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Continuous electricity generation with piggery wastewater treatment using an anaerobic baffled stacking microbial fuel cell

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ABSTRACT

Anaerobic baffled stacking microbial fuel cells (ABSMFCs) consisting of four individual MFCs with total volume of 6.4 L was constructed to generate electricity from piggery wastewater in present study. Anode materials (carbon paper, carbon fiber felt, and graphite granule) and anode connecting modes of the four MFCs (in series or parallel) could affect the bioelectricity generation and wastewater treatment effects. When they were connected in series, voltage loss occurred and voltage reversed with increase in current between some MFCs. The influent COD loadings showed significant relationship to the performance of ABSMFC, as it increased from 0.2 to 4.0 g/L d, voltage output across an external resistance of 1,000 Ω decreased by 71.7% (in series) and 30.7% (in parallel), respectively; coulombic efficiency decreased rapidly by 96.7% (in series) and 94.3% (in parallel), while COD removal efficiency initially increased and then decreased. This study demonstrated that the large volume ABSMFC can realize stable power output associated with piggery wastewater treatment and is suitable to scale-up for actual application.

Keywords: Anaerobic baffled stacking microbial fuel cells (ABSMFCs); Anode materials; COD removal; Piggery wastewater

1. Introduction

Microbial Fuel Cell (MFC) is an electrochemical device that can directly generate electricity from organic matters using micro-organisms as biocatalysts [1–3]. All biodegradable substrates, even agricultural waste, domestic, and industrial wastewater, can be used as fuels in MFC. A typical MFC consists of an anode chamber and a cathode chamber separated by a proton exchange membrane, namely two-chamber MFC, which usually produces low-power densities due to its high internal resistance and insufficient oxygen in the cathode. Single-chamber MFC is exploited by exposing the cathode directly to the air [3–5], which avoids the requirement of aeration and offers simpler and cheaper designs. This continuous flow, single-chamber and membrane-less MFC [2,6,7] is most favored for wastewater treatment and electricity generation because it is relatively easy to be scaled-up.

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Piggery effluent is one of the most highly produced wastewaters throughout the world [8], with high concentrations of particulates, organics, and nutrients [9]. The main removal methods of piggery wastewater include physicochemical methods and biological methods. Physicochemical methods are always parts of pretreatment but the effluent quality cannot reach the standard. Comparing with the former, biological methods are effective to treat piggery wastewater, while the process was complicate. In another aspect, MFC has great potential to treat wastewater and generate electricity simultaneously, while it has been rarely employed to treat piggery wastewater as it is newly developed [10].

Additionally, application of MFC for wastewater treatment with energy recovery is limited by several technical challenges, such as low-power densities due to energy utilization by bacteria, electrode over potentials, and high resistance. The treatment capacity is relatively lower as most laboratory MFCs are conducted in the level of several milliliters, which is too small for practical application. Thus, an MFC with great power output and large volume that can treat high-concentration piggery wastewater should be exploited.

On the other hand, MFC units can be stacked together or linked together in series, which can result in an additive increase in total voltage and treatment capacity [11], while anaerobic baffled reactor (ABR) [12,13] comprises a series of vertical baffles to force wastewater to flow under and over them, so that wastewater can flow through activated sludge beds in every compartment. Although the mass transfer efficiency may be limited without stirring, it can be compensated by sufficient contact between activated sludge and wastewater. Moreover, different microbial populations exist in different compartments, and the ABR system has better resilience to hydraulic and organic shock loadings and enhances the stability of the reactor during shock loadings. Li et al. [14] found that a steady increase in the feed loading from 4.8 to 18 kg COD/m^3 d did not significantly decrease the substrate removal efficiency in a three-compartment ABR system.

To combine the advantages of ABR and MFC, an anaerobic baffled membrane-less microbial fuel cell (ABSMFC) was constructed with four identical MFC units in present study to (1) scale-up the reactor of MFC, (2) investigate power output by connecting several MFC units in series and parallel connections, and (3) deal with piggery wastewater.

