

# Enhanced Coulombic efficiency and power density of air-cathode microbial fuel cells with an improved cell configuration

Yanzhen Fan, Hongqiang Hu, Hong Liu\*

Department of Biological and Ecological Engineering, Oregon State University, 116 Gilmore Hall, Corvallis, OR 97331, USA

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## Abstract

Single chamber air-cathode microbial fuel cells (MFCs) that lack a proton exchange membrane (PEM) hold a great promise for many practical applications due to their low operational cost, simple configuration and relative high power density. One of the great challenges for PEM-less MFC is that the Coulombic efficiency is much lower than those containing PEM. In this study, single-chamber PEM-less MFCs were adapted by applying a J-Cloth layer on the water-facing side of air cathode. Due to the significant reduction of oxygen diffusion by the J-Cloth, the MFCs with two-layers of J-Cloth demonstrated an over 100% increase in Coulombic efficiency in comparison with those without J-Cloth (71% versus 35%) at the same current density of  $0.6 \text{ mA cm}^{-2}$ . A new cell configuration, cloth electrode assembly (CEA), therefore, was designed by sandwiching the cloth between the anode and the cathode. Such an MFC configuration greatly reduced the internal resistance, resulting in a power density of  $627 \text{ W m}^{-3}$  when operated in fed-batch mode and  $1010 \text{ W m}^{-3}$  in continuous-flow mode, which is the highest reported power density for MFCs and more than 15 times higher than those reported for air-cathode MFCs using similar electrode materials. This study indicates that the Coulombic efficiency and power density of air-cathode MFCs can be improved significantly using an inexpensive cloth layer, which greatly increases the feasibility for the practical applications of MFCs.

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**Keywords:** Microbial fuel cell; Coulombic efficiency; Power density; Cloth electrode assembly

## 1. Introduction

Microorganisms in microbial fuel cells (MFCs) can utilize a diverse range of environmental fuels, such as sugar [1,2], protein [3], organic matters in ocean sediment [4], even human and animal wastes [5,6] to generate electricity. This provides MFCs great application potentials in the following areas: (1) renewable energy generation from biomass, (2) wastewater treatment and energy recovery, and (3) power supply for sensors using indigenous fuels. However, increasing the Coulombic efficiency (CE) and volumetric power density of an MFC and simplifying its design are critical for the successful applications of this technology [7].

The single chamber air-cathode MFC [5,8] provides great advantages over the two chamber system for practical applications because of the following reasons: (1) passive air can

be used thus no aeration is needed, (2) no recycle or chemical regeneration of catholyte is required thus the operation is simplified, and (3) smaller cell volume, thus higher volumetric power density, can be easier to achieve. Air-cathode MFCs that lack a proton exchange membrane (PEM) hold a greater promise due to their low cost, simple configuration, and relatively high power density [9]. However, there are two major challenges for a membrane-free MFC. One is that its CE is much lower than that of MFC containing a membrane when a mixed culture is used due to the consumption of substrate by oxygen diffused through the cathode [9]. The other is that the anode and cathode distance in a membrane-free MFC is limited to a certain range (about 1–2 cm) due to the potential negative effect of oxygen on the activity of the anaerobic bacteria on the anode [10,11] and the risk of short circuit. Therefore, it is of great interest to develop new membranes that can reduce the oxygen diffusion without greatly affect the internal resistance or power density.

In this study, cloth was applied between the electrodes to reduce the oxygen diffusion rate while maintaining the high

\* Corresponding author. Tel.: +1 541 737 6309; fax: +1 541 737 2082.

E-mail address: [liuh@engr.orst.edu](mailto:liuh@engr.orst.edu) (H. Liu).

power density of MFCs. J-Cloth was used as an example to study the effect of different cloth layers on the performance of MFCs. The MFCs with a cloth electrode assembly (CEA) structure were also constructed and evaluated under both fed-batch and continuous-flow operations. Significantly increased CE and volumetric power density have been achieved in such MFCs.

