

Modeling of Microbial Fuel Cells with the Aim of Energy Production and Wastewater Treatment

Abstract

The present study was conducted to model the fuel cells with the aim of energy production and wastewater treatment. In the present study, the obtained equations for the model were solved using numerical methods in MATLAB software. First, the series of ordinary differential equations (ODE) for C_B were solved in the time interval of t_n and $t_n+\Delta t$. The new C_B values obtained in each step were used to obtain other new C_B values in the time interval of t_n to $t_n+\Delta t$, and C_L concentration values at a time of t_n were used to calculate r_L and C_E rates to obtain r_E . Non-linear methods were used to solve partial differential equations (PDE). The geometry of the microbial fuel cell including the volume and surface area of the anode, operating conditions including pH, biomass concentration, and initial concentrations of substrate and medium, and parameters related to the electrical circuit including external resistance and cathode potential was directly adapted from the article by Delaney et al. Some kinetic parameters such as the rate constant for the reduction of thionine and its standard potential had been also calculated in another article by Roller et al. The results showed that by combining the current intensity-time curves obtained from the study by Delaney et al, a total value of 12.6 C was obtained. It means that only 74.8 C was the result of glucose oxidation. Using these data, the value of $YQ=0.37$, and accordingly the real value of glucose efficiency on $Y_s=0.22 \text{ mol.mol}^{-1}$ was obtained. These values provide a better fit of current intensity-time and electric charge-time curves to experimental data.

Keywords: *Microbial fuel cells, Energy production, Wastewater treatment.*

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Introduction

Microbial fuel cells (MFC) are considered one of the important potentials in the supply of clean and renewable energy in the future. In addition to the supply of electrical energy, which is the most widely used and flexible among other types of energy, MFCs cause the slightest pollution to the environment and play a major role in the treatment and eliminating the environmental pollution such as urban wastewater and leachate from urban solid wastes. The technology of microbial fuel cells (MFCs), which convert the energy stored in the bonds of the organic compound into electrical energy through catalytic reactions by microorganisms, has recently received special attention (Allen and Bennetto, 1993; Gil et al., 2003; Moon et al., 2006; Choi et al., 2003).

The bacteria used in MFCs can produce electricity and at the same time, they decompose wastewater and organic matter biologically (Park and Zeikus, 2000; Oh and Logan, 2005). Mediator-less MFCs are very suitable for wastewater treatment and power generation since the mediator cost is eliminated in these MFCs. The electron transfer mechanism in the anode chamber is a key factor in understanding how MFCs work. As stated above, microbes transfer electrons to the anode through an electron transport system, which itself includes a series of compounds in the bacterial extracellular matrix or along with electron shuttles that have been dissolved in the solution. *Geobacter* belongs to the group of metal-reducing microorganisms that produce energy in the ATP form that is biologically useful and is obtained during the reduction of metal oxides under anaerobic conditions in soil and sediments.

Electrons are transferred to the final electron acceptor, such as Fe_2O_3 . It is mainly done through direct contact with inorganic oxides and metal-reducing microorganisms (Lovley et al., 2004; Vargas et al., 1998). The anodic reaction in a mediator-less MFC that consists of a metal-reducing bacterium and mainly belongs to the *Shewanella*, *Rhodospirillum rubrum*, and *Geobacter* families is similar to this process since the anode acts as the last electron acceptor and this is just like solid metal oxides (Lovley et al., 2004; Vargas et al., 1998; Holmes et al., 2004). An MFC consists of an anode chamber and a cathode chamber separated by a PEM. In a single-component MFC, the cathode chamber has been removed, since the cathode is "directly in contact with air" (Logan et al. colleagues, 2006; Rabaey and Verstraete, 2005; Bullen et al., 2006; Lovley, 2006).

