

Innovative microbial fuel cell for electricity production from anaerobic reactors

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Abstract

A submersible microbial fuel cell (SMFC) was developed by immersing an anode electrode and a cathode chamber in an anaerobic reactor. Domestic wastewater was used as the medium and the inoculum in the experiments. The SMFC could successfully generate a stable voltage of 0.428 ± 0.003 V with a fixed 470Ω resistor from acetate. From the polarization test, the maximum power density of 204 mW m^{-2} was obtained at current density of 595 mA m^{-2} (external resistance = 180Ω). The power generation showed a saturation-type relationship as a function of wastewater strength, with a maximum power density (P_{max}) of 218 mW m^{-2} and a saturation constant (K_s) of 244 mg L^{-1} . The main limitations for achieving higher electricity production in the SMFC were identified as the high internal resistance at the electrolyte and the inefficient electron transfer at the cathode electrode. As the current increased, a large portion of voltage drop was caused by the ohmic (electrolyte) resistance of the medium present between two electrodes, although the two electrodes were closely positioned (about 3 cm distance; internal resistance = $35 \pm 2 \Omega$). The open circuit potential (0.393 V vs. a standard hydrogen electrode) of the cathode was much smaller than the theoretical value (0.804 V). Besides, the short circuit potential of the cathode electrode decreased during the power generation in the SMFC. These results demonstrate that the SMFC could successfully generate electricity from wastewater, and has a great potential for electricity production from existing anaerobic reactors or other anaerobic environments such as sediments. The advantage of the SMFC is that no special anaerobic chamber (anode chamber) is needed, as existing anaerobic reactors can be used, where the cathode chamber and anode electrode are immersed.

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1. Introduction

The concept of a microbial fuel cell (MFC) has been of interest again to researchers around the world in response to high demand of sustainable energy. An MFC is a device that can convert chemical energy from organic matter to electrical energy via both biotic- and abiotic-catalysts [1–3]. Electron transport between microorganisms (biocatalyst) and an electrode has been known to be possible without any additions of artificial mediators [4–8].

An MFC is generally composed of two chambers; an anode chamber where the oxidation of organic compounds takes place by microorganisms under anaerobic condition and a cathode chamber where an oxidant (electron acceptor) such as oxygen

or ferricyanide is reduced under aerobic condition [6,8–16]. The main potential advantage of the MFC technology for power generation from biodegradable organic matter is that electricity is produced directly without any additional step, as for other bioenergy conversion processes, where first an energy carrier is produced and subsequently the energy carrier is used for electricity generation with efficiency loss in each step. Additionally, MFCs can be used as a green treatment technology for removal of organic matter with simultaneous renewable energy production [17–20].

One promising application of the current MFC technology will be to generate electricity from wastewaters [11,13,21–23]. However, MFC technology is still in its infancy in laboratory experimental stage operation, and far from field application. Several limiting factors of the current MFCs have been identified: high internal resistance, high costs of materials, potential diffusion of oxygen from cathode chamber to anode chamber, and difficulty of scaling up [12–16,21,24–27]. It has been

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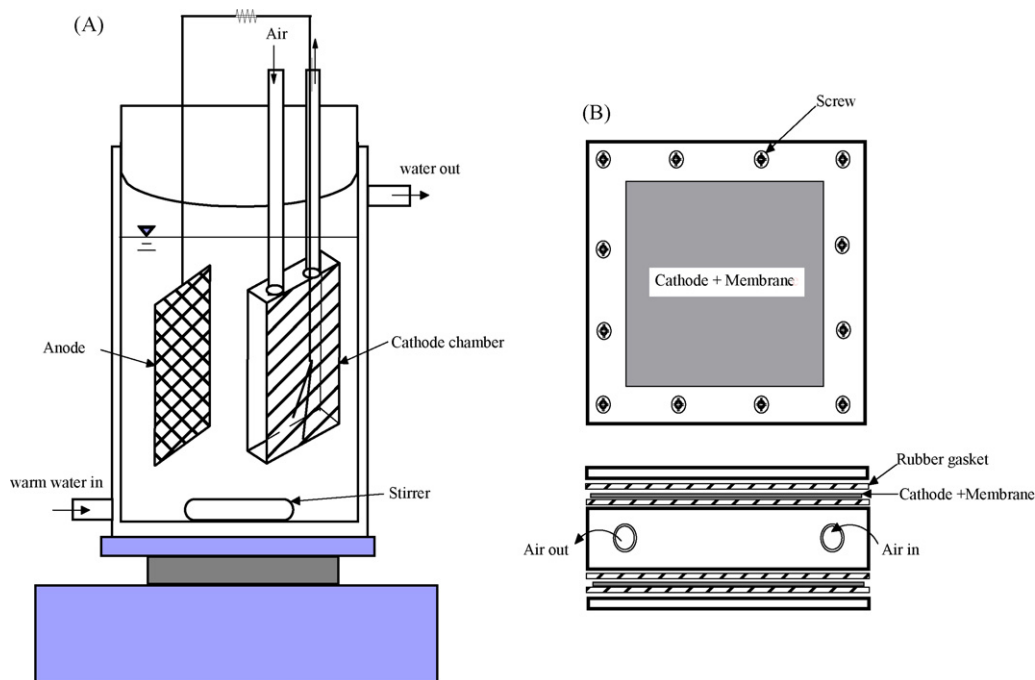