2. Data sources and methodology

2.1. ABSMFC configuration

The ABSMFC was in rectangular shape, consisting of four compartments, which were subdivided into down-flow and up-flow sections by a series of vertical high/low baffles (5 mm thick). The down-flow and up-flow sections are of 2 cm and 8 cm wide, respectively. The baffles forced wastewater to flow over and under as it progressed through the reactor. The baffles had a 45 turnout angle and 5 mm separation from the base of the reactor (Fig. 1), which diverged the main flow to the center of the up-flow section. Each compartment represented a MFC unit, which was an air-cathode MFC. To optimize the suitable electrode materials for the ABSMFC, four different composing modes of carbon paper (Toray company, Japan), carbon fiber felt (3 mm thick, Beijing Evergrow Resources Co., Ltd, China), graphite granules (diameter between 3 and 5 mm, Beijing North Xin Yuan Electrical Carbon Products Co., Ltd, China), and proton exchange membrane (PEM, Nafion 112, DuPont, USA) were tested in this study. Whatever carbon material without chemical catalyst was used as anode. The carbon paper cathodes were made of wet proofed carbon paper with a surface loading of 0.5 mg Pt/cm^2 (Beijing LN-Power Sources Co., Ltd, China) and a surface area of 20 cm^2 in all four modes. Mode 1, carbon paper anode of 25 cm^2 without graphite granules in the anode chamber and the carbon paper/PEM cathode manufactured by bonding the PEM directly onto the carbon paper cathode (Beijing LN-Power Sources Co., Ltd, China). Mode 2, carbon paper anode without graphite granules in the anode chamber with PEM-less carbon paper cathode. Mode 3, carbon fiber felt anode without graphite granules in the anode chamber and PEM-less carbon paper cathode. Mode 4, carbon fiber felt anode with graphite granules in the anode chamber and PEM-less carbon paper cathode. In the ABSMFC, cathodes were pressed on the same side of four compartments and the distance between anode and cathode was about 2 cm. The total volume of four anode chambers was 6.4 L, and the available volume was about 4.8 L. Four sample ports were placed on the top of each reactor at the identical location.

2.2. Operating conditions

The ABSMFC was inoculated with mixed anaerobic sludge and operated under room temperature of 18–22˚C. Two types of anaerobic sludge were obtained

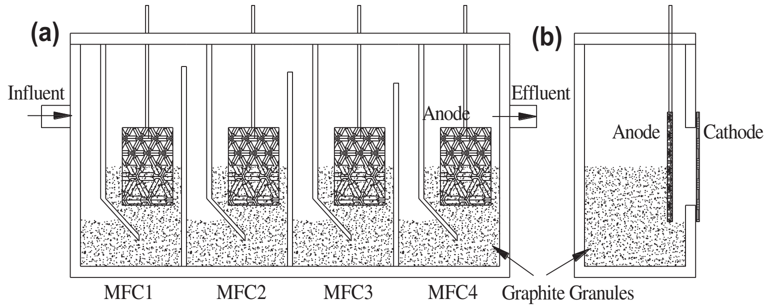

Fig. 1. Schematic prototype of the anaerobic baffled stacking MFC. (a) Transverse section and (b) vertical section.

from anaerobic digester of Beijing Wastewater Treatment Plant (China) and starch wastewater anaerobic treatment reactor, respectively. The piggery wastewater used in this study as fuel was obtained from a septic tanks at a large pig farm in the suburbs of Beijing, China. The supernatant liquid of the septic tank consisting fresh urine, dung, and rinse water was chosen as the experiment influent, with COD concentrations of 3,998 ± 13 mg/L. The raw wastewater was diluted to form different organic loading rates when necessary. As the voltage output reached stable, piggery wastewater was continuously pumped into the first compartment, and then flowed to the next three compartments one by one. It was at a flow rate of 3.1 mL/min, with hydraulic retention time (HRT) of 24 h and different influent COD loading rates of 0.2–4 g/L d. Copper wire was used to connect the circuit with an external load of 1,000 Ω for each MFC unit, as employed in previous research for molasses wastewater treatment in similar ABSMFC [15].

