-\frac{(X-1)^2}{4T}} - (X-1)\, erfc\left(\frac{X-1}{2\sqrt{T}}\right)\right](X-1) \end{aligned} \tag{16}$$

$$v(X,T) = 1 - erfc\left(\frac{X-1}{2\sqrt{T}}\right) + \frac{k_3}{2}\left[2\sqrt{\frac{T}{\pi}} e^{-\frac{(X-1)^2}{4T}} - (X-1)\, erfc\left(\frac{X-1}{2\sqrt{T}}\right)\right](X-1) \tag{17}$$

$$\begin{aligned} w(X,T) = 1 &+ \frac{k_5}{2}\left\{ T erfc\left(\frac{X-1}{2\sqrt{T}}\right) + \sqrt{T}\,(X-1)\left[\left(\frac{X-1}{2\sqrt{T}}\right) erfc\left(\frac{X-1}{2\sqrt{T}}\right)\right.\right. \\ &\left.\left. - \frac{1}{\sqrt{\pi}} e^{-\frac{(X-1)^2}{4T}}\right]\right\} \\ &+ \frac{k_5}{2}\left[2\sqrt{\frac{T}{\pi}} e^{-\frac{(X-1)^2}{4T}} - (X-1)\, erfc\left(\frac{X-1}{2\sqrt{T}}\right)\right](X-1) \end{aligned} \tag{18}$$

Equations (16–18) are the analytical solutions for the dimensionless concentrations as a function of dimensionless distance X and time T. The current density is

$$\psi = \frac{0.56419}{\sqrt{T}} + k_3\sqrt{\frac{T}{\pi}} \tag{19}$$

Equation (19) represents the new approximate analytical expression for the current for short and medium time and small values of parameter k_3.

4 Numerical simulation

The system of non steady-state non-linear differential Eqs. (9–11) are also solved by numerical methods. The function pdex4 in Scilab software which is a function of solving partial differential equations (PDE) is used to solve these equations. Its numerical solution is compared with the solution obtained using HPM and it gives a suitable result. The Scilab program is also given in Appendix C.

5 Result and discussion

The approximate analytical expressions of concentration of u, v and w [solutions of Eqs. (9), (10) and (11)] are given in the Eqs. (16), (17) and (18). The Eqs. (9), (10) and (11) are also solved by numerical methods. The function pdex4 in Scilab software is used for solving the initial-boundary value problems for dimensionless non-linear reaction diffusion equations for planar electrode. The obtained analytical results are compared with the numerical results for various values of dimensionless parameters. In all cases the concentration of the species A, B and C gives good agreement with the numerical results. The concentration u and v are represented in Figs. 1 and 2. From these figures, it is evident that the value of concentration gradually decreases as T increases. The concentration increases as the distance increases and attains the maximum value 1. Figure 3 represent the concentration profile of w. It is clear that as time increases the value of concentration is also increases and the concentration decreases as the distance increases.

The analytical expression for dimensionless non-steady state current is given in Eq. (19). The dimensionless current ψ versus T for various values of k_3 is given in Fig. 4. From this figure the value of the current increases when the rate constant k_3 increases. The value of the current decreases with increasing value of time T.

6 Conclusions

In this work, the time dependent non-linear differential equations has been restudied and solved analytically. In this paper, we have derived the analytical expressions of the concentrations of the species A, B and C in terms of the parameters k_1, k_2, k_3, k_4, k_5 and k_6 using Homotopy perturbation method. In addition, we have also presented an analytical expression for the non-steady state current for short time and small values

