Models for microbial fuel cells: a critical review

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Abstract: Microbial fuel cells (MFCs) have been widely viewed as one of the most promising alternative renewable energy source. MFCs have attracted a large amount of interest in the past decade and much scientific effort has been dedicated to making this technology more efficient. MFCs are complex bioelectrochemical system that generates electrical energy by the catalytic reaction of organic substrates. The study of MFCs requires an interdisciplinary approach. Experimental method is time-consuming and uneconomical. Modelling of MFCs is an effective way not only for thorough understanding the effects of operation conditions on the power generation performance, but also for the successful implementation of MFCs. The state of the art of MFCs modelling is present in this article. Models and modelling methods are elaborated based on the classification of mechanism-based models and application-based models. Furthermore, mechanism, advantages, drawbacks, and application fields of different models are illustrated.

1 Introduction

Microbial fuel cells (MFCs) as a novel renewable energy source have received extensive attention in recent years. The substrate in anaerobic anode of MFCs produces electrons by bacteria catalytic oxidation, and the electrons are transferred to anode electrode through an external circuit to the aerobic cathode where the reduction reaction is performed [1]. The MFCs can be divided into mediator-based MFCs that used mediator to transfer electrons to anode and conduction-based MFCs in which the electrons transfer is carried out by biofilm. Although MFCs have the huge advantage in wastewater treatment with simultaneous electricity generation [2], the lower power output and the costly materials of MFCs both result in reducing its competitiveness compared with other energy [3]. Therefore, how to improve the power generation performance and find the inexpensive catalyst materials are the pronounced challenges in MFCs application.

The non-linear and hysteresis characteristic due to plenty of bio-electrochemical coupling reactions makes MFCs difficult to control and optimise the power generation through the direct experimental means. Conversely, mathematical model is considered as a fast and effective approach to comprehend the major influence factors of the system and distinguishes the main bottlenecks to improve the power generation performance of MFCs. In general, the MFCs models primarily represent the biochemical reaction process, mass transport process, and electricity generation process of the system [4]. With the development of MFC modelling, the conflict between the exhaustive expressing degree and practical application has appeared in different models. Many efforts have been directed to the fields related with MFCs modelling.

In this paper, MFCs models in literatures have been classified into two groups: mechanism-based models and application-based models. Mechanism-based models focus on the main reactions processes occurred in MFCs. These models with the purpose of presenting the MFC’s reaction mechanism in detail are applied to optimise the MFC’s configuration and power generation performance fundamentally. Application-based models divide into the electrical models and the learning models. The electrical models are able to be employed to configure the circuit components parameters to perform the practical application. The learning models are mainly used to optimise the output power via the enormous experimental data or the state equations instead of the whole reacting processes. Fig. 1 shows the classification of the MFCs models that will be discussed in the following sections.

2 Mechanism-based models for MFCs

Mechanism-based models aim to present the reactions and principles of MFCs in detail with the kinetics equations and bioelectrochemical features. These models are of four groups: bulk liquid models, electrochemical models, biofilm models, and special factor models.

2.1 Bulk liquid model

Bulk liquid model contains the basic reactions of MFCs. The rate of substrate consumption and mass balances are illustrated in this section. Substrate is a significant electricity production element as the electrons donor. Mass balances represent the variation of primary components in MFCs, including the solute chemical species and biomass, are commonly expressed by the ordinary differential equations.

Picioreanu et al. [5] introduced a double-limited Monod equation to characterise that the substrate consumption was limited both by the concentrate of electron donor (ED) and electron acceptor (EA). This equation is applied to a specific occasion that the add-mediators as the EA. Pinto et al. [6] used this expression in the case of the substrate competition of two microbial groups: the electrogenesis and methanogenic. This approach was widely used regarded to the different microbial populations competed for the same substrate [7–9]. Zeng et al. [10] developed a model for a two-chamber MFC and considered the process of consuming substrate was affected by the anode potential. The Monod equation was combined with the Butler–Volmer/Tafel equation afterwards to establish an equation of substrate consumption rate and the cathodic reaction rate referenced this form as well. The equations were generally applied to two-chamber MFCs models [11–13]. In the model of MFCs based on conductive mechanism, Kato Marcus et al. [14] mentioned a double-limited Monod equation, considering the...
biofilm as the EA rather than the extra mediator. The expression of specific consumption rate was deduced by the concentration of EA and the anode potential calculated with the Nernst equation, recombined the Monod equation into the Monod–Nernst equation and was subsequently used in the later models of conduction-based MFCs. It indicates that the Monod-type and Nernst–Monod equation both remake the double-limited Monod equation. The ED concentration is ought to combine with electrode potential when the mechanism of electron transfer has changed.