Due to the complex configuration of two-component MFCs, they cannot be scaled up easily. Park et al. (DATE) designed a single-component MFC that consisted of a rectangular anode chamber and a porous cathode that was in the vicinity of air. Protons were transferred from the analyte solution to the porous cathode in the vicinity of air (Park and Zeikus, 2003). Logan and Lin (2004) designed an MFC with the anode inside a plastic cylinder chamber and the cathode outside the cylinder. Jang et al. (2004) designed another type of MFC that operated continuously. It was a Plexiglas cylinder divided into two parts using glass wool glass beads. The MFC considered in this study for modeling consists of two chambers that are separated by an ion exchange membrane that is selective to cations. The anode consists of cross-linked carbon and the cathode consists of platinum foil. N_2 gas was injected into both chambers to

remove oxygen and create mixing. The analyte is a solution of an electron exchange mediator such as thionine and glucose, which has been used in a phosphate buffer (pH=7) to maintain the polarization in a stable state. Fresh microorganisms kept in suspension in phosphate buffer are added to the solution in a certain amount. The desired MFC is kept in a 30 °C water bath to maintain the proper temperature.

Model development

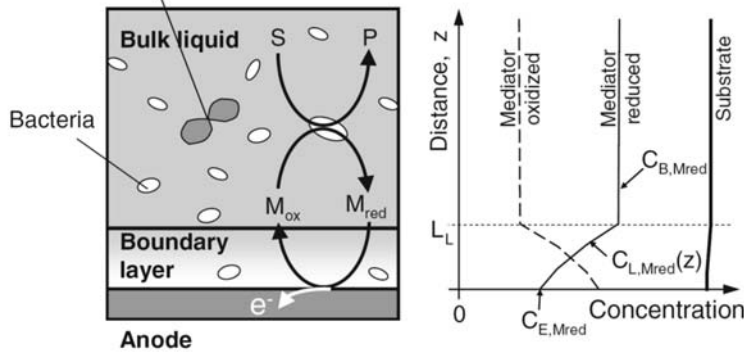
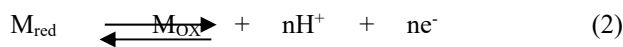


Figure 1: Schematic view of anode chamber and mass exchange boundary layer

It is assumed that the bulk liquid in the V_B volume is completely homogeneously mixed and contains biomass in a stable state. Dissolved components in the bulk solution penetrate the anode and vice versa through a mass transfer boundary layer attached to the anode with a thickness of L_L . To facilitate the calculations, a flat surface with an area of A_E is considered for the electrode surface. The reactions are considered simple so that only one reaction takes place in the bulk liquid, in which the organic substrate S is oxidized by microorganisms using mediators that are in the oxidized state, M_{ox} :



The biochemically reduced mediator, M_{red} , is electrochemically oxidized at the anode surface:



The stoichiometric coefficient of the substrate in equation (1) depends on the number of electrons in the substrate. For example, if glucose is completely oxidized to produce CO_2 , 24 electrons must be absorbed by the mediator. If each mole of the oxidized mediator accepts 2 electrons, the theoretical

$$\frac{k}{K_s + C_s} \frac{C_x}{K_{M_{ox}} + C_{M_{ox}}} \quad (3)$$

k

Where, k_1 is the maximum rate coefficient [$kmol$ mediator g biomass $^{-1}$ day $^{-1}$], C_x biomass concentration [gm^{-3}], K_s and

The mathematical model of the desired microbial fuel cell is obtained to predict the intensity of the produced current in the short term (several hours). For this model, two separate spaces are considered: the bulk liquid in the anode chamber and the mass exchange boundary layer that is connected to the anode. The schematic view of this model is shown in Figure 1.

efficiency of glucose per mediator will be: $Y_S^{max} = 1/12 = 0.0833$ mol/mol.

However, the real Y_s efficiency is less than this value because all the electrons in the substrate do not reach the electrode surface. Therefore, Coulomb's efficiency will be $Y_Q < 1$. The value of Y_s is determined from the real current intensity, which will be explained further. Since the nutrients required for the growth of microorganisms have not been added to the anode solution, the growth of microbes has been ignored. Similarly, other reaction products will not be involved in the calculations and analysis discussed. Thus, only three components dissolved in the solution will be discussed in this model: substrate (glucose), oxidized mediator, and reduced mediator. Assuming complete mixing in the system, the pH is assumed to be constant. The stoichiometry of an electrochemical reaction (2-3) indicates the number of electrons transferred, n . In this model, this value is assumed to be 2 because the mediator used in this model will be Thionin.

