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Short communication

Surface floating, air cathode, microbial fuel cell with horizontal flow for continuous power production from wastewater

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ABSTRACT

A surface floating, air cathode, microbial fuel cell (MFC) with a horizontal flow is devised and characterized using glucose-based synthetic wastewater. The performance of the MFC is significantly affected by the current-collector of the electrodes. When graphite foil ribbon (150 cm) serves as the current-collector, the respective specific internal resistance and maximum power density are 0.362 $\Omega\,m^{-2}$ and 124.0 $W\,m^{-3}$. The internal resistance can be reduced by increasing the length of the current-collector. For a graphite ribbon current-collector 256 cm long, the specific internal resistance is only 0.187 $\Omega\,m^{-2}$ and the maximum power density markedly increases to 253.6 $W\,m^{-3}$; however, the maximum power density is affected by the current-collector material. When the current-collector is changed to a stainless-steel wire, the maximum power density is reduced to approximately 100 $W\,m^{-3}$ because of its high liquid|solid interfacial impedance. During three continuous months of operation, issues such as leaking are not observed and as such, the MFC could be easily scaled-up for wastewater treatment by increasing the electrode size and stacking a number of cells without additional ohmic resistance.

1. Introduction

Microbial fuel cells (MFCs) directly convert chemical energy contained in organic matter into electric power using electrochemically active microorganisms as a catalyst. The cells are an emerging green technology for the treatment of wastewater [1–6]. By adopting MFCs for wastewater treatment, the expensive aeration for aerobic biological treatment is precluded as the organic pollutant is degraded under anaerobic conditions, and biological sludge production is reduced by the low yield of electrochemically active microorganisms [4–7]. Therefore, it is possible to reduce the operational cost of wastewater treatment plants and produce electric power from wastewater.

The earliest MFC concept was demonstrated by Potter in 1910, who produced electrical energy from living cultures of *Escherichia coli* and *Saccharomyces* using platinum electrodes [8]. Interests in MFCs for wastewater treatment, however, began in earnest when microbes capable of directly transferring electrons without the addition of an external mediator to the anode were found in 1999 [9]. Since then, numerous studies of MFCs have been performed, including electron-transfer mechanisms, environmental conditions affecting the performance of electric power generation, electrode materials, and design and configurations [10–15].

Recently, MFC performance (power density and coulomb efficiency) has been significantly improved by an increased understanding of the electron-transfer mechanisms and the adoption of new electrode materials. Ten years ago, the power density from a square meter of anode was only several $\mu W\,m^{-2}$, but has now increased to 5850 $mW\,m^{-2}$ [16,17]. Regarding MFC design and configuration for wastewater treatment, two chamber MFCs with liquid cathode were redesigned to a single chamber with an air cathode, including tubular, flat plate, horizontal flow, and cassette type MFCs [6,10,18–22]. A number of hurdles, however, have still to be overcome before effective wastewater treatment is achieved [4,5,22–24], namely: (i) higher power production, compared with existing anaerobic digestion that recovers methane or hydrogen from organic waste; (ii) new design and configuration that promises ease of scale-up, operation and maintenance; (iii) process stability under perturbation conditions to cope with high variations in wastewater characteristics and flow rate; (iv) better effluent quality to meet the discharge standard for the wastewater; (v) reduction in the cost for construction, equipment, and materials. Specifically, the power density produced by a continuous-flow MFC designed for wastewater treatment is currently as small as 100 $W\,m^{-3}$, which is too low to compete with existing anaerobic digestion [4,5,21,23]. Furthermore, it is difficult to find a MFC design

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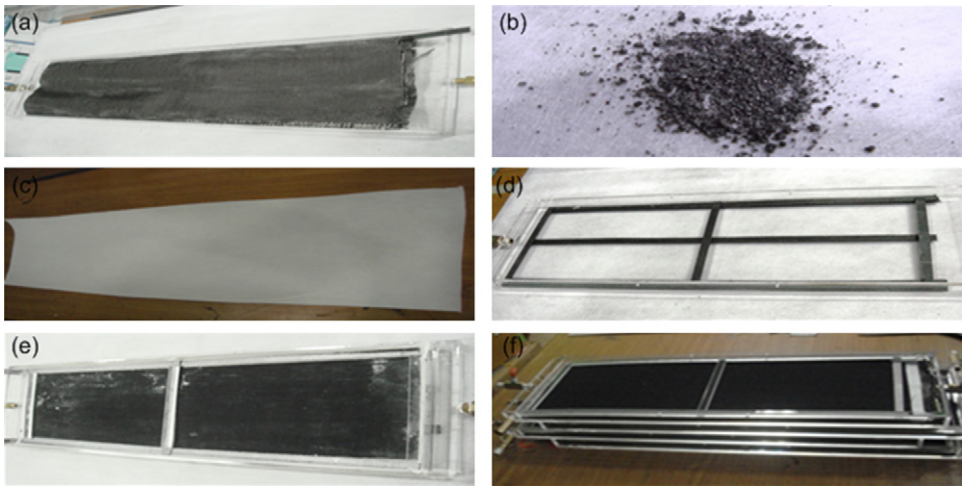

