



Influence of Coulombic Efficiency in Air-Cathode Microbial Fuel Cell by Temperature and Baffle-microfiltration Membrane Barrier

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Coulombic efficiency is an important measurement of energy recovery, which is very low in the air-cathode microbial fuel cell. Temperature variation and baffle-microfiltration membrane barrier installation were performed to investigate the effect on coulombic efficiency of air-chamber microbial fuel cell. The results under three temperature stages (28-32 °C, 16-20 °C and 6-12 °C) showed that coulombic efficiency can be increased by lowering operating temperature appropriately as a price for power generation loss, however, decreased seriously under excessively low temperature. The baffle-microfiltration membrane barrier trapped bacteria in the bottom part near anode, avoided the contact between bacteria and substrate in the top part with high oxygen concentration near cathode, which can decrease the loss of substrate for aerobic respiration and effectively increase coulombic efficiency from 4.5 % (control) to 9.9 %, without obvious reduction in power generation. Furthermore, the reformed reactor even showed higher performance than the control at the higher initial COD concentration range.

Key Words: Microbial fuel cell, Coulombic efficiency, Temperature, Baffle, Microfiltration membrane.

INTRODUCTION

The depletion of energy sources is a critical problem for more and more concern. Microbial fuel cells (MFCs) provides new opportunity for the energy production¹, which can convert organic matter directly into electricity with microorganisms as biocatalysts under ambient temperature/pressure.

Microbial fuel cell is a complex system which involves biological, chemical, electrical and physical processes. Therefore, the performance of microbial fuel cells is influenced by many factors, such as electrogenic biofilm^{2,3}, substrate (fuel)⁴, system design⁵ and various operating conditions^{6,7}.

In the anode chamber of microbial fuel cell, substrate was oxidized in the anode chambers, afterwards the produced electrons and protons end up in the cathode chamber to form H₂O, through the external electrical circuit and proton exchange membrane (PEM), respectively.

Electrons are generally transferred through the circuit, but can also be utilized *via* several other pathways, such as in bacterial growth in the anode chamber, or metabolite generation. Lots of researches focused on increasing microbial fuel cell power outputs, however, due to the limitations of microbial metabolic processes in microbial fuel cell, energy recovery efficiency is very low in microbial fuel cell.

Proton exchange membrane divides the microbial fuel cell into two chambers to maintain anoxic and aerobic condition in anode and cathode chamber respectively, unfortunately, these membranes are permeable to oxygen⁸. Therefore, the substrates added to microbial fuel cell anode could be lost through aerobic respiration by bacteria⁹, rather than be used for bioelectricity generation near anode, low coulomb efficiency (CE) was revealed.

Many methods had been done to fix the problem of low coulombic efficiency. The oxygen scavengers, such as cysteine¹⁰ was used to maintain a low redox potential in anode chamber¹¹, since cysteine reacts with oxygen to form a disulfide dimer (cystine). 2-Bromoethanesulfonate (BES) can be used to inhibit the activity of methanogens, which reduce the substrate utilization for methanogens, hence increase the utilization for exoelectrogens¹². Watson *et al.*¹³ applied nafion-coated cathode to reduce oxygen diffusion into the anode chamber and increased the coulombic efficiency compared to uncoated cathode, but resulted in decreased power production. By configuration reform, a two-stage process used by a dual anode microbial fuel cell, was constructed by Kim *et al.*¹⁴, to enhance the coulombic efficiency from 33 to 59 %, compare with one anode microbial fuel cell.

These methods effectively increased the coulombic efficiency of microbial fuel cell, however, most of them required

complex processes, high cost or may be as a price for power generation decreasing¹³, which hindered their practical application.

Air-cathode single chamber microbial fuel cell attract more attention in recent study, because the removal of the proton exchange membrane causes a reduction in the internal resistance of the system, which greatly increases the power densities⁹. However, due to unobstructed diffusion of oxygen, the air-cathode microbial fuel cell especially has to face the problem of low coulombic efficiency. Besides, diffusion of oxygen to the anoxic anode raised the redox potential, halts cell electrochemistry respiration and hinders the electrochemistry activity bacteria growth on anode.

Operating temperature in microbial fuel cell, as a main factor in practical application, can affect microbial fuel cell performance in many ways. The growth, metabolism activity and the way of electrons utilization of microorganism in microbial fuel cell varies with the temperature change. It was suggested that microbial fuel cell can accommodate a widely range of temperature and lowering the temperature within limit is an effective way to increase the coulombic efficiency without reducing power output in microbial fuel cell¹⁵.