Reaction rate

To obtain the kinetics of the biological reaction (1), the Monod model was used as a function of the concentration of the substrate and the concentration of the mediator in the oxide state, MOX :

$K_{M_{ox}}$ are the coefficients of half-saturation (Monod) [$kmol$ m^{-3}] for the substrate and oxidized mediator, respectively. In a special case, in low concentrations ($C_s \ll K_s$ and

$C_{Mox} \ll K_{Mox}$), the second-order reaction ($r_1 = k_1 \cdot C_X \cdot C_S \cdot C_{Mox}$) can be used. This work was done in the article of Benneto et al. Using equation (3), the stoichiometry of reaction (1), and substrate concentration in bulk liquid, $C_{B,s}$ and mediator concentration in bulk liquid $C_{B,Mox}$, [kmol m⁻³] and biomass concentration C_X [g m⁻³], the net reaction rate of glucose and two mediator forms are obtained in this way:

$$r_{B,S} = -Y_s r_1 \left(\frac{C_X}{C_{B,Mox}} \right) C_{B,S}, \quad (4)$$

$$r_{B,Mred} = r_1 \quad (5)$$

$$r_{B,Mox} = -r_1 \quad (6)$$

$$r_{E,Mred} = - \frac{i(C_{E,Mred}, C_{E,Mox})}{nF} \quad (7)$$

$$r_{E,Mox} = \frac{i}{nF} \quad (8)$$

The intensity of the current produced during oxidation of the medium (at constant pH) can be expressed by the Butler-Volmer equation:

$$i = i_{0,ref} \left(\frac{C_{E,Mred}}{C_{ref,Mred}} \right) \left(\frac{C_{E,Mox}}{C_{ref,Mox}} \right)^{-1} \left[\exp \left(\frac{2.303}{b} \eta_{A,act} \right) - \exp \left(-\frac{2.303}{b} \eta_{A,act} \right) \right] \quad (9)$$

Where, b is the slope of the Tafel equation [V/decade of current] and $i_{0,ref}$ [A m⁻²] is the reference current intensity density calculated in a reference concentration of reactants and products, C_{ref} . It should be noted that different equations can be obtained for the current intensity density depending on the reaction mechanism that occurs on the electrode. Thus, the order of reactions (here, 1 for M_{red} and 1 for M_{ox}) is not necessarily the same as the stoichiometric coefficients. To calculate the current intensity density based on equation (9), the activation overpotential, $\eta_{A,act}$, [V] of the electrochemical reaction of the anode must be known. $\eta_{A,act}$ can be obtained from the potential balance around the MFC. When the microbial fuel cell is connected to an external resistance, R_{ext} , using Ohm's law, the voltage ratio of the microbial fuel cell, V_{cell} , and the intensity of the current passing through the external resistance can be expressed as an equation:

Also, an equation should be obtained for the rate of the electrochemical process. Electric current is created when a chemical substance dissolved in a solution (mediator) is oxidized on the surface of the anode and another substance is reduced on the surface of the cathode. The rate per unit area of the electrochemical reactions that occur on the surface of the anode can be expressed as a function of the current intensity density, i , [A m⁻²]. Thus, the net rate for the mediator conversion at the electrode surface (kmol m⁻² day⁻¹) will be a function of the current intensity density, stoichiometry, and the material concentration values at the anode surface, $C_{E,Mred}$ and $C_{E,Mox}$ [kmol m⁻³] (Faraday's law):

$$V_{cell} = IR_{ext} \quad (10)$$

By aggregating all the polarization losses in the anode and

cathode, the voltage of the microbial fuel cell is represented by the following equation:

$$V_{cell} = E_{cell} - \eta_{act} - \eta_{ohm} = (E_c - \eta_{C,act}) - (E_A + \eta_{A,act}) - \eta_{ohm} \quad (11)$$

In the above equation, the concentration overpotentials have not appeared since the equilibrium potentials of the cathode and anode, E_c and E_A , are directly calculated with the C_E concentration on the surface of the electrodes. C_E concentrations depend on mass transfer conditions and reaction rates and vary over time. Also, if we assume Ohm's law to be true for the electrolyte and electrodes, $\eta_{ohm} = IR_{int}$, where R_{int} includes the resistances of the electrons, ions, and the contact surface.