Fig. 1. Schematic diagram of (A) a submersible MFC (SMFC) and (B) a cathode chamber in the SMFC (upper, side view; lower, top view).

recognized that improvement of the MFC configurations could significantly contribute to overcome some of the present limitations [12,15,24]. Therefore, several types of MFCs have been developed and operated in either batch or continuous mode using natural or artificial wastewater [11,12,21,24–26]. For example, Liu et al. developed a single compartment MFC, which has a centered cathode surrounded by several anode electrodes. More detail information about the types of the MFCs can be found in the reference by Logan et al. [15]. Although researchers have intensively looked for new configurations, the current designs of MFC still suffer of high internal resistance limitations and are not suitable for in situ utilization of compounds from natural environment.

In this study, we developed a novel configuration of MFC, which is called a submersible MFC (SMFC), by immersing an anode electrode and an air-cathode chamber in an anaerobic reactor. In this innovative design, there was no necessity of a specially designed anode chamber which is the main difference from other MFC configurations. We expect that SMFC can be applied easily to the existing anaerobic reactors or natural anaerobic environments without additional constructions to recover maximum electrical energy from reduced compounds. The performance and characteristics of the SMFC was investigated in terms of individual electrode potential and power generation from organic substrate in wastewater medium.

2. Materials and methods

2.1. Inoculum and medium

Domestic wastewater was collected after a fine screen process of the Lundtofte Wastewater Treatment Plant (Lyngby,

Denmark). The wastewater was first sparged with nitrogen gas and then was placed in a temperature-controlled room at 4 °C prior to being used. Wastewater was used both as inoculum and as medium for power generation in the SMFC without addition of nutrients, buffer, vitamins, or minerals. During the startup period, acetate was added to wastewater as additional substrate at a final concentration of 1.6 g L⁻¹. In some experiments, different initial concentrations of acetate were tested.

2.2. SMFC configuration

The SMFC was composed of an anode electrode and a rectangular cathode chamber, which were submersed in a glass reactor (total volume = 600 mL, liquid volume = 550 mL) (Fig. 1A). The reactor was covered by a rubber stopper having several openings for liquid and gas samples, air supply to the cathode chamber, a thermometer, and the anode electrode. The reactor was submerged in a thermostatic water bath for temperature control. The anode electrode was a 4 cm × 4 cm piece of not wet proofed plain carbon paper (Toray carbon paper, E-TEK division, USA). The cathode electrode was a 5% wet proofed carbon paper (4 cm × 4 cm), and one side of the electrode contained Pt catalysts (0.5 mg cm⁻² with 20% Pt, E-TEK). The Pt-coated side was hot-pressed with a proton exchange membrane (NafionTM 117, DuPont Co., USA) as prepared in other study [21]. The cathode compartment was made of non-conductive polycarbonate plate enclosing a chamber of dimensions 4 cm × 4 cm × 1 cm (16 cm³) (Fig. 1B). The length and width (4 cm × 4 cm) of the cathode chamber were chosen in order to have the same size of the cathode as the anode electrode. There were two 3-mm diameter holes on the top of the chamber for airflow

and electrical outlet. The two sides of the cathode chamber were covered by a membrane-electrode assembly and an open frame was sealed with rubber gasket as shown in Fig. 1B. All metal parts were covered using silicon glue. The distance of the anode and cathode electrode was approximately 3 cm in the reactor.

