



Short communication

Power generation from furfural using the microbial fuel cell

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ARTICLE INFO

Article history:

Received 19 May 2009

Received in revised form 16 June 2009

Accepted 17 June 2009

Available online 24 June 2009

Keywords:

Furfural

Electricity generation

Microbial fuel cell

ABSTRACT

Furfural is a typical inhibitor in the ethanol fermentation process using lignocellulosic hydrolysates as raw materials. In the literature, no report has shown that furfural can be utilized as the fuel to produce electricity in the microbial fuel cell (MFC), a device that uses microbes to convert organic compounds to generate electricity. In this study, we demonstrated that electricity was successfully generated using furfural as the sole fuel in both the ferricyanide-cathode MFC and the air-cathode MFC. In the ferricyanide-cathode MFC, the maximum power densities reached 45.4, 81.4, and 103 W m⁻³, respectively, when 1000 mg L⁻¹ glucose, a mixture of 200 mg L⁻¹ glucose and 5 mM furfural, and 6.68 mM furfural were used as the fuels in the anode solution. The corresponding Coulombic efficiencies (CE) were 4.0, 7.1, and 10.2% for the three treatments, respectively. For pure furfural as the fuel, the removal efficiency of furfural reached up to 95% within 12 h. In the air-cathode MFC using 6.68 mM furfural as the fuel, the maximum values of power density and CE were 361 mW m⁻² (18 W m⁻³) and 30.3%, respectively, and the COD removal was about 68% at the end of the experiment (about 30 h). Increase in furfural concentrations from 6.68 to 20 mM resulted in increase in the maximum power densities from 361 to 368 mW m⁻², and decrease in CEs from 30.3 to 20.6%. These results indicated that some toxic and biorefractory organics such as furfural might still be suitable resources for electricity generation using the MFC technology.

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1. Introduction

Furfural (2-furaldehyde) is the widely distributed simple furan in the nature and a well-known inhibitor for ethanologenic microorganisms [1,2]. It has been reported that furfural can inhibit the growth and fermentation of microorganisms at relatively low concentrations of 1–12 mM [3]. Furfural can severely decrease the rate of ethanol production and final conversion via the effects on the cell membrane function, as well as the growth and the glycolysis of ethanol-producing yeast and bacteria [4–6]. Therefore, removing furfural is a key step to increase the ethanol production.

The microbial fuel cell (MFC) technology has gained a great attention attributable to its ability in generating electricity directly from and potentially enhancing biodegradation of contaminants [7]. It has been reported that microorganisms in the MFC can utilize many kinds of organics, such as acetate, glucose, cysteine, xylose, and petroleum contaminants to generate electricity [8–12]. Catal et al. [13] investigated furfural as the substrate in a single-chamber air-cathode MFC for electricity generation. Their experimental results showed that furfural severely inhibited electricity generation even at a concentration lower than 0.2 mM [13]. Nevertheless, it has

been reported that furfural can be transformed, degraded, and even utilized by bacteria as the sole substrate under aerobic conditions after a period of acclimation [14–17]. Degradation of furfural under anaerobic conditions by sulfate-reducing bacteria (SRB) has also been reported [18,19]. Zarnt et al. [20] showed that furfural biodegradation into carboxylic acid was carried out by an aldehyde dehydrogenase. Some tolerant bacteria to furfural may have high activity of those enzymes after a period of acclimation and can work on biodegradation of the inhibitor [21]. Many microbes in the MFC have been identified to be capable of transferring the electrons directly or by electron shuttles to the electrode [22–24], and the electrons are possibly produced from the degradation of furfural. Based on the research results of [22–24], we hypothesize that furfural used as the sole fuel in the MFC may generate electricity, although no previous report has shown that electricity is generated from the biodegradation of furfural in the MFC.