Finally, by integrating equations (10) and (11), we can obtain the activation overpotential of the anode:

$$\eta_{A,act} = (E_C - \eta_{C,act}) - E_A - I(R_{int} + R_{ext}) \quad (12)$$

Based on the examination of various reports, it can be concluded that the cathode potential can be assumed to be constant and independent of the current intensity, I , $V_C = E_C - \eta_{C,act}$. This assumption is only to simplify the calculations since the value of cathode activation polarization, $\eta_{C,act}$, compared to anode activation polarization is negligible.

Moreover, a certain value must be obtained for the equilibrium potential of the anode reaction. This value can be represented by the standard reduction potential, E_A^0 , which can be calculated with the real values of the concentrations at the electrode surface. For the mediator oxidation reaction at the surface of the anode, the relevant equation can be written as follows:

$$\eta_{A,act} = V_C - I(R_{int} + R_{ext}) - \left(E_{Mox/Mred}^0 - 0.06\text{pH} + \frac{0.06}{2} \lg \frac{C_{E,Mox}}{C_{E,Mred}} \right) \quad (14)$$

Based on equations (4) to (8) and assuming that the MFC works as a batch and the constant volume of the anode chamber

$$E_A = E_{Mox/Mred}^0 + \frac{RT}{nF} \ln \frac{C_{E,Mox} C_{E,H^+}^2}{C_{E,Mred}} \quad (13)$$

Finally, at constant pH and constant temperature of 30 °C and knowing the values of internal and external resistances and the reduction potential of the mediator in standard conditions, $E_{Mox/Mred}^0$, the activation overpotential can be obtained as a function of the current passing through the fuel cell, I , in this way:

solution, V_B , the mass equilibrium of the dissolved substances in the bulk liquid will be as follows:

$$\frac{dC_{B,S}}{dt} = r_{B,S}, \quad C_{B,S} \Big|_{t=0} = C_{0,S} \quad (15)$$

$$\frac{dC_{B,Mox}}{dt} = r_{B,Mox} + r_{E,Mox} \frac{A_E}{V_B}, \quad C_{B,Mox} \Big|_{t=0} = C_{0,Mox} \quad (16)$$

$$\frac{dC_{B,Mred}}{dt} = r_{B,Mred} + r_{E,Mred} \frac{A_E}{V_B}, \quad C_{B,Mred} \Big|_{t=0} = C_{0,Mred} \quad (17)$$

Due to the limitations in the boundary layer on the electrode surface, to calculate the reaction rates according to equations (7) and (8), the values of C_E concentrations should be used, which are different from the C_B concentrations in the bulk liquid. C_E concentrations can be calculated using the one-

dimensional mass equilibrium equation in the steady state for the mass transfer boundary layer, taking into account diffusion and local concentration-dependent reactions in the mass transfer boundary layer as follows:

$$D_S \frac{d^2 C_{L,S}}{dz^2} + r_{L,S} = 0, \quad C_{L,S} \Big|_{z=L_L} = C_{B,S}, \quad \frac{dC_{L,S}}{dz} \Big|_{z=0} = 0 \quad (18)$$

$$D_{\text{Mred}} \frac{d^2 C_{\text{L,Mred}}}{dz^2} + r_{\text{L,Mred}} = 0, \quad C_{\text{L,Mred}} \Big|_{z=L_L} = C_{\text{B,Mred}}, \quad D_{\text{Mred}} \frac{dC_{\text{L,Mred}}}{dz} \Big|_{z=0} + r_{\text{E,Mred}} = 0 \quad (19)$$

$$D_{\text{Mox}} \frac{dC_{\text{L,Mox}}}{dz^2} r_{\text{L,Mox}} = 0, \quad C_{\text{L,Mox}} \Big|_{z=L_L} = C_{\text{B,Mox}}, \quad D_{\text{Mox}} \frac{dC_{\text{L,Mox}}}{dz} \Big|_{z=0} + r_{\text{E,Mox}} = 0 \quad (20)$$