Fig. 1. Surface floating, air cathode, MFC system with horizontal flow and its parts. (a) Graphite fibre fabric sheet for anode. (b) Graphite granules (dia. <1 mm) for anode. (c) Separator between anode and cathode. (d) Graphite ribbon for current-collector. (e) Surface floating, air cathode, MFC. (f) Stacked MFC system.

and configuration capable of simple scale-up and maintenance during continuous wastewater treatment.

In the present study, a new MFC design with a horizontal flow has been devised to ensure high power density and ease of scale-up and operation. The influence of some operational conditions of the MFC, such as influent chemical oxygen demand (COD), horizontal flow velocity, and current-collector on the power production is also examined.

2. Materials and methods

2.1. MFC construction

A new surface floating, air cathode, MFC with a horizontal flow is shown in Fig. 1. The device consists of a thin rectangular box type acrylic reactor (15 cm × 50 cm × 2.5 cm), anode, cathode, and separator. For the anode, a graphite fibre fabric (projected area = 750 cm^2) was prepared and placed on the bottom of the reactor. Graphite granules (dia. 0.3–1.0 mm) were then spread out on the carbon fabric to provide pores for the passage of wastewater and to increase the surface area of the anode. A graphite cloth with platinum (5.0 g m^{-2}, GDE LT250EW, E-TEK) served as the cathode material. The cathode was floated on the wastewater surface with one side (bearing the platinum) in contact with water and the other side exposed to air. In order to avoid short-circuiting, an unwoven polyethylene sheet was inserted as a separator between the anode and cathode. The thickness of the anode chamber between the reactor bottom and the separator was approximately 0.25 cm. A current-collector was placed on the surface of the anode and cathode to collect electrons from the electrodes (Fig. 1d).

Each of the three arrangements of the graphite ribbon and the meshes of the stainless-steel wire were used as the current-collector. The current-collectors for the anode and cathode were connected to make a circuit; a variable resistance was installed in the circuit.

2.2. Artificial wastewater and MFC operation

During MFC start-up, effluent from another MFC operated over three months was continuously added into the anode chamber for inoculation. After five days, the effluent was switched with synthetic wastewater composed of glucose, macro- and micronutrients, and sodium bicarbonate for alkalinity. The COD, electric conductivity and pH of the influent wastewater were 300–1000 mg l^{-1}, 21 mS cm^{-1} and 7.0–7.2, respectively. A portion of the MFC effluent that collected in a settling tank was recycled into the MFC. The recycle ratio of the effluent, compared with the feeding wastewater, was 0–2.96. The MFC system was operated at 30 °C in a constant temperature box and the hydraulic retention times (HRTs) were controlled to 31.4–42.4 min. The detailed operating conditions for the MFC are summarized in Table 1.

2.3. Calculation and analysis

During MFC operation, both the voltage and current were monitored using a DMM (digital multimeter, Keithley 2700) connected to a computer. Other physico-chemical parameters for the effluent, such as pH, COD, and electro-conductivity were also analyzed. The open-circuit voltage (OCV) and short-circuit current (SCC) were intermittently measured with the DMM either after disconnecting the copper wire circuit between the electrodes or removing the resistance load from the circuit. The power (W) generated from the MFC was estimated by the product of current (A) and voltage (V). The specific internal resistance (R_{int}, Ω m^{-2}) based on projected area of anode (m^{-2}) was obtained from the linear slope of the voltage *versus* current curve (polarization curve), which was obtained by changing the external resistance. The internal resistance was characterized by electrochemical impedance spectroscopy using a Nyquist plot. The impedance measurements were taken from 1.0 to 10^5 Hz by applying a sine wave (10 mV rms) with an impedance analyzer (CompactStat, Ivium Technologies, Eindhoven, The Netherlands).

Table 1
Experimental conditions for surface floating, air cathode, MFC with horizontal flow.