2.3. SMFC operation

The reactor was operated in batch mode at 30 °C generally over 1 or 2 weeks before the solution was replaced with a freshly prepared medium. Wastewater was filled into the reactor and was purged with nitrogen gas for approximately 30 min prior to being tightly sealed inside an anaerobic glove box (Coy Laboratory products Inc.). Replacement of the wastewater in the SMFC was performed in an anaerobic glove box all the times. The reactor mixing was provided by magnetic stirring (300 rpm). The cathode chamber was aerated at a flow rate of approximately 10 mL min⁻¹ in all experiments. The electric current between two electrodes was generally measured with a fixed resistor of 470 or 180 Ω, except for the measurement with various loads using a resistance box.

2.4. Analysis and calculations

The strength of wastewater amended with acetate was expressed as the average soluble COD (SCOD) based on duplicate samples. All samples were filtered through a 0.2-μm diameter syringe membrane (Sartorius Minisart®), and in most cases the filtered samples were used immediately for the COD measurement except some samples were stored at a -20 °C refrigerator before the measurement. The determination of COD was performed using cell tests (Merck, Germany) on the basis of Standard Methods (method 5220) [28]. The potential of each electrode was measured by immersing an Ag/AgCl reference electrode (Bioanalytical Systems Inc.) into solutions, and electrolyte resistance between two electrodes was determined by an impedance spectroscopy instrument (HIOKI 3522-50 LCR HiTESTER).

The voltage difference between two electrodes was measured across a fixed load every 10 or 30 min, and the data were collected automatically by a data acquisition program and a personal computer. In some test, the external resistor was varied ranging 43–22 kΩ to determine the maximum power density and individual electrode potential as a function of different electric current. Current (I) was calculated on the basis of Ohm's law ($I = V/R$), where V is voltage and R resistance, and current density, i (A m⁻²), was calculated as $i = I/A$, where A is the projected surface area of the anode electrode. Power density, P (W m⁻²), was calculated by multiplying the current density by voltage, $P = IV/A$. Coulombic efficiency (CE) was calculated based on $CE = (C_G/C_T) \times 100\%$, where C_G is the total coulombs calculated by integrating the current generated over time, and C_T is the theoretical amount of coulombs available based on the measured COD removal in the MFC.

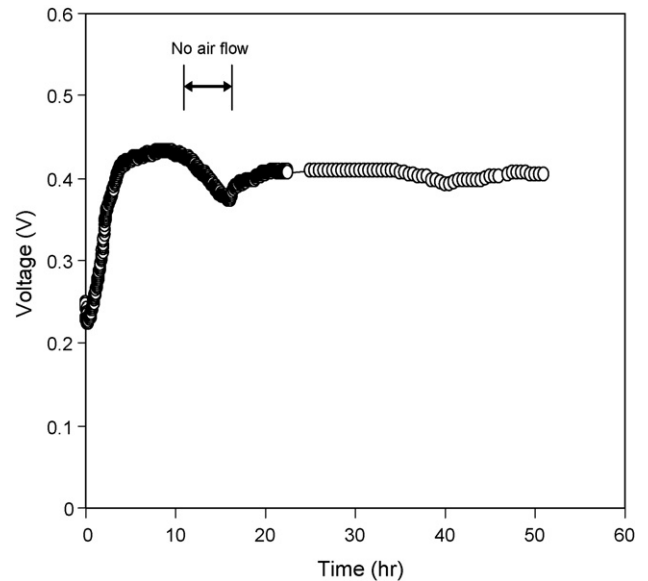


Fig. 2. Voltage generation from the modified wastewater containing acetate (1.6 g L⁻¹; SCOD = 1694 ± 20 mg L⁻¹) with a 470-Ω resistor in the SMFC. No airflow to the cathode chamber between 12 and 16 h.

