Mathematical Modeling of Suspended Microbial Fuel Cells and Electron Transfer Mediator using Homotopy Perturbation Method

P. Jeyabarathi, M. Kannan, L. Rajendran

Abstract: The theoretical model of microbial fuel cells with suspended cells and an additional electron transfer mediator is analyzed. Many biological, chemical and electrochemical reactions occur in the bulk liquid and on the surface of the electrode with the substrate (glucose), oxidized mediators and reduced mediators. The homotopy perturbation method (HPM) is used to solve the nonlinear diffusion equations in microbial fuel cells. Direct and approximate polynomial expressions of a substrate (glucose), oxidized mediator and reduced mediator concentration are obtained at the mass transfer balance layer. The results of the experiment are compared with the results of the analytical and simulation and satisfactory agreement is noted.

Keywords: Mathematical modeling, Numerical simulation, Microbial fuel cell, Non-linear diffusion equation, Homotopy perturbation method.

I. INTRODUCTION

An MFC can use complex organic substrates as a source of power, including domestic, industrial and agricultural wastewater, drawing attention as a promising technology that combines sustainable energy with waste treatment [1,2]. In this study, in addition to the electrochemical reactions that occur in the MFC, similar to those developed for chemical fuel cells, a steady-state, one-dimensional model is provided that accounts for coupled heat, charge and mass transfer, and biofilm formation. The model predicts the right trends in the effect of current density on cell voltage, as well as the impact of substrate concentration and temperature on the output of MFC and the thickness of biofilm. The proposed design is implemented quickly and easily, making it ideal for use in MFC calculations at the real-time system level. Microbial fuel cells (MFCs) are a type of fuel cell using bacteria as a biocatalyst to transform the chemical energy found in organic matter into electricity. Previously described the potential benefits of biological systems over standard chemical systems [3].

First, MFCs that can perform under very mild conditions are feasible to create. Second, any biodegradable material that the micro-organisms can oxidize can serve as a gas. Several recent reviews devoted to understanding the microbiology of MFC processes [4,5] as well as developments in technical aspects [6-10] detail the design configurations for MFCs. Mathematical modeling rigor provides a framework for evaluating hypothesis and a tool for incorporating information from multiple disciplines and from researchers with backgrounds ranging from biology to engineering. Zhang and Halme’s [11] preliminary research attempted to simulate only with suspended cells and an added mediator the currents generated by MFC. Kato-Markus et al [12] models are dedicated to biofilm-based MFC, the design which microbiologists and engineers are currently most experimentally researching. In testing more complex MFC models, including biofilms, recently proposed by Piccioreau et al [13], parameters obtained in this study were used. In this paper, for all values of parameters, analytical expressions of the concentration of substrate (glucose), oxidized mediator, reduced mediator are obtained by solving the non-linear equation using the homotopy perturbation method.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Meanings</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_{S,b}$</td>
<td>Concentration of bulk substrate (glucose)</td>
<td>-</td>
<td>mM</td>
</tr>
<tr>
<td>$C_{S,b,red}$</td>
<td>Concentration of bulk reduced mediator</td>
<td>-</td>
<td>mM</td>
</tr>
<tr>
<td>$C_{S,b,ox}$</td>
<td>Concentration of bulk oxidized mediator</td>
<td>-</td>
<td>mM</td>
</tr>
<tr>
<td>$C_{S,0}$</td>
<td>Initial concentration of substrate (glucose)</td>
<td>j</td>
<td>mM</td>
</tr>
<tr>
<td>$C_{S,0,red}$</td>
<td>Initial concentration of reduced mediator</td>
<td>j</td>
<td>mM</td>
</tr>
<tr>
<td>$C_{S,0,ox}$</td>
<td>Initial concentration of oxidized mediator</td>
<td>j</td>
<td>mM</td>
</tr>
<tr>
<td>$S$</td>
<td>Biomass concentration in bulk liquid</td>
<td>$1500$</td>
<td>g m$^{-3}$</td>
</tr>
<tr>
<td>$D_{med}$</td>
<td>Diffusion coefficient of mediator</td>
<td>$0.00009$</td>
<td>m$^2$ day$^{-1}$</td>
</tr>
</tbody>
</table>