2.3. Analysis and calculations

Voltage, soluble chemical oxygen demand (SCOD), and pH were monitored in this study. Voltage was measured using a data acquisition system (PMD-1608LS, MCC Corporation, USA), which can be transferred to the computer at an interval of 10 min. All samples were filtered through a 0.45-μm pore diameter syringe filter to remove bacteria and suspended solid before measuring SCOD. SCOD was measured using Standard Methods (HACH COD system, HACH Company, Loveland, CO.) and pH was measured using pH 201 (Hanna, Italy). Power density (P, mW/m^2), was obtained according to:

$$P = IV/A \tag{1}$$

where I (A) is the current, V (V) is the voltage, and A (m^2) is the projected surface area of the cathode, as showed in previous study [15].

The Coulombic efficiency was calculated as:

$$E_c = \left(C_p/C_{Ti}\right) \times 100 \tag{2}$$

where C_p (C) is the total coulombs calculated by integrating the current over time. C_{Ti} (C) is the theoretical amount of Coulombs that can be produced from either wastewater ($i = w$) or glucose ($i = g$), calculated as:

$$C_{Ti} = Fb_iS_iv/M_i \tag{3}$$

where F is the Faraday's constant (96,485 C/mol of electrons), b_i is the number of mol of electrons produced per mol of substrate ($b_w = 4$, $b_g = 24$), S_i (g/L) is the substrate concentration, v (l) is the liquid volume, and M_i is the molecular weight of the substrate ($M_w = 32$, COD basis; $M_g = 180$). For continuous flow through system, E_c can be calculated on the basis of current generated under steady conditions as:

$$E_c = M_iI/Fb_iS_iQ \tag{4}$$

where Q (L/d) is the volumetric influent flow rate. The maximum power density can be obtained from the polarization curve which was determined by varying the external resistance over a range of 20,000–10 Ω (in disconnection) or 20,000–1 Ω (in series and parallel connections).

3. Results and discussion

3.1. Voltage generation from ABSMFC with carbon paper anode and carbon fiber felt anode

When different composing modes of electrode materials were studied, the bioreactor was fed with piggery wastewater (800 mg/L) continuously, while the OCV of four MFCs were detected by the data acquisition system. As the OCV approached stable, an external resistance of 1,000 Ω was connected in each closed circuit. The voltages across the external resistance of four MFCs were shown in Fig. 2. Data showed that the average voltage was only stable at 12.2 mV in Mode 1 which was almost 47 times less than the voltage output of traditional single-chambered air-cathode MFC (570 mV) [16], indicating that the internal resistances of four MFCs were much higher than others [16,17]. In Mode 2, the PEM-less carbon paper cathodes were used to reduce internal resistance, but it only resulted in an insignificant increase of average voltage to 34.2 mV. This suggested that the internal resistance was not mainly attributed to the resistance to the ions through PEM. In Mode 3, as soon as the carbon paper anodes were replaced by carbon fiber felt, the average voltage increased abruptly to 210 mV. In Mode 4, four anode chambers were filled with graphite granules to increase power output of the ABSMFC (Fig. 2). Data showed that the average voltage increased rapidly to 319.8 mV, and it reached constant at around 400 mV after eight days. It could be concluded that the internal resistances of four MFC units had been reduced greatly and consequently increased the power output.

