  Water Science & Technology | 69.12 | 2014

High speed municipal sewage treatment in microbial fuel cell integrated with anaerobic membrane filtration system

Y. Lee and S. W. Oa

ABSTRACT

A cylindrical two chambered microbial fuel cell (MFC) integrated with an anaerobic membrane filter was designed and constructed to evaluate bioelectricity generation and removal efficiency of organic substrate (glucose or domestic wastewater) depending on organic loading rates (OLRs). The MFC was continuously operated with OLRs 3.75, 5.0, 6.25, and 9.38 kg chemical oxygen demand (COD)/(m^3·d) using glucose as a substrate, and the cathode chamber was maintained at 5–7 mg/L of dissolved oxygen. The optimal OLR was found to be 6.25 kgCOD/(m^3·d) (hydraulic retention time (HRT) 1.9 h), and the corresponding voltage and power density averaged during the operation were 0.15 V and 13.6 mW/m^3. With OLR 6.25 kgCOD/(m^3·d) using domestic wastewater as a substrate, the voltage and power reached to 0.13 V and 91 mW/m^3 in the air cathode system. Even though a relatively short HRT of 1.9 h was applied, stable effluent could be obtained by the membrane filtration system and the following air purging. In addition, the short HRT would provide economic benefit in terms of reduction of construction and operating costs compared with a conventional aerobic treatment process.

Key words | bioelectricity, membrane filter, microbial fuel cell, organic loading rate, wastewater

Y. Lee
S. W. Oa (corresponding author)
Department of Railroad Civil & Environmental Engineering,
Woosong University,
Daejeon 300-718,
Republic of Korea
E-mail: *swoa@wsu.ac.kr*

INTRODUCTION

Microbial fuel cell (MFC) technology has a merit in terms of it being possible to treat contaminants and to produce electrical energy simultaneously. MFC is one of the promising technologies for wastewater treatment. Application of a MFC system for wastewater treatment is comparable to a conventional aerobic wastewater treatment process in terms of energy production and energy saving. Approximately 1 kWh of electricity is consumed for aeration to treat 1 kg of BOD (biochemical oxygen demand) and the aeration accounts for half of the operating cost in a conventional wastewater treatment process, while the MFC system is able to treat wastewater without energy-intensive aeration needed for aerobic treatment (Oswald 2003). In addition, sludge production yield from the anaerobic process is about 20% compared to the aerobic process, thereby reducing operating cost for sludge handling (Tchobanoglous *et al.* 2003).

However, electric performances of the MFC such as voltage generation or coulombic efficiency should be improved to apply in wastewater treatment. And also economic feasibility of the MFC technology should be secured. To achieve the optimal MFC process, identification of various operating parameters affecting the reaction mechanisms of the MFC is required. The operating parameters are known as the carbon source to be supplied to an anode chamber, oxidation rate of the substrate, electron transfer efficiency onto an electrode, external resistance, movement velocity of protons via a proton exchange membrane, oxygen supply, reduction rate in cathode chamber, size of an electrode and so on (Davila *et al.* 2008). Over the past 20 years, power production yield has been increased through intensive research regarding improvement of electrode materials, system design, and change of configuration; however, research on field application is still narrow.

One of the important parameters in MFC for application in wastewater treatment in a field is determining organic loading rate (OLR) to supply appropriate carbon source, or hydraulic retention time (HRT) to ensure the capacity of the MFC system in continuous operation. Another is securing high quality effluent. MFC is basically

doi: 10.2166/wst.2014.179

an anaerobic process; however, an anaerobic process fundamentally has a long retention time which is unsuitable for domestic sewage with low concentration and high flux. To resolve these issues, combining the anaerobic membrane bioreactor (AnMBR) process to the MFC system could be considered, in terms of securing the anaerobic reaction to produce bioelectricity and high quality effluent in a continuous system. AnMBR is an attractive option for treating low strength wastewater such as domestic sewage in terms of providing a short HRT, a long solid retention time (SRT), low sludge production, high loading rate, and high quality effluent resulting from complete solid–liquid separation by filtration through a membrane (Lew *et al.* 2009; Gao *et al.* 2010).