The present study aimed to investigate the feasibility of bioelectricity generation utilizing furfural as the fuel through a ferricyanide-cathode MFC and an air-cathode MFC employing mixed bacteria collected from anaerobic and aerobic sludge. The MFCs performances were evaluated via power density curve, Coulombic efficiency (CE), and chemical oxygen demand (COD) removal. We also determined the effect of concentrations of furfural and furfural–glucose mixtures on electricity generation. Moreover, the degradation of furfural in the ferricyanide-cathode MFC was also investigated.

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2. Materials and methods

2.1. MFCs set up

A ferricyanide-cathode MFC consisted of an anode and a cathode, which were separated by a proton exchange membrane (PEM, Nafion 212, Dupont Co., USA). The electrodes were made of carbon fiber brush with 7 cm in length and 3.6 cm in width. The cathode compartment of the MFC contained 50 mM ferricyanide, used as the electron acceptor. Copper wires were used to connect the circuit with an external resistance of 1000 Ω and all wire contacts were sealed with epoxy material. The net volumes of the anode and cathode chambers were 18 mL, respectively. The anode (or cathode) chamber was connected with a brown bottle (250 mL capacity) as the anode (or cathode) groove. A flow rate of 20 mL min⁻¹ was controlled by a peristaltic pump for solution circulation between each pair of the brown bottle and the chamber. The MFC was operated at a constant temperature (30 \pm 1 $^{\circ}$ C).

To compare with the results of Catal et al. [13], a single-chamber air-cathode MFC was also constructed as described by Catal et al. [13]. Both the anode and cathode electrodes were made of carbon-cloth (TGP-H-060, Toray, Japan) with an area of 7 cm² for each. The PEM was boiled in H₂O₂ (30%), deionized water, 0.5 M H₂SO₄, and deionized water sequentially, during which each step took about 1 h. The carbon electrode-PEM cathode (CE-PEM) was constructed by bonding the PEM directly onto a flexible carbon-cloth electrode containing around 0.5 mg cm⁻² Pt catalyst. Then the PEM was hot-pressed directly onto the carbon-cloth electrode using a method by Liu et al. [10]. In the single-chamber MFC, oxygen was used as the electron acceptor. Copper wires were used to connect the circuit with an external resistance of 1000 Ω . The net volume of the MFC was 13 cm³. Other operation conditions were the same as the ferricyanide-cathode MFC described above.

2.2. Microbial inoculum and operation

Anaerobic and aerobic sludge (1:1, 10 mL) was taken from Liede Municipal Wastewater Treatment Plant, Guangzhou City, and inoculated in the ferricyanide-cathode MFC with a glucose solution of 1000 mg L⁻¹ and an anodic solution. The anodic solution contained (in 1 L deionized water): 4.0896 g Na₂HPO₄, 2.544 g NaH₂PO₄, 0.31 g NH₄Cl, 0.13 g KCl, and 12.5 mL trace metal solution and 12.5 mL vitamin solution [25]. After stable voltage outputs were achieved and kept for more than 2 cycles, furfural–glucose mixtures were used to replace the solutions in the MFC. The furfural–glucose mixtures included 0.1 mM furfural (Sigma–Aldrich, USA) + 1000 mg L⁻¹ glucose, 0.2 mM furfural + 1000 mg L⁻¹ glucose, 0.5 mM furfural + 1000 mg L⁻¹ glucose, 2.0 mM furfural + 500 mg L⁻¹ glucose, and 5.0 mM furfural + 200 mg L⁻¹ glucose. Again after stable voltage outputs were achieved and kept for more than 2 cycles, we used a furfural solution with a concentration 6.68 mM as the sole fuel to operate the MFC. The choice of the furfural concentration of 6.68 mM was based on that the COD value of the furfural solution (COD = 1100 mg L⁻¹) was almost the same as that of 1000 mg L⁻¹ glucose (COD = 1060 mg L⁻¹).

The air-cathode MFC was inoculated with active microorganisms taken from the anode of the ferricyanide-cathode MFC, which was incubated with the 1000 mg L⁻¹ glucose medium. When a stable power output was obtained from the MFC, we used a furfural solution with a concentration 6.68 mM as the sole fuel to incubate the active microorganisms. The substrates in the MFCs were replaced when the voltage decreased below 90 mV. Before operations, the anode compartment of the ferricyanide-cathode MFC was flushed with N₂ for 10 min to ensure an anaerobic environment.

