

Sustainable Approach for Leachate Treatment: Electricity Generation in Microbial Fuel Cell

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Electricity generation from landfill leachate was examined by using both a dual-chamber microbial fuel cell (MFC) and a single chamber MFC. Experimental results showed that the maximum power density of 2060.19 mW/m³ in the dual-chamber MFC and that of 6817.4 mW/m³ in the single chamber MFC were obtained. It was recognized that the difference in internal resistance for two MFC systems was the main reason for resulting in the difference of power generation. Power generation as function of chemical oxygen demand (COD) in single chamber MFC showed a Monod-type relationship with P_{\max} of 5920.96 mW/m³ and K_s of 251.39 mg/L at an external resistance of 500 Ω . Cyclic voltammetry showed that electrons were directly transferred onto the anode by bacteria in biofilms, rather than self-produced mediators of bacteria in the solutions. At low COD concentration, electricity generation was limited by the anode due to kinetic limitation; while at high COD concentration, the cathode was shown to have more significant effects on the electricity generation than the anode. COD in leachate could be removed when it increases, mainly because oxygen diffused from the cathode was substantially reduced by aerobic or anoxic bacteria in the anode chamber, leading to the substrate loss. Removal of ammonium-nitrogen was not observed in the single chamber MFC. This novel technology provides an economical route for electricity energy recovery in leachate treatment.

Key Words: Leachate; Dual-chamber microbial fuel cell; Single chamber microbial fuel cell; Power density; Electron recovery.

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INTRODUCTION

Sanitary landfill has been widely adopted as one of the most economical processes of solid waste disposal for years. For example, it is recently estimated that about 209 tons of municipal solid waste are produced annually in the United States, of which 60% is disposed by sanitary landfills.^[1] One of environmental concerns related to landfill is the discharge of leachate with high concentration of complex organic matters [e.g., chemical oxygen demand (COD) of 5000–20,000 mg/L] and ammonium-nitrogen (e.g., 3000–5000 mg/L), distinguishing it from municipal wastewater, which may cause some serious problems such as contaminations to ambient ground water, eutrophication of water body or odor release. Many methods that are currently used to treat leachate may have several drawbacks. For example, physical methods such as sedimentation,^[2] adsorption^[3] and membrane filtration,^[4] or chemical processes such as coagulation^[2] and chemical precipitation^[5,6] are commonly regarded as higher cost or lower effectiveness. Aerobic biological processes are effective for removing organic carbon but do remain inefficient due to large amounts of excessive sludge production, odor generation and high energy consumption.^[7] Anaerobic approach, for example, upflow anaerobic sludge blanket (UASB) system was reported to be able to remove COD from leachate with efficiency up to 71%–92%,^[8] but methane gas released during this process may contribute a lot to global warming.^[9] Thus, more economical pathways for efficient treatment of leachate remain necessary to be developed.

Recently, microbial fuel cells (MFCs) have been drawn increasing attentions in directly generating electricity from carbohydrates.^[10–13] Additionally, even domestic wastewater can also be used as substrate to generate electricity, with the realization of simultaneous wastewater treatment.^[14] MFCs that function organic wastewater as a result of electricity generation show some advantages in comparison with other wastewater treatment processes because they can (i) directly convert substrate energy to electricity with high conversion efficiency; (ii) operate at ambient, even low temperatures; (iii) produce less amounts of excessive sludge and thus result in the reduction of sludge treatment or disposal costs. Both dual-chamber and single chamber MFCs can be available for producing electricity from different kinds of organic matters. Among the MFCs reported until now, single-chamber air-cathode MFC showed the greatest potentials in practical applications due to its higher power generation, low cost and simple design.^[13,15] Several studies have demonstrated that a wide range of organic matters can be used as substrate to generate electricity in single chamber MFC such as glucose,^[13] acetate,^[16] butyrate^[16] and complex domestic wastewater.^[13] Leachate belongs to high concentration of organic wastewater, in which organic substances are possibly recovered into electricity in a MFC; however, there is no report on electricity generation from leachate until now.