Therefore, in this study, a MFC integrated with an anaerobic membrane filter was designed and operated in continuous mode to provide fundamental data for field application. As one of the operating parameters, OLR was evaluated in terms of voltage generation and removal efficiency of organic compounds. The purposes of this study are: (1) to develop a composite system using a MFC and an AnMBR; (2) to effectively treat low strength domestic sewage, capable of guaranteeing the effluent limits; and (3) to directly use potential energy produced by the MFC, resulting in increased economic value.

MATERIALS AND METHODS

Configuration of MFC

A cylindrical two chambered MFC integrated with a membrane filter was constructed. The anode chamber was made of a 22 cm long by 11 cm outer diameter waterproof acryl with an effective volume of 800 mL, and the cathode chamber was 22 cm long by 1.5 cm inner diameter with volume of 50 mL. A commercial filter membrane of 5 μm pore size using pure polypropylene (KASCO Inc.) which is made of a 22 cm long by 6.2 cm outer diameter and 2.9 cm of inner diameter was embedded in the anode chamber. The cathode chamber was located inside the filter membrane within the anode chamber. Each detailed electrode configuration and construction photographs are shown in Figure 1. The anode electrode was made by wrapping carbon fibers (Panex35 50 K, Zoltek) onto the surface of the filter membrane. A silver wire (ø 0.7 mm) was put between the filter membrane and carbon fibers to deliver electrons produced by substrate degradation. The cathode electrode was constructed using carbon fibers without a catalyst and a silver wire onto the surface of the cathode chamber. And a waterproof cloth was put onto the cathode electrode to prevent release of effluent from the anode chamber. The waterproof cloth consisted of

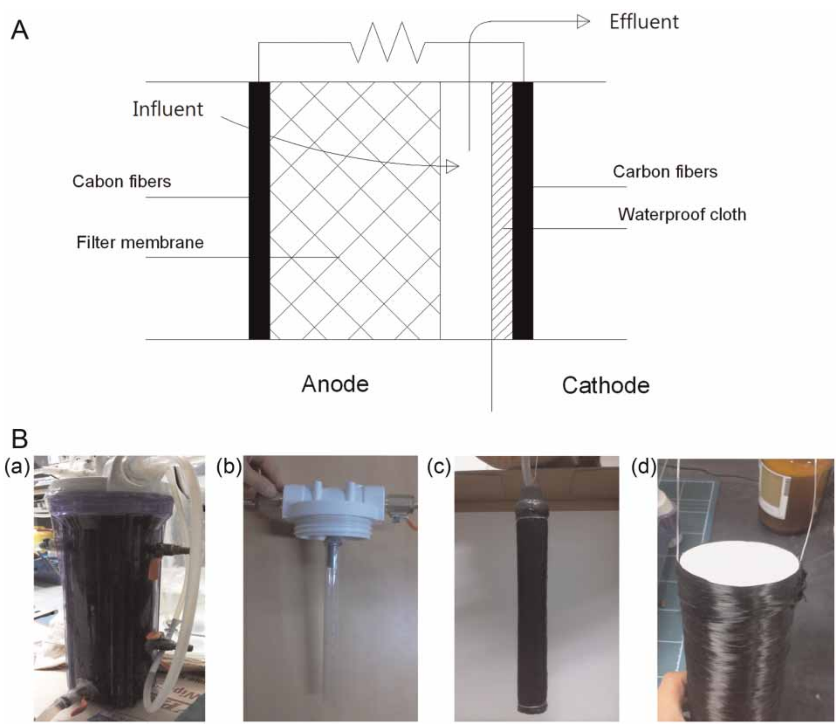

Figure 1 | Details of constructed MFC reactor inside (A) and photographs of MFC constructed in this study (B): (a) entire reactor, (b) cathodic part, (c) waterproof cloth on cathode electrode, and (d) filter membrane and carbon fiber.

breathable hipora cloth (water pressure 2,000 mm/H_2O, water vapor permeability 936 g/(m²·d), and ø 0.1–3 μ) able to accept the movement of hydrogen ions from anode to cathode chamber. The two electrodes were connected by silver wires carrying an external load of 2,000 Ω resistance unless otherwise stated. The spacing between the two electrodes was 2.1 cm.