## 2. Materials and methods

### 2.1. MFC construction

The MFCs with different anode and cathode spacing were constructed using plastic (Plexiglas) cylindrical chambers as the main bodies following a previous design [9]. The anode electrode was made of type A carbon cloth (no wet proofing; E-Tek, USA). The cathode was made of type B carbon cloth (30% wet-proofing; E-Tek, USA) and treated following the procedures reported previously [12] to achieve a  $0.5 \text{ mg cm}^{-2}$  Pt loading. Polytetrafluoroethylene (PTFE) layers were coated on a carbon/PTFE base layer to the air-side of the cathode to reduce the water loss through the cathode [12]. In some of MFCs, the anode and cathode (3 cm in diameter) were placed on opposite sides of the chamber at a distance of 1.7 cm with 1–3 layers of J-Cloth (Associated Brands LP.) on the water-facing sides of the cathodes (Fig. 1B). The J-Cloth does not contain any antibacterial coating and is about 0.3 mm in thickness. The MFCs without J-Cloth but same cell structure (Fig. 1A) were used as control. In the other MFCs, J-Cloth (2 layers) was sandwiched between the anode and the cathode at one end (Fig. 1C) or both ends (Fig. 1D) of the cylindrical chamber. Similar to membrane electrode assembly (MEA), we refer this type of configuration as cloth electrode assembly (CEA).

### 2.2. MFC test

The MFCs were inoculated with a mixed bacterial culture from the anode of a single chamber MFC, which was originally inoculated with domestic wastewater (Corvallis Wastewater Treatment Plant, Corvallis, OR) and has been operated for about 1 year. Acetate (20–30 mM) was used as the substrate and the

medium solution contained the following (per liter):  $\text{NH}_4\text{Cl}$ , 0.31 g;  $\text{KCl}$ , 0.13 g;  $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$ , 5.84 g;  $\text{Na}_2\text{HPO}_4 \cdot 7\text{H}_2\text{O}$ , 15.5 g; and mineral (12.5 ml) and vitamin (12.5 ml) solutions as reported by Lovley and Phillips [13].

A series of experiments were conducted to study the effects of different (1, 2, 3) layers of J-Cloth applied to the water-facing side of cathode on the performance of MFCs using cells with 1.7 cm electrode spacing (Fig. 1B). The performance of MFCs with single CEA (Fig. 1C) and different empty bed volumes (3 ml and 6 ml), as well as double CEAs (Fig. 1D) and 2.5 ml volume, were also evaluated. Cell voltages were measured at various external resistances from  $1000 \Omega$  to  $25 \Omega$  in a decreasing order. For batch experiments, MFCs ran for at least two batches at each resistance to ensure repeatable power output can be achieved. The MFC with double CEAs was also continuously operated in an up-flow mode at a constant flow rate of  $0.6 \text{ ml min}^{-1}$  maintained through a peristaltic pump (Master-Flex 7550-10, Cole-Parmer Instrument Co.) to further evaluate the performance of the CEA-MFC.

Polarization curves were made by using hourly averaged voltages at the stable stage of each batch. Electrode potentials were measured by using the multimeter with a data acquisition system (2700, Keithly, USA), and a reference electrode ( $\text{Ag}/\text{AgCl}$ ; RE-5B, Bioanalytical systems, USA) located in the middle of the fuel cell chamber. Experiments were conducted under constant temperature of  $30^\circ\text{C}$  in duplicate.

### 2.3. Analyses

Voltage ( $V$ ) was recorded using a multimeter mentioned above, and used to calculate volumetric power densities based on empty bed volumes and surface power densities based on projected surface area of electrodes. The projected surface area of all the MFCs was  $7 \text{ cm}^2$  except MFCs with double CEAs (Fig. 1D), which was  $14 \text{ cm}^2$ .

CE was obtained by calculating the ratio of total recovered coulombs by integrating the current over time to the theoretical amount of coulombs that can be produced from acetate. Detail information can be found in a previous report [9].