Microbes in the inoculated sludge mainly contained Firmicutes, Chlorobi, and Proteobacteria as discovered in our previous with the similar sludge, and the electrochemically active bacteria mainly classified as Proteobacteria [18]. The sludge tended to settle at the bottom due to large volume of the reactor. On the other hand, the carbon paper was parallel to the cathode in the middle part of up-flow sections, resulting in insufficient electrical contact between suspended bacteria and anode. Therefore, time required for biofilm establishment was prolonged which consequently decreased the electron transfer efficiency. As the carbon fiber felt and graphite granules worked as anode, the improvement in power generation could be a consequence of high surface area carbon fiber felt which increased the adhesion of bacteria on anode surface and the presence of graphite granules which can facilitate electron transfer. Previous study also indicated that carbon fiber felt and graphite granules accumulated more electrochemically active bacteria than common carbon paper [17]. The specific bacterial communities on the selected anode in present study could be further investigated afterwards. When the ABSMFC was operated in continuous mode, the direct water flow to the anode can increase power output since the oxygen diffusion into the anode chamber can be restricted, which has been demonstrated by Cheng et al. [19].

The internal resistances of four MFCs in Mode 3 and Mode 4 were much lower than those in Mode 1 and Mode 2 as reflected from the voltage outputs under the same external resistance. It is known that the voltage of MFC is controlled by numbers of factors including activation polarization loss, ohmic loss, and concentration polarization. Activation losses can be reduced by increasing the electrode surface area and the establishment of enriched biofilm on the electrode [1]. Carbon fiber felt can increase the pore volume, surface area, and the internal aperture of porous electrodes which can increase biomass on the anode and decrease the anodic internal resistance. On the other hand, the graphite granules in the anode chamber could complement the electrical contact between bacteria and anode in large volume reactor. As a result, carbon fiber felt with graphite granules in the anode chamber is capable of being scaled-up and used in larger size MFC reactors.

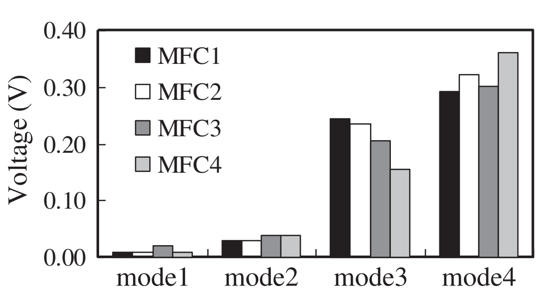

Fig. 2. Voltage outputs under different modes. *X*-axis: composing modes of electrodes. *Y*-axis: voltage outputs (external resistance was 1,000 Ω).

3.2. *Polarization curves of four individual MFCs with carbon fiber felt anodes*

With a flow rate of 3.1 ml/min and influent COD concentration of 2000 mg/L, polarization curves (Fig. 3) can be obtained by varying the circuit external resistance between 20,000 and 10 Ω. The maximum power densities of each MFC (from MFC 1 to MFC 4) were 165.3, 160.0, 164.7, and 297.6 mW/m^2 (normalized by the carbon paper cathode projected surface area of 20 cm^2), respectively. The corresponding currents were 524.8, 632.5, 641.8, and 996.0 mA/m^2, respectively. As the value of the internal resistance of MFC is the slope of polarization curve, the internal resistances of four individual MFCs were determined (Table 1) and the results showed that the increase in power output was consistent with an overall reduction in internal resistance.