Substrates

Experiments were started with artificial wastewater to examine system security and to maintain a steady state using glucose medium (500 mg chemical oxygen demand (COD)/L glucose, 0.31 g/L NH_4Cl, 0.13 g/L KCl and 50 mM phosphate buffer solution using 4.58 g/L Na_2HPO_4, and 2.45 g/L $NaH_2PO_4 \cdot H_2O$, pH 7) supplemented with 12.5 mL/L Wolfe's vitamin solution and 12.5 mL/L Wolfe's mineral solution. Domestic wastewater was directly collected from a sewerage manhole in Deajeon, Republic of Korea, and extra glucose was added to supply appropriate substrate concentration for activation of the system. The influent concentration was adjusted to around 500 mg/L using supplemental glucose medium.

Source of inoculum

Anaerobic digested sludge as a seed was collected from a wastewater treatment plant in Daejeon and pre-treated after removing impurities using a 2 mm sieve. According to Sung *et al.* (2002), heat treatment as a pre-treatment method increased hydrogen production by inactivation of non-spore-forming hydrogen-consuming bacteria. In most cases, heat treatment at 75–80 °C for 15–20 min is used to inactivate vegetative cells (Doyle 1989). In this study, the anaerobic digested sludge was heat-treated in 80 °C for 10 min to inactivate the heat-sensitive methane-producing bacteria, which resulted in providing more hydrogen ions rather than methane as a final product. The anaerobic sludge, up to as much as 50% of reactor volume, was added into the anode chamber to maintain anaerobic condition, and oxidation reduction potential (ORP) was measured during the operation to check the anaerobic condition.

Operating conditions

Decision of optimal OLR

The effects of OLR on voltage production and wastewater treatment efficiency with OLR of 3.75, 5.0, 6.25, and 9.38 kg COD/(m^3·d) (feeding rate of 6, 8, 10, and 15 L/d) were evaluated. Glucose as an anodic electrolyte was continuously injected from an inflow water bath into the anode chamber depending on OLRs using a Master flex pump (No.7523-80). And tap water as a cathodic electrolyte was aerated using an air stone and circulated to the cathode chamber. Dissolved oxygen (DO) was maintained between 5 and 8 mg/L. Temperature for the operation was maintained between 20 and 30 °C.

Voltage generation from municipal sewage

Carbon fibers for the anode electrode were pre-treated to increase electron transfer efficiency by creating the hydrophilic condition and to remove impurities (Huang *et al.* 2002). The carbon fibers were flushed by 20% nitric acid for 24 h and then washed using tap water until pH 7. The cathode system was changed from aeration to air cathode system. OLR 6.25 kgCOD/(m^3·d) was applied and the corresponding HRT was 1.9 h. The external resistance of 200 Ω was used.

Analytical methods

Influent and effluent liquid samples were collected from the MFC to measure pH, DO, suspended solids (SS), volatile suspended solids (VSS), and soluble COD (SCOD). The pH, DO, and ORP were measured using Thermo Scientific water analysis instrument Orion 5star (Model Meter, 1119000 Serial Number B36702, USA). SCOD was analyzed by closed reflux method (Spectronic 20D +), and SS and VSS were measured by total suspended solid dried method (Standard Method 2540D, B (APHA 2005)). Cell voltage generated from the MFC was recorded at interval of 10 min by a multi-volt meter with a data acquisition system (Keithley 2700, USA).

RESULTS AND DISCUSSION

Continuous voltage production and substrate removal by OLRs

The effects of OLR on voltage production and removal efficiency of organic compound were evaluated in the continuous MFC system. OLRs were adjusted to operating conditions in the range of 3.75–9.38 kgCOD/(m^3·d) (HRT 3.2–1.3 h) by changing feeding rate. The results of voltage production as a function of OLR in the continuous MFC are shown in Figure 2. With OLR 3.75 kgCOD/(m^3·d), voltage