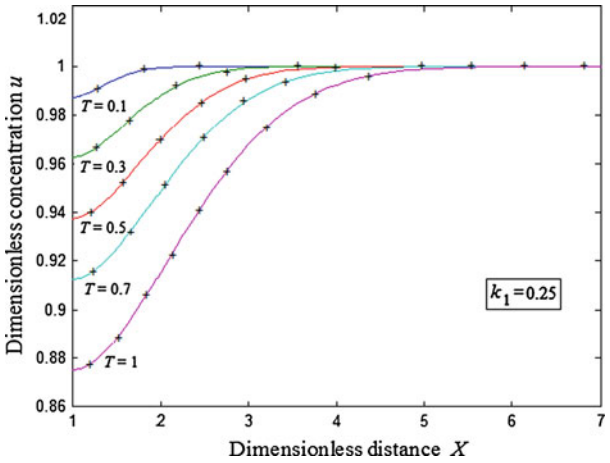

Fig. 1 Plot of the two-dimensional case diagram of the concentration u versus the dimensionless distance X. The concentrations were computed using Eq. (16) for various values of T and the parameter $k_1 = 0.25$. The key to the graph (*solid line*) represents the Eq. (16) and (*dotted line*) represents the numerical simulation

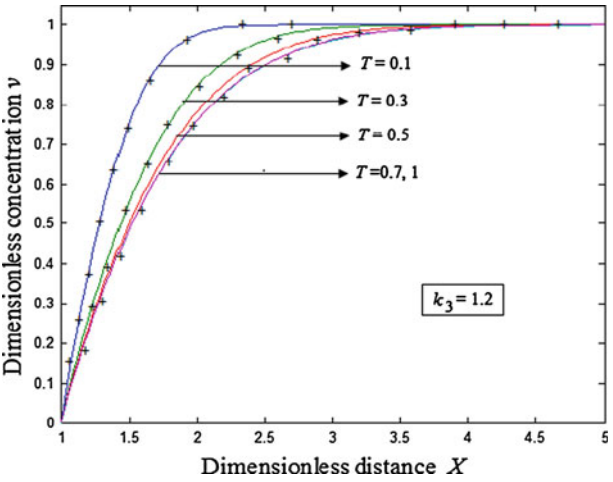

Fig. 2 Plot of the two-dimensional case diagram of the concentration v versus the dimensionless distance X. The concentrations were computed using Eq. (17) for various values of T and the parameter $k_3 = 1.2$. The key to the graph (*solid line*) represents the Eq. (17) and (*dotted line*) represents the numerical simulation

of rate constants. The kinetics of this homogeneous step can in principle be studied by observing how the limiting current responds to changes in electrode size. It must be mentioned here that the physical situation as well as the boundary conditions employed herein represent the simplest possible description whereas, in several recent investigations, more complicated but realistic problems [28–30] have been explored. Further, based on the outcome of this work it is possible to calculate the concentration and current for the CE mechanism for all microelectrodes for various complex boundary conditions.

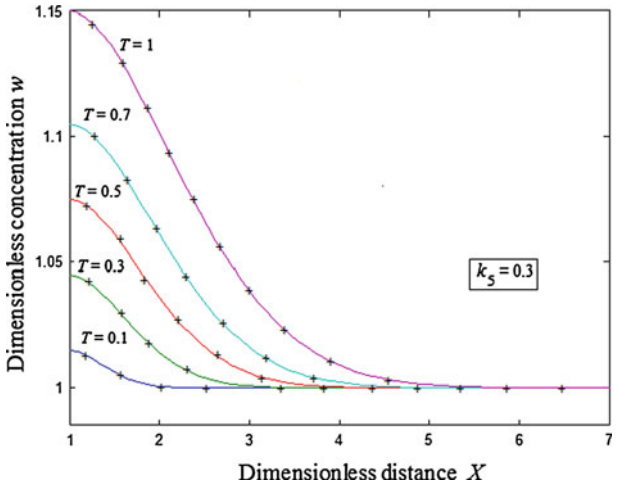

Fig. 3 Plot of the two-dimensional case diagram of the concentration w versus the dimensionless distance X. The concentrations were computed using Eq. (18) for various values of T and the parameter $k_5 = 0.3$. The key to the graph (*solid line*) represents the Eq. (18) and (*dotted line*) represents the numerical simulation