2.3. Analyses and calculation

Samples from the anode solutions in the air-cathode MFC and the ferricyanide-cathode MFC were treated by filtered through a membrane (with a pore diameter of 0.22 μ m) to remove cells. Glucose concentrations of the samples were determined using the anthrone method [26]. Chemical oxygen demand (COD) was measured according to the standard method [27]. Furfural concentrations were analyzed using HPLC (Agilent 1100), in which 70% methanol and 30% water were used as the mobile phase and a flow rate of 1 mL min⁻¹ was maintained. A UV spectrophotometric detector was employed with a wavelength of 275 nm.

Cell voltages were measured at a time interval of 30 s across the external resistance using a data acquisition system (DT50). The current was calculated from the cell voltage and the external resistance based on the Ohms law. The area power density (P_A , W m⁻²) and volumetric power density (P_V , W m⁻³) were calculated as follows:

$$P_A = \frac{UI}{A} \quad (1)$$

$$P_V = \frac{UI}{V} \quad (2)$$

where I is the current (A), U is the voltage (V), A is the projected surface area of the electrode (m²), V is the available volume of anodic compartment. The volumetric power density indicates how much power is generated from unit volume of wastewater.

Coulombic efficiency (CE) is defined as the ratio of total coulombs actually transferred to the anode from the substrate to maximum possible coulombs if all substrate removal produces current. The CE (%) is calculated by:

$$CE = 100\% \frac{\sum_{i=1}^n U_i t_i}{RFb\Delta SV} M \quad (3)$$

here U_i is the output voltage of MFC at time t_i , F is Faraday's constant (96485 C mol⁻¹ electrons), b is the number of moles of electrons produced per mol of the COD (4 mole⁻ mol⁻¹ COD), ΔS is the removal of COD concentration (g L⁻¹), V is the liquid volume (L), and M is the molecular weight of oxygen (32 g mol⁻¹).

Polarization curves were generated by varying the external resistances from 20 to 8000 Ω . For each resistance, at least two sets of the same setup of each of the MFCs were operated to ensure that repeatable voltage outputs were achieved. Averaged voltages from the outputs were used to calculate the power density (W m⁻³ and W m⁻²).

3. Results

3.1. Power generation from the ferricyanide-cathode MFC

The ferricyanide-cathode MFC was operated with different concentrations of glucose and furfural, including (1) 1000 mg L⁻¹ glucose, (2) a mixture of 1000 mg L⁻¹ glucose and 0.1 mM furfural, (3) a mixture of 1000 mg L⁻¹ glucose and 0.2 mM furfural, (4) a mixture of 1000 mg L⁻¹ glucose and 0.5 mM furfural, (5) a mixture of 500 mg L⁻¹ glucose and 2 mM furfural, (6) a mixture of 200 mg L⁻¹ glucose and 5 mM furfural, and (7) 6.68 mM furfural. Voltage curves of 14 operation cycles are shown in Fig. 1, in which the letters A–G indicate the voltage cycles for substrates 1–7, respectively. All the maximum voltage outputs were in the range from 670 to 710 mV. The average operation period was less than 50 h (30–45 h) for each case of A–D. In the cases of E–G, the concentrations of glucose decreased from 500, 200 to 0 mg L⁻¹ and the furfural concentrations increased from 2, 5, to 6.68 mM. The curves of power generation of cases E–G were distinctive from those of cases A–D. The average operation period was prolonged to more than 50 h and the longest period was nearly 70 h.

