Integrated Experimental Investigation and Mathematical Modeling of a Membrane Bioelectrochemical Reactor with an External Membrane Module

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Abstract:

Membrane bioelectrochemical reactors (MBER) integrate membrane filtration module into microbial fuel cells (MFCs) to achieve simultaneous wastewater treatment, bioenergy production, and high-quality effluent. Previous MBERs usually have membrane modules as a part of the MFC reactors that creates challenges for membrane cleaning. In this study, an MBER with an external membrane module was investigated through both experiments and mathematical modeling. This MBER produced a current density of $7.1 \pm 0.5 \text{ A m}^{-3}$ with an anolyte recirculation of 90 mL min^{-1} ; reducing the anolyte recirculation rate had a negligible effect on MBER's electrical performance but resulted in a positive energy balance of $0.003 \pm 0.002 \text{ kWh m}^{-3}$. Periodic backwashing (1 min-backwashing/15 min-operation) was demonstrated as an effective method for fouling control. A mathematical model was developed and validated using the experimental data. The model could predict the influence of key parameters such as influent organic concentration and anolyte flow rate on the current generation, and identify the maximum organic loading rate for current generation. Those results encourage further investigation and development of this MBER towards system scaling up.

Keywords: Membrane bioelectrochemical reactor; microbial fuel cell; membrane separation; mathematical modeling; wastewater treatment

1. Introduction

Membrane bioelectrochemical reactors (MBERs) are a newly emerged technology for sustainable wastewater treatment and resource recovery [1]. An MBER is a combination of bioelectrochemical system (BES), such as microbial fuel cells (MFCs), with membrane separation. Thus, the technology inherits the advantages of both BES and membrane treatment (e.g., membrane bioreactors), and can directly generate electricity and produce a high-quality effluent. MBERs have been demonstrated in various studies. For example, biofilm was formed on the stainless steel mesh as a filtration material to achieve high organics and nutrient removal [2, 3]. Commercial ultrafiltration hollow fiber membrane was integrated into a tubular MFC, either in the anode or the cathode [4-6]. The MBER operation can generate energy and require low energy input, thereby creating an energy neutral treatment system. It was estimated that net electricity could be recovered from treating a low-strength synthetic wastewater after eliminating aeration process in the cathode chamber of an MBER [7]. Increasing organic loading rate by decreasing hydraulic retention time (HRT) can also diminish the gap between energy consumption and production because of more energy production [4].

Membrane fouling poses a great challenge to MBER operation, especially when membranes are integrated with BES in an internal configuration. It was reported that foulants could accumulate on the membrane surface rapidly when the membrane was installed in the anodic compartment of an MBER, and severe fouling occurred in two weeks with a rapid TMP increase from 0 to 50 kPa [4]. Coating polydopamine on membrane surface to increase hydrophobicity [8] or lowering

particle zeta potential to increase electro-repulsion force [9] have been demonstrated to control membrane fouling effectively, but a long-term operation should be examined on membrane fouling. Fluidized bed granular activated carbon (GAC) was applied in the anodic compartment of an MBER to reduce transmembrane pressure [10], but the high pumping load due to the high recirculation and clogging issue after long-term abrasion of GAC could cause maintenance issues and high energy demand.

Periodic cleaning of membranes using physical and chemical methods is necessary to maintain a functioning membrane module, but with an internal membrane installation, *in situ* cleaning becomes very difficult because of potential effects on biological and electrochemical reactions. Thus, external linkage between BES and membrane module may provide a flexible system in which the membrane module can be cleaned/replaced without affecting the BES. Such a linkage was reported in two recent studies. In one study, an air-cathode MFC was connected to an anaerobic fluidized membrane bioreactor (AFMBR), forming a two-staged MFC-AFMBR [11]. This system achieved the removal of more than 90% total COD and almost 100% total suspended solids; biogas production in the AFMBR may compensate for energy consumption by the system. The other study used an MFC-MBR system to treat the waste from a fermentation process that helped recycle the water and nutrients back to the fermentation process [12]. The challenge of those systems is that the external membrane module involves biological reactions that have high requirement for operation and maintenance; in addition, the collection of methane (and dissolved methane in the effluent) in the AFMBR [11] and aeration in the MBR [12] may limit their application. In contrast, an abiotic membrane module could be easier to manage.