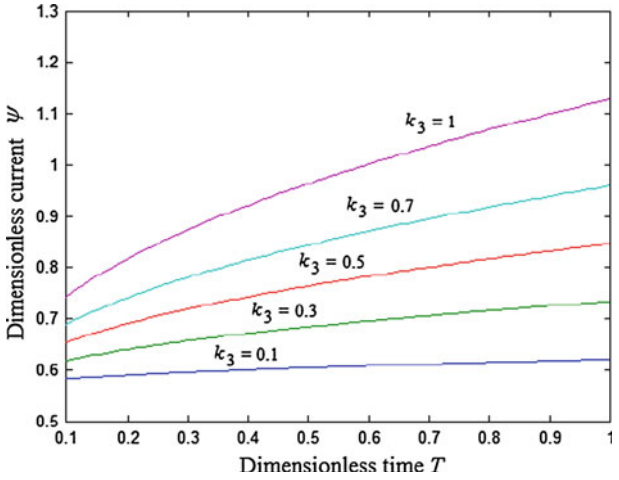

Fig. 4 Variation of dimensionless non-steady state current response ψ with dimensionless time T. The curve is computed using Eq. (19) for various values of the parameter k_3

Acknowledgments This work was supported by the University Grants Commission (F. No. 39-58/2010(SR)), New Delhi, India. The authors are thankful to Mr. M. S. Meenakshisundaram, The Secretary, Dr. R. Murali, The Principal and Mr. S. Thiagarajan, Head of the Department of Mathematics, The Madura College, Madurai for their encouragement. The author K. Indira is very thankful to the Manonmaniam Sundaranar University, Tirunelveli for allowing to do the research work.

Appendix A

Solution of the Eqs. (9)–(11) using Homotopy perturbation method

In this Appendix, we indicate how Eqs. (16)–(18) in this paper are derived. To find the solution of Eqs. (9)–(11) we first construct a Homotopy as follows:

$$(1-p)\left[\frac{\partial^2 u}{\partial X^2}-\frac{\partial u}{\partial T}\right]+p\left[\frac{\partial^2 u}{\partial X^2}-k_1 u+k_2 vw-\frac{\partial u}{\partial T}\right]=0 \tag{A1}$$

$$(1-p)\left[\frac{\partial^2 v}{\partial X^2}-\frac{\partial v}{\partial T}\right]+p\left[\frac{\partial^2 v}{\partial X^2}+k_3 u-k_4 vw-\frac{\partial v}{\partial T}\right]=0 \tag{A2}$$

$$(1-p)\left[\frac{\partial^2 w}{\partial X^2}-\frac{\partial w}{\partial T}\right]+p\left[\frac{\partial^2 w}{\partial X^2}+k_5 u-k_6 vw-\frac{\partial w}{\partial T}\right]=0 \tag{A3}$$

The boundary conditions are

$$T=0,\ u=1,\ v=1,\ w=1 \tag{A4a}$$

$$X=1,\ \frac{\partial u}{\partial X}=0,\ v=0,\ \frac{\partial w}{\partial X}=0 \tag{A4b}$$

$$X\to\infty,\ u=1,\ v=1,\ w=1 \tag{A4c}$$

The approximate solutions of (A1), (A2) and (A3) are

$$u=u_0+pu_1+p^2u_2+p^3u_3+\cdots\cdots \tag{A5}$$

$$v=v_0+pv_1+p^2v_2+p^3v_3+\cdots\cdots \tag{A6}$$

$$w=w_0+pw_1+p^2w_2+p^3w_3+\cdots\cdots \tag{A7}$$

Substituting Eqs. (A5), (A6) and (A7) into Eqs. (A1), (A2) and (A3) and comparing the coefficients of like powers of p