The internal resistance of MFCs operated in batch mode,  $R_{\text{int}}$ , was calculated from the slope of plots of voltage ( $V$ ) versus

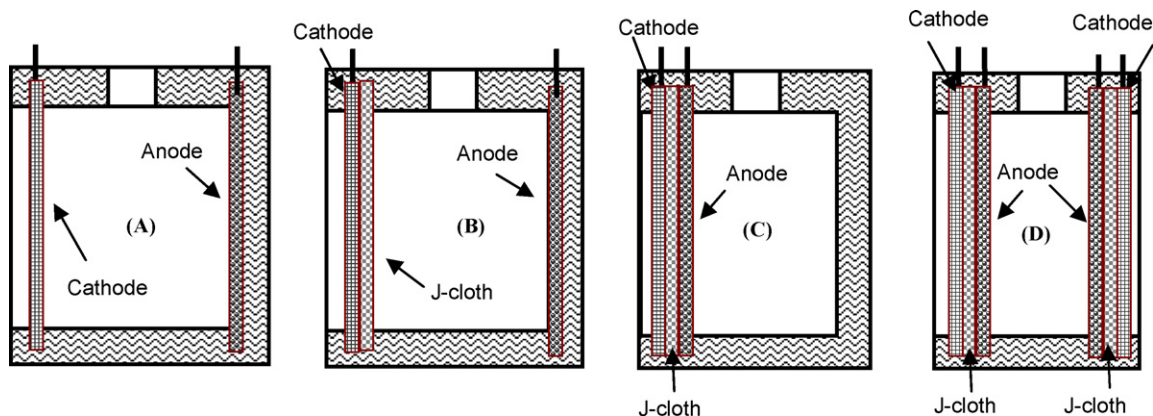


Fig. 1. Schematic of MFCs (A) without J-Cloth, (B) with J-Cloth, (C) with single CEA (cloth electrode assembly), and (D) with double CEAs.

current ( $I$ ) using:

$$V = E_{\text{cell}} - IR_{\text{int}} \quad (1)$$

where  $E_{\text{cell}}$  is the electromotive force of the cell.

The internal resistance of MFC in continuous-flow operation was analyzed by electrochemical impedance spectroscopy (EIS). Impedance measurements were conducted using a potentiostat (G300 potentiostat, Gamry Instrument Inc.) at constant potential of 300 mV with a sinusoidal perturbation of 5 mV amplitude between the anode and cathode (two-electrode mode) over a frequency range of  $3 \times 10^5$  to 0.005 Hz. Before starting each impedance measurement the MFC was prepolarized for at least 15 min at the measuring potential provided by the potentiostat to reach steady state conditions. On a Nyquist plot of the impedance spectra, the ohmic resistance was determined by the intercept of the curve with the real impedance ( $Z_{\text{re}}$ ) axis (imaginary impedance  $Z_{\text{im}} = 0$ ) [14,15].

### 3. Results

#### 3.1. Effect of J-Cloth on power generation and electrode potentials

MFCs (Fig. 1A and B) with different layers of J-Cloth and same electrode spacing (1.7 cm) were used to study the effects of J-Cloth on power density. In the absence of J-Cloth, the MFC generated a maximum power density of  $80 \text{ W m}^{-3}$  (Fig. 2). A slight decrease of power densities was obtained in the MFCs with one ( $71 \text{ W m}^{-3}$ ) and two layers of J-Cloth ( $68 \text{ W m}^{-3}$ ). Further addition of J-Cloth layers (3 layers) resulted in a 30% of decrease in power density ( $55 \text{ W m}^{-3}$ ). This decrease in power density corresponded to an increase of internal resistance from  $82 \Omega$  in the absence of J-Cloth to  $88 \Omega$  (1 layer),  $92 \Omega$  (2 layers), and  $105 \Omega$  (3 layers).