Given much interest in MBERs and in light of the above mentioned issues of managing membrane module, an MBER system consisting of an MFC and an abiotic membrane module was developed and investigated through an integrated approach of experimental investigation and mathematical modeling. The membrane module was linked to the MFC through a unique approach to take advantage of the catholyte dripping from the MFC (Figure 1). This type of tubular MFCs with dripping catholyte has been extensively demonstrated in previous studies including a long-term operated system with actual wastewater in a wastewater treatment plant [13-17]. A dynamic mathematical model for MBER has been recently developed [18], and is applied with proper modification for further understanding of the present MBER. The objectives of this study were: (1) to demonstrate feasibility of this MBER in an energy-neutral operation; (2) to maintain low membrane fouling with an enhanced flexibility; and (3) to apply a mathematical model to predict MBER performance affected by organic loading rate (OLR).

2. Materials and methods

2.1 MBER Setup

The proposed MBER system consisted of two units, an MFC and a (external) membrane module (Figure 1). The MFC was constructed as a tubular reactor (38 cm long and 5 cm in diameter) made of anion exchange membrane (AEM-Ultrex AMI 7001, Membrane International. Inc, Glen Rock, New Jersey, USA). A carbon brush was installed in the anodic chamber as an anodic electrode, resulting in a net liquid volume of 750 mL. Before use, the carbon brush was soaked in acetone overnight, and then heated for 30 min at 450 °C [19]. The cathode electrode was one piece of carbon cloth (Zoltek Corporation, St. Louis, MO, USA) coated with Pt/C powder (Etek,

Somerest, NJ, USA) with a loading rate $0.2 \text{ mg Pt cm}^{-2}$. The cathode electrode wrapped the AEM membrane tube. The anode and cathode electrodes were connected to a 10Ω resistor. The external membrane module was made of thirteen 18-cm PVDF hollow fiber ultrafiltration membranes (15,000 Dalton, Litree Purifying Technology Co. China) that were installed in a 150 mL rectangular container, and was set up under the MFC to collect the catholyte.

2.2 Operating condition

The MBER system was operated at room temperature. Its anode was fed with a synthetic solution containing (per L of tap water): glucose 0.25 g; NH_4Cl 0.15 g; NaCl 0.5 g; MgSO_4 0.015 g; CaCl_2 0.02 g; KH_2PO_4 0.53 g; K_2HPO_4 1.07 g; and 1mL trace elements [20]. The anolyte was recirculated at 90, 50 and 20 mL min^{-1} on day 9, 55 and 74, respectively. The MBER was operated in a full loop mode, in which the synthetic solution was first fed into the anodic compartment and then the anodic effluent flowed to the cathode acting as a catholyte. There was no recirculation applied to the catholyte. The catholyte was collected by the membrane module, and the final permeate was extracted from the hollow-fiber membranes. The hollow fiber membrane module was operated in 4-min working/1-min relaxing mode; 1-min backwashing was applied in every 30-min from day 22 and in every 15-min from day 40, respectively. The flow rate of 1.25 mL min^{-1} was controlled by peristaltic pumps to achieve the desired anodic hydraulic retention time (HRT) at 10 h.

2.3 Measurements and analysis

The MFC voltage was recorded every 5 min by a digital multimeter (2700, Keithley Instruments, Cleveland, OH). The current and power density was normalized to the anode liquid volume. The pH was measured using a benchtop pH meter (Oakton Instruments, Vernon Hills, IL, USA). The conductivity was measured by a benchtop conductivity meter (Mettler-Toledo, Columbus, OH, USA). The concentration of chemical oxygen demand (COD) was measured according to the manufacturer's procedure (Hach DR/890, Hach Company, Loveland, CO, USA).