The concept of one of the boundary conditions is that at a certain distance from the electrode, which is equal to the thickness of the penetration layer ($z=L$), the concentrations are equal to the concentration values in the bulk liquid. The concept of another boundary condition is that in stable conditions, the reaction rate at the electrode surface ($z = 0$) is equal to the penetration rate ($C_L = C_E$). If the current intensity density, i , is assumed to be uniform on the entire surface of the electrode, the current produced by the fuel cell at any moment will be proportional to the surface of the electrode:

$$I = iA_E \quad (21)$$

Since the equation (21) is absolute with respect to I , the problem will be complicated since the intensity density of the current expressed by the Butler-Volume equation (Equation 9) is a function of the overpotential, which itself is a function of the current intensity, I (Equation 14). The amount of charge produced, QC (Coulombs), is obtained by integrating the cell current intensity over time. Coulomb efficiency (Y_Q) is defined as the ratio of the real produced charge QC to the maximum amount of charge, in theory, QC_{max} . The maximum amount of charge in theory is the number of available electrons (γ) for the reduction reaction in the entire oxidizable substrate (nS mol). The Coulomb efficiency can be obtained using the following equation:

$$Y_Q = \frac{Q_c}{Q_{c,\text{max}}} = \frac{Q_c}{\sum \gamma_i n_{si}} \quad (22)$$

Problem Solving

The obtained equations for the model were solved using numerical methods in MATLAB software. First, the series of ordinary differential equations (ODE) (15 to 17) were solved for C_B in the time interval of t_n and $t_n + \Delta t$. The new C_B values obtained in each step were used to obtain other new C_B values in the time interval of t_n to $t_n + \Delta t$. C_L concentration values at a time of t_n were used to calculate r_L and C_E rates to obtain r_E . Non-linear methods were used to solve partial differential equations (PDE) (18 to 20), since calculating the current intensity requires solving the implicit equation for I , obtaining the solutions for C_B , $C_L(z)$, and C_E at any moment continued until a solution for I was obtained that is applied in the equation for the current intensity, I (21).

Calculation of parameters

To quantitatively compare the model results with the experimental values presented in Delaney et al.'s article, various parameters need to be calculated. The geometry of the microbial fuel cell including the volume and surface area of the anode, operating conditions including pH, biomass concentration, and initial concentrations of substrate and medium, and parameters related to the electrical circuit including external resistance and cathode potential was directly adapted from the Delaney et al.'s article. Some kinetic parameters such as the rate constant for the reduction of thionine and its standard potential were also calculated in another article by Roller et al. Some unknown parameters should also be extracted from other experimental data. First, by fitting the current intensity-time and electrical charge-time curves, the real values of substrate conversion efficiency Y_S and Coulomb efficiency Y_Q were calculated (Figures 2 and 3).