The electrode potentials were also measured at various current densities to investigate the effect of J-Cloth on electrode performance. Similar anode potentials were observed for the

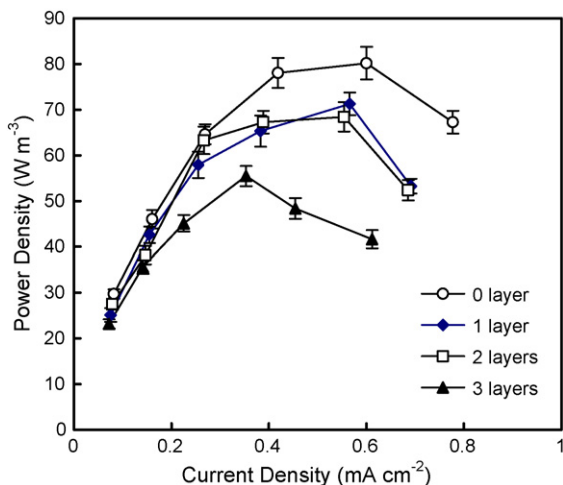


Fig. 2. Effect of J-Cloth layers on power generation of MFCs with 1.7 cm electrode spacing.

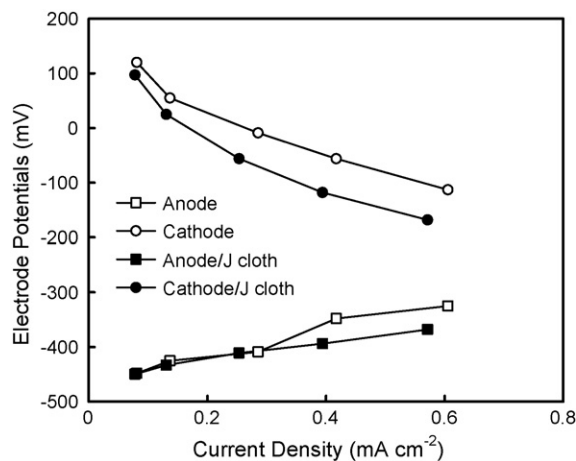


Fig. 3. Anode and cathode potentials (vs. Ag/AgCl reference electrode, 195 mV vs. NHE) as a function of current density in MFCs with 1.7 cm electrode spacing.

MFCs with and without J-Cloth at low current densities (less than  $0.3 \text{ mA cm}^{-2}$ ) (Fig. 3). Further increase of current density from  $0.3 \text{ mA cm}^{-2}$  to  $0.6 \text{ mA cm}^{-2}$  resulted in a much quicker increase in anode potentials for the MFC without J-Cloth possibly due to the effect of oxygen on the activity of anaerobic bacteria on anode. More oxygen might be diffused through the cathode in the absence of J-Cloth than that with J-Cloth. At low current densities, when the electron transfer was limited by the external resistance, such effect was negligible [9,12].

The cathode potentials decreased with the increase of current density for both MFCs. However, the potentials without J-Cloth were higher than that with 1-layer J-Cloth. A potential difference of 50–60 mV was observed at the high current density of  $0.3\text{--}0.6 \text{ mA cm}^{-2}$  possibly due to the increase of internal resistance when J-Cloth was applied to the surface of cathodes. Although adding a J-Cloth layer resulted in the lower cathode potential, the overall power output was not significantly affected due to the positive effect of J-Cloth on anode potential. The working cathode potentials obtained here (from 27 mV to 315 mV versus NHE) were much lower than the theoretical cathode potential of 804 mV at standard conditions [7], indicating that further improvements in power density could be made through the modification of cathode.