Transmembrane pressure (TMP) was recorded manually and the average value was reported in this study. Turbidity was measured using a turbidimeter (DRT 100B, HF Scientific, Inc, Fort Meyers, FL, USA). Energy recovery was evaluated by normalized energy recovery (NER) in kWh m⁻³ [21, 22], a parameter for evaluating amount of energy can be generated for treating each m³ of wastewater. The estimated energy consumption by the pumping system (for feeding, recirculating and membrane extracting) was calculated by the following equation [23]:

$$P = \frac{Q\gamma E}{1000}$$

where P is power requirement (kW), Q is flowrate (m³ s⁻¹), γ is 9800 (N m⁻³) and E is head loss (m H₂O). The energy consumption was expressed based on the volume of treated wastewater (kWh m⁻³). The energy balance is the gap between energy recovery and consumption, which was also expressed based on the volume of treated wastewater (kWh m⁻³).

2.4 Model formulation

A mathematical model is formulated by applying multiplicative Monod kinetics for microbial growth [24, 25]. Intracellular redox mediator is assumed to be a key factor to aid electron

transfer from substrate to an electrode, and a well-mixing condition is assumed in the anodic compartment. The overall cell voltage in MBER system is calculated by the difference between open circuit potential (V_{oc}) and potential loss, which includes overpotential from anode and cathode electrode, concentration overpotential and internal resistance. Because of the sufficient buffer solution and oxygen supply, the overpotential of the anode and the cathode electrodes has been neglected in this model. The estimated internal resistance includes mass transfer resistance, ohmic resistance and activation resistance. More information about mass balance for substrate consumption, microorganism concentration, electron mediators and electrical production can be found in Supplemental Data.

3. Results and discussion

3.1 MBER performance of electricity generation and organic removal

The MBER system was fed with a synthetic solution for 101 days, and current generation is shown in Figure 2A. The anolyte recirculation rate was gradually decreased from 90 (day 9), to 50 (day 55), and then to 20 mL min⁻¹ (day 74). The MBER generated 7.1±0.5 A m⁻³ with the recirculation of 90 mL min⁻¹. Comparing to a previous MBER that was operated under a similar HRT [6], the present MBER produced about 65.6% less electricity, possibly because that glucose used here is more complex than acetate used in the prior study and a lower COD concentration (250 vs. 350 mg L⁻¹) limits substrate transfer. Decreasing the anolyte recirculation rate to 50 mL min⁻¹ resulted in current generation of 7.1±1.0 A m⁻³, and further decrease to 20 mL min⁻¹ produced current of 7.4±1.4 A m⁻³. Unlike the prior study that shows improved electricity generation with the anolyte recirculation [13, 26], the present system was not obviously affected

by the tested anolyte recirculation rates, most likely because of those recirculation rates are generally low. Significant improvement of the electricity generation was obtained with a high recirculation rate of 4-12 times of the anolyte volume per minute [13], which is equivalent to 3,000 to 9,000 mL min⁻¹ in the present system. Electrolyte recirculation has been identified as a major energy consumer in a bioelectrochemical system [6, 27], and thus low recirculation rates are preferred from an aspect of low-energy operation. In fact, in our ongoing large MFC system (200-L), no anolyte recirculation is applied (data not shown).

The present MBER could effectively remove organic compounds (COD), through a staged removal in the anode, the cathode and the hollow-fiber membranes module. At the anodic HRT of 10 h and a recirculation rate of 90 mL min⁻¹, the anodic compartment removed 43.3±7.7% of total COD, and the cathode removed 48.2±7.2%; a final COD concentration in the membrane permeate was 4.0±3.8 mg L⁻¹, resulting in an overall COD removal efficiency of 98.4±1.5 % (Figure 2B). The dripping cathode functioned as a secondary aerobic degradation process that polished the anode effluent by removing much of residual COD; such a removal would benefit the down-stream membrane module with less foulant. Thus, the membrane module acted mainly as a solid-liquid separation process without much biological reaction. Decreasing the anolyte recirculation rate to 50 or 20 mL min⁻¹ actually enhanced the overall COD removal efficiency to 100.0±0.0%, benefited from the improved removal by the anode (TCOD removal of 74.9±0.6 or 72.4±18.4%). Such improvement might be related to the reduced oxygen leak into the anode through recirculation tubing at a lower rate, but the exact reasons are not clear at this moment and warrant further investigation. The turbidity of the membrane permeate was under 1 NTU.