$$p^0:\frac{\partial^2 u_0}{\partial X^2}-\frac{\partial u_0}{\partial T}=0 \tag{A8}$$

$$p^1:\frac{\partial^2 u_1}{\partial X^2}-\frac{\partial u_1}{\partial T}-\kappa_1 u_0+k_2 v_0 w_0=0 \tag{A9}$$

$$p^0:\frac{\partial^2 v_0}{\partial X^2}-\frac{\partial v_0}{\partial T}=0 \tag{A10}$$

$$p^1:\frac{\partial^2 v_1}{\partial X^2}-\frac{\partial v_1}{\partial T}+\kappa_3 u_0-k_4 v_0 w_0=0 \tag{A11}$$

$$p^0:\frac{\partial^2 w_0}{\partial X^2}-\frac{\partial w_0}{\partial T}=0 \tag{A12}$$

$$p^1:\frac{\partial^2 w_1}{\partial X^2}-\frac{\partial w_1}{\partial T}+\kappa_5 u_0-k_6 v_0 w_0=0 \tag{A13}$$

Subjecting Eqs. (A8–A13) to Laplace transformation with respect to T we have,

$$p^0 : \frac{\partial^2 \overline{u}_0}{\partial X^2} - s\overline{u}_0 + 1 = 0 \tag{A14}$$

$$p^1 : \frac{\partial^2 \overline{u}_1}{\partial X^2} - s\overline{u}_1 - k_1\overline{u}_0 + k_2\overline{v}_0\overline{w}_0 = 0 \tag{A15}$$

$$p^0 : \frac{\partial^2 \overline{v}_0}{\partial X^2} - s\overline{v}_0 + 1 = 0 \tag{A16}$$

$$p^1 : \frac{\partial^2 \overline{v}_1}{\partial X^2} - s\overline{v}_1 + k_3\overline{u}_0 - k_4\overline{v}_0\overline{w}_0 = 0 \tag{A17}$$

$$p^0 : \frac{\partial^2 \overline{w}_0}{\partial X^2} - s\overline{w}_0 + 1 = 0 \tag{A18}$$

$$p^1 : \frac{\partial^2 \overline{w}_1}{\partial X^2} - s\overline{w}_1 + k_5\overline{u}_0 - k_6\overline{v}_0\overline{w}_0 = 0 \tag{A19}$$

The boundary conditions are

$$X = 1,\ \frac{\partial u}{\partial X} = 0,\ v = 0,\ \frac{\partial w}{\partial X} = 0 \tag{A20}$$

$$X \to \infty,\ u = \frac{1}{\mathrm{s}},\ v = \frac{1}{\mathrm{s}},\ w = \frac{1}{s} \tag{A21}$$

where s is the Laplace variable and an overbar indicates a Laplace-transformed quantity. Solving the Eqs. (A14–A19), and using the boundary conditions (A4a), (A4b) and (A4c) we can find the following results

$$\overline{u}_0(X,\ \mathrm{s}) = \frac{1}{s} \tag{A22}$$

$$\overline{u}_1(X,\ \mathrm{s}) = -\frac{k_1}{2}\left[\frac{e^{-\sqrt{s}(X-1)}}{s^{3/2}}\right](X-1) - \frac{k_1}{2}\left[\frac{e^{-\sqrt{s}(X-1)}}{s^2}\right] \tag{A23}$$

$$\overline{v}_0(X,\ \mathrm{s}) = \frac{1}{s} - \frac{e^{-\sqrt{s}(X-1)}}{s} \tag{A24}$$

$$\overline{v}_1(X,\ \mathrm{s}) = \frac{k_3}{2}\left[\frac{e^{-\sqrt{s}(X-1)}}{s^{3/2}}\right](X-1) \tag{A25}$$

$$\overline{w}_0(X,\ \mathrm{s}) = \frac{1}{s} \tag{A26}$$

$$\overline{w}_1(X,\ \mathrm{s}) = \frac{k_5}{2}\left[\frac{e^{-\sqrt{s}(X-1)}}{s^{3/2}}\right](X-1) + \frac{k_5}{2}\left[\frac{e^{-\sqrt{s}(X-1)}}{s^2}\right] \tag{A27}$$