In this study, the electricity generation from landfill leachate in MFC was investigated for the first time with the major contents as stated as follows. First, a lab-scale dual-chamber MFC was used to study the feasibility of generating electricity from leachate. Second, some tests were carried out mainly focusing on a single chamber MFC fed with a given concentration of leachate as the substrate. Third, cyclic voltammetry was plotted to examine the mechanisms of electron transfer on the anode. Fourth, the potential variation of the electrode at different COD concentration was studied. Last, the power generation, electron recovery as well as removal of COD as a function of leachate concentration in the single chamber MFC was respectively examined.

MATERIALS AND METHODS

MFC Apparatus

A dual-chamber MFC was constructed by connecting two plastic bottles (both total volume of 125 mL) with a column-shaped tube (length of 4 cm and diameter of 2 cm) containing proton exchange membrane (PEM, Nafion 112), clapped with two rubber loops. Preparations for PEM were performed based on the method as described by Bond and Lovley.^[12] The anode was made by filling particle activated carbon (average diameter from 0.5 mm to 2.0 mm, porosity of 0.44, China) into one bottle that was sealed by a rubber stopper with two drilled holes used for connecting the circuit and sampling respectively. A graphite rod was inserted into the carbon matrix to conduct electrons. The other bottle with the same size to the anode was used as the cathode and 50 mM of potassium ferricyanide (pH 7.0, adjusted by adding 100 mM NaH_2PO_4 as buffer solution) was used as electron acceptor. Copper wire was wrapped around the two graphite rods to connect the circuit through a rheostat (10–10,000 Ω).

The single chamber air-cathode MFC consisted of an anode and cathode sealed by rubber loop that was placed on the opposite sides of cylindrical chamber (13 cm² of sectional area, and 3 cm of length; 40 mL of empty volume) with sampling port on the middle top. The anode was made of Toray carbon paper (4 cm × 4 cm, effective area of 12.6 cm², E-TEK) without catalyst coated and was fixed on one side of MFC. Carbon cloth (effective area of 12.6 cm², 30% wet proofing, E-TEK) containing platinum catalysts (0.15 mg/cm², 5% Pt) was placed on the other side used as cathode. Connections between two electrodes were made with copper wire through a rheostat (10–1000 Ω).

MFC Incubations and Leachate Preparation

Anaerobic activated sludge produced during bioprocesses for wastewater treatments has been verified to be the best inoculums for MFCs.^[14,17]

Table 1: Mean values of physical-chemical parameters in leachate.

Parameters	Values	Parameters	Values
pH	6.8	Cu (mg/L)	0.08
COD _{Cr} (mg/L)	9800	Zn (mg/L)	2.6
BOD ₅ (mg/L)	4116	Fe (mg/L)	23
BOD ₅ /COD _{Cr}	0.42	Pb (mg/L)	0.056
TOC	2883	Cr (mg/L)	0.019
NH ₄ ⁺ -N (mg/L)	1182	Ca (mg/L)	460
Na (mg/L)	401	Mg (mg/L)	133

Activated sludge (MLSS of 3500 mg/L) was collected from the secondary clarifier of Wenchang Wastewater Treatment Plant in Harbin City, and then maintained in an anaerobic bottle of 1000 mL by adding glucose (2 g/L) once a week. Diluted anaerobic sludge (MLSS of 500 mg/L) was mixed with a medium (pH 7.0) containing glucose (COD of 500 mg/L), then pumped into the two reactors using a peristaltic pump to inoculate both MFCs.