3.2 Energy balance

The energy performance of the present MBER was evaluated through establishing an energy balance. The NER of the MBER was $0.004 \pm 0.001 \text{ kWh m}^{-3}$ under the recirculation of 90 mL min^{-1} and the system would require 0.020 kWh energy to treat 1 m^3 of the synthetic solution; as a result, the overall energy balance was negative at $-0.016 \pm 0.001 \text{ kWh m}^{-3}$ (Figure 3). Decreasing the anolyte recirculation rate to 50 mL min^{-1} did not obviously change the NER but significantly decreased the energy consumption to 0.007 kWh m^{-3} , thereby achieving a final energy balance of $-0.003 \pm 0.001 \text{ kWh m}^{-3}$. When the recirculation rate was further reduced to 20 mL min^{-1} , the NER was estimated to be $0.004 \pm 0.002 \text{ kWh m}^{-3}$, and the energy consumption was 0.001 kWh m^{-3} , which was only 5.0 and 14.3% of the two previous operational modes. Correspondingly, a slightly positive energy balance of $0.003 \pm 0.002 \text{ kWh m}^{-3}$ was achieved. This result indicates that a low anolyte recirculation rate such as 20 or 50 mL min^{-1} could have been sufficient for substrate distribution in the anode chamber of the MBER, likely benefiting from its brush-containing space that has much room for liquid flow. Despite the positive energy balance at 20 mL min^{-1} , energy recovery was very low and must be further improved through adding more anode electrode materials (which could result in more impact of the anolyte recirculation rate on energy recovery), improving the catholyte catalysts, and optimizing the operating conditions (e.g., temperature). It is worth noting that the energy consumption due to backwashing for membrane fouling control was small and negligible in the energy balance calculation (estimated at $1.1 \times 10^{-4} \text{ kWh m}^{-3}$). The energy consumption by the MBER is significantly lower than that of activated sludge (0.6 kWh m^{-3} [28]), or aerobic MBRs ($1.2\text{--}2.0 \text{ kWh m}^{-3}$ [29]). The anaerobic MBRs (AnMBRs) could be energy positive, and it was estimated that a maximal 0.16 kWh

energy can be produced to treat 1 m³ wastewater by AnMBR [30]; however, a strict temperature control and proper strategies for extracting the dissolved methane will be important to maximize its energy recovery with biogas.

3.3 Membrane performance

The TMP of the membrane module is shown in Figure 4A, and the fouling control was conducted by membrane relaxation, backwashing, and offline cleaning. The offline cleaning was conducted by first applying soft brush to remove the attached foulants, and then rinsing the membrane surface with tap water until the cake layer mass was detached completely (based on visual judgment). The membrane module was connected to the MFC on day 12 of the operation under a 4-min work/1-min relax mode, and the serious fouling of the membrane was observed after only three days, when the TMP increased to 30 kPa on day 14, indicating a cake layer could accumulate rapidly by only applying a periodic relaxation to membrane. Offline cleaning was applied to reduce the TMP, which increased repeatedly in the subsequent weeks, which required offline cleaning on day 18 and 20. From day 22, a regular backwashing flow at 2.5 mL min⁻¹ (as double as membrane filtration flowrate) was applied for 1 min in every 30-min operation. This strategy has clearly delayed the TMP increase, which took about 10 days to reach 40 kPa on day 31, and after offline cleaning increased to 45 kPa on day 40. The frequency of backwashing was adjusted to 1 min in every 15-min permeation cycle and water flux was recorded from day 40 (Figure 4B). Such a change allowed the MBER to maintain its TMP under 40 kPa in the following 46 days without any offline cleaning. The water flux began to drop from 6 L m⁻² h⁻¹ (LMH) on day 40, due to the cake mass accumulation, to 2.8 LMH as the minimum on day 64.

The TMP decrease and water flux increase on day 80 was related to the detachment of the cake mass due to the operational reasons.