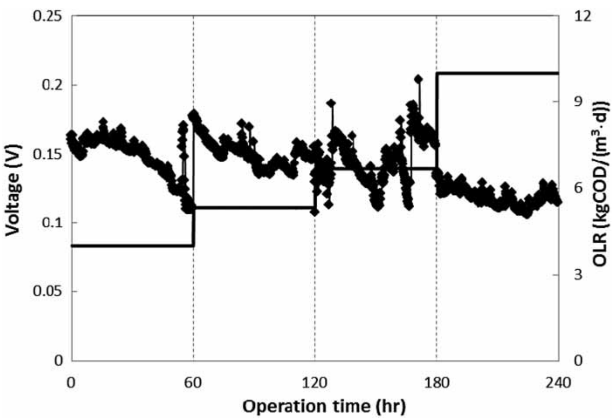

Figure 2 | Voltage production according to the OLRs.

and power density produced from the MFC averaged at 0.14 V and 12.2 mW/m^3. When the OLRs were increased to 5.0 and 6.25 kgCOD/(m^3·d), averaged voltage and power were also slightly increased to 0.15 V and 13.6 mW/m^3. Even though OLR was increased from 3.75 to 6.25 kgCOD/(m^3·d), we did not observe a significant voltage variation. This indicates that readily available substrate was sufficiently supplied and rapidly contributed to voltage generation by the electroactive biofilm in spite of increased loading rate. This result agreed with a previous study that there was no significant voltage variation when OLR increased from 1.92 to 4.80 gCOD/(L·d) using fermented wastewater (Nam *et al.* 2010). However, these authors mentioned that bacterial activity, internal resistance, and cathode reaction limitation could be different at different OLRs. Our study also presented different internal resistance of 2,091, 1,755, 1,728, and 2,374 Ω at OLR of 3.75, 5.0, 6.25, and 9.38 kg/(m^3·d), respectively. The internal resistance was calculated from the slope of a plot of current vs voltage generation (Freguia *et al.* 2008). However, the averaged voltage and power was decreased to 0.12 V and 8.7 mW/m^3 at OLR 9.38 kgCOD/(m^3·d) (HRT 1.3 h). It was considered to be inhibited by shorter HRT. Too short a HRT might disturb the transferring of electrons or hydrogen ions from anode to cathode electrode. Consequently, if the power produced from the MFC is similar, a shorter HRT has more economic merits. Therefore, the optimal OLR and HRT based on the voltage and power production were decided as 6.25 kgCOD/(m^3·d) and HRT 1.9 h. A common HRT in a general municipal wastewater treatment plant is 6–8 h (Tchobanoglous *et al.* 2003). Considering that, the approximately 2 h HRT derived from this study is effective in terms of decreasing construction cost by reducing the reactor volume to 25%.

Averaged COD concentration, as organics treatability, during each operating period (60 h) for influent and effluent from the anode chamber was analyzed for the varied OLRs. Removal efficiencies of glucose were found to be 84–66% with OLR 3.75–9.38 kg/(m^3·d). The removal efficiency was the highest with OLR 3.75 kg/(m^3·d) and similar efficiencies were found in range of 66–69% with other OLR conditions. The organics treatability was deteriorated by increased feeding rate, especially below the 2.4 h of HRT. The removal efficiency of the organic substrate at optimal OLR 6.25 kg/(m^3·d) was 67%. The pH was decreased from around 7 to 6.5 with OLR 3.75, 6.25, and 9.38 kg/(m^3·d). The decreased pH is inferred to be caused from VAs (volatile acids) produced by anaerobic fermentation in the anode chamber. We confirmed that the effluent of the MFC reactor included considerably high VAs of 190 mg/L within HRT 1.9 h. The acidified effluent is expected to be easily volatilized by air purging. Therefore, the COD removal efficiency would be expected to be increased by the following air purging.

Power production from domestic sewage

An air cathode system was employed in the original MFC designed in this study and the external resistance changed to 200 Ω. In Figure 3(a), the voltage produced from domestic sewage increased steeply until around 70 mV during the first 3 h; thereafter, the voltage was gradually increased and averaged at around 100 mV. The peak power density was 56 mW/m^3 and the power output was averaged at 51 mW/m^3 after stabilization (11 h). The averaged voltage and power density were increased to 0.13 V and 80 mW/m^3 after removing the cap of the cathode compartment (46 h). And the maximum power output reached 91 mW/m^3 at external resistance of 200 Ω via a polarization curve test (Figure 3(b)). The internal resistance was calculated as 255 Ω by the polarization slope method. In Table 1, produced power in this study was relatively low compared to that from previous researches in continuous operation. Liu *et al.* (2004) reported significant power output using domestic wastewater and an air cathode system, unlike our study. We operated at relatively short HRT with a non-catalyst cathode system. Also high internal resistance or large systemic loss (open cell voltage of 0.46 V) would result in low power production. Besides that, Jang *et al.* (2004) used glucose and glutamate as a carbon source and obtained maximum power of 10 mW/m^3 in continuous operation, which was a similar result to our study using glucose.