For the mass balances of MFCs, the full-cell simulation \([10–12, 15]\) at a large-scale aspect comprehensively considers the changes in important chemical components, but the diffusion between the biofilm and substrate are often ignored. Additionally, some models aimed at the half-cell simulation mostly discussed the mass balances at microscale. Picioream et al. \([5]\) established the mass balances for solute and biomass components for MFCs, and the equations included the electrochemical reaction rate in the bulk liquid and biofilm. After that, Picioream et al. \([16]\) used this equation and established mass balances for each relevant redox production composition. While in conduction-based models, Kato Marcus et al. \([14]\) proposed a model for biofilm anode of an MFC, correlated mass balances with reactions occurred in the biofilm and the substrate diffusion phenomenon from the bulk liquid to biofilm. The following conduction-based models all used the analogous approach in modelling. Karimi Alavijeh et al. \([17]\) considered more species biomass and expanded the mass balances expression in \([14]\) to different types of substrate conditions. Ismail and Habeeb \([18]\) added the diffusion process to full-cell simulation, developed the diffused substrate mass balances in biofilm, and other balance equations also accounted for the dynamic reaction process in bulk liquid. Kazemi et al. \([19]\) presented a model for a reverse-MFC aimed at the cathodic compartment, considering the diffusion to biofilm cathode, and the mass balances were employed for the variation of the chemical component in catholyte. Zeng et al. \([10]\) focused on the competition behaviour and proposed relatively simple mass balances equations, calculated the change of two microorganisms and substrate concentration. The form of equation is generally different according to the operation modes of the MFC and the emphasis of simulation.

2.2 Electrochemical model

In electrochemical models, the relevant electrical performance like the output voltage and current density of MFCs is expressed through the basic theory of electrochemical.

Current density links to the rate of electrochemical reaction and were introduced in some models to describe the polarisation curve of MFCs. The Butler–Volmer equation was used in \([20]\) to describe the relationship between the current density and activation overpotential in anode ethanol oxidation and cathode oxygen reduction reactions. Pinto et al. \([8]\) applied the mass balances and the current expression to discuss the effects of operation current on the existence of two species microorganisms, proposed a theoretical operation current with optimal production. Hamelers et al. \([21]\) applied a Butler–Volmer–Monod (B–V–M) equation to calculate the current density. The B–V–M equation has a better ability in coherence with experimental data as well as predicts the zero current and describes the polarisation curve. On this basis, Stein et al. \([22]\) established a model for a biosensor based on an MFC and considered the effect of the presence of toxic component, modified the B–V–M model with enzyme inhibition kinetics for four types of toxicity. For models involving the biofilm, the gradient in the local potential of biofilm drives the current density. The model in \([14]\) referenced Ohm’s law and Fick’s law combining the other parameters to express the current density. Besides the conventional expression of current density based on the Butler–Volmer equation, Ou et al. \([23]\) developed a cathode chamber model and characterised the current density of cathodic biofilm and cathode catalyst layer.

The output voltage of MFCs is determined by the cathode and anode potential. In view of the fact that the effect of polarisation leads to the overpotential which results in the voltage losses (activation overpotential, concentration overpotential, ohm overpotential) in a cell, there is not apparent discrimination among the different MFCs models and differs from the degree of overpotential calculation merely. A basic output voltage calculation equation is expressed as

\[
V_{\text{cell}} = V_c - V_a - \eta_{\text{act}} - \eta_{\text{con}} - \eta_{\text{ohm}} \tag{1}
\]

where \(V_c\) is the anode potential; \(V_e\) the cathode potential; \(V\) the theoretical thermodynamic potential; \(\eta_{\text{act}}\) the activation overpotential; \(\eta_{\text{con}}\) the concentration overpotential; \(\eta_{\text{ohm}}\) the ohm overpotential (Table 1).