Revised Manuscript Received on November 15, 2019

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Retrieval Number: D7979118419/2019©BEIESP

Published By: Blue Eyes Intelligence Engineering & Sciences Publication
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<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>D_L</td>
<td>Diffusion coefficient of substrate (glucose)</td>
<td>$0.0001 \text{ m}^2 \text{ s}^{-1}$</td>
</tr>
<tr>
<td>$r_L$</td>
<td>Thickness layer</td>
<td>$50 \mu \text{m}$</td>
</tr>
<tr>
<td>V_B</td>
<td>Bulk liquid volume</td>
<td>$0.0002 \text{ m}^3$</td>
</tr>
<tr>
<td>A_E</td>
<td>Electrode area</td>
<td>$25 \text{ cm}^2$</td>
</tr>
<tr>
<td>V_C</td>
<td>Cathode potential</td>
<td>$0.65 \text{ V}$</td>
</tr>
<tr>
<td>K_M</td>
<td>Total cell resistance</td>
<td>$0.0001 \Omega$</td>
</tr>
<tr>
<td>$i_{ref}$</td>
<td>Exchange current density in reference conditions for mediator oxidation</td>
<td>$0.0002 \text{ A m}^{-2}$</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
<td>$-$</td>
</tr>
<tr>
<td>$E_{0,\text{red}}$</td>
<td>Standard reduction potential for the pair of mediator</td>
<td>$0.477 \text{ V}$</td>
</tr>
<tr>
<td>$b$</td>
<td>Tafel coefficient for mediator oxidation</td>
<td>$0.12 \text{ V}$</td>
</tr>
<tr>
<td>$k_2$</td>
<td>Rate constant</td>
<td>$0.0002 \text{ (L mol^{-1}) s^{-1}}$</td>
</tr>
<tr>
<td>$r_2$</td>
<td>Yield substrate</td>
<td>$0.22 \text{ S Med^{-1}}$</td>
</tr>
<tr>
<td>$K_M$</td>
<td>Monod coefficient for substrate</td>
<td>$2 \text{ mL}$</td>
</tr>
<tr>
<td>$K_{\text{mon}}$</td>
<td>Monod coefficient for oxidized mediator</td>
<td>$2 \text{ mL}$</td>
</tr>
<tr>
<td>$d$</td>
<td>Distance from anode surface</td>
<td>$50 \mu \text{m}$</td>
</tr>
</tbody>
</table>

II. MATHEMATICAL FORMULATION OF THE PROBLEM

The reaction scheme of mediator and substrate oxidation is

$$Y_S + M_{\text{ox}} \rightarrow M_{\text{red}} + \text{products}$$

(1)

$$M_{\text{red}} + e \rightarrow M_{\text{ox}} + nH^+ + ne^-$$

(2)

This reaction scheme is represented in Figure 1.

The mass balance equations in the bulk liquid for soluble components are [14]:

$$\frac{dC_{B,S}}{dt} = r_{B,S}$$

(3)

$$\frac{dC_{B,M}}{dt} = r_{B,M}$$

(4)

The corresponding initial conditions are

at $t = 0$, $C_{B,S} = C_{0,S}$

(6)

at $t = 0$, $C_{B,M} = C_{0,M}$

(7)

at $t = 0$, $C_{B,Med} = C_{0,Med}$

(8)

where $C_{B,S}, C_{B,Med}$ and $C_{B,Med}$ represent concentration of bulk substrate, oxidized mediator and reduced mediator.