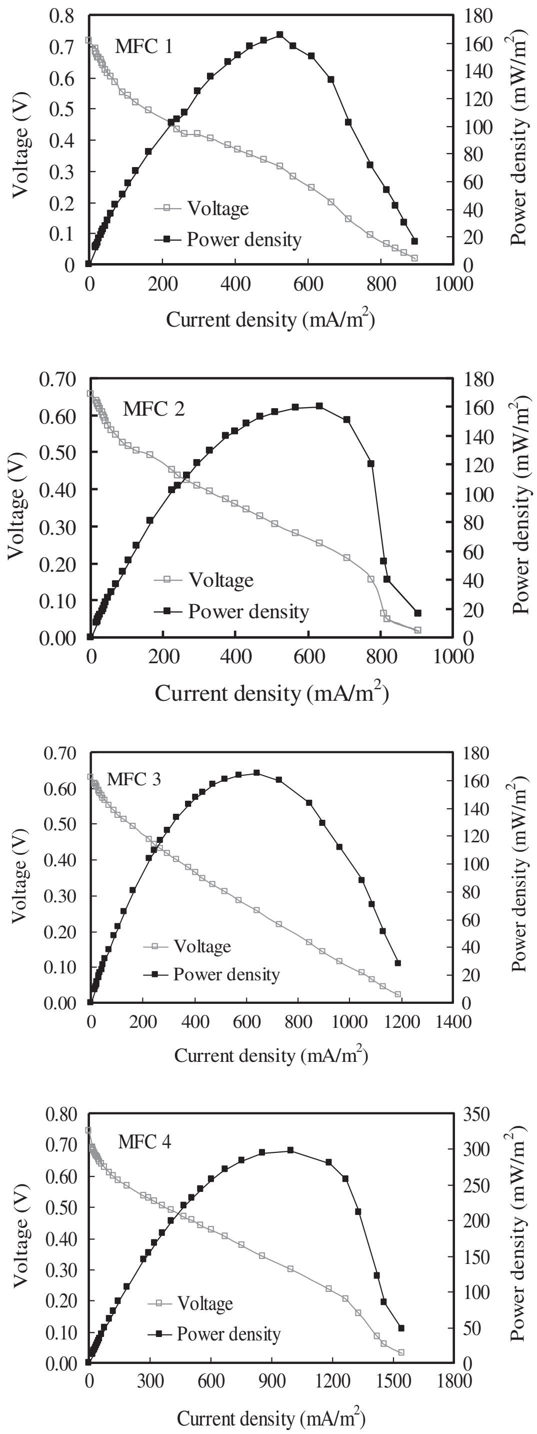

Fig. 3. Polarization curves of four individual MFCs at the influent COD loading rate of 2.0 g/L d.

3.3. Power output by series connection and by parallel connection

Four MFCs were connected in series to increase the overall voltage and in parallel to increase current, respectively. With the influent COD loading rate of 2.0 g/(L d), the total OCVs were 1.963 V in the case of series connection and 0.683 V for parallel connection. However, the individual OCV of four MFCs (from MFC1 to MFC4) were 0.625, 0.459, 0.417, and 0.439 V in series connection lower than the values (0.723, 0.654, 0.635, and 0.742 V, respectively) of four individual MFCs when they were disconnected. Thus, the actual total OCV was much less than theoretical value 2.754 V and a great voltage loss occurred in series connection, which was mainly due to the simple cells connection by a copper wire. The point contact between the electrode plate and the copper wire causes non-uniform potential distribution on the electrode surface, and potential drop occurs between an anode in one single cell and a cathode in the other single cell [20]. The average power output of four individual MFCs was 178.1 mW/m^2 (698.8 mA/m^2) while they were 160.1 mW/m^2 (182.5 mA/m^2) and 195.4 mW/m^2 (736 mA/m^2) in series and parallel connections, respectively. According to the polarization curves, the internal resistances were 724.1 Ω (series) and 61.4 Ω (parallel) at the influent COD loading rate of 2.0 g/(L d). With different influent COD concentrations, the power densities of parallel connection were higher than series connection, which was mainly due to the low internal resistance for the case of parallel connection. Though the stacked MFCs will not produce higher power densities than the individual MFCs, an average power at more practical voltage could be achieved by combining series and parallel connections.