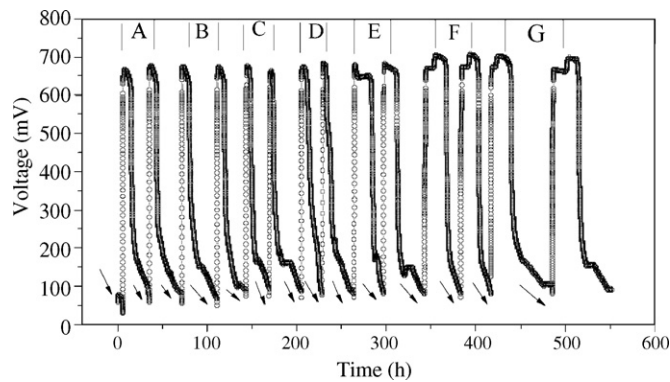


Fig. 1. Electricity voltage output of the ferricyanide-cathode MFC using glucose, glucose-furfural mixtures, and furfural as the fuels. (A) 1000 mg L⁻¹ glucose; (B) 1000 mg L⁻¹ glucose + 0.1 mM furfural; (C) 1000 mg L⁻¹ glucose + 0.2 mM furfural; (D) 1000 mg L⁻¹ glucose + 0.5 mM furfural; (E) 500 mg L⁻¹ glucose + 2 mM furfural; (F) 200 mg L⁻¹ glucose + 5 mM furfural; (G) 6.68 mM furfural. The arrows indicate the replacement time of the fuels.

Fig. 2 compares the differences among cases A, F, and G in details. When using 6.68 mM furfural as the sole fuel (case G), the time period for voltages higher than 600 mV was 30 h. However, when using 1000 mg L⁻¹ glucose as the sole fuel (case A), the time period was only 6 h. Interestingly, with almost the same values of COD of glucose, furfural could produce a much longer operation period for voltages higher than 600 mV than glucose in the ferricyanide-cathode MFC.

3.2. Power generation from the air-cathode MFC

After the stable power generation was achieved with 1000 mg L⁻¹ glucose, 6.68 mM furfural was replaced in the air-cathode MFC as the sole fuel. As shown in **Fig. 3**, in the first cycle, the maximum voltage was lower than 150 mV and the operation time was less than 20 h. However, the power generation gradually improved as time went on. After operation for five cycles, the maximum voltage output reached 420–450 mV and the longest period time keeping the high voltages was about 25 h. The cultivated time for furfural, i.e., the period from adding furfural to generating stable power, was about 140 h. The last cycle in **Fig. 3** was the result by replacing the furfural medium solution with a glucose solution of 1000 mg L⁻¹. Within 3 h of the replacement, the maximum voltage output reached 400–420 mV, which was totally different from the result reported by Catal et al. [13].

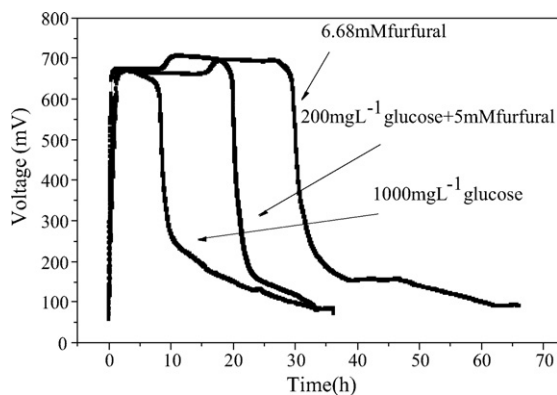


Fig. 2. Electricity voltage output obtained from the ferricyanide-cathode MFC using 1000 mg L⁻¹ glucose, a mixture of 200 mg L⁻¹ glucose and 5 mM furfural, and 6.68 mM furfural as the fuels. The three treatments have almost the same COD concentrations.