The rates of reaction are as follows:

$$r_1 = k_1 C_S \frac{C_{B,Med}}{K_M + C_{B,Med} + C_M}$$

(9)

$$r_2 = -Y_{B,S} r_1$$

(10)

$$r_{B,M} = \frac{[C_{B,Med} - C_{B,M}]}{nF} = r_{B,M} - \frac{1}{nF}$$

(11)

where $k_1$ is a maximum rate coefficient, $K_M$ and $K_{\text{mon}}$ are Monod coefficients for substrate and oxidized mediated, $C_s$ is the biomass concentration, current density $j (\text{A m}^{-2})$, and the electrode surface concentration values, $C_{B,Med}$ and $C_{B,Med}$ [mol m$^{-2}$](Faraday’s law)

By using the Butler-Volmer equation[15] shown in the below formula:

$$i = i_{a,c} \frac{C_{B,Med} + C_{B,M}}{C_{B,Med}} \frac{C_{B,Med} + C_{B,M}}{C_{B,Med} + C_M}$$

(12)

with parameters being the Tafel slope and the current density transfer function of the current density $i_{ref}$ measured at certain reactant and product reference concentrations $C_{ref}$. The over- potential activation $\eta_{a,c}$ ($V$) is a function of the current $i$ passing through the fuel cell:

$$\eta_{a,c} = \frac{1}{V} \left[ V_C - 0.622 + R_{\text{ext}} \right] - \frac{2.303}{F} \frac{C_{B,Med} - C_{B,M}}{C_{B,Med} + C_M}$$

(13)

where $V_C$ is cathode potential, $R_{\text{ext}} + R_{\text{int}}$ is the mediator's standard reduction potential, $R_{\text{ext}} + R_{\text{int}}$ is total cell resistance, and $t$ is current.

A. Bulk liquid mass balances

The mass balance equation of $C_{L,S}, C_{L,Med}$ and $C_{L,Med}$ are

$$D_L \frac{d^2C_{L,S}}{dz^2} + r_{L,S} = 0$$

$$D_L \frac{d^2C_{L,Med}}{dz^2} + r_{L,Med} = 0$$

$$D_L \frac{d^2C_{L,Med}}{dz^2} + r_{L,Med} = 0$$

B. Diffusion layer

The mass balance equation of $C_{L,S}, C_{L,Med}$ and $C_{L,Med}$ are

$$D_L \frac{d^2C_{L,S}}{dz^2} + r_{L,S} = 0$$

$$D_L \frac{d^2C_{L,Med}}{dz^2} + r_{L,Med} = 0$$

$$D_L \frac{d^2C_{L,Med}}{dz^2} + r_{L,Med} = 0$$
\[
\frac{d^2 C_{L,Mod}}{dz^2} + \nu C_{L,Mod} = 0
\]  
(14)

\[
\frac{d^2 C_{L,Med}}{dz^2} + \nu C_{L,Med} = 0
\]  
(15)

the corresponding boundary conditions is

\[ C_{L,Ex} = C_{E,Ex} \rightleftharpoons C_{L,Med} = C_{E,Med} \text{ for } z = L_e \]  
(16)

\[ C_{L,Ex} = C_{E,Ex} \rightleftharpoons C_{L,Med} = C_{E,Med} \text{ for } z = 0 \]  
(17)

IV. NUMERICAL SIMULATION

Numerically, the non-linear equations (3-5) are solved for the initial condition equations (6-8). For ordinary differential equations, the function ode 45 in Scilab software is used to problems with two-point boundary value (BVPs). Appendix(C) for will also be provided to the Scilab program. The numerical results

III. ANALYTICAL EXPRESSION OF CONCENTRATION USING HOMOTOPY PERTURBATION METHOD.

In recent years, many more numerical methods have been used in physical and chemical sciences to solve a wide range of linear and nonlinear problems. We will use the homotopy Perturbation Method (HPM) in this article to solve such differential equation systems. This article confirms the method's strength, simplicity, and efficiency compared to other methods, JH. He [16-20] first introduced the HPM technique. The concentrations of bulk liquid can be obtained using HPM (see Appendix (A)) as follows:

\[ C_{E,Ex} = C_{E,Med} \rightleftharpoons C_{E,Med} \]  
(18)

\[ C_{E,Ex} = C_{E,Med} \rightleftharpoons C_{E,Med} \]  
(19)