In our experiment, temperature effect on power generation and coulombic efficiency of single chamber microbial fuel cell was studied. Also another simple method by application of microfiltration membrane (with 0.2 μm pore) was applied in the single-chamber microbial fuel cell to promote the coulombic efficiency. A "baffle-microfiltration membrane" barrier was installed in the microbial fuel cell chamber, which can obstruct bacteria through by, in order to isolate the O_2 , bacteria and substrates effectively in space.

Therefore, this study involves: (1) investigate the effects of temperature on coulombic efficiency in single chamber microbial fuel cell; (2) wonder if this "baffle-microfiltration membrane" barrier could promote the coulombic efficiency or power generation of the microbial fuel cell.

EXPERIMENTAL

Air-cathode single-chamber microbial fuel cell made of plexiglass was constructed as a cylinder with a height of 9 cm, radius of 4 cm (the total working volume is 450 cm^3). Carbon felt was used as anode which was placed at the bottom of cylinder. Polytetrafluoroethylene (PTFE) coated (the upper side contact air) carbon cloth was used as cathode¹⁶, which floated at the top of microbial fuel cell. The project area of both anode and cathode was 50.2 cm^2 .

A hollow baffle, with a radius of 2 cm hole, was installed (as a transverse) at the 6 cm height from the bottom (3 cm near the top cathode) in order to divide the single chamber into two parts (interlinked): bottom part with volume of 300 cm^3 and top part with volume of 150 cm^3 .

Cellulose acetate microfiltration membrane (with pore diameter of 0.2 μm) was fixed at the hole on the baffle. Theoretically, the microorganisms were confined in the bottom part of single chamber microbial fuel cell, while the top part was free of bacteria after the integrated barrier installed. Microfiltration membrane was stored in deionized water and gently rinsed before use.

General procedure: All the experiments operated in batch mode in single chamber microbial fuel cell. The glucose was used as the sole substrate for bacteria and temperature was controlled in water bath or in room temperature (6-12 $^\circ\text{C}$, in winter).

The anode medium contained (per litre): KCl (0.13 g), NaH_2PO_4 (4.22 g), Na_2HPO_4 (2.75 g), $(\text{NH}_4)_2\text{SO}_4$ (0.56 g), $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ (0.2 g), CaCl_2 (15 mg), $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (1 mg), $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ (20 mg) and 1 mL/L of trace elements solution¹¹. The anaerobic digester sludge which was taken from a pharmaceutical, was inoculated in the anode compartment (15% v/v, in the bottom part if barrier installed) as biological catalyst.

First, the single chamber microbial fuel cell without the barrier, was operated to check the temperature effect on power output and coulombic efficiency. The temperature was controlled at three stages: 28-32 $^\circ\text{C}$, 16-20 $^\circ\text{C}$ by water bath and at 6-12 $^\circ\text{C}$ in room temperature in winter. Each temperature stage operated for two repeated fed-batch cycles with 300 mg/L COD initially, except for the first start-up circle.

After temperature experiment, three identical reactors: non-barrier (R1), partial-barrier (R2) with baffle only and integrated-barrier (R3) with both baffle and microfiltration membrane, operated at the same time in parallel under the same operating conditions (temperature was fixed at 28-32 $^\circ\text{C}$).

At the end of each fed-batch cycles, glucose (with 300 mg/L COD) was added in the anode chamber (or in the top part of the anode chamber of microbial fuel cell).

Detection method: The external resistance R was fixed at 1000 Ω . U is the voltage between anode and cathode (V), I is the current (A), Voltage was measured using a data acquisition system every 1 h and converted to power density, power density (PD, mW/m^2), according to $P = U \times I$ and normalized by anode project area. The internal resistance of the cell R_{int} , was calculated from the slope of V and I.

COD was determined according to the standard methods. The coulombic efficiency (CE) is calculated as:

$$\text{Coulombic efficiency (\%)} = \frac{M \int_0^t I dt}{F b V \Delta \text{COD}} \times 100 \%$$

where M represented the molecular weight of substrate (180 for glucose); F is Faraday's constant (96,485 C); b represented number of electrons exchanged per mole of substrate utilized; V was the working volume of microbial fuel cell.

At the end of operation, the carbon felt anode at the bottom was cut into small pieces and characterized by scanning electron microscopy to observe the surface image of anode biofilm.