3. Results and discussion

3.1. Voltage generation from the modified wastewater in the SMFC

Within 5 days after a startup period, the SMFC could successfully generate voltage up to a maximum 492 mV with a fixed 470 Ω resistor from modified wastewater amended with acetate to a concentration of 1.6 g L⁻¹ (data not shown). This result suggests that the domestic wastewater could support microbial growth as it contained the necessary electrolytes, nutrients and microorganisms necessary for power generation during the starting up period of the SMFC. This is in accordance with previous studies [11,13]. After two more additional loadings, the SMFC produced a stable voltage of 0.428 ± 0.003 V (±standard deviation) for 7 h (5–12 h) from the acetate-fed wastewater (SCOD = 1694 ± 20 mg L⁻¹; SCOD of wastewater itself = 222 ± 3 mg L⁻¹; Fig. 2). When air supply to the cathode chamber was stopped for about 4 h, a voltage decrease to 0.373 V was noticed (Fig. 2). This indicated that continuous air supply is necessary for oxygen reduction in the completion of the electrical circuit. Re-establishing the air supply to the cathode chamber restored the voltage up to average 0.406 ± 0.003 V over the next 31-h period.

3.2. Cell voltage and power output as a function of current density

In order to measure maximum power output, loadings from 43 to 22 kΩ resistances were applied between the anode and cathode electrode. The voltage and power density were plotted as a function of current density as shown in Fig. 3. An open circuit voltage (OCV, without a circuit load) of 0.720 V was first obtained with the wastewater of SCOD = 1672 ± 6 mg L⁻¹. However, as cur-

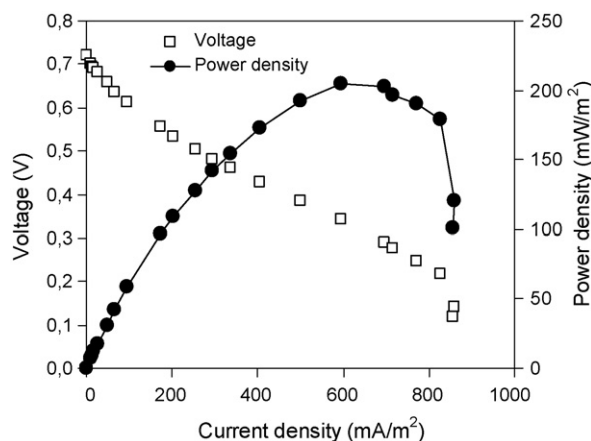


Fig. 3. Power density and cell voltage as a function of current density by varying an external resistor between the anode and cathode electrodes ranging from 43 Ω to 22 k Ω (SCOD of wastewater = 1672 ± 6 mg L $^{-1}$).

rent between the electrodes was allowed, the voltage decreased sharply to approximately 0.6 V (at around 96 mA m^{-2}) possibly showing the presence of a kinetic limitation at the lower current generation [29]. At higher current production, voltage decreased almost linear from 0.612 to 0.217 V until a current density of about 827 mA m^{-2} was obtained. This result indicated that the electrolyte resistance of a real wastewater medium still affected significantly the power output even though the anode and cathode electrodes were placed close together in this study (3 cm). The internal resistance of the SMFC was measured at $35 \pm 2 \Omega$ from the electrochemical impedance test. The significant effect of the electrolyte resistance on power generation was also reported in other MFC studies [12,25,30]. A maximum power density of 204 mW m^{-2} was obtained at a current density of 595 mA m^{-2} at 180 Ω resistance. This maximum power output in the SMFC containing acetate in the wastewater medium is approximately five times higher than the values (range of 38–43 mW m^{-2}) obtained in the two-chamber membrane MFCs with the artificial nutrient medium containing acetate [10,12,14]. However, this power generation was lower in comparison with the one obtained by membrane-less single-chamber MFC showing a maximum power density of 506 mW m^{-2} from acetate in a nutrient synthetic medium [31]. Possible explanations for the lower power output compared to the single chamber MFC could be due to lower conductivity of the wastewater medium and the presence of a membrane in the SMFC [12,30,31]. The cell voltage in the SMFC further decreased sharply at the range of high current density (approximately 850 mA m^{-2}) probably due to mass transport limitation at both anode and cathode chambers.