According to the HPM, we can conclude that

$$\overline{u_0}(X,s) = \lim_{p\to 1}\overline{u}(X,s) = \overline{u}_0 + \overline{u}_1 + \cdots \tag{A28}$$

$$\overline{v}_0(X,s) = \lim_{p\to 1} \overline{v}(X,s) = \overline{v}_0 + \overline{v}_1 + \cdots \tag{A29}$$

$$\overline{w}_0(X,s) = \lim_{p\to 1} \overline{w}(X,s) = \overline{w}_0 + \overline{w}_1 + \cdots \tag{A30}$$

We put Eqs. (A14) and (A15) into Eqs. (A28), (A16) and (A17) into Eqs. (A29) and (A18) and (A19) into Eqs. (A30). Then using inverse Laplace transform, the final results can be described by Eqs. (16–18) in the text. The remaining components of $u_n(X)$ and $v_n(X)$ can be completely determined such that each term is determined by the previous term.

Appendix B

Nomenclature and units

Symbol	Meaning	Usual dimension
a	Concentration of the species A	mole cm^{-3}
b	Concentration of the species B	mole cm^{-3}
c	Concentration of the species C	mole cm^{-3}
a_0	Bulk concentration of the species A	mole cm^{-3}
b_0	Bulk concentration of the species B	mole cm^{-3}
c_0	Bulk concentration of the species C	mole cm^{-3}
l	Thickness of the planar electrode	cm
κ_f	Forward rate constant	s^{-1}
κ_b	Backward rate constant	s^{-1}
D_a	Diffusion coefficient of the species A	$cm^2\ s^{-1}$
D_b	Diffusion coefficient of the species B	$cm^2\ s^{-1}$
D_c	Diffusion coefficient of the species C	$cm^2\ s^{-1}$
F	Faraday constant	C
n	Number of electrons	None
t	Time	s
u	Dimensionless concentration of the species A	None
v	Dimensionless concentration of the species B	None
w	Dimensionless concentration of the species C	None
T	Dimensionless time	None
k_1, k_2, k_3 k_4, k_5 and k_6	Dimensionless rate constants	None
ψ	Dimensional current	None

Appendix C (Numerical simulation program)

```
function pdex4
m = 0;
x = linspace(1,5);
t = linspace(0, 1);
sol = pdepe(m,@pdex4pde,@pdex4ic,@pdex4bc,x, t);
u1 = sol(:, :, 1);
u2 = sol(:, :, 2);
u3 = sol(:, :, 3);
```

```
figure
plot(x,u1(end,:))
title('u1(x, t)')
xlabel('Distance x')
ylabel('u1(x,3)')
figure
plot(x,u2(end,:))
title('u2(x, t)')
xlabel('Distance x')
ylabel('u2(x,3)')
figure
plot(x,u3(end,:))
title('u3(x, t)')
xlabel('Distance x')
ylabel('u3(x,3)')
function [c, f, s] = pdex4pde(x, t, u, DuDx)
c = [1;1;1];
f = [1;1;1].*DuDx;
k = 0.1;
k1 =0.001;
k2=0.001;
F=−k*u(1)+k*u(2)*u(3);
F1=k1*u(1)−k1*u(2)*u(3);
F2=k2*u(1)−k2*u(2)*u(3);
s = [F;F1;F2];
function u0 = pdex4ic(x)
u0 = [1; 1;1];
function [pl, ql, pr,qr] = pdex4bc(xl,ul, xr, ur, t)
pl = [0; ul(2);0];
ql = [1;0;1];
pr = [ur(1)−1;ur(2)−1;ur(3)−1];
qr = [0;0;0];
```

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