RESULTS AND DISCUSSION

SEM of anode: After operation of the microbial fuel cell, the carbon felt anode at the bottom was cut into pieces and subjected to scanning electron microscopy (SEM).

Fig. 1 showed a biofilm was formed on the electrode surface by bacteria on the carbon felt anode when microbial fuel cell were stably operated. Anode biofilm is considered as the bio-catalyst which plays the significant role in electron transfer between anode and electrode. In air-cathode microbial fuel cell, due to partial aerobic respiration of bacteria, the

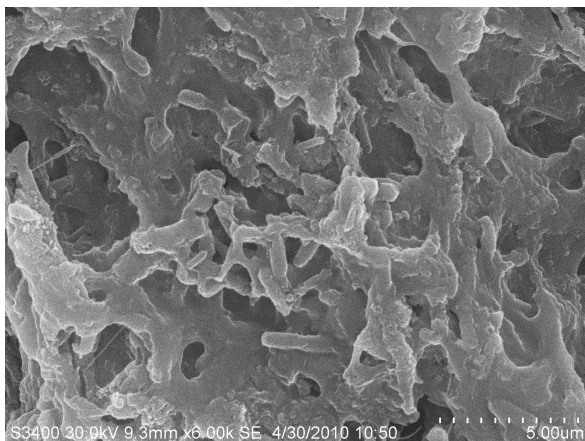


Fig. 1. Scanning electron microscopy image of biofilm on the carbon felt anodic surface of microbial fuel cells

anode biofilm grows faster and thicker. The image demonstrated that bacteria attached to the carbon felt surface and created a multilayer and thick biofilm. And from morphology, generally, most of the microorganisms on the anode were rod-shaped.

Temperature effect: Fig. 2 showed the voltage-time profile of the single chamber microbial fuel cell (without barrier) under different temperature. Seven cycles was performed totally and the obtained results were demonstrated in Table-1.

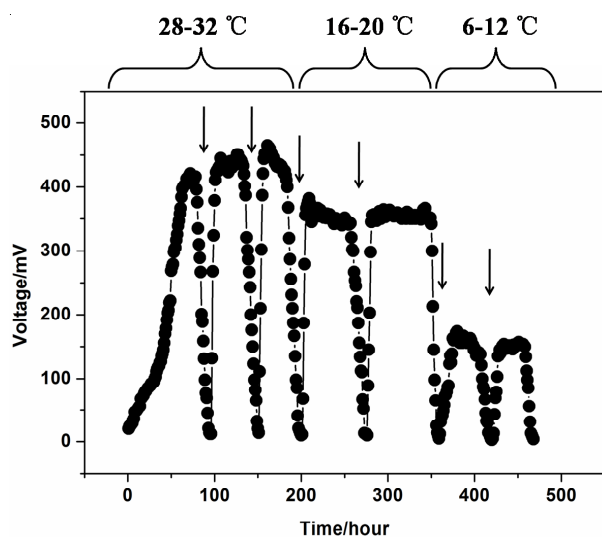


Fig. 2. Comparison of cell voltage output under different operation temperature for repeated fed-batch cycles. Arrows showed the addition of glucose (COD = 300 mg/L)

TABLE-1
LIST OF RELEVANT INFORMATION FOR EACH CYCLE
AT DIFFERENT OPERATION TEMPERATURE

Cycle number	Temp. (°C)	Average voltage (mV)	Maximum power density (mW/m ²)	Cycle time (h)	COD removal (%)	CE (%)
2	28-32	450	41.4	45	96	4.4
3	28-32	445	42.5	44	95	4.2
4	16-20	370	29.1	64	90	5.3
5	16-20	358	26.4	67	90	5.4
6	6-12	151	6.0	40	73	1.6
7	6-12	150	4.7	39	75	1.3

The first circle showed the voltage evolution in start-up period. Microbial fuel cell (without barrier) was started by addition of glucose with 1000 mg/L COD in the first batch circle under temperature of 28-32 °C. For single chamber microbial fuel cell, the start-up speed is very fast (compare to two-chamber microbial fuel cell). After a very short lag phase (almost no), the microbial fuel cell voltage began to rise quickly at the 20th h and finally reached the platform around 430 mV at the 75th h.

The following two cycles proceeded under the same temperature 28-32 °C. The maximum power density was 42.5 mW/m² and the coulombic efficiency was 4.4 %, which was relative lower, but with a high COD removal (about 95 %). In air-cathode microbial fuel cell, due to no obstacle of O₂ diffusion to anode, much substrate was consumed by aerobic respiration of bacteria.