Leachate was collected from Taiping Waste Landfill Plant (1 year old) in Harbin City, then stored in a glass bottle at temperature of 4° before each use. The characteristics of leachate were summarized in Table 1. Because heavy metals present in the leachate sample were very low or even not detectable, the effects of heavy metals on power output were not carried out. During the tests, leachate was mixed with nutrient solution containing NaH₂PO₄ (4.2 g/L) and Na₂HPO₄ (2.75 g/L), or in some cases leachate was diluted into lower COD concentration with tap water. All the experiments were conducted under the constant environmental conditions (temperature of 30° and pressure of 1.013 MPa). In order to compare the performance of two systems, both MFCs were operated under batch mode, with leachate replacement performed once the voltage output was below about 0.08 V at a given external load.

Calculations and Analyses

Measurements of voltage produced during experiments for long-term studies were recorded directly from potentiostat output every 60 seconds using a dual-channel voltage collection instrument (12 bit A/D conversion chips, US) connected with a personal computer via universal serial bus (USB, Intel) interface and calibrated with a digital multimeter (Agilent HP 34970, US) before each test. In order to compare electricity generation in different configuration of MFCs, voltage obtained was converted to power density P (mW/m³) based on reactor volume.^[18] Electron recovery in term of Coulomb efficiency (CE) was calculated on the basis of the approach reported by Liu et al.^[16] Internal resistance of MFCs was determined through linear regression deduced from Ohm law expressed as $U = V - Ir$, where U (V) was the voltage drop

at external load; $V(V)$ was the electromotive force; $I(A)$ was the current and r (Ω) was the internal resistance of the cell, which could be read from the slope of the regression line, respectively.^[15]

Cyclic voltammetry curve was introduced to study the redox reaction on the anode surface using a potentiostat (model 263a; Princeton Applied Research) connected to a personal computer (SoftCorr III; Princeton Applied Research) by detecting current response to the applied potential ranged from -600 mV to 400 mV at a scan rate of 5 mV/s. The anode was used as working electrode; the cathode was the counter electrode and the reference electrode was Ag/AgCl electrode (195 mV vs standard hydrogen electrode, SHE).^[16]

Measurements for working potential of the anode and the cathode were taken by a Ag/AgCl reference electrode connected to a multimeter (Agilent HP 34970, US). The working potential of the electrode was versus Ag/AgCl reference electrode (19 mV vs SHE). Chemical oxygen demand (COD) and ammonium-nitrogen were measured according to the standard methods.^[19] Metallic elements in leachate were directly analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES; model 5300 DV, Perkin Elmer Company, US).

RESULTS AND DISCUSSION

Electricity Generation from Leachate in Dual-Chamber MFC

Dual-chamber MFC with the anode and the cathode compartments filled with activated carbon as the electrodes was inoculated with anaerobic sludge and the prepared medium containing glucose (COD of 500 mg/L) at an external resistance of 5000 Ω . Incubation of the system took about 10 days (data not shown), and when voltage began to increase with each replacement of mediums, leachate without dilution (COD of 9800 mg/L) was fed into the MFC reactor, resulting in an instant increase of voltage and power output with the stable value of 0.398 V and 1602 mW/m³ respectively (Fig. 1). This demonstrated that it was possible to generate electricity not only from glucose,^[11,13] acetate, butyrate^[16] and municipal wastewater,^[14] but also from leachate.

In order to examine power output as function of current in dual-chamber MFC, polarization curve was made by varying external resistance from 1000 to 6000 Ω , once the repeatable power cycle was obtained. The maximum power density of 2060.19 mW/m³ based on the anodic liquid volume was obtained at current of 0.149 mA (external load of 1400 Ω) as shown in Figure 2. Although high power output could be achieved by increasing COD in the wastewater,^[20] such high strength of organic matters in leachate did not increase the power density substantially. This was mainly attributed to large internal resistance