3.4. Voltage reversal in series connection

As four individual MFCs were connected in series, the anode of first cell was connected to the cathode of the second cell using copper wire. At all influent COD loading rates, polarization curves of the stacked MFC were obtained by varying the circuit resistance between 20,000 and 1 Ω. When current increased at an influent COD loading rate of 0.2 g/(L d), voltage of MFC1 reversed at 150 Ω of external resistance while MFC4 reversed at 45 Ω (Fig. 4(a)). At higher influent COD loading rate of 4.0 g/(L d), voltage reversal remained in MFC 2 and MFC 3 (Fig. 4(b)). Data showed that voltage reversal occurred in different MFCs at different influent COD loading rates (0.2, 1.0, 2.0, and 4.0 g/(L d)) and it occurred as current increased. First, at lower resistance, the electrons move through the closed circuit more easily than at higher resistance and more oxidation of substrates at the anode is expected. But at low-loading rate of influent

Table 1
Internal resistances (R_i) of four MFCs at different influent COD loading rates

COD loading rate (g/L d)	R_i of MFC1 (Ω)	R_i of MFC2 (Ω)	R_i of MFC3 (Ω)	R_i of MFC4 (Ω)
0.2	197.5	318.6	424.2	143.1
1.0	174.8	169.7	297.3	176.12
2.0	357.7	321.7	249.3	197.6
4.0	692.7	669.7	989.5	469.0

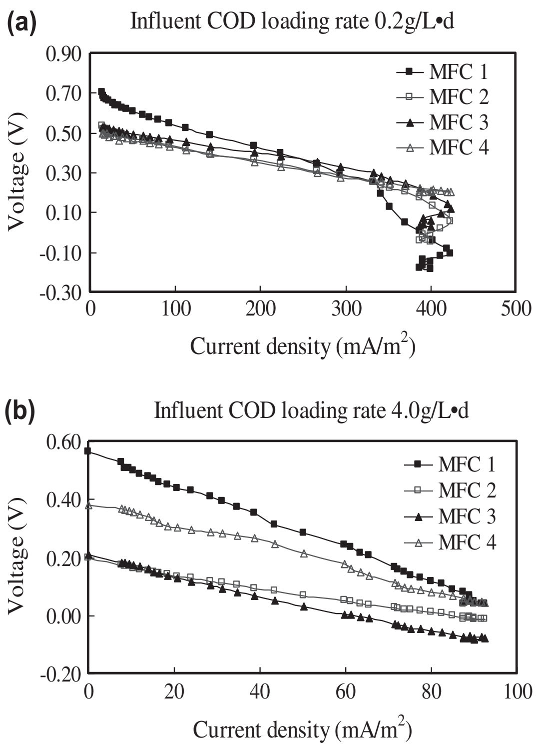

Fig. 4. Voltages of four individual MFCs in series connection by lowering the external resistance from 20,000 to 1 Ω.

COD (such as 0.2 g/(L d)), organic matter is insufficient and the microbes will suffer starvation. In this case, excessive current from a MFC unit at a rate higher than its fuel delivery supports will result in an increase of anode potential and lead to voltage reversal [21]. Second, although the influent COD loading rate was as high as 4.0 g/(L d) and the fuel delivery was adequate, cell reversal still occurred in MFC2 and MFC3 at higher currents. It was most likely due to poor performance at low pH value and limited catalytic substrate conversion efficiency of some electricity-producing microbial consortia. Third, voltage reversal may be involved with the internal resistance of individual MFCs. Data showed (Table 1) that the MFCs that suffered voltage reversal mostly have the maximum internal resistance. Moreover, both anode and cathode potentials of each MFC were also monitored and they kept relatively stable during the operation, due to the hydrodynamic isolation of each MFC unit.