RUN	GR-1	GR-2	GR-3	SSW
COD_{in} (mg l^{-1})	300	600	600	1,000
Flow rate (l d^{-1})	8.6	7.7	7.7	6.37
Recycle ratio (Q_i/Q_r)	1.0	2.78	2.96	0–2.42
HRT (min)	31.4	35.07	35.07	42.4
Horizontal flow velocity (cm min^{-1})	3.18	5.39	5.65	1.18–4.03
Current-collector type	GFR	GFR	GFR	SSW
Current-collector length (cm)	150	189	265	–

GFR: graphite foil ribbon with 10 mm in width; SSW: stainless-steel wire mesh with 0.5 mm in wire diameter and 5 mm mesh opening.

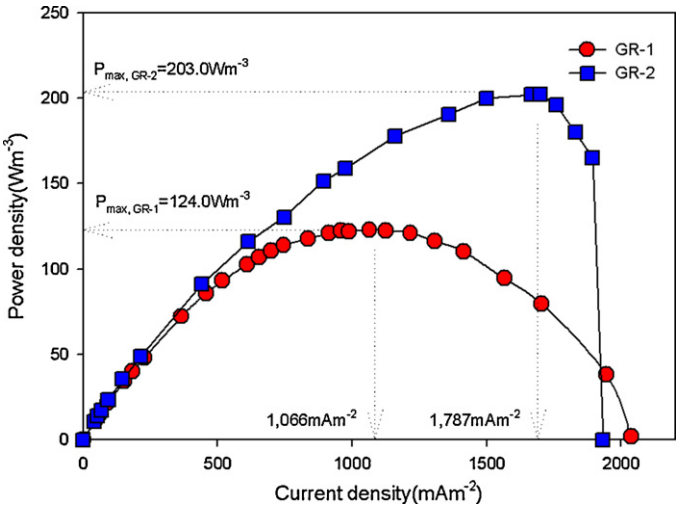

Fig. 2. Power output of surface floating, air cathode, MFC with horizontal flow in GR-1 and GR-2.

3. Results and discussion

3.1. Performance of surface floating, air cathode, MFC with horizontal flow

During three-month continuous operation of the MFC (GR-1), both the voltage and current remained stable after a start-up period of a couple of weeks. In order to investigate the dependence of voltage and power on the current, a polarization curve was obtained at a steady state (Figs. 2 and 3). The OCV was 0.65 V, and the maximum power density 124.0 W m^{-3} at a current density of 1066 mA m^{-2}. It is possible that the maximum power density of the MFC could be affected by the OCV. With oxygen as an electron acceptor in the cathode, the OCV is theoretically estimated to be approximately 0.82 V, but from the experiment, it is usually reported to have a smaller value between 0.5 and 0.8 V [4–6]. The substrate crossover and unwanted side-reactions on both the anode and cathode, collectively called parasitic losses, account for the difference between the measured OCV and its theoretical value [25]. It is likely that the small OCV of 0.65 V in GR-1 might be due to an increase in anode potential caused by oxygen diffusion from the air cathode, and a reduction in cathode performance by chemical deposits or biofilm

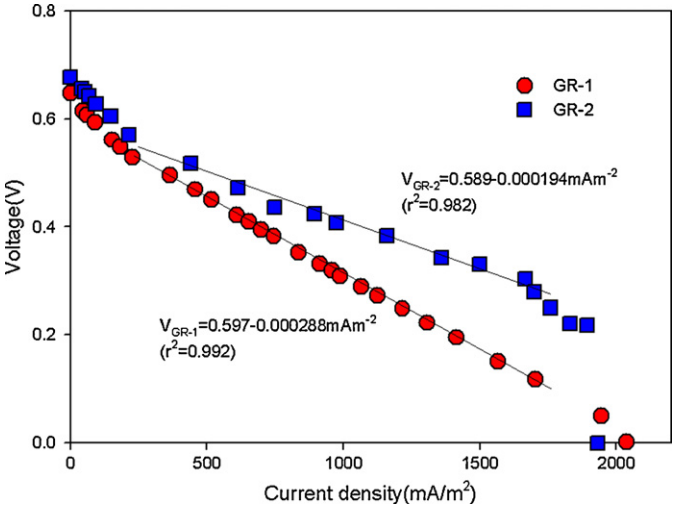

Fig. 3. Polarization of surface floating, air cathode, MFC with horizontal flow in GR-1 and GR-2.