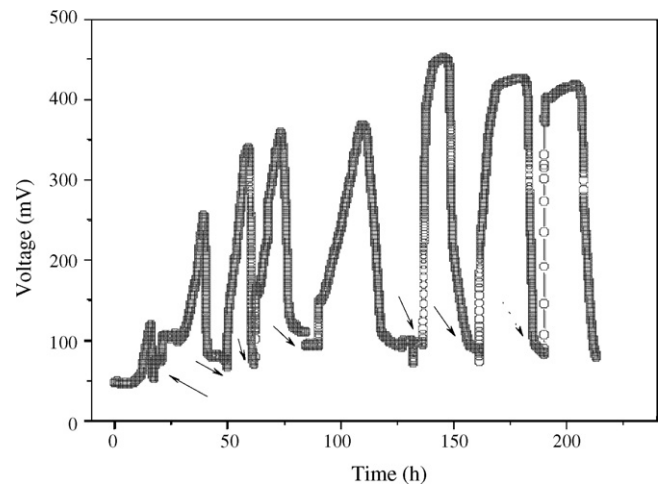


Fig. 3. Electricity voltage output obtained from 6.68 mM furfural and 1000 mg L⁻¹ glucose in the air-cathode. The solid arrow indicates the replacement time of the furfural medium and the dashed arrow indicates the replacement time of the furfural medium with 1000 mg L⁻¹ glucose medium.

3.3. Power density and coulombic recovery

The highest power density achieved from 6.68 mM furfural as the sole fuel in the air-cathode MFC was 361 mW m⁻² (18 W m⁻³) at a current density of 0.16 mA cm⁻² (**Fig. 4**). Correspondingly, the electrical charge and the CE were 34.6 C and 30.3%, respectively. The highest power density was higher than that from 1000 mg L⁻¹ glucose as the sole fuel (298 mW m⁻²). In the ferricyanide-cathode MFC, 6.68 mM furfural as the sole fuel resulted in the highest volumetric power density of 103 W m⁻³ at a current density of 169.1 A m⁻³ (**Fig. 5**). The total electrical charge and the CE obtained from the MFC were 86.6 C and 10.2%, respectively. The power density for the mixture of 200 mg L⁻¹ glucose and 5 mM furfural was 81.4 W m⁻³ at a current density of 150.4 A m⁻³. The electrical charge and the CE were 65.7 C and 7.1%, respectively. The lowest power density of 45.4 W m⁻³ was generated with 1000 mg L⁻¹ glucose at a current density of 79.4 A m⁻³. The electrical charge and the CE were 35.1 C and 4%, respectively. For the three fuels of 6.68 mM furfural, the mixture of 200 mg L⁻¹ glucose and 5 mM furfural, 1000 mg L⁻¹ glucose, their COD values were almost the same. However, the highest volumetric power density, the total electrical charge, and the CE values increased with increasing of the furfural concentrations (or decreasing of the glucose concentrations).

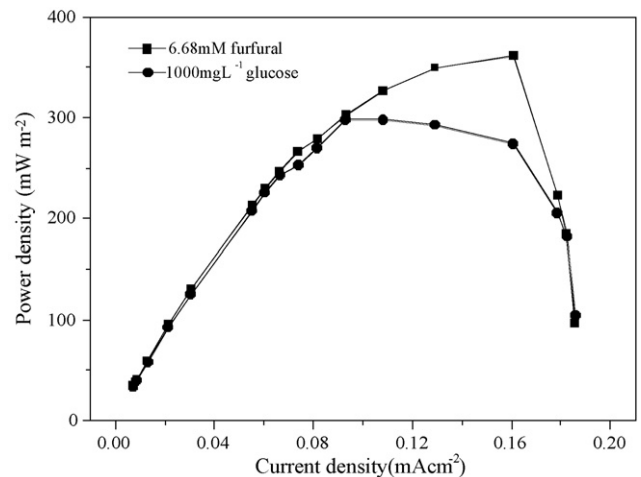


Fig. 4. Power densities as a function of current density using furfural and glucose as the fuels in the air-cathode MFC.