3.5. *Influence of organic load on electricity generation from ABSMFC units*

After 150-d incubation, the power output of ABSMFC approached stable, the influence of COD loading rates on the performance of the ABSMFC was studied. First the reactor was fed with tap water for 15 d to reduce the interference of remained substrates from the previous operation. Then the influent COD loading rate varied from 0.2 to 4.0 g/(L d) at a constant hydraulic retention time of 24 h and an external resistance of 1,000 Ω in series and parallel connections. When a stable COD removal efficiency of 60% was achieved, the influent COD loading rate was increased. Fig. 5 showed the results of voltages in series and parallel connections at different influent COD loading rates indicating that the influent COD loading rates had significant influence on power yield, i.e. the higher COD was, the poorer performance of the cell was. The voltages decreased noticeably with increased

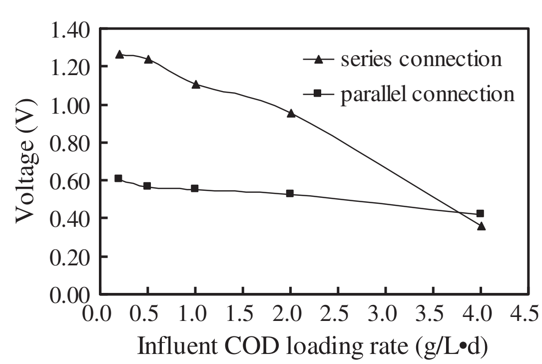

Fig. 5. Influence of the COD loading rates on the stacking voltages in series and parallel connections (external resistance was 1,000 Ω).

COD concentration both in series and parallel connections, which was not consistent with the previous observation [22] in a single-chamber air-cathode MFC.

The poor performance of ABSMFC would be due to the decrease of pH with increased influent COD loading rates. The pH values of influent, four MFC compartments, and effluent of ABSMFC in series connection were measured and shown in Fig. 6. Data showed that the pH in each MFC compartment decreased with increased concentration of COD. At each COD concentration, MFC1 had the lowest pH value and it increased gradually from MFC2 to MFC4 and the effluent and its variation trend was same as the pH variation trend in batch mode same as the function of time [23]. Mohan et al. [24] showed that variation of pH values was consistent with the variation of volatile fatty acids (VFA; represented as the total of all acids generated during acidogenic fermentation step), which may lower the voltage generation in the case of high concentration of VFA. As the COD loading rate was increased, VFA accumulation that contributed to pH decrease occurred in the reactors, especially in MFC1. It is known that each microbial consortium engaged in anaerobic degradation can develop in a specific pH range and have an optimum pH. As shown in Fig. 5, when the influent COD loading rate was 4.0 g/(L d), pH of four MFC compartments was below 6, and there was a rapid decrease in voltage in series connection across an external resistance of 1,000 Ω. This result was similar to Gil's study [25] which stated that the optimal pH value of electricigens was 7 and the power output will significantly decrease when pH was below 6 and above 9. At pH 6, the microbial activity of electricigens was inhibited, and much less electrons and protons were produced, resulting in lower voltage output. On the other hand, according to Eq. (4) (take acetate oxidation as example):

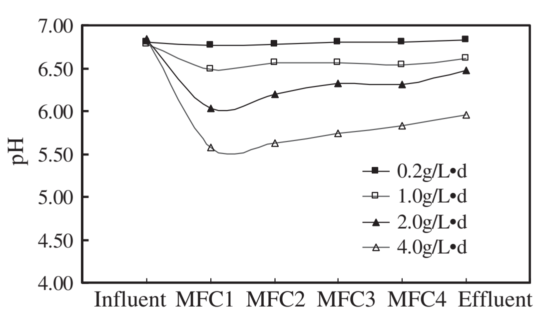

Fig. 6. Variation in pH of influent, four MFC compartments, and effluent at different influent COD loading rates.

$$E_{\mathrm{An}} = E^0_{\mathrm{An}} - \frac{RT}{8F}\ln\left(\frac{[\mathrm{CH_3COO^-}]}{[\mathrm{HCO_3^-}]^2[\mathrm{H^+}]^9}\right) \tag{5}$$

the anode potential of four MFC units will increase with the decreased pH, leading to a decrease in the voltage output of MFC. pH should be adjusted and VFA should be consumed during the following operation to maintain higher electricity generation as well as organic removals.