Since it was demonstrated that microbial fuel cells can effectively be operated over a wide range of temperatures¹⁷, after two repeated cycles, the operation temperature was controlled to descend to 16-20 °C by water bath. The results showed that power output decreased from 42.5 to 29.1 mW/m² compare to the temperature 28-32 °C with a little of COD removal decreasing (from 96 to 90 %). This might due to the reduction of metabolic activity of microorganisms in microbial fuel cell at relative lower temperature range, which influenced the utilization of substrates of all of the microorganisms in microbial fuel cell.

However, longer period of time to maintain stable power generation was observed at lower temperature range 16-20 °C (Fig. 2) and as a result, higher coulombic efficiency was obtained (Table-1). This may be due to prevention of some non- electrochemistry active bacteria, such as methanogens, at unfavourable temperature. The temperature stress successfully inhibited the methanogens, while slightly suppressing the exoelectrogens, which reduced the loss of substrate *via* other pathways, increased the substrate utilization for electrochemistry active bacteria on anode, which lead to a higher coulombic efficiency (5.4 %).

This lower operating temperature range demonstrated that the electrochemically active bacteria could remain high capability converting substrate to electrical energy even at lower temperature below 20 °C¹⁸. Therefore, in the single-chamber microbial fuel cell, lower the operating temperature properly, did not influence the power generation obviously, on the contrary, the coulombic efficiency can be raised.

Since higher temperature favoured higher COD removal efficiency (Table-1), while lower temperature created higher coulombic efficiency, the proper operating temperature should be selected according to the specific needs in practical application.

Nevertheless, high coulombic efficiency did not always increase with the decrease in temperature. When the operating temperature reached a much lower range of 6-12 °C, (by placing in room temperature in winter), the results in cycle 6 and 7, showed both power output and coulombic efficiency reduced obviously (Table-1 and Fig. 2). The period of time to maintain power generation also decreased, which meant excessive low temperature greatly reduced the total microbial fuel cell performance.

Enhanced coulombic efficiency by baffle-microfiltration membrane barrier: Fig. 3 showed three repeated cycles for each reactor after startup period. Similar power generations were obtained in all the three reactors. The average voltage was 430 mV in R3, which showed a bit lower than R1 (450 mV) and R2 (448 mV).

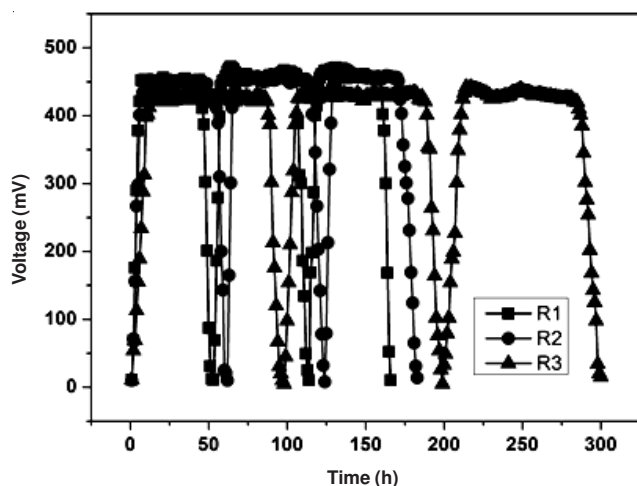


Fig. 3. Three repeated fed-batch cycles for each reactor. (R1: control, R2: with baffle, R3: with baffle and 0.2 μm microfiltration membranes). Glucose with 300 mg/L COD was added at each end of the batch cycle

It was observed that much longer period of maintain power generation for R3, compared to R1 and R2. The same quantity of electron donor (glucose with 300 mg/L COD) was applied, but maintained different power generation time, for about 50 h, 60 h and 100 h in R1, R2 and R3 separately in each circle. As a result, the coulombic efficiency of R3 (9.9 %) was much higher than R1 (4.5 %) and R2 (5.3 %).

In single chamber microbial fuel cell, the diffusion of O_2 raises the redox potential of anode and halts cell electrochemistry respiration. Although recent study on the effect of O_2 presence in the anode compartment concluded that the dissolved oxygen (DO) level did not affect the power output since the O_2 is scavenged by aerobic digestion¹⁹, the energy in electron donor is still wasted by the aerobic digestion, rather than electrochemistry respiration.