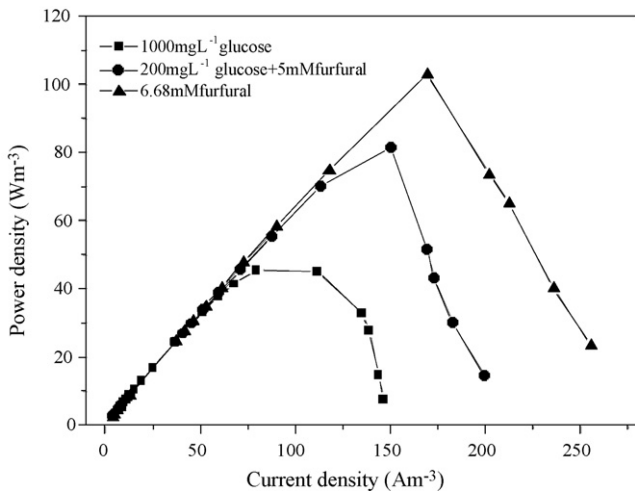


Fig. 5. Power densities as a function of current density using glucose, glucose–furfural mixture, and furfural as fuels in the ferricyanide-cathode MFC.

3.4. Substrate utilization and furfural degradation

The COD removal reached up to 68% using the air-cathode MFC with 6.68 mM furfural as the sole fuel. The COD removal rates were in the range of 82–96% in the ferricyanide-cathode MFC (Fig. 6). It took more than 60 h to completely degrade 6.68 mM furfural but the removal efficiency reached up to 95% within 12 h. The 1000 mgL⁻¹ glucose as the sole fuel was completely degraded between 30 and 35 h in the ferricyanide-cathode MFC. However, the complete degradation time for 1000 mgL⁻¹ glucose decreased to less than 8 h

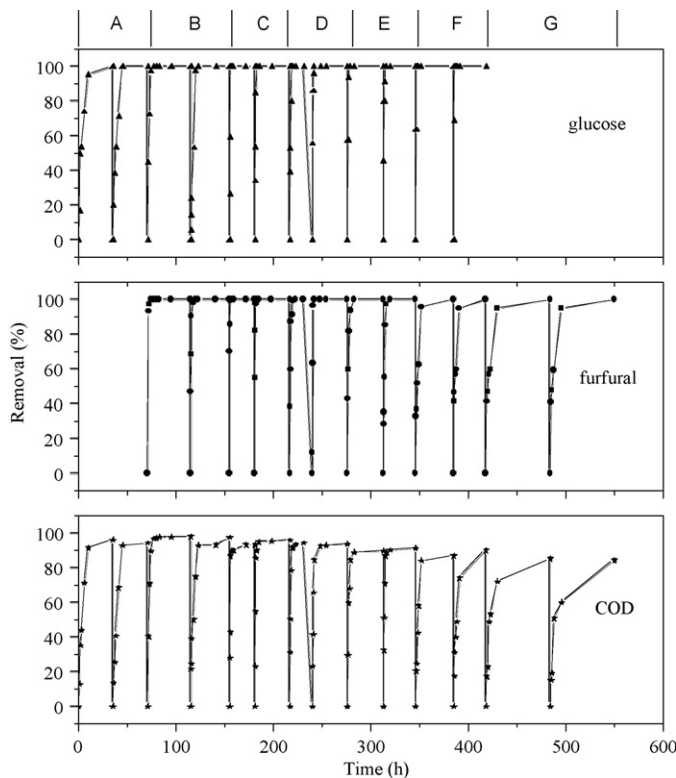


Fig. 6. The removal of glucose, glucose+furfural mixture, and furfural obtained from the ferricyanide-cathode MFC (A) 1000 mgL⁻¹ glucose; (B) 1000 mgL⁻¹ glucose + 0.1 mM furfural; (C) 1000 mgL⁻¹ glucose + 0.2 mM furfural; (D) 1000 mgL⁻¹ glucose + 0.5 mM furfural; (E) 500 mgL⁻¹ glucose + 2 mM furfural; (F) 200 mgL⁻¹ glucose + 5 mM furfural; (G) 6.68 mM furfural).