3.6. SCOD removal and coulombic efficiency

Soluble chemical oxygen demand (SCOD) removal efficiency and CE, as indicators for substrate degradation and electron recovery, were calculated to determine the relationship between organic substrate and bioelectricity generation (Table 2). All tests were conducted with an external resistance of 1,000 Ω in series and parallel connections. As shown in Table 2, CE decreased from 1.21 to 0.04% in series connection and from 0.7 to 0.04% in parallel connection as the COD loading rates increased from 0.2 to 4 g/(L d). The highest SCOD removal efficiency was obtained at the loading rate of 0.5 g/(L d) both in series and parallel connections. Because of low internal resistance, the parallel connection produced higher SCOD removal efficiency than series connection, which was consistent with other studies [7,26]. Several factors may be responsible for such low CE of the ABSMFC both in series and parallel connections. First, oxygen transfer into the anode chamber through the cathode was believed to be a consequence of PEM removal, which may induce alternative respiration of organic substrates by other community instead of electron-transfer bacteria. Second, large volume of the reactor resulted in insufficient contact between bacteria and electrode, and long distance for electron transfer and proton diffusion. Third, the difference of low CE and high SCOD removal efficiency indicated that the majority of SCOD was biodegraded by methanogens or other microbes. It could be attributed to the fact that the anode chambers of four MFCs acted as an anaerobic baffled reactor for treating wastewater, and some bacteria presented in the sludge without contacting with the anode could degrade a portion of SCOD through fermentation. Considering practical application, methane inhibitors can be added to the ABSMFCs to restrain methane production for limitation of anaerobic digestion thus, obtaining higher CE.

The COD removal efficiency showed similar trend with CE as COD concentration increased (over

Table 2
Influence of COD loading rate on COD removal efficiency and coulombic efficiency (external resistance was 1,000 Ω)

COD loading rate (g/L d)	COD removal efficiency (%)		Coulombic efficiency (%)	
	Series connection	Parallel connection	Series connection	Parallel connection
0.2	78.80	81.04	1.21	0.70
0.5	84.05	86.30	0.57	0.22
1.0	76.70	82.53	0.22	0.10
2.0	68.02	70.96	0.11	0.06
4.0	42.70	50.40	0.04	0.04

0.5 g/(L d)), which may be a consequence of insufficient amounts of bacteria in the anode chamber. Another possible reason was that the high COD loading rates may lead to pH decrease, which would inhibit the microbial activity including electron-transfer bacteria. At the loading rate of 0.5 g/(L d), the ABSMFC was operated in series connection for three months and stable power outputs as well as effective COD removals were obtained, implying the ABSMFC could be scaled-up for practical application.

4. Conclusions

An ABSMFC containing four individual MFCs with a total volume of 6.4 L was developed to produce electricity. The ABSMFC could successfully generate electricity from piggery wastewater with carbon fiber felt as anode when anode chambers were filled with graphite granules. The average maximum power generation of four individual MFCs was 178.1 mW/m^2 while they were 160.1 mW/m^2 in series and 195.4 mW/m^2 in parallel at a COD loading rate of 2.0 g/(L d). Although stacked MFCs could not increase power density, it could enhance the overall voltage in series and current output in parallel. The voltage loss was significant in series connection because of the simple cells connection by a copper wire. Voltage reversal occurred in some MFCs with increase in current at all influent COD loading rates of this study, which may be a consequence of fuel starvation, poor performance of electricity bacteria, different internal resistance, and so on. The voltage output across an external resistance decreased with increased influent COD loading rate, which may be resulted from the poor microbial activity at low pH. Therefore, future studies should explore means to control voltage reversal in series connection, and select enriched mixed inoculum adapted to acidophilic conditions. In summary, the ABSMFC has great potential for practical application for effective electricity production from wastewater treatment in the future.

Acknowledgments

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