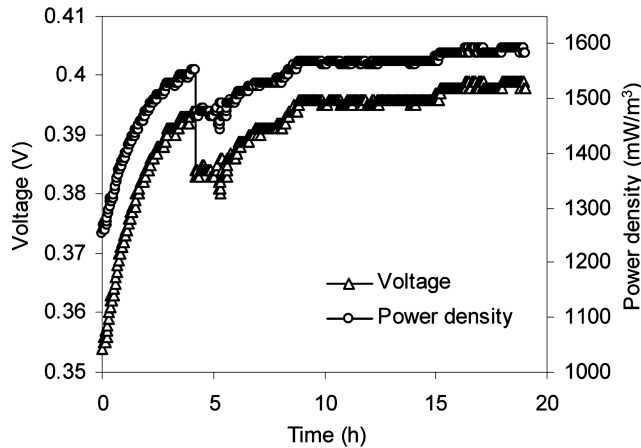


Figure 1: Voltage and power density generation with time when leachate was used as substrate in dual-chamber MFC (based on external resistance of 5000 Ω).

(up to 2100 Ω) in such system because of the use of proton exchange membrane (PEM) and mass transfer limitation in activated carbon MFC.

Electricity Generation from Leachate in Single Chamber MFC

In order to achieve higher power generation from leachate, another test was performed for the single chamber MFC fed with leachate. Following the inoculation of the MFC reactor with the same procedures as described to dual-chamber MFC, diluted leachate (1:5, COD of 1960 mg/L; $\text{NH}_4^+\text{-N}$ of 236.4 mg/L) was added into the reactor so that the reacting time of each cycle might be reduced. After feeding the leachate sample into the MFC, it could be observed that power output rapidly reached up to 4437.6 mW/m^3 within

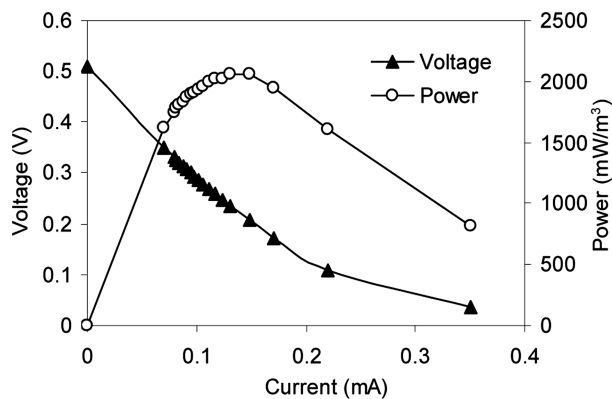


Figure 2: Power and voltage output as function of current in dual-chamber MFC.

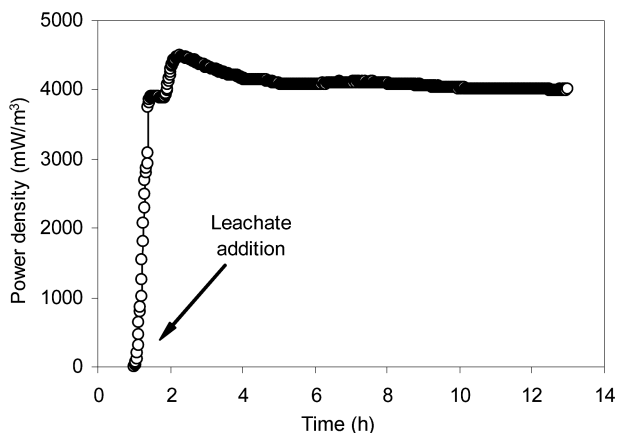


Figure 3: Voltage generation when leachate (COD of 1960 mg/L) used as substrate in single chamber MFC.

2 hours, and subsequently a stable power generation of 4166 mW/m^3 could be maintained on the basis of external resistance of 1000Ω (Fig. 3).