The cathode potential was assumed as a constant value commonly in the half-cell simulation based on anode. Pinto et al. \([6]\) considered that the activation loss dominated little under the optimal electrical load and was calculated by an equation resembled the Butler–Volmer equation. The model referred to a more accurate concentration loss that was expressed by additional boundary conditions at high current density and a Monod term at low reduction concentration. The same expressions were used in \([7, 24]\). Besides that the anode activation and anode voltage were characterised by the Butler–Volmer and Nernst equation, respectively, the model of a continuous MFC anode of \([25]\) considered that the concentration overpotential appeared at the interface of liquid electrode and consequently neglected the concentration overpotential to reduce the complexity of the model. The model \([26]\) aimed at cathode chamber expressed the anode concentration losses based on the Nernst form with several variables, and calculated the activation overpotential of anode subsequently. Oliveira et al. \([11]\) modelled the cathode reaction with Tafel equation, linked the cathode overpotential to the current density, and considered the membrane resistance as internal resistance. The model \([27]\) used the Nernst equation to calculate the cathode voltage, proposed a half-max-rate anode voltage and the local voltage in biofilm which were both applied to calculate anode voltage. In addition, the electron balance equation was used in the full-cell simulation normally to calculate the activation and concentration losses \([10]\). The activation overpotential could be expressed by Tafel equation with polarisation curve known. In \([28]\), the natural logarithm form of Tafel equation was used to represent the activation losses, which
Table 1 Equations applied to MFCs models

| Application object       | Form                                                                                                           | Calculation occasion | Rate of substrate consumption | Bacteria growth | Application object | Form                                                                                                           | Calculation occasion | Rate of substrate consumption | Bacteria growth |
|--------------------------|----------------------------------------------------------------------------------------------------------------|----------------------|-------------------------------|----------------|-------------------|----------------------------------------------------------------------------------------------------------------|----------------------|-------------------------------|----------------|
| Monod                    | $q = \frac{q_{max} \cdot \phi_0 \cdot S_0}{K_s + S_0 + S_1}$                                                   |                      |                               |                | Monod             | $q = q_{max} \cdot \phi_0 \cdot S_0 / K_s + S_0$                                                            |                      |                               |                |
| double-limited Monod     | $q = \frac{q_{max} \cdot \phi_0 \cdot S_0}{K_s + S_0 + S_1}$                                                   |                      |                               |                | double-limited    | $q = q_{max} \cdot \phi_0 \cdot S_0 / K_s + S_0$                                                            |                      |                               |                |
| Monod-type               | $q = q_{max} \cdot \phi_0 \cdot S_0 / (K_s + S_0 + S_1)$                                                       |                      |                               |                | Monod-type        | $q = q_{max} \cdot \phi_0 \cdot S_0 / (K_s + S_0 + S_1)$                                                    |                      |                               |                |

2.3 Biofilm model

The biofilm is derived by the bacteria accumulation and carry out the conduction of electrons. Models require establishing the biofilm model to estimate the formation and influence of biofilm for MFCs. Therefore, the biofilm model is normally considered with the biomass distribution and variation of thickness, gradually extended from anode biofilm to cathode biofilm in models.

Kato Marcus et al. [14] presented a one-dimensional biofilm model at the first time of MFCs with the definition of two species microorganisms: the active biomass and inactive biomass. The former oxidised the substrate and the latter formed the conductive materials to transfer the electrons directly. The dynamic thickness of biofilm was calculated by the advective velocity of biofilm matrix and the detachment of microorganism. The model contributed to the conduction-based mechanism modelling approach, revealed that the biomass growth and death leded to the change of biofilm. Such main idea was referenced and expanded in the later work [17, 31–33]. Jayasinghe et al. [33] modified the biofilm model with the extracellular, intracellular components and a genome-scale metabolic model integrated. The model expanded the microbial population in biofilm of [14] into three kinds of microorganisms: the active, the inert, and the respiring microorganisms. The characteristics and interconversion of respective biomass were mentioned in the model. What is more, the death and lysis of bacteria were used to characterise the variation of biofilm, which made the modelling process more exhaustive. A two-dimensional biofilm model based on [14] was proposed in [31] within three regions: the bulk, the biofilm, and anode electrode. Two interfaces were defined and the position evolution was calculated by a price-wise function. Mardanpour et al. [30] introduced a model developed from Kato Marcus et al. [14] for microfluidic MFCs. The model explained how the bacteria accumulated to form a biofilm. Delivering the principle of the random motion and chemotaxis process of sensing molecules, the model employed the Keller-Segel model and mass balances to describe the attached bacteria accumulation as biofilm and the bacteria distribution, respectively.