#### 3.2. Effect of J-Cloth on Coulombic efficiency

CE increased with current density for all MFCs (Fig. 4). A CE of 71% was obtained at a current density of  $0.6 \text{ mA cm}^{-2}$  in the MFC with two-layers of J-Cloth, which is over 100% higher than that without J-Cloth (35%) at the same current density. However, there was no significant difference in CE in terms of different layers of J-Cloth: 64%, 71%, and 63% for 1, 2 and 3 layer(s) of J-Cloth, respectively. The much higher CE achieved by MFCs with J-Cloth layers indicates that J-Cloth can effectively reduce the oxygen diffusion, which was considered as the main reason for lowering the CE in air-cathode MFC

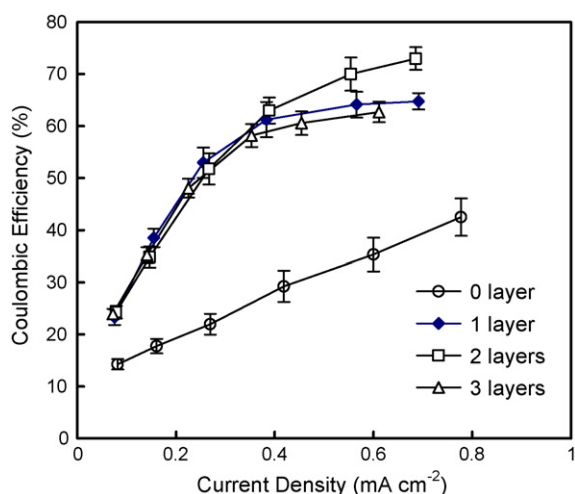


Fig. 4. Effect of J-Cloth layers on the Coulombic efficiency of MFCs with 1.7 cm electrode spacing.

using a mixed culture [9]. As a comparison, when no oxygen is involved in the reaction, CE can be as high as 89% [2] and 95% [16] for two chamber MFCs using ferricyanide as electron acceptor.

### 3.3. Performances of CEA-MFCs in fed-batch operation

Due to the effective blocking of oxygen by J-Cloth, which was indicated by the CE increase in the above study, CEA-MFCs were constructed (Fig. 1C and D) to investigate if improved volumetric power density can be achieved through increasing the electrode area to cell volume ratio. A maximum power density of  $170 \text{ W m}^{-3}$  was generated by the MFC with a single CEA and an empty bed volume of 6 ml (Fig. 5). The decrease of cell volume from 6 ml to 3 ml resulted in a 78% increase in volumetric power density ( $303 \text{ W m}^{-3}$ ). When the electrode area to cell volume ratio was further increased by using double CEAs and a cell volume of 2.5 ml, a power density of  $627 \text{ W m}^{-3}$  was achieved at a current density of  $0.28 \text{ mA cm}^{-2}$  (Fig. 5).

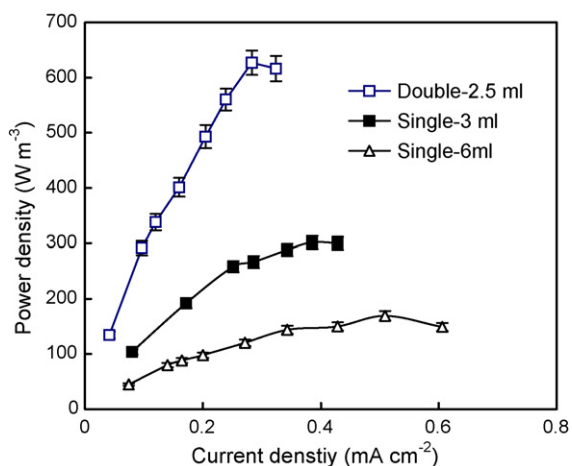


Fig. 5. Power density as a function of current density using MFCs with single CEA or double CEAs and different working volumes.

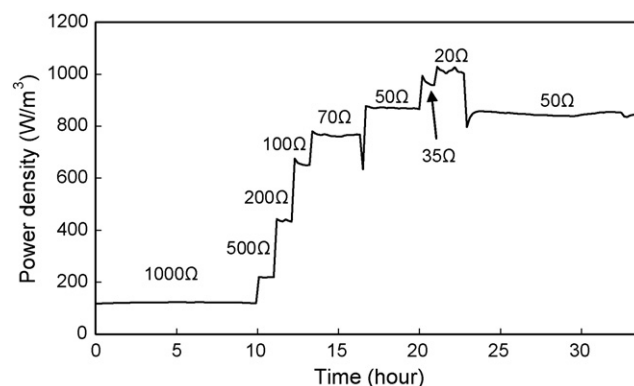


Fig. 6. Power generation of the double CEAs MFC operated in a continuous-flow mode.