3.3. Individual electrode potential at different current densities

Individual electrode potential was measured against an Ag/AgCl reference electrode (0.195 V vs. standard hydrogen electrode, SHE) in order to examine the performance of each electrode at different current densities. The open circuit potentials (OCPs vs. an Ag/AgCl reference) of the anode and cathode

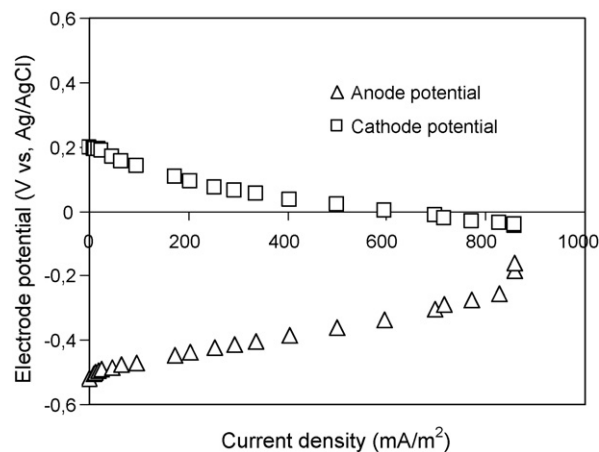


Fig. 4. Individual electrode potential vs. an Ag/AgCl reference electrode (0.195 V vs. standard hydrogen electrode, SHE) as a function of current density in the SMFC fed with wastewater.

were -0.518 V (-0.323 V vs. SHE) and 0.198 V (0.393 V vs. SHE), respectively. The cathode OCP ($\text{OCP}_{\text{cathode}}$) of 0.393 V vs. SHE was much lower than the theoretical value (0.804 V) calculated by the Nernst equation on the base of pH 7, 0.2 atm. O_2 , and 30°C (51% potential reduction) [15]. The obtained $\text{OCP}_{\text{cathode}}$ from the SMFC was higher than the value (0.358 V vs. SHE) for the membrane two-chamber MFC [12], but was lower than 0.413 V for the single chamber MFC without membrane [31]. The lower $\text{OCP}_{\text{cathode}}$ in the SMFC would be one of the factors causing the lower power generation in comparison with the single chamber MFC. One of the reasons for the lower $\text{OCP}_{\text{cathode}}$ in MFC could be that other cations than protons (Na^+ , K^+ , Ca^{2+} , etc.) are crossing the membrane from the anode side [32]. However, the OCP of the anode ($\text{OCP}_{\text{anode}}$) could not be compared directly to the theoretical value because there was no information about the redox pairs for electron transport in microorganisms in this study. Instead, if considering the redox potential of the input substrate (assuming acetate = 20 mM, $\text{HCO}_3^- = 1 \text{ mM}$ at pH 7 and 30°C), the observed value of $\text{OCP}_{\text{anode}}$ (-0.323 V vs. SHE) was similar to the theoretical value of 0.318 V [15]. The $\text{OCP}_{\text{anode}}$ (-0.323 V) observed in this study is higher than the values (from -0.285 to -0.214 V) from other studies using different types of the MFCs [12,31].

The overvoltage (internal resistance) at both anode and cathode increased with current generation between the two electrodes of the SMFC. The $\text{OCP}_{\text{anode}}$ of -0.518 V increased quickly to -0.492 V at the lower current density possibly indicating that there were kinetic limitations (Fig. 4). As higher current was applied, the anode potential further increased almost linearly up to -0.254 V at a current density of around 827 mA m^{-2} . Possible mass transport limitation on the anode was observed at higher current density ($> \sim 827 \text{ mA m}^{-2}$). The same trend was seen for the potential of the cathode, that in this case, decreased almost linearly from 0.198 V of $\text{OCP}_{\text{cathode}}$ down to -0.043 V with an increase of the cell current generation without a sudden change of the potential as shown in the anode potential.