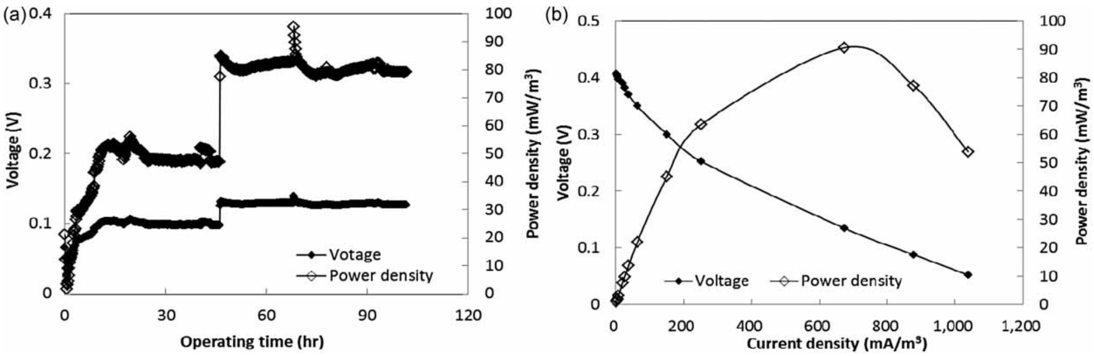

Figure 3 | Voltage and power density produced from domestic sewage (a) and polarization curve (b).

Table 1 | Comparison of MFC performance in continuous operation

Cathode system	Substrate	OLR (kg/(m^3·d))	HRT (h)	Maximum power output (mW/m^3)	COD removal efficiency (%)	Coulombic efficiency (%)	Reference
Air cathode	Domestic wastewater	0.13–0.70	3–33	1600	40–80	3–12	Liu *et al.* (2004)
Air cathode	Fermented wastewater	3.84	1.04	3000	93	<1.0	Nam *et al.* (2010)
Aeration	Glucose & glutamate	0.3	1	10	>90	<10	Jang *et al.* (2004)
Aeration	Glucose	3.75–9.38	3.2–1.3	13.6	>99	<1.0	This study
Air cathode	Domestic wastewater	6.25	1.9	91	87	1.8	This study

Averaged COD removal efficiency during the operation was 43% and increased up to 87% by subsequent air purging. And the coulombic efficiency was very low at 1.8%, which indicated that substantial reduction of COD was not contributing to power production. Liu *et al.* (2004) also reported that a large percent of COD removal was not associated with power generation. It is difficult to clearly explain the correlation between COD reduction and power generation, while COD reduction could be possibly caused by oxygen transmitted from the cathode part or DO in the influent. And a short HRT would accelerate the inflow of the DO. Even though the power generated was still low, the wastewater could be treated with high efficiency. And the amount of electrical energy which can be recovered from a large wastewater treatment plant would be significant. This anaerobic membrane filtration system would be about three to four times more cost-effective than a conventional aerobic treatment process with an energy-intensive aeration system, by reducing reactor volume because of the very short HRT. In comparison with MFC aerobic MBR combination system in which the MFC is located inside the aerobic MBR (Wang *et al.* 2012), our system has significantly short HRT which is 4.5 times less, and could reduce the facility cost and aeration cost, which accounts for almost all the energy and operating cost in the wastewater treatment plant. Moreover, we used air purging to increase removal efficiency of organics in this study, but we are studying the availability of the organics as a carbon source for denitrification of the MFC effluent without aeration. In this case, the operating cost can be further reduced. Therefore, ultimately, the system would be effective in terms of low investment and operating cost.