For electrons-shuttles mechanism, the concentration gradient also characterises the biofilm domain in MFCs [5, 18, 34]. Picoreau et al. [5] established a model for MFC with extra diffusible mediator. The attached biomass related to the biofilm and the suspended biomass were both discussed in terms of their kinetic reactions and mass balances through a biofilm model whose main framework referenced [35]. The modified version defined the spatial concentration gradients for two species to calculate the variation of concentration in biofilm. Ismail and Habeeb [18] presented a two-dimensional biofilm model and assumed that the concentration of solutes and biomass varied over time and position in biofilm. The gradient information was applied to the mass balance equation to represent the concentration in biofilm region.

While some models paid close attention to the cathode biofilm in recent works [19, 23]. Experimental work validated the cathode biofilm is a transition area that accepted electrons and affected the reaction of cathode [36]. A one-dimensional transient model in [23] did not involve the variation of cathode biofilm thickness or the concentration distribution, but rather the reaction of oxygen consumption that partly occurred in biofilm. Moreover, based on [14], the model for a reverse MFC of [19] took into account that the biofilm formed in cathode and its thickness change.
2.4 Special factor model

Some special factors that affect the performance of MFCs are discussed in this section. These models were less mentioned in literature and were not considered as the main influential element in the system.  

Vast majority of models assumed the constant pH of liquid in the reactor chamber ignoring the effect of pH. While the model of [34] developed from [5] characterised the MFCs at the macroscale and microscale. The water dissociation reaction and acid–base equilibrium for other types of acid were established to calculate the pH distribution. Similarly, regarding to the effect of pH value to the biochemical reaction, PerazaBaeza et al. [37] presented a mathematical model to predict the influence of oscillatory of the pH. The model referenced the performance of LRC electrical circuit to the model to predict the influence of oscillatory of the pH. The model referenced the performance of LRC electrical circuit to the pH value in the anodic chamber as an underdamping second-order oscillatory system and related to the consumption rate of substrate. Furthermore, Oliveira et al. [11] established a model of a two-chamber MFC based on [10] without the biofilm due to the short computing time, but added the heat conduction process which consisted of fundamental heat balance equations to the model.

3 Application-based models for MFCs

Although the mechanism-based models are able to optimise MFCs from basic principles, the computing time and complexity always limit the practical application. Hence, for engineering designers, some models based on the electrical characters of MFCs expressed by equivalent circuit (EC) model [38–41] and are used to achieve the electrical application. There are also some models developed from several typical reactions and experimental data of MFCs to acquire the best parameters and maximum power [42–46].

3.1 Electrical model

In electrical models, the EC model and power output curve are presented by classical electrical parameters of MFCs. The electrical model provides a novel analytical method particularly aims at the application in electronic and electrical way. For example, the EC model is applied to design the post-electrical circuit to achieve the energy harvesting or the maximum power tracking of MFCs (e.g. [47, 48]).