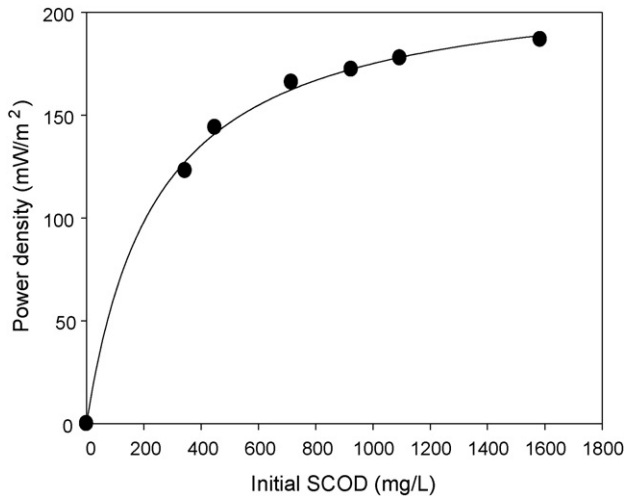


Fig. 5. Electricity generation in the SMFC as a function of initial SCOD of wastewater ($R=180\ \Omega$). The power density showed a saturation curve with respect to different SCODs, with $P_{\max}=219\ \text{mW m}^{-2}$ and $K_s=244\ \text{mg L}^{-1}$.

3.4. Power output with different concentrations of the modified wastewater

Power generation by the SMFC, from different initial acetate concentrations in wastewater, was tested. The voltage measurements were conducted with a fixed resistor of $180\ \Omega$, at which the maximum power was obtained at the previous polarization test. Power generation was immediately developed without a lag period after receiving the acetate amended wastewater ($344\text{--}1584\ \text{mg L}^{-1}$ SCOD), and average power density was calculated on the basis of values obtained during the operation of stable power generation. The power generation as a function of wastewater strength showed a saturation-type relationship over the range of the tested initial acetate concentrations (Fig. 5). This observation is consistent with the findings reported by other studies regardless of reactor configuration and substrates [21,31,33]. The maximum power density, P_{\max} , in the equation for this hyperbola was $218\ \text{mW m}^{-2}$ with a half-saturation constant, K_s of $244\ \text{mg L}^{-1}$.

3.5. Cell voltage and individual cell potential as a function of time

Cell voltage decreased slowly with operation time at some experiments, especially after approximately a week of the SMFC operation. For elucidating the reason for this decrease, short circuit potentials (SCP, at $470\ \Omega$) of both anode and cathode were measured separately vs. an Ag/AgCl reference electrode as a function of operation time (Fig. 6A). The cell voltage of the SMFC was calculated based on the SCPs of the two electrodes ($\text{SCP}_{\text{cathode}} - \text{SCP}_{\text{anode}}$). For about 20 h of operation, the cell voltage and cathode potential decreased from 0.432 to 0.373 V and from 0.094 to 0.008 V, respectively. However, the performance of the anode was improved slightly as a consequence of the decrease of the potential from -0.344 to -0.364 V during that period. This result indicated that optimization of a cath-

ode in the SMFC is necessary for increasing power generation, especially for stable power generation in a continuous operation of the SMFC. The SCOD of the wastewater decreased from 968 ± 3 to $826 \pm 23\ \text{mg L}^{-1}$ for 20 h of operation, and Coulombic efficiency was 7% during that period. The low CE was likely due to substrate loss by suspended bacteria from wastewater, in their non-power generation processes such as aerobic respiration, biomass production, and methanogenesis [12,31].

Water flooding at the cathode was thought to cause the deterioration of the cathode performance by hindering accessibility of gaseous oxygen to platinum catalysts [34]. Therefore, the cathode electrode was dried almost completely, and then installed again at nearly identical conditions to the previous operation. However, the performance of the cathode did not improve, even became worse compared to the previous set-up (Fig. 6B). Cathode potential of 0.034 V was initially developed, and then decreased sharply down to -0.131 V within about 21 h. However, this time, the anode potential decreased from -0.402 to -0.419 V indicating improvement of the anode performance. As a result, the significant decrease in cathode potential resulted in decreasing the cell voltage from 0.435 to 0.297 V. The SCOD was decreased from 1004 to $980\ \text{mg L}^{-1}$ during 21 h of operation, and at the end of the experiment, it decreased to $940 \pm 6\ \text{mg L}^{-1}$ after about 52 h (CE = 28%).