A. Analytical expression of concentration in diffusion layer using homotopy perturbation method.

By solving the Eqs. (14-16) using HPM (Appendix (B)), we can obtain the concentration of substrate (glucose), oxidized mediator and reduced mediator at MTBL as follows:

\[ C_{E,Mod}(z) = \frac{D_a}{D_{E,Ex}} \left( \frac{C_{E,Mod}}{C_{E,Med}} \right) \left( 1 - e^{-\frac{D_{E,Ex}}{D_a}} \right) \]  
(20)

\[ C_{E,Med}(z) = \frac{D_a}{D_{E,Ex}} \left( \frac{C_{E,Mod}}{C_{E,Med}} \right) \left( 1 - e^{-\frac{D_{E,Ex}}{D_a}} \right) \]  
(21)

where \( k_e \) = \( \frac{D_a C_{E,Mod}}{k_{B,Ex} C_{E,Med} + k_{E,Ex} C_{E,Med}} \) + \( \frac{D_a C_{E,Med}}{k_{E,Ex} C_{E,Med} + k_{B,Ex} C_{E,Med}} \)

\[ C_{E,Med}(z) = \frac{D_a}{D_{E,Ex}} \left( \frac{C_{E,Mod}}{C_{E,Med}} \right) \left( 1 - e^{-\frac{D_{E,Ex}}{D_a}} \right) \]  
(22)

\[ C_{E,Med}(z) = \frac{D_a}{D_{E,Ex}} \left( \frac{C_{E,Mod}}{C_{E,Med}} \right) \left( 1 - e^{-\frac{D_{E,Ex}}{D_a}} \right) \]  
(23)

\[ C_{E,Med}(z) = \frac{D_a}{D_{E,Ex}} \left( \frac{C_{E,Mod}}{C_{E,Med}} \right) \left( 1 - e^{-\frac{D_{E,Ex}}{D_a}} \right) \]  
(24)

\[ C_{E,Med}(z) = \frac{D_a}{D_{E,Ex}} \left( \frac{C_{E,Mod}}{C_{E,Med}} \right) \left( 1 - e^{-\frac{D_{E,Ex}}{D_a}} \right) \]  
(25)

parameter values are also opposed with the analytical expressions equations obtained (19-21).
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Fig 2. Comparison of analytical and numerical solutions of concentration of substrate (glucose) for some fixed experimental values of parameters $k_1 = 0.002, C_x = 1500, Y = 0.22, K_x = 1, K_{Mox} = 2, C_{SO} = 1, C_{Mox} = 1$. The key to the plot: (***) Eqn. (19) and (---) numerical simulation of Eqn. (3).
Fig 3. Comparison of analytical and numerical solutions of concentration of oxidized mediator for some fixed experimental value of parameters $k_i = 0.002, C_x = 1500, K_x = 1, K_{Med} = 2, C_{Med} = 1, C_{Med,i} = 1, b_{ref} = 0.001, C_{E,Med} = 0.5, C_{E,Med,i} = 1, V_c = 0.68, V_p = 0.0002, R_{int} + R_{ext} = 0, b = 0.12, n = 24, F = 96480$
$E_{Med,Med,i} = 0.477, A_E = 25$. The key to the plot: (***) Eqn.(20) and (---) numerical simulation of Eqn. (4). $b_{ref}=0.0$ for (a), (b), (f) and (d) and $b_{ref}=0.5$ for (c).

Fig 4. Comparison of analytical and numerical solutions of concentration of reduced mediator for some fixed experimental value of parameters $k_i = 0.002, C_x = 1500, K_x = 1, K_{Med} = 2, C_{Med} = 1$
$C_{Med,i} = 1, b_{ref} = 0.001, C_{E,Med} = 0.5, C_{E,Med,i} = 1, V_c = 0.1V_p = 0.002, R_{int} + R_{ext} = 0, b = 0.12, n = 24, F = 96480$
$E_{Med,Med,i} = 0.477, A_E = 25$. The key to the plot: (***) Eqn.(21) and (---) numerical simulation of Eqn. (5).