The maximum surface power densities, on the other hand, were slightly decreased with the empty bed volume. A power density of  $1460 \text{ mW m}^{-2}$  was generated by the MFC with single CEA and a bed volume of 6 ml, and it decreased to  $1300 \text{ mW m}^{-2}$  for the MFC with 3 ml volume. For the MFC with double CEAs and 2.5 ml volume, the power density was  $1120 \text{ mW m}^{-2}$  based on the total anode surface of  $14 \text{ cm}^2$ . Such a power density (based on anode surface area) decrease was possibly due to the limited total substrate caused by the reduced volume when operated at relatively high current densities. Since same initial substrate concentration was used for all the MFCs, it was relatively difficult for the MFCs with smaller volumes to maintain the power output plateau due to the rapid substrate consumption, resulting in lower hourly averaged power density.

### 3.4. Performances of CEA MFCs in continuous-flow operation

Operating MFC in a continuous-flow mode holds potential advantages over a batch mode by maintaining a more stable substrate supply, more controllable environmental conditions for bacteria, and better mass transfer conditions. Fig. 6 shows the performance of the MFC with double CEAs operated in a continuous-flow mode. A maximum power density of  $1010 \text{ W m}^{-3}$  ( $1800 \text{ mW m}^{-2}$ ) was generated at a current density of  $0.9 \text{ mA cm}^{-2}$ , which was over 60% higher than that operated in the fed-batch mode. The ohmic resistance of the electrolyte of double-CEA MFC was  $3.9 \Omega$  as measured using EIS.

## 4. Discussion

### 4.1. Membranes/cloth and Coulombic efficiency of air-cathode MFCs

Membrane is needed in a two-chamber MFC system to separate the anode and cathode chambers. Studies have been conducted on various membranes with their effects on power generation and CE, including bipolar membrane [17], Nafion

[18–23], cation, anion and ultrafiltration membrane [19], and nanoporous polymer filters [24]. Among the various membranes used in MFCs, Nafion membrane is most commonly used in two chamber MFC systems [18–23], possibly due to its popularity in chemical fuel cells for excellent proton conductivity and thermal and mechanical stability. However, the expensive Nafion does not show great advantage over other exchange membranes in MFC application because of the low proton concentration and moderate reaction condition in MFCs [18,19,25].

In an air-cathode MFC, the major purpose of using membrane is to reduce or block the oxygen diffusion from the cathode to anode. Nafion membrane was bonded directly onto the air cathode of an MFC in a previous study [9]. Although improved CE was obtained due to the reduced oxygen diffusion from the cathode, the addition of Nafion membrane also lowered the power density [9]. In this study, the addition of J-Cloth layers significantly increased the CE without greatly affecting the power output. The CE of 71% achieved by using two-layers of J-Cloth is about 100% higher than that without a J-Cloth, and also higher than the 40–55% of air-cathode MFCs containing PEM [9], indicating that the J-Cloth adjacent to the cathode can greatly reduce the oxygen diffusion to the cell chamber.

The rate of oxygen diffusion ( $W$ ) through a J-Cloth layer can be calculated as

$$W = -DA\Delta C/\delta_m \quad (2)$$

where  $D$  is the diffusion coefficient of oxygen in the cloth;  $\delta_m$  the cloth thickness (two layers of J-Cloth is about 0.6 mm);  $A$  the cross-sectional area ( $7\text{ cm}^2$ );  $\Delta C$  the concentration difference. The maximum  $\Delta C$  equals to the saturation of oxygen in the water ( $7.62\text{ mg l}^{-1}$  at  $30^\circ\text{C}$ ) assuming no oxygen in the anode side of the cloth.