When the same leachate sample with COD of 1960 mg/L was applied into the single chamber MFC, the maximum power density reached 6817.4 mW/m^3 at an external resistance of 150Ω (current of 1.64 mA) (Fig. 4), which was almost 3.4 times higher than that obtained in the above-mentioned dual-chamber MFC. As shown in Figure 4, voltage decreased with electricity current increase in the circuit, then the internal resistance of the cell was determined to be 155Ω . This indicated that the maximum power generation was achieved when external load was approximately equivalent to internal resistance (approximate 150Ω). Such low internal resistance in single chamber MFC was recognized to be the main reason for higher power output over the dual-chamber MFCs (2060.19 mW/m^3 , seen the section above).

Cyclic Voltammetry

Cyclic voltammetry curve was plotted to examine the presence or the absence of electron mediators in the MFC during voltage generation (0.38 V , 1000Ω) and at the end of voltage generation (0.008 V , 1000Ω). During electricity generation, an obvious oxidation peak was found at -314 mV (vs SHE; $706 \mu\text{A}$) in the forward scan and an additional weak oxidation peak was found at -321 mV (vs SHE; $122 \mu\text{A}$) in the reverse scan (Fig. 5A). The potential at this two peaks was close to redox potential of -320 mV for NAD^+/NADH couple (pH 7.0). At the end of electricity generation, no active oxidation peak was observed during either forward or reverse scan (Fig. 5B). This partially suggested that the electrons released from electron donors were carried away from a respiratory enzyme at certain point during NAD^+/NADH redox process,

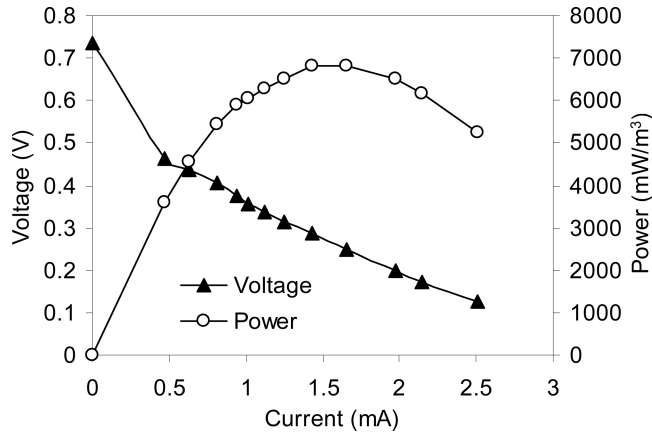


Figure 4: Power generation as function of current in single chamber MFC (leachate with COD of 1960 mg/L.)

and these respiratory enzyme were likely present in the biofilm, but not in the mixed solutions.

Performance of the Single Chamber MFC as Function of Leachate Concentration

Power Output. In order to determine the effect of substrate concentration on power generation from the single chamber MFC, the maximum power density on the basis of external load of 500Ω was measured when diluted leachate with a COD concentration range from 9.8 mg/L to 4900 mg/L was applied into the single chamber MFC. It was assumed that power generation in MFC could be written as the form of $P = P_{\max}S/(K_s+S)$, where P (mW/m^3)

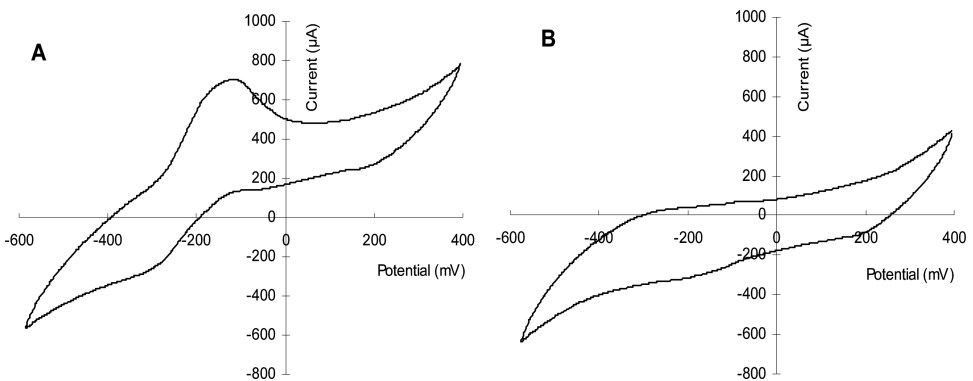


Figure 5: Cyclic voltammetry curve of the anode (A) during stable voltage output and (B) at the end of the electricity generation at COD 1000 mg/L in the single chamber MFC.