Fig 5. The local concentration of substrate (glucose) MTBL versus time using Eqn. (23) for some fixed experimental value of the parameters $k_i = 0.002, C_x = 1500, K_x = 1, K_{Med} = 2, C_{Med} = 1, C_{Med,i} = 1, C_{E,Med} = 1, C_{E,Med,i} = 1, D_x = 0.0001$ for the various value of electrode surface $z$, thickness $l_x$, $L_x = 50$ for (a) and $z = 50$ (b).
Mathematical Modeling of Suspended Microbial Fuel Cells and Electron Transfer Mediator using Homotopy Perturbation Method

Fig 6. The local concentration of reduced mediator MTBL versus time using Eqn. (24) for some fixed experimental value of parameters $k_i = 0.0002, C_x = 1500, K_s = 1, K_{Med} = 2, C_{Ox} = 1, C_{Med} = 1$, $b_o = 0.1, C_{E,Med} = 1, C_{E,Med} = 1, V_C = 0.68, V_0 = 0.1, R_M + R_{M,int} = 0, b = 0.12, n = 24, F = 96480$

$E_{Mox/Med}^0 = 0.477, A_E = 25, D_{Med} = 0.0099, z = 1, L_z = 20$ for various values of electrode surface $z$ and thickness $L_z$.

Fig 7. The local concentration of oxidized MTBL versus time using Eqn. (25) for some fixed experimental value of parameters

$k_i = 0.0002, C_x = 1500, K_s = 1, K_{Med} = 2, C_{Ox} = 1, C_{Med} = 1, b_o = 0.0001, C_{E,Med} = 0.5, C_{E,Med} = 1, V_C = 0.68, V_0 = 0.0001, R_M + R_{M,int} = 0, b = 0.1225, n = 24, F = 96480$

$E_{Mox/Med}^0 = 0.477, A_E = 25$ for various values of electrode surface $z = 1$ for (a) and thickness $L_z = 20$.
Fig 8. Concentration of oxidized mediator versus current were computed using Eqn. (20) the experimental values $C_x = 1500, K_x = 1, K_{Max} = 2, C_{0,3} = 1, i_{0,ref} = 0.001, C_{E,Max} = 0.5, C_{E,Mod} = 1$ $V_C = 0.68, V_E = 0.1, R_{ext} + R_{ref} = 0.001, F = 96480$ for various value of parameters $k_j = 0.000002t = 1$ for (a), $k_j = 0.002, t = 0.1$ for (b), $k_j = 0.002, t = 100$ for (c) and $k_j = 0.002, t = 100$ for (d).

Fig 9. The concentration of reduced mediator versus current was computed using Eqn. (21) for some fixed experimental value of a parameters $k_j = 0.0002, C_x = 1500, K_x = 1, K_{Max} = 2, C_{0,3} = 1$ $C_{0,3} = 1, C_{E,Mod} = 0.001, C_{E,Max} = 0.5, C_{E,Mod} = 1, V_C = 0.68, V_E = 0.1, b = 0.12, n = 24, F = 96480, E_{E,Mod} = 0.477, A_E = 25, t = 10, R_{ext} + R_{ref} = 0.01$ for (b) and (c), $R_{ext} + R_{ref} = 0.001$ for (c).

V. DISCUSSION

Equations (19) to (21) represent the generally closed type of analytical expression for substrate (glucose) concentrations, oxidized mediator and reduced mediator for non-steady state conditions and different system parameters. Influence of parameters $Y_g, C_x, k_j, K_x, K_{Max}$ on the concentration of substrate (glucose) of some experimental values of parameters is shown in figure 2. The concentration of the substrate is decreased when the yield $Y_g$ decrease and rate constants $k_j$ increase. Monod coefficient for substrate and oxidized mediator $K_S, K_{Mox}$ and biomass concentration $c_g$ lead to decrease the substrate concentration.

The effect of the parameters $V_E^0, A_E, E_{E,Mod}, V_C, V_{E,ref}, b$ on the concentration of oxidized...
mediator is shown in figure 3(b),(d) and (f). From the figure, it is observed that the concentration of oxidized mediator increases with the decreases in volume, standard reduction potential, tafel coefficient. But the concentration of oxidized mediator decreases on increasing in electrode area, cathode potential and exchange current density. Figure 4 represents the effect of cathode potential $\nu_c$ and electrode area $A_g$ on the concentration of reduced mediator. From this figure, it is inferred that there is no significant difference in the concentration of reduced mediator for the values of parameters $\nu_c = 0.1, 1, 10$ and $A_g = 0.1, 1, 10$.