The oxygen diffusion coefficient ( $D$ ) through a porous J-Cloth should be close to but less than that of bulk water, which is  $2.26 \times 10^{-5}\text{ cm}^2\text{ s}^{-1}$  at  $30^\circ\text{C}$  [26]. Therefore, the maximum oxygen flux through a  $7\text{ cm}^2$  two-layer J-Cloth can be calculated as  $0.07\text{ mg h}^{-1}$  based on Eq. (2). This value is comparable to the oxygen flux through a  $0.194\text{ mm}$  thick Nafion membrane under the similar conditions, which was measured as  $0.05\text{ mg h}^{-1}$  in a previous study [9], but much lower than the  $0.187\text{ mg h}^{-1}$  in a membrane-free MFC with the same size and similar cathode material [9]. This explains why a significant improvement of CE can be achieved in the MFCs with J-Cloth layers. With the development of biofilm on the J-Cloth within several days of operation, the total thickness of two-layer of J-Cloth, as measured by an optical microscope, can be as much as 1–2 mm. The oxygen flux, therefore, can be further decrease to about  $0.01\text{--}0.02\text{ mg h}^{-1}$  in such an MFC by considering that the effective diffusion coefficient in the biofilm is about 60% of that in water [27]. The significantly decreased oxygen flux but slightly changed internal resistance makes it possible to further reduce the electrode spacing. Although a Nafion membrane with a biofilm can also be effective in blocking oxygen, it might increase the internal resistance, resulting in a lower power density and current density [9]. The lower cur-

rent density may prolong the overall reaction time, yielding a higher amount of total oxygen transferred and therefore a lower CE.

While J-Cloth is an inexpensive, widely used material, other porous and non-conductive materials may also achieve similar results. Basic requirements for the materials are non-conductive, durable, low cost, high anion permeability and low oxygen permeability.

#### 4.2. Power density of CEA–MFCs

MEA, a common practice in chemical fuel cells, may not be suitable to be applied to an air-cathode MFC directly due to the oxygen diffusion through Nafion membrane without a biofilm [9]. The blocking of oxygen diffusion by a cloth/membrane layer and a biofilm makes a similar configuration (CEA) possible in an MFC, which can result in a reduced spacing between electrodes, a lower internal resistance, and an enhanced volumetric power density.

A significant difference in volumetric power density may be obtained for the MFCs with a similar surface power density (based on electrode surface area) due to the variety of MFC configurations [2,10,15,30]. While surface power density is suitable to compare the electrode characteristics, volumetric power density can be used to evaluate the whole cell performance. An hourly averaged power density of  $627\text{ W m}^{-3}$  was achieved in fed-batch operation. The power density was further increased to  $1010\text{ W m}^{-3}$  when operated in a continuous-flow mode. Such a power density is about 53 folds higher than the value reported for a 4 cm spacing MFC system with same substrate, electrode material and surface area [12] and about 15 times higher than that obtained by 2 cm spacing MFC systems reported previously [10,11] and in this study (1.7 cm spacing) (Table 1). It is also over 100% higher than  $500\text{ W m}^{-3}$  obtained in a miniature two-chamber system [28] with a 1.2 ml anode chamber (the volume of cathode chamber was not considered in power calculation) using ferricyanide as catholyte, which is not sustainable in practice since it must be chemically regenerated [7,29].

The surface power densities, on the other hand, varied slightly for the air-cathode MFCs when similar electrode materials were used (Table 1). The significant increase in volumetric power density in this study was due to the CEA configuration, which decreased the electrode spacing and increased the surface/volume ratio. In a previous study, Cheng et al. [11] reported that there was slim change in volumetric power densities from  $41\text{ W m}^{-3}$  to  $42\text{ W m}^{-3}$  (calculated based on reported surface power densities) when the electrode spacing was reduced from 2 cm to 1 cm, while the internal resistance reduced from  $35\ \Omega$  at 2 cm to  $16\ \Omega$  at 1 cm. This is most likely a result of oxygen diffusion from the cathode into the surface of anode, as  $0.05\text{--}0.1\text{ mg l}^{-1}$  dissolved oxygen (DO) was measured at a distance 1 cm from cathode. With the help of the J-Cloth layer in the reduction of oxygen diffusion, the electrode spacing of CEA–MFCs was able to be reduced to about 0.06 cm without significantly affecting of the surface power density, resulting in a greatly increased volumetric power density.