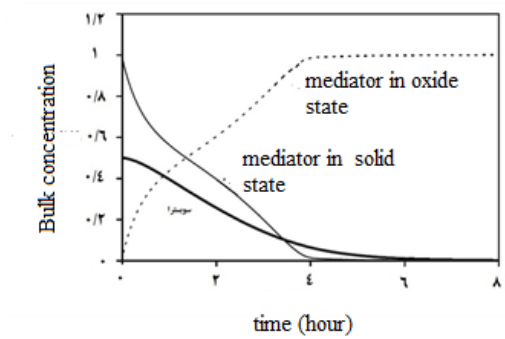


Figure 2: Substrate and mediator concentration changes in standard conditions

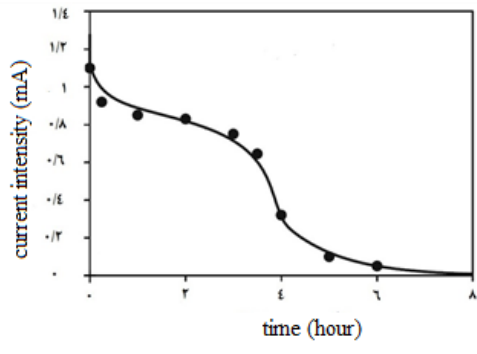


Figure 3: Current intensity changes over time

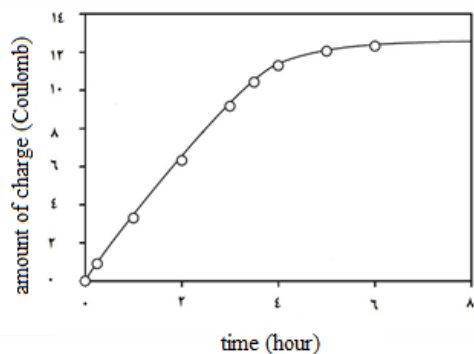


Figure 4: Changes in the amount of charge produced over time

The total electric charge is obtained as a result of the oxidation of the substrate and the mediator. For this purpose, by applying Faraday's law in the ideal state, that is, when all the electrons of glucose are converted to current (QC, S=C0, SVBnF), the value of C charge was obtained at 23.16 (assuming $n=24$ mole e^- mol $^{-1}$ for glucose). Similarly, with the mediator oxidation until the completion of the process, 3.86 C was obtained (assuming $n=2$ mole e^- mol $^{-1}$ for thionin), and a total of 27.01 coulombs was obtained. The same amount of electric charge was also obtained in the simulation assuming the efficiency of glucose on the mediator is $Y_S=0.0833$ mol $^{-1}$ (Figures 5 and 6).

In practice, by combining the current intensity-time curves obtained from the study of Delaney et al, a total value of 12.6 C was obtained. It means that only 8.74 C was the result of glucose oxidation. Using these data, the value of $Y_Q=0.37$, and accordingly the real value of glucose efficiency on $Y_S=0.22$ mol.mol $^{-1}$ is obtained. These values provide a better fit of the current-time and electric charge-time curves to the experimental data (Figures 3 and 4), since less real current intensity compared to the ideal state per one mole of glucose is produced (Figures 5 and 6).

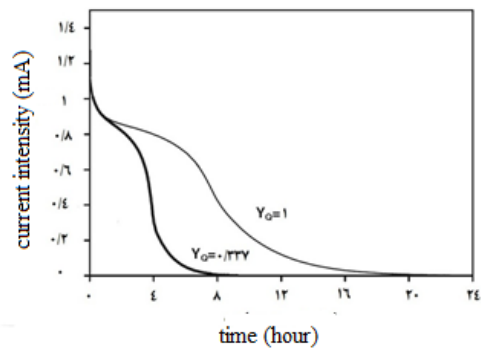


Figure 5: Modeled curve of current intensity with time in the ideal condition of $Y_Q=1$ and experimental condition of $Y_Q=0.337$

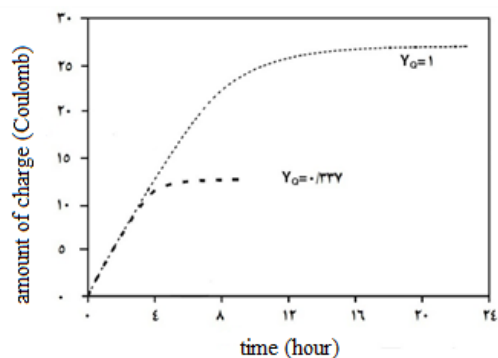


Figure 6: The modeled curve of the amount of charge produced with time in the ideal conditions of $Y_Q=1$ and the experimental conditions of $Y_Q=0.337$

To calculate the mass transfer parameters for the medium and the exchange current intensity $i_{0,ref}$, the experimental current intensity-voltage curves obtained from the study of Delaney et al who used thionine and *Proteus Vulgaris* were used. Higher values of the intensity of the exchange currents increase the values displayed in the V_{cell} -I curve (7). An acceptable value is obtained in the range of 10^{-3}Am^{-2} . After obtaining these

values, the mass transfer rate of the mediator to the surface of the anode will be the limiting factor for the intensity of the produced current, which was obtained at 25 mA. Larger values of mass transfer resistances, such as thicker transfer layers or lower penetration coefficients, will reduce the level of limitation in current intensity (7) and produced power (8).