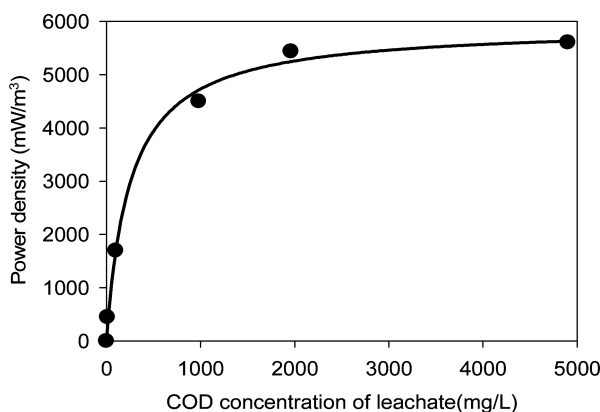


Figure 6: Power density as a function of COD concentration for leachate in single chamber MFC (on the basis of external resistance of 500 Ω .)

was the power output; P_{\max} (mW/m^3) the maximum power; S (mg/L) the COD concentration and K_s (mg/L) the affinity constant for substrate. Thus, the results revealed that power output followed a Monod-type relationship with respect to COD concentration of leachate with $P_{\max} = 5920.96 \text{ mW}/\text{m}^3$ and $K_s = 251.39 \text{ mg}/\text{L}$ ($R^2 = 0.9958$; $P < 0.001$ for t -test) as illustrated in Figure 6. The result obtained here was observed to be similar to previous literatures using glucose^[13] or acetate as substrate in a single chamber MFC.^[16]

Electrochemical Characteristic of the Electrodes

In order to study the effect of substrate concentration on the electrochemical performance of the electrodes in single chamber MFC, the anodic and cathodic potentials as function of external resistance at different initial COD concentration were compared after stable electricity generation. As illustrated in Figure 7A and 7B, when initial COD was kept at relative low level in the reactor (150 mg/L), changing external load from 0.002 $\text{k}\Omega$ to 5 $\text{k}\Omega$ in the circuit resulted in a decrease of anodic potential from -116 mV to -470 mV (vs Ag/AgCl electrode, by 305.2%); whereas the cathode potential increased from -85 mV to 91 mV (by 207.1%). In this case, biological kinetics related to the anode was proven to be the limiting factor for voltage output. In contrast, when leachate with initial COD 500 mg/L or 1000 mg/L was fed, slight difference in anode potential variation (both from about -446 mV to -492 mV , by 10.3%) was observed; however, cathode potential increased from -353 mV to 106 mV (by 130.0%) for COD 500 mg/L , and from -320 mV to 123 mV (by 138.4%) for COD 1000 mg/L . This demonstrated that substrate was sufficient and there was no more limiting factor for the anodic biological reactions at higher COD conditions, thus the voltage output mainly depended on the performance of