Table 1  
Comparison of power density generated in mediator-less MFCs

Cell type	Substrate	Anodes	Spacing/catholyte	$P_s$ (mW m <sup>-2</sup> )	$P_v$ (W m <sup>-3</sup> )	Reference
Double CEAs (continuously flow)	Acetate	Carbon cloth	0.4 cm <sup>a</sup>	1800	1010	This study
Double CEAs	Acetate	Carbon cloth	0.4 cm <sup>a</sup>	1120	627	This study
Single chamber (continuously flow)	Glucose	Carbon cloth	1 cm	1540	51	[11]
Single chamber	Glucose	Carbon cloth	1 cm	423	42	[11]
Single chamber	Acetate <sup>b</sup>	NH <sub>3</sub> -treated carbon cloth	2 cm <sup>c</sup>	1970	115	[31]
Single chamber	Glucose	Carbon cloth	2 cm	811	41	[11]
Single chamber	Acetate	Carbon paper	2 cm	1210	61	[10]
Single chamber	Acetate	Carbon cloth	4 cm	766	19	[12]
Single chamber	Acetate	Carbon paper	4 cm	506	13	[32]
Two chamber (miniature)	Lactate	Graphite felt	Ferricyanide	3000	500	[28]
Two-chamber	Glucose	Plain graphite	Ferricyanide	3600	216	[2]
Two-chamber (bipolar membrane)	Acetate	Graphite felt	Ferric iron	860	42	[23]
Two chamber (upflow)	Sucrose	Activated carbon	Ferricyanide	NA	29	[15]

<sup>a</sup> Spacing between the anodes of two CEAs, the anode and cathode spacing in each CEA is about 0.06 cm initially.

<sup>b</sup> 200 mM phosphate buffer.

<sup>c</sup> The spacing should be 1.7 cm based on  $P_s/P_v$ .

The steady status maintained in a continuous-flow operation made it suitable to measure the internal resistances of the cell using EIS due to the long time requirement (>1 h) for a complete scan [14]. The ohmic resistance of the electrolyte of double-CEA MFC was only 3.9 Ω, which was lower than the 31 Ω for a single chamber carbon cloth MFC (7 cm<sup>2</sup> anode and 7 cm<sup>2</sup> cathode, 4 cm electrode spacing, 200 mM PBS) and the 8 Ω for a brush MFC (7 cm<sup>2</sup> cathode, 200 mM PBS) [30]. It was also lower than the 8.6 Ω in an upflow MFC with interior cathode (membrane surface area 188 cm<sup>2</sup>) [15]. The small electrode spacing (0.06 cm) was attributed to the low internal resistance of the system. A great advantage of the CEA configuration is that the internal resistance per surface area of such an MFC can remain the same during the scale-up due to the fixed electrode spacing.

## 5. Conclusions

Coulombic efficiency of single chamber MFCs can be substantially enhanced by applying two layers of J-Cloth on the water facing side of the air cathode (71% versus 35%). The significant reduction of oxygen diffusion by the cloth layer made it possible to greatly reduce the spacing between electrodes and form a cloth-electrode-assembly configuration. A volumetric power density of 1010 W m<sup>-3</sup> was obtained with double-CEA MFCs operated in continuous-flow mode, which is over 15 times higher than that of MFCs using similar electrode materials. Such a power density is higher than 1000 W m<sup>-3</sup>, a critical value that was considered to make a 10-year payback possible for wastewater treatment [7,29]. The high volumetric power density achieved in an air-cathode MFC will also provide a great promise for other applications, such as potable power supplies and power sources for remote sensors using indigenous fuels.

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