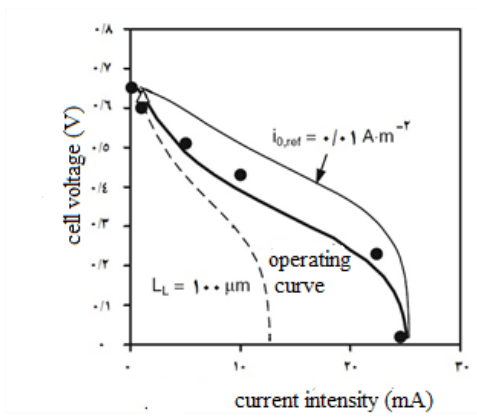


Figure 7: The modeled curve of voltage with current intensity in standard conditions, high current intensity exchange rate $i_{0,ref}=0.01$ A/m², and high resistance in mass transfer rate $L_L=100\text{m}$

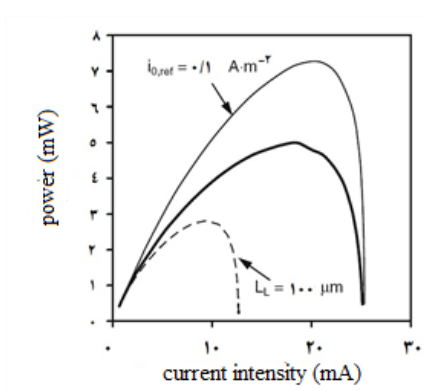


Figure 8: The modeled curve of the produced power with current intensity in standard conditions, high current intensity exchange rate $i_{0,ref}=0.01$ A/m², and high resistance in mass transfer rate $LL=100m$

The half-saturation coefficients of the Monod equation for the substrate and mediator affect the current intensity-time curves when they are reduced to small values, for example, $C_S \ll K_S$ and $C_{Mox} \ll K_{Mox}$. This mathematical modeling aims to evaluate the effect of different operating parameters of the microbial fuel cell on the intensity of the produced current and the level of substrate consumption in relation to time.

Conclusion

The present study was conducted to model microbial fuel cells with the aim of energy production and wastewater treatment. The slow metabolism of microbes and the slow transfer of electrons from the substrate to the surface of the anode and protons from the electrolyte of the anode to the cathode chamber (through penetration in the membrane or salt bridge) are among the most important barriers to the progress and commercialization of the mass production of microbial fuel cells. If the amount of power produced by microbial fuel cells is optimized to the extent that it can be used at a commercial level, mathematical modeling in defining appropriate configurations with the highest efficiency and the lowest power loss for various applications of these cells will find a special place.

Mathematical modeling as a precise tool provides an opportunity for a designer to determine the energy required for a specific application of influencing parameters such as the type, material, and dimensions of the electrodes, the required surface of the electrodes, the porosity of the electrodes, the type of microbes used in the anode electrolyte, the type of mediators suitable for transporting the produced electrons, the dimensions of the anode and cathode chambers, the way microbial fuel cell works (batch, semi-batch, or stable current), and the number of fuel cells required. Mathematical models that are currently presented by various researchers are considered not as tools, but as recommended solutions for mathematical modeling of microbial fuel cells, where the theoretical data obtained from them have the highest compatibility with the existing experimental data. With technology progress of microbial fuel cell technology, mathematical models are also corrected by other researchers and adapted to the new conditions of these cells. The model presented in this study is not considered a complete model and with new developments in MFC technology, it will undergo changes and corrections in the parameters and assumptions used in the model presentation.

Acknowledgments

None.

Conflict of interest

None.

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None.

Ethics statement

None.

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