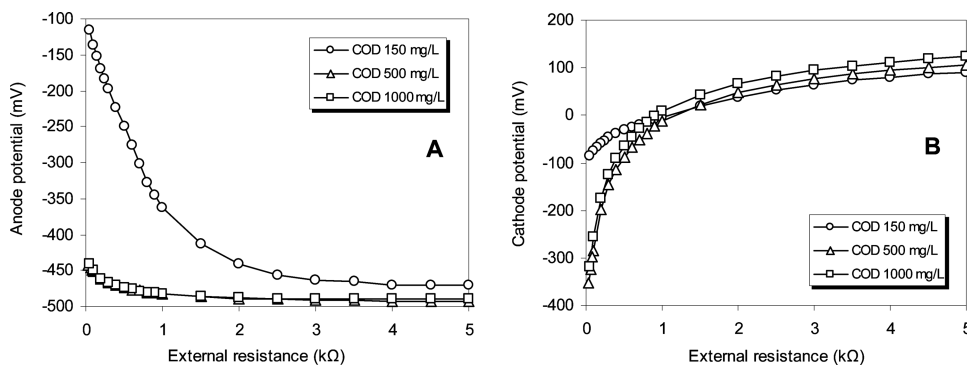


Figure 7: Working potential of the (A) anode and (B) the cathode as function of applied external resistance.

the cathode. A previous study^[21] indicated that the current was commonly limited by the anode in a dual-chamber MFC even substrate concentration in the anode chamber was high enough, and this was possibly ascribed that the anode and the cathode separated by membrane in a dual-chamber MFC was individual equipped. However in our case, a single chamber MFC without membrane allowed for direct substrate arrival at the cathode.

Electrons Recovery and COD Removal

Electron recovery can be described by the relative experimental coulombs to theoretical coulombs in term of Coulomb efficiency (CE). All the tests were carried out on the basis of external resistance of 500 Ω . As indicated in Figure 8, CE decreased from 21.2% to 3.4% when COD increased from 9.8 mg/L to 4900 mg/L for the single-chamber MFC. Similarly, Liu et al.^[15] also found the same reverse relationship between CE and substrate concentration. Such low electron recovery was believed to result from alternative respiration of organic matters by other community of aerobic or anoxic bacteria instead of electron-transfer bacteria, which could be verified by the difference of CE and COD removal. For instance, when initial COD of leachate was 1960 mg/L, only 6.63% of COD was converted into the form of electricity. In contrast, COD removal efficiency up to 69.54% (Fig. 8) suggested almost 60% loss of substrate in a non-electricity generation process. Furthermore, when COD in leachate was kept at relatively low level (less than 100mg/L), almost all the organic substances could be removed in the reactor (above 98%); but higher COD removal at high concentration of COD was limited by the insufficient amounts of bacteria in the system. Besides the examination of COD removal, ammonia-nitrogen elimination from leachate in the single chamber MFC was also examined. Experimental results showed that no nitrogen removal was obtained during the reaction period. For example, when the leachate with

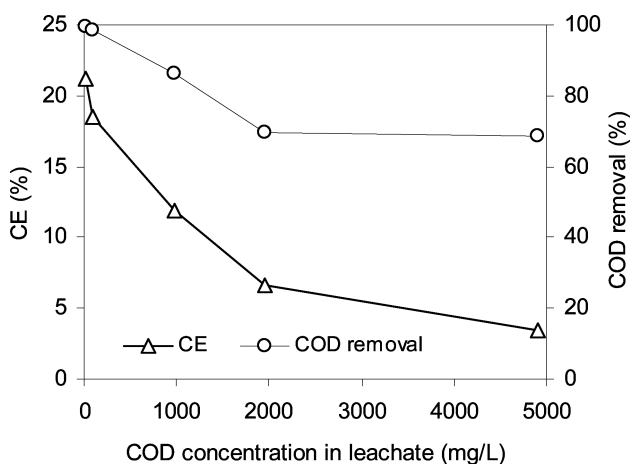


Figure 8: Electron recovery and COD removal as function of COD in leachate in single chamber MFC.

ammonia-nitrogen of 1182 mg/L was diluted into 236.4 mg/L, the ammonia-nitrogen in the treated effluent remained to be 235 ± 3.2 mg/L, demonstrating that no reaction associated with nitrogen removal such as simultaneous nitrification denitrification (SND) or anammox process^[20] took place in such a system. However, such high concentration of ammonia-nitrogen in leachate was not observed to affect electricity generation apparently in this study.