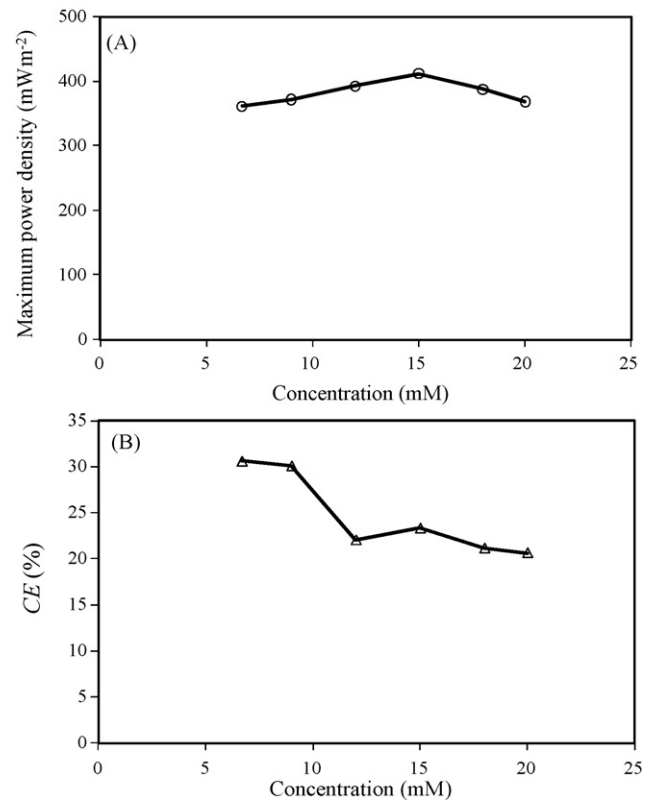


Fig. 7. Maximum power densities (A) and Coulombic efficiencies (CE) (B) as a function of furfural concentrations in the air-cathode MFC.

when a furfural solution with concentrations of 0.1, 0.2, and 0.5 mM was added into the anode solution (cases B–D in Fig. 6). The experimental results suggested that the co-substrates between furfural and glucose could accelerate the glucose degradation significantly. Fig. 6 also shows that the degradation time for furfural increased with the furfural concentrations. For example, it took only 6 h for 2 mM furfural to be completely degraded in the MFC, while it took more than 60 h for 6.68 mM furfural to be completely degraded.

In order to ascertain the effect of furfural on the characteristics of MFC, furfural concentrations increased from 6.68 to 20 mM in the air-cathode MFC. Higher furfural concentrations resulted in longer operation periods. For example, the operation period was 30 h for 6.68 mM furfural, whereas the operation period was 58 h for 20 mM furfural. Furthermore, the maximum power densities increased from 6.68 to 15 mM (Fig. 7A). Further increase in furfural concentration to 20 mM led to the slight reduction of the maximum power density to 368 mW m⁻². The CE values were significantly decreased from 30.3 to 20.6% when the furfural concentration increased from 6.68 to 20 mM (Fig. 7B). The experimental results demonstrated that furfural could be used as the fuel of the MFC even if the concentrations were high up to 20 mM.

4. Discussions

Catal et al. [13] reported that voltage output was severely inhibited when added 0.2 mM furfural to 1200 mgL⁻¹ glucose medium in a single-chamber air-cathode MFC. They further showed that electricity generation was not recovered upon a replacement of the furfural medium with a glucose solution. However, in our study, furfural was utilized as the sole fuel in the single-chamber air-cathode MFC to generate electricity successfully. Within 3 h of the replacement of the furfural medium solution with a glucose

solution of 1000 mg L⁻¹, the maximum voltage output reached 400–420 mV. The result indicated that microorganisms in the MFC could adapt glucose quickly after adapting furfural. Nevertheless, the cultivated time period for furfural (140 h) was longer than those for arabinose (60–70 h), arabitol (45 h), and xylitol (70 h) [28,29], which suggested that microorganisms in the MFC needed longer time to adapt furfural. Various bacteria have been reported to degrade furfural. For example, the enteric bacteria including the genera *Klebsiella*, *Enterobacter*, *Escherichia*, *Citrobacter*, *Edwardsiella* and *Proteus*, are able to degrade furfural [15]. Boopathy [30] shows that the metabolic activities of both *Desulfovibrio strain B* and *Methanosarcina barkeri 227* are essential for the complete degradation of furfural. Sulfate-reducing bacterium *Desulfovibrio strain B* can convert furfural to acetic acid, which can be converted to methane by *Methanosarcina barkeri 227*. In the current study, the furfural utilization by bacteria might be attributable to the use of mixed bacterium culture from the anaerobic and aerobic sludge after a period of acclimation in MFCs. It is also demonstrated that the mixed bacteria generate power from recalcitrant contaminants such as phenol [7].