As the substrate, glucose was the most favourable substrate generating the greatest power density, however, in terms of coulombic efficiency, it was the lowest one compared with other substrates²⁰. Because it is a fermentable substrate, which implies its consumption by diverse competing metabolisms such as fermentation and methanogenesis that cannot produce electricity. However, glucose was used in this experiment to better discriminate the coulombic efficiency influence factors of the single chamber microbial fuel cell.

In order to overcome these limitations and optimize microbial metabolic processes in single chamber glucose-fed microbial fuel cell, the new reforms in configuration was conducted in this experiment and play their own role in promoting the microbial fuel cell performance.

Floating cathodes on the top of microbial fuel cell chamber were applied for all the microbial fuel cell in this experiment, which can reduce the hydrostatic pressure of

cathode and decrease the water seepage near cathode. The contact area of cathode can remain the same, even the working electrolyte loss, which benefit to the stable operation of the microbial fuel cell.

Compare R2 to R1, the baffle which was installed in the chamber of R2 played the role of obstructing oxygen diffusion to anode with a certain extent. Since it was interlinked between the top and bottom parts of the chamber, the internal resistance did not change much. So the power output of R2 remained almost same as R1. Higher dissolved oxygen concentration exists near the top cathode. The baffle partially divided the chamber into the top part (with higher dissolved oxygen) and bottom part (with lower dissolved oxygen), which reduced the contact between bacteria and oxygen to a certain degree. Aerobic degradation with substrates for bacteria was decreased and coulombic efficiency increased (from 4.4 to 5.4 %). The result of coulombic efficiency increment was consistent with the research of Hu²¹ who also applied a baffle in single chamber microbial fuel cell.

Compare R3 to R2, the coulombic efficiency was further increase remarkably to 9.9 %. The 0.2 μm microfiltration membrane was fixed to better separate bacteria, substrates and oxygen. The 0.2 μm microfiltration membrane owed much larger pore than PEM in two-chamber microbial fuel cell. The electrolyte, ion, substrate (glucose) and macro-molecule can pass through freely, which do not contribute much internal resistance for microbial fuel cell (the average voltage only reduced a little). Only the bacteria, with large bulk, were trapped in the bottom part (near anode, far from cathode), cannot suspend in the top part (near cathode), since the membrane (with 0.2 μm pore) did not allow traverse for bacteria.

The dissolved oxygen concentration reduces gradually from top (cathode) to bottom (anode) in the microbial fuel cell chamber, Therefore, the integrated "baffle-microfiltration membrane" barrier divided the microbial fuel cell chamber into two parts: the top part with higher dissolved oxygen close to cathode and no bacteria (theoretically), while the bottom part with lower dissolved oxygen and much bacteria (near anode).

A relative anaerobic condition was maintained in the bottom part, which was fit for electrochemistry respiration. When glucose was added to the top part of R3, it gradually permeated to the bottom part to feed the bacteria for power generation. Although higher dissolved oxygen existed in top part near the cathode, the substrates were not consumed due to sterile environment in top part. So the electron recovery increased.

In other words, the top part in R3 (1/3 of the total working volume) can be considered as a dissolved oxygen buffering zone which help the bottom part to maintain more anaerobic condition for electrochemistry respiration by anode bacteria. Also the top part acted as a slow-release region for substrates. The substrates were slowly feed to bacteria in the bottom and were protected from depleting by aerobic respiration of bacteria (high dissolved oxygen and substrate concentration, but no bacteria in the top part). It was noticed that the rising speed of R3 for each circle was slower than R1 and R2, which indicated that substrate utilization speed was slow due to the storage effect in the top part.

To summarize, the baffle-microfiltration membrane barrier killed two birds with one stone (two benefit effects): 1) buffering O_2 , reducing O_2 diffusion to anode in order to maintain a stable redox potential for anode; 2) protecting substrate (glucose), which reduces its waste for aerobic respiration and increases coulombic efficiency.

It is known that aerobic respiration consumes substrates much faster than anaerobic respiration (including electrochemistry respiration). So the barrier in R3 inhibited the growth of aerobic bacteria, which not only promoted the competitive power of electrochemistry active bacteria for substrate and increased coulombic efficiency, but also benefited for selective growth of bacteria on anode. The more anaerobic condition in the bottom part near anode, guaranteed the growth of obligate anaerobes such as *Geobacteraceae* and forced the facultative anaerobes such as *Shewanella* to perform electrochemistry respiration. So, it is an effective method to promote coulombic efficiency with little loss in power density of microbial fuel cell.