Applications of MFCs for Leachate Treatment

High concentration of organic matters present in leachate is undoubtedly to require high capital investment and consume a substantial amount of energy for its effective treatment. MFC, by which electricity can be directly generated from organic substances in wastewater, representing a fully novel process in reproducing energy from wastewater and reducing the overall wastewater treatment cost. Here, we for the first time showed that complex organic pollutants present in leachate could also be served as substrate for a dual-chamber or a single-chamber MFC to generate electricity. In the single chamber MFC, the maximum power density of 7000 mW/m^3 could be obtained with simultaneous COD removal in the range of 69.54%–98%, depending upon the initial COD concentrations.

In spite of the failure in removing ammonium-nitrogen from leachate, there is still a prospect for MFC to be used in leachate treatment for several reasons. Firstly, complex organic matters in leachate can be directly converted into electricity, which may provide a potential in the application for domestic or uptown electricity supply. Thus, a landfill plant can be possibly reconstructed to a small-scale power plant in the future. Although power obtained here is

relative low, but several breakthroughs have been made with an achievement of increasing power up to 90 W/m^3 .^[22] It is believed that these technologies can also be used to recycle electricity with high power output from leachate. Secondly, COD removal stands for partial treatment of leachate, mainly resulting from power conversion and anoxic biodegradation for organic matters in MFC reactor system. For certain, if necessary, MFC can be followed by some other treating processes such as activated sludge, biofilm units or physical-chemical facilities with the purpose of full treatment of leachate. Thirdly, in order to remove ammonium-nitrogen in leachate to meet the discharge standard, some pretreatment processes such as chemical precipitation of ammonium nitrogen as $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$,^[5,6] might be recommended as one of the most efficient methods in ammonium-nitrogen removal followed by MFC process to produce electricity.

Although MFC has its unique merits as stated above, there are still several problems in MFC application to practical leachate treatment. For instance, even if near 7000 mW/m^3 of power density could be obtained from leachate, much higher power generation was actually expected. This could be realized by developing the more efficient configuration of MFCs, improving the anode performance including biological factors and the materials, using alternative electron acceptors with higher redox potential, as well as reducing the internal cell resistance. Moreover, the problem of low electron recovery in single chamber MFC system should also be taken into account. Considering that aerobic biological respiration contributes a lot to substrate loss, the procedures available for increasing electron recovery should focus on the decrease of oxygen diffusion from the cathode, including reducing the cathode area, adding oxygen scavengers,^[23] changing the design mode of MFC or increasing the microorganisms amount in the anode chamber and so forth. Consequently, more efforts are needed for MFC to be applied in generating electricity from leachate within the not-too-distant future.

CONCLUSION

On the basis of experimental results obtained from the dual-chamber and single-chamber MFCs fed with landfill leachate, the following conclusions can be drawn: Electricity could be generated from leachate in the designed MFCs. The maximum power density of 2060.19 mW/m^3 for dual-chamber MFC and 6817.4 mW/m^3 for single chamber MFC were obtained, respectively. The difference in power generation for the two MFCs was mainly due to the difference of internal resistance. Power generation as function of organic substrate in single chamber MFC obeyed the Monod-type relationship with P_{\max} of 5920.96 mW/m^3 and K_s of 251.39 mg/L based on external load of 500Ω . Cyclic voltammetry suggested that the self-produced mediators of bacteria in solutions were absent and electrons were more likely transferred directly by

the bacteria in biofilms. Low COD concentration limited biological reactions on the anode and high COD concentration limited the chemical reactions on the cathode, respectively. COD in leachate could be eliminated when electricity was generated, but electron recovery in term of CE was relative low and decreased with COD because of substrate loss resulted from oxygen diffusion from the cathode. Removal of ammonium-nitrogen was not obtained in the single-chamber system. MFC showed a prospect in simultaneous leachate treatment and power generation but there remained further investigation into further increase of power generation.

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