CONCLUSIONS

The effects of OLR on voltage production and removal efficiency of organic substrate were evaluated using the MFC integrated with an anaerobic membrane filter. The optimal OLR was found to be 6.25 kgCOD/(m^3·d) (HRT 1.9 h), and the corresponding voltage and power produced from domestic sewage was found as 0.13 V and 91 mW/m^3. Even though a relatively low HRT was applied to the system, stable effluent quality could be obtained by the membrane filtration system and the simple air purging. The wastewater was securely removed within the short HRT of 1.9 h using the MFC designed in this study, and the MFC had economic benefit in terms of reduction of construction and operating cost compared to that in conventional activated sludge plants. In addition, the acidified effluent from the MFC can be used for denitrification as a carbon source; thereby operating costs can be more reduced.

ACKNOWLEDGEMENT

This research was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology (No. 2011-0013589).

REFERENCES

APHA 2005 *Standard Methods for the Examination of Water and Wastewater*, 21st edn. American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC, USA.

Davila, D., Esquivel, J. P., Vigues, N., Sanchez, O., Garrido, L., Tomas, N., Sabate, N., Campo, F. J., Munoz, F. J. & Mas, J. 2008 Development and optimization of microbial fuel cell. *Journal of New Materials for Electrochemical Systems* **11**, 99–103.

Doyle, M. P. 1989 *Foodborne Bacterial Pathogens*, 10th edn, Marcel Dekker, New York.

Freguia, S., Rabaey, K., Yuan, Z. & Keller, J. 2008 Sequential anode-cathode configuration improves cathodic oxygen reduction and effluent quality of microbial fuel cells. *Water Research* **42**, 1387–1396.

Gao, D.-W., Zhang, T., Tang, C.-Y. Y., Wu, W.-M., Wong, C.-Y., Lee, Y. H., Yeh, D. H. & Criddle, C. S. 2010 Membrane fouling in an anaerobic membrane bioreactor: differences in relative abundance of bacterial species in the membrane foulant layer and in suspension. *Journal of Membrane Science* **364**, 331–338.

Huang, W. Z., Zhang, X. B., Tu, J. P., Kong, F. Z., Ma, J. M., Liu, F., Lu, H. M. & Chen, C. P. 2002 The effect of pretreatments on hydrogen adsorption of multi-walled carbon nanotubes. *Materials Chemistry and Physics* **78**, 144–148.

Jang, J. K., Pham, T. H., Chang, I. S., Kang, K. H., Moon, H., Cho, K. S. & Kim, B. H. 2004 Construction and operation of a novel mediator-and membrane-less microbial fuel cell. *Process Biochemistry* **39**, 1077–1012.

Lew, B., Tarre, S., Beliavski, M., Dosoretz, C. & Green, M. 2009 Anaerobic membrane bioreactor (AnMBR) for domestic wastewater treatment. *Desalination* **243**, 251–257.

Liu, H., Ramnarayanan, R. & Logan, B. E. 2004 Production of electricity during wastewater treatment using a single chamber microbial fuel cell. *Environmental Science & Technology* **38**, 2281–2285.

Nam, J. Y., Kim, H. W., Lim, K. H. & Shin, H. S. 2010 Effects of organic loading rates on the continuous electricity generation from fermented wastewater using a single-chamber microbial fuel cell. *Bioresource Technology* **101**, 533–537.

Oswald, W. J. 2003 My sixty years in applied algology. *Journal of Applied Phycology* **15**, 99–106.

Sung, S., Raskin, L., Duangmanee, T., Padmasin, S. & Simmons, J. J. 2002 Hydrogen production by anaerobic microbial communities exposed to repeated heat treatments. *Proceedings of the 2002 U.S. DOE Hydrogen Program Review NREL/CP-610-32405*, Golden, CO, USA.

Tchobanoglous, G., Burton, F. L. & Stensel, H. D. 2003 *Wastewater Engineering, Treatment and Reuse*, 4th edn, Metcalf & Eddy, Inc., McGraw-Hill, Boston.

Wang, Y.-P., Liu, X.-W., Li, W.-W., Li, F., Wang, Y.-K., Sheng, G.-P., Zeng, R. J. & Yu, H.-Q. 2012 A microbial fuel cell–membrane bioreactor integrated system for cost-effective wastewater treatment. *Applied Energy* **98**, 230–235.

First received 21 November 2013; accepted in revised form 31 March 2014. Available online 12 April 2014