TABLE-2
COMPARISON OF POWER OUTPUT AND COULOMBIC EFFICIENCY AMONG THE THREE REACTORS

Reactor	Average voltage (mV)	Maximum power density (mW/m^2)	Coulombic efficiency (%) [*]
R1	450	44.2	4.5 ± 0.05
R2	448	43.8	5.3 ± 0.05
R3	430	38.6	9.9 ± 0.08

^{*}For each reactors, n = 3, three cycles in Fig. 3.

COD concentration effect on three reactors: All the repeated cycles was fed by 300 mg/L COD (glucose) in this experiment since it had the optimal concentration²². In order to accommodate various operating condition in practical application, the initial COD concentration was increased stepwise to investigate its influence to the three reactors.

In R1 and R2, the power generation decreased remarkably with the initial COD concentration increased from 300 to 1200 mg/L. However, R3 showed less sensitivity for the COD variation, even increased its power generation at the 600 mg/L stage. Instead, at higher COD concentration range, better performance was obtained in R3, than R1 and R2 (Fig. 4).

In a traditional single chamber microbial fuel cell (R1, control), higher COD concentration may lead a relative lower power generation²². Although higher glucose concentration could release more electrons for longer duration of power output, it performs substrate inhibition effect on conversion to power generation. It is more noticeable, in single chamber microbial fuel cell, excessive substrate near cathode would consume much oxygen by aerobic respiration of bacteria (even microbial biofilm formation on the cathode), which reduces oxygen tension and lowers the redox potential of cathode. So, higher COD concentration is not suitable in single chamber microbial fuel cell.

As the barrier applied in R3, the top part protected oxygen from excessive consuming (high dissolved oxygen and substrate concentration, but no bacteria in the top part), which provided a stable oxygen tension near cathode. Although more substrates added, it did not contact with bacteria in this higher dissolved oxygen region. Therefore, due to the oxygen store

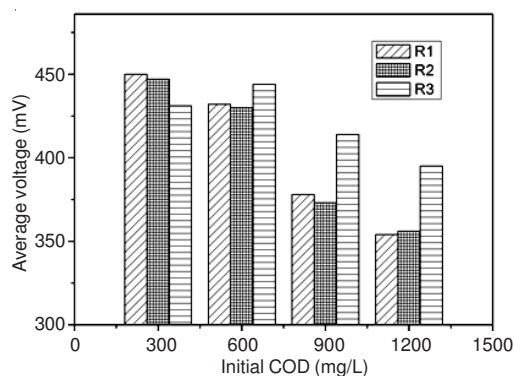


Fig. 4. Power output comparison of the three reactors with different initial COD concentration. (Four stages: 300, 600, 900, 1200 mg/L)

effect in the top part, at higher initial COD concentration range (≥ 600 mg/L in this experiment), the barrier installed can even improve power generation, besides coulombic efficiency in single chamber microbial fuel cell.

The baffle blocked oxygen diffusion to anode, while the 0.2 μ m microfiltration membrane restricted bacteria at the bottom part. The synergy effect of them can not only greatly promote coulombic efficiency without reducing power generation obviously, but also make the single chamber microbial fuel cell more tolerant with relative higher COD concentration. This is a simple method, with both economical and environmental benefits, which promote energy sources utilization ratio in microbial fuel cell. Furthermore, the reformed single chamber microbial fuel cell is still apt to scale-up in the practical application and facilitated in other multiple-stage wastewater process, which showed broad prospect for application.

Conclusion

Two methods for coulombic efficiency promotion in single chamber microbial fuel cell were discussed in this paper. Lower operating temperature moderately can slightly increase coulombic efficiency, as a price for lower power generation. However, both power generation and coulombic efficiency greatly decreased at the excessive low temperature. The "baffle-microfiltration membrane (with 0.2 μ m pore)" barrier is the more effective method, which greatly increased coulombic efficiency (from 4.4 to 9.9 %) and less affect on power generation. Meanwhile, at higher COD concentration (≥ 600 mg/L in this experiment), the barrier installed microbial fuel cell even showed better performance than non-barrier ones. These superiorities owed to the barrier installation, which can benefit for: 1) buffering O_2 , in order to maintain anaerobic condition for the bottom anode; 2) storing substrate, which reduces its waste for aerobic respiration and increase coulombic efficiency; 3) protecting O_2 from excessive consuming to maintain a stable redox potential for cathode.

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