From figure 5, it is observed that an increase in thickness layer $l_t$ or decrease in anode distance $z$ results in an increase in the local concentration of substrate in the mass transfer balance layer. The concentration of oxidized mediator and reduced mediator for different values of anode distance $z$ and thickness layer $l_t$ are displayed in figure 6 and 7. From the figure it is noticed that a decrease in $z$ and $l_t$ leads to decrease in the concentration of reduced mediator in mass transfer boundary layer. From figure 7 it is noted that an increase in anode distance $z$ leads to decreases in the oxidized mediator in mass transfer boundary layer.

Figure 8 represents the concentration of oxidized mediator versus current for different value of parameters total cell resistance, standard reduced potential, cathode potential and bulk liquid volume. From this figure, it is clear that decrease in the concentration of oxidized mediator when the $R_{ct} + R_{oi} = E_{Mox/Med}$ and $V_c$ decreases. Figure 9 depicts the concentration of reduced mediator versus current for different value of parameters total cell resistance, standard reduced potential, cathode potential and bulk liquid volume. Clearly as $V_c$ increases the concentration of reduced mediator increases. From the figure it is observed that $R_{ct} + R_{oi} = E_{Mox/Med}$ and $V_c$ is decreases when the concentration of reduced mediator decreases.

VI. CONCLUSION

The non-linear reaction-diffusion equation in the modeling of suspended microbial fuel cells and electron transfer mediator was solved analytically. The approximate analytical expression of concentration of substrate (glucose), oxidized mediator and reduced mediator were obtained for all experimental values of parameters using the homotopy perturbation method. A satisfactory agreement with numerical simulation (Scilab program) is noted. These analytical expressions can be used to analyze the effect of various parameters such as thickness layer, yield substrate, biomass concentration, standard reduction potential, total cell resistance, Tafel coefficient, Monod coefficient for substrate and oxidized mediator, cathode potential, exchange current density and rate constant.

APPENDIX

A. Approximate analytical solution of nonlinear Eqn. (3) using HPM for bulk liquid mass balances [14].

In this Appendix, we derive the general solution of the nonlinear reaction equation (14) using the new approach homotopy perturbation method. We begin by constructing the homotopy for Eqn. (23) as follows:

$$\left( 1 - r \right) \frac{d^2 C_{S,t}}{dz^2} + \frac{Y_c k_c C_S}{C_{Mox,t}} \left( \frac{C_{S,t}}{C_{Mox,t}} \right)_{|z=0} = 0$$  \hspace{1cm} (B1)

The approximate solution of the Eq. (B1) is

$$C_{S,t}(z) = C_{S,t}(0) + C_{S,t}^{rel}(z)$$  \hspace{1cm} (B2)

Substituting equation (B2) into equation (B1) and equate the terms with identical power of $r$, we obtain

$$r^0 : \frac{d^2 C_{S,t}}{dz^2} + \frac{Y_c k_c C_S}{C_{Mox,t}} \left( \frac{C_{S,t}}{C_{Mox,t}} \right)_{|z=0} = 0$$  \hspace{1cm} (B3)

with boundary conditions for Eqn. (B3) given by

$z=0, C_{S,t} = C_{S,t}$

$z=L, \frac{dC_{S,t}}{dz} = 0$

(B4)

(B5)

Solving Eqn. (B3) with boundary conditions (B4) and (B5) we get

$C_{S,t}(z) = C_{S,t}(0) + C_{S,t}^{rel}(z)$

(B6)

C. Scilab program for the numerical solution of Eqns. (3-5)

function main2
options= odeset('RelTol',1e-6,'Stats','on');
Xo = [1; 1; 1];
tspan = [0,100];
tic
[t,X] = ode45(@TestFunction,tspan,Xo,options);
toc
end

function y= TestFunction(t,x)

end
REFERENCES


ACKNOWLEDGMENT

The Authors are also thankful to Shri J. Ramachandran, Chancellor, Col. Dr. G. Thiruvasagam, Vice-Chancellor, Academy of Maritime Education and Training (AMET), Deemed to be University, Chennai, for their constant encouragement.

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