When using 6.68 mM furfural as the sole carbon source, the maximum power density achieved in the air-cathode MFC was 361 mW m⁻², which was higher than these reported with glucose (262 mW m⁻², 600 mg L⁻¹) and butyrate (305 mW m⁻², 1000 mg L⁻¹) as the carbon sources in the MFC [31,32]. Furthermore, with almost the same values of COD, the maximum power density generated from furfural was even higher than that from glucose using the ferricyanide-cathode MFC and the air-cathode MFC. The results may be explained as follows. Firstly, some microbes, including sulfate-reducing bacteria *Desulfovibrio* sp [19,30], in the anode of the MFC are capable of converting furfural to acetate, which is also good substrate for electricity-generating microbes [8,32]. Secondly, furfural might be utilized by a more broad range of electricity-generating bacterial species than glucose, which resulted in higher maximum power density. It is conceivable that certain microbes in the mixed culture used in this particular study had the ability to adapt to furfural better than to glucose. Nevertheless, further research is needed to identify the microbial community and to analyze the mechanism of electricity generation from furfural.

The result of the *CE* values decreasing with the furfural concentrations in the air-cathode MFC was most probably attributable to the oxygen diffusion. Higher furfural concentration resulted in longer operating time, therefore, much more oxygen diffusing through the cathode and entering into the anode solution. Consequently, the limited electrons were consumed by the excess oxygen and then the *CE* decreased [11,33].

In addition to power generation, the MFCs also significantly degraded substrates during the operations. In the ferricyanide-cathode MFC, 100% of furfural was removed at the end of each cycle (Fig. 6). When 6.68 mM furfural was used as the sole fuel, the COD removal in the ferricyanide-cathode MFC was 15% higher than that in the air-cathode MFC. The higher COD removal achieved by the ferricyanide-cathode MFC mainly related to three factors: (1) the brush fiber material adsorbed more microbes than the carbon-cloth because of its larger surface area [34]; (2) ferricyanide as the cathode electron acceptor reduced the reaction activity in the MFC due to its higher oxidizing potential than oxygen; (3) the MFC operated with ferricyanide as catholyte avoided the influence of oxygen to the substrate removal.

A comparison of Figs. 1 and 6 indicated that the operation periods in the MFC sustained for a long time after most of the COD and furfural were removed quickly. Freguia et al. [35] reported that the acetate and glucose in the MFC were loaded as pulse carbon, stored inside the cells during initial high substrate conditions, and consumed during starvation. In our study, the substrates in

the MFCs were likely stored quickly inside the cells, resulting in the fast removal of the COD and furfural. Afterward, the substrates were consumed gradually to sustain electricity generation. In addition, some possibly intermediates from furfural degradation, such as acetate [19,30], also played an important role in sustaining electricity generation.

5. Conclusions

Electricity was successfully generated using furfural as the sole fuel in the ferricyanide-cathode MFC and the air-cathode MFC for the first time. Furfural at a concentration up to 20 mM could also be utilized to produce electricity in the air-cathode MFC. The maximum power density from furfural as the sole fuel was even higher than that from glucose using the two different MFCs. Moreover, 100% furfural was degraded at the end of each operation cycle in the ferricyanide-cathode MFC. These results indicate that the MFC technology should be a potential method for the biological treatment of furfural-containing wastewater and energy output.

Acknowledgements

This work was partially supported by grants from the Natural Science Foundation of China (Nos. 50608070 and 50779080) and the Research Fund Program of Guangdong Provincial Key Laboratory of Environmental Pollution Control and Remediation Technology (No. 2006K0007).

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