The internal resistance influences the output performance of MFCs to a large extent has been reported and the most direct impact is reflected in the ohm losses [49, 50]. To figure out the effect of internal resistance, electrochemical impedance spectroscopy (EIS) is an effective tool to measure the value of internal resistance of an MFC [51]. However, the material of electrode is one of the factors that determine the internal resistance. Hernández-Flores et al. [52] introduced a numerical relationship between the volumetric power of MFCs and specific surface area of anode, related the anode region to the output power, as well as internal resistance in a succinct expression based on Tafel equation. It is noticeable that the model was firstly proposed as alternative way to analyse the electrical performance of MFCs with anode surface area. Sindhija et al. [38] proposed two EC models for different materials of electrode. A lumped EC model for activated charcoal electrode and fitting EC model for graphite electrode were established both through the EIS measurement of cathode and anode and the calculation of anode overpotential. Yin et al. [39] modelled the MFC operated under static magnetic field (MF) that contributed to induce several biological reactions in a microbial system [53]. The EIS was used to investigate the electrochemical reactions of the MFC and presented the EC model of anode, cathode, and entire system, respectively. The work of [41] improved the defect of EIS so that the cell disconnect to a potentiostat for the duration of measurement, which was a novel method to determine the value of charge transfer resistance and double-layer capacitor in MFCs. An EC model was introduced with the effect in terms of capacitor within electrode considered. Coronado et al. [40] conducted the experiment that the MFC connected to the external resistance (R-PMW). The existence of fast and slow dynamic component of the MFC was obtained via the analysis of the result, thus utilising the capacitor and resistance to establish the EC model. Although the electrochemical reactions were negligible in this model, it provided the dynamic process of relevant electrical performance.

3.2 Learning model

As the increasingly development of artificial intelligence and modern control theory, some models are established without much in-depth understanding of mechanism of MFCs; instead, a few significant reactions and/or massive experimental data plays an essential role in modelling. These models facilitate designers to maximum the MFCs output and configure the MFCs with less bio-electrochemical knowledge, which contributes to improve the development of MFCs application.

In fact, MFCs have the slow dynamics character due to much electrochemical and biochemical reactions occurred, which requires a long period to acquire the experimental result. The experiment design approach has been therefore proposed to shorten the time of experiments. Fang et al. [42] developed a modelling approach to optimise the power generation of MFCs with multivariable. The experiment was carried out by the uniform design method which determined four input variables with five levels and regarded the power density and coulombic efficiency as output variables. The model adopted relevance vector machine to acquire a mathematical model between the input and output variables through the result from 16 experiments. To speed up the rate of optimisation of model’s parameters, accelerating genetic algorithm was selected to seek the maximum output variables and optimal reacting conditions. Another experimental design approach: the response surface design methodology (RSM), which was also employed to design the experiment of MFCs in [46]. The work introduced a Box–Behken design approach to optimise the temperature, external resistance, the concentration of feed fuel, and the pH in anode chamber, maximised output power density simultaneously. The results from experiments could derive a quadratic mathematical model for an MFC by RSM; meanwhile, the output power density achieved the maximum. To improve the shortcoming of the model that has long computational time, He and Ma [43] proposed a model based on data-driven Gaussian process regression for MFCs. The experiment design approach was firstly used to obtain a series of experiment data as the initial sample. The model adopted the concentration of substrate, anodic feed rate, and current density as the input variables while the voltage as the output variable, and the accuracy and reliability of the model was validated by operating both in online and offline mode with different requirement of training samples and executive procedures.

Besides, the state estimation method was introduced with state-space model as well. A control-oriented model based on state space equation of an MFC was presented in [44]. The model set up the substrate concentration, biomass concentration, hydrogen ion concentration, and bicarbonate ion concentration as the state variables, combined the mass balances and voltage equation to establish the state-space equations. It was worth noting that the approach offered a way to deduce the model that is likely to employ the state estimation method (e.g. Kalman filter) to observe the parameters. In view of the more complexity of the MFC model, the more difficult it is for the process controlling and the parameter estimation, the model in [45] proposed a model combined the previous model mentioned competitive substrate consumption in different microbial population [6] and the EC model [38] into a bioelectrochemical–electrical model which could be applied to both the fast calculation process and the slow dynamic simulation.
4 Conclusion
The mathematical models and modelling methods of the MFCs are summarised in the paper. The representative models to present are discussed and classified based on the mechanism and the application. The mechanism-based models based on the reactions of these systems fall into several submodels on the basis of the normal modelling domains. From the result of summary, the different electrons transfer mechanisms and the requirement of modelling emphasis lead to the difference among the mechanism-based models. For the application-based models, the EC models basically have a fixed form and the accuracy of learning models remains requiring continuous improvement. With the deeper insights of MFCs and the development of computing speed and intelligent method of modelling, the more precise and practical models of MFCs will contribute to the development of MFCs in future.

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