3.4 Model validation and prediction

The data used for parameter estimation were obtained under the experimental condition of varying either the anolyte COD concentration from 100 to 250 mg L⁻¹, or the anolyte flow rate from 1.3 to 2.1 mL min⁻¹ (250 mg L⁻¹ COD). The predicted current generation and organic concentration in the anode effluent were plotted along the experimental data in Figure 5. When varying the COD concentration, the model output of current generation generally followed the trend of experimental data that current generation was enhanced along with the increased substrate concentration (Figure 5A). The relative root mean square error (RMSE) showed that the simulated current generation was within 16.4% of their maximum value, affected by the fluctuation of experimental data. The model output of substrate concentration shared the same trend with the experimental measurement (Figure 5B), with a relative RMSE of 36.9%, indicating large deviation existed between the simulation and the experimental data. Such a discrepancy can be from mismatch of current generation that at the early stage, the modeled current endured a sharp decrease and a temporal lagging period of bacteria growth could occur due to insufficient substrate supply; consequently, a lower COD concentration in the anode effluent was observed. The model output of the substrate concentration in the anode effluent began to increase as a response to the enhanced COD concentration in the influent, but overestimation was observed from the model simulation data, possibly due to the overestimated current generation from model simulation results with larger COD removal.

The model output of current generation affected by the anolyte flow rate had a high RSME of 27.8%, because of the poor sensitivity of the MBER experiment (Figure 5C). When the anolyte flow rate was changed from 1.3 to 2.1 mL min⁻¹, the model current increased from 5.0 to 8.2 mA but the experimental data did not obviously respond. A similar phenomenon was reported in a model for microbial desalination cells (MDCs), suggesting that mathematical model has higher sensitivity to the change of substrate concentration when current generation is in a low range [25]. Likewise, the experimental data of the COD concentration did not well respond to the change of the anolyte flow rate like that of the modeled value (Figure 5D). The simulated substrate concentration shared a similar trend to the experimental data, with a relative RSME of 32.1%. Enhancing the organic loading rate (OLR) from 0.6 to 1.0 kgCOD m⁻³ day⁻¹ by increasing the influent flowrate from 1.3 to 2.1 mL min⁻¹ did not significantly change the COD concentration in the anode effluent, suggesting that more organics might have been consumed by the microorganisms other than electrogens at a higher COD loading rate. The modeled substrate concentration was lower than the experimental data, because of the overly estimated current generation.

The variation of TMP was also simulated and the model output could give a good prediction on TMP change (Figure 6). The model simulation has a relative RSME of 16.0%, affected by inaccurate estimation of suspended solids concentration that could yield a deviated increase rate on membrane resistance. It is worth noting that membrane fouling is a consequence of a dynamic process, which includes both attachment and detachment of the foulants. The attached

foulants includes the suspended solids from the influent (from the MFC) and newly produced biomass within the membrane module; while the detachment occurs when physical fouling control such as air-bubble scrubbing or surface cross-flowing is provided. Water flux (J) and suspended solids (X) are two major components for simulating TMP (indicating membrane fouling) in the developed model, and result in a linear increase of TMP with time as shown in Figure 6.

The model was used to predict the performance (current generation and COD removal) of the MBER affected by two factors, the influent COD concentration and the feeding rate of the anolyte (anodic HRT), both of which can affect the organic loading rate (OLR). It was found that current generation began to decline after the anodic HRT was extended (Figure 7A) or influent COD was reduced (Figure 7B). Insufficient organic supply for anodophilic bacteria was assumed to be a reason for such interactive effect. It also demonstrated that the anodic HRT has little impact on current generation after the influent COD is beyond 2150 mg L^{-1} , indicating that the conversion rate of the substrate-to-electricity may have reached its maximum and additional substrate could not be converted to electricity. The relationship between current generation and organic loading rate (OLR) confirmed that the current generation could increase along with OLR in a logarithmic way with R^2 of 0.928 (Figure 7C): current generation of 16.9 mA could be achieved after the OLR was increased to $11.4 \text{ kg COD m}^{-3} \text{ day}^{-1}$, and further enhancing OLR had a marginal effect on current generation at a cost of worse organic removal performance. It is worth noting that to achieve same OLR, coupling high influent organic concentration with a longer anodic HRT could result in more current generation than that of low influent organic concentration with a shorter anodic HRT (Figure 7D). For example, at an OLR of $2.4 \text{ kg COD m}^{-3}$