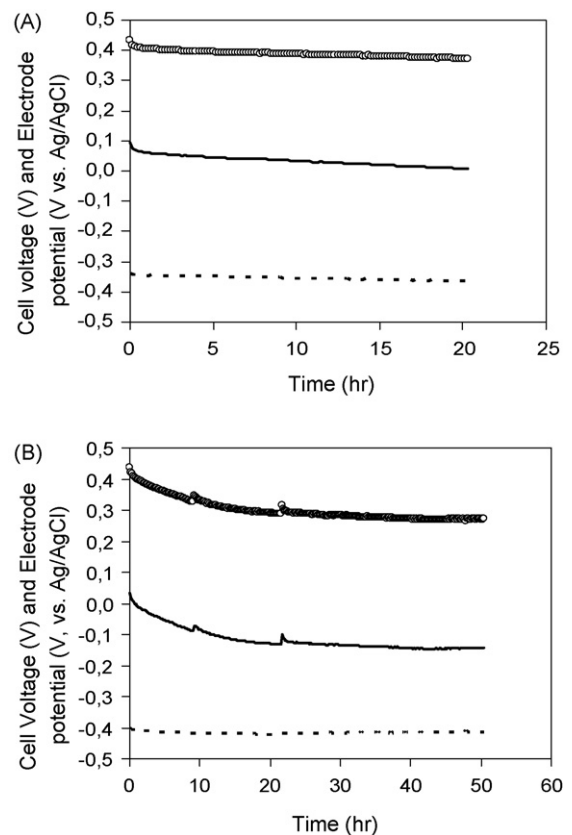


Fig. 6. Cell voltage (\circ) and two-electrode potentials (anode (\cdots) and cathode (---)) as a function of time. (A) With a cathode electrode used for several runs of the SMFC, (B) after drying out the cathode used in the previous experiment.

Another possible reason of the deterioration of the cathode performance during the operation could be due to the Nafion-type membrane used, which was hot-pressed with a cathode electrode in this study [11,16,32,35]. The membrane is typically used as solid electrolyte for transport of only protons in hydrogen fuel cell. However, in MFC, there are other kinds of cations (e.g. Na⁺) at high concentration in anode chamber, which also can cross through the membrane to cathode chamber [35]. Therefore, permeability of other cations through Nafion membrane in MFC will inhibit the accessibility of proton ions to the cathode, which are necessary for oxygen reduction, and finally decrease oxygen reduction rate. The slow permeability of proton ions would also increase pH at the surface of the cathode, and decrease the theoretical potential of the cathode of 0.804 (pH 7) to 0.677 V (e.g. pH 9) vs. SHE at 30 °C [15,16]. The decreased theoretical potential will further limit the possibility of higher power generation in the SMFC operation.

Dimensioning of the SMFC is also important. Several considerations are important for the size of the cathode chamber, such as the relative volume of the cathode chamber to the anaerobic reactor volume. A small cathode chamber volume would permit a larger number of cathode chambers immersed in the anaerobic reactor and thus facilitate easier electron transfer to the anode. Other considerations are the practical aeration of the cathode chamber as well as construction costs. However, in the present study the dimensions of the cathode chamber were not optimized.

4. Conclusions

It has been determined from several previous studies that the MFC configuration is very important factor for power generation. In this study, an innovative MFC configuration (the SMFC) was developed. The SMFC was constructed by immersing both anode and cathode chamber in one bioreactor. The SMFC could successfully generate electricity from wastewater amended with acetate. The maximum power generation was 204 mW m⁻² with current density of 595 mA m⁻² at a circuit resistance of 180 Ω (SCOD of wastewater = 1672 ± 6 mg L⁻¹). The power output showed a saturation-type relationship as a function of wastewater concentration (SCOD), with a half saturation coefficient of $K_s = 244 \text{ mg L}^{-1}$ and a maximum power density of $P_{\max} = 244 \text{ mW m}^{-2}$. The OCP of the anode electrode was 0.323 V vs. SHE, which was a similar to theoretical values or the observations from other studies. However, the cathode OCP of 0.393 V (vs. SHE) was much less than the theoretical value (0.804 V, by 51% reduction), and even smaller than the value (~0.413 V) from other types of air cathode MFCs. Besides, the cathode potential decreased after a long period of operation. These results indicated that more optimization of the SMFC, especially the cathode chamber, is essential for higher and stable power generation. The SMFC configuration has however great potential for effective electricity generation from artificial or even natural environment, and can be also used for other applications like biosensors or bioremediation in the future.

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