$\text{m}^3 \text{ day}^{-1}$, current generation of 10.7 mA could be achieved under the influent COD of 800 mg L^{-1} at an HRT of 8 h, but a higher current of 16.2 mA can be generated after the influent COD is changed to 1400 mg L^{-1} at HRT of 14 h (with the same OLR of $2.4 \text{ kg COD m}^{-3} \text{ day}^{-1}$). The model indicated that such improvement could be related to more stimulated anodophilic bacteria activity.

3.5 Perspectives

This is an early attempt to integrate experimental results with a mathematical model to optimize an MBER system. The present MBER is different from the prior MBERs in several aspects: (1) the external installation of the membrane module (through one-direction liquid flow connection) improves the flexibility of membrane module and decreases the effects of its operation/cleaning on the MFC unit; (2) the dripping method for supplying the cathode effluent can bring dissolved oxygen to the membrane module for additional aerobic treatment, which will be important when organic loading rate becomes higher; (3) aeration is eliminated from the cathode with its special configuration exposing the cathode surface to the air; and (4) the present MBER could theoretically achieve energy neutral. Both the findings of the influence of the anolyte recirculation rates and the mathematical model developed here can provide more insightful information on optimizing MBER system. Future studies will investigate the improvement of energy recovery from organic conversion, possibly through adding the anode electrode materials and optimizing the system operating conditions such as temperature. Despite the low fouling of the present system, membrane fouling will be expected during a long-term operation with actual wastewater, and thus appropriate anti-fouling methods will still be needed. One of the interesting

approaches for fouling control is to use in situ generated hydrogen peroxide for reducing fouling, and this will require precise control of oxygen reduction (via two electron reduction pathway for hydrogen peroxide production) on the cathode. In addition, system scaling up will remain a great challenge for developing MBERs.

4. Conclusions

This study has presented an MBER consisting of an MFC with an external membrane module, and the performance of this system was demonstrated through both the experiments and mathematical modeling. The external configuration allowed more flexibility in membrane cleaning without affecting the MFC unit. The anolyte recirculation was a major energy consumer in this system, and by decreasing the anolyte recirculation to 20 mL min^{-1} , the MBER could potentially be a net energy producer. Mathematical model was validated with experimental data and predicated that current generation by the MBER could reach a plateau of 16.9 mA after the OLR is increased to $11.4 \text{ kgCOD m}^{-3} \text{ day}^{-1}$. The model may be further improved by considering heterogeneous substrate and microbial distribution.

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Figure 1. Schematic of the membrane electrochemical reactor (MBER) consisting of a tubular microbial fuel cell (MFC) and an external membrane module under the MFC.

Figure 2. The performance of the MBER fed with synthetic wastewater: (A) current density with 10 ohm resistor, and (B) COD concentrations in different liquid stream. The arrows (a), (b) and (c) indicate the anolyte recirculation rate of 90, 50 and 20 mL min^{-1} on day 9, 55 and 74, respectively.

Figure 3. The energy production, consumption, and balance of the MBER at different anolyte recirculation rates.

Figure 4. The variation of transmembrane pressure –TMP (A) and water flux (B). R: rinsing membrane; BW1: backwashing 1 min in every 30 min; BW2: backwashing 1 min in every 15 min.

Figure 5. Model fitting under different testing conditions: (A) current generation and (B) substrate concentration in the anode effluent, when the COD concentration was changed from 100 to 250 mg L⁻¹; (C) current generation and (D) substrate concentration in the anode effluent, when the anolyte feeding rate was changed from 1.3 to 2.1 mL min⁻¹. Arrow means the changing point for operational condition.

Figure 6. Model fitting of transmembrane pressure (TMP).

Figure 7. Model prediction for current generation affected by (A) anodic HRT (h); (B) influent COD (mg L⁻¹); (C) organic loading rate (kg COD m⁻³ day⁻¹); and (D) COD removal efficiency (%).