growth on the electrode surface. The volumetric maximum power density of 124.0 W m^{-3} in GR-1 is higher than the 50.2 W m^{-3} of a membrane-less MFC with graphite granules using an air cathode [18], and comparable with 129 W m^{-3} of a cassette electrode MFC with the cathode [19]. Although the statistical significance of the power density is not provided, it is highly reproducible because the polarization for the continuous MFC system is obtained at a steady state. The surface power density in the study is only 311.2 mW m^{-2}, indicating that the ratio of the surface area of the cathode to the anode volume of the MFC (4.0 m^2 m^{-3}) in this study is a very important design factor for high power density; this finding is similar to that of a previous report [26]. The maximum power density of a MFC is generally determined by internal losses, including activation, concentration, bacterial metabolism and ohmic losses, as well as the OCV [6,23]. For the polarization curve, the voltage was a linear function of the current over a wide range (182.4–1945.6 mA m^{-2}). The total specific internal resistance obtained from the polarization curve slope is 0.362 Ω m^{-2} (Fig. 3), i.e., smaller than other types of air cathode MFCs reported in the literature [18,20]. Activation losses appear vaguely at a low current density range, whereas the concentration losses appear are manifested at high current density. The low activation losses are achieved by a large electrode surface, a well-developed active biofilm on the anode surface, and a high oxygen reduction rate on the cathode by a Pt catalyst [6].

In addition, a horizontal flow velocity of 3.18 cm min^{-1}, provided by both the short HRT (31.4 min) and recycling of the effluent, is attributed to small concentration losses. It is well known that ohmic losses are determined by the transport of both electrons and protons [1,6], and that small ohmic losses can be described by details considered in the design and configuration of the MFC; First, in order to ensure closure of the space between the electrodes, the cathode is floated over the separator that covers the anode, with a thin porous unwoven polyethylene sheet used as a separator instead of a proton-exchange membrane. Second, a graphite ribbon is used as a current-collector to reduce the resistance to electron flow in the electrodes. Generally, ohmic losses depend significantly on the resistance of the electrode materials if a large electrode is used. Third, a high electric conductivity of feed wastewater (20 mS cm^{-1}) is adopted as a good proton transport in the liquid phase. Good performance of the MFC in GR-1 implies that several of the above considerations in the design and operation are effective for a small internal resistance.

In the GR-2, the COD of the feed wastewater and the recycle ratio are increased to 600 and 2.78 mg l^{-1}, respectively, and a slightly longer current-collector is employed, i.e., nearly 26% longer than the graphite ribbon used GR-1 (Table 1). The OCV is 0.68 V, which is slightly higher than the GR-1, but the maximum power density is significantly increased to 203.0 W m^{-3} at an increased current density of 1787 mA m^{-2}, compared with the GR-1 (Fig. 2). The specific internal resistance obtained from the polarization curve is 0.258 Ω m^{-2}, namely, only 71% of the GR-1. It is possible that the increased maximum power density in the GR-2 is obtained by the increased OCV and the reduced internal resistance. In a previous study [27], adopting Monod kinetics for describing the bioelectrochemical reaction, the saturation constant was 115–144 mg COD l^{-1}. Therefore, a substrate concentration of 300–600 mg COD l^{-1} in both GR-1 and GR-2 is sufficient to maintain the metabolic activity of the bacteria; however, mass transfer on the anode surface might be enhanced by a horizontal flow velocity of 5.39 cm min^{-1} in GR-2, which is grater than the 3.18 cm min^{-1} of GR-1. In the MFC anode chamber, the oxygen could be generally diffused from an air cathode or combine with the feed wastewater; nevertheless, the oxygen could be removed during the substrate degradation activity of aerobic or facultative bacteria if an organic

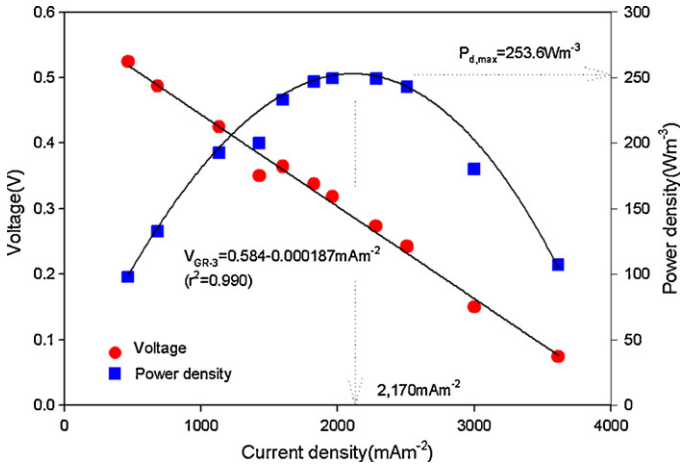

Fig. 4. Polarization of surface floating, air cathode, MFC with horizontal flow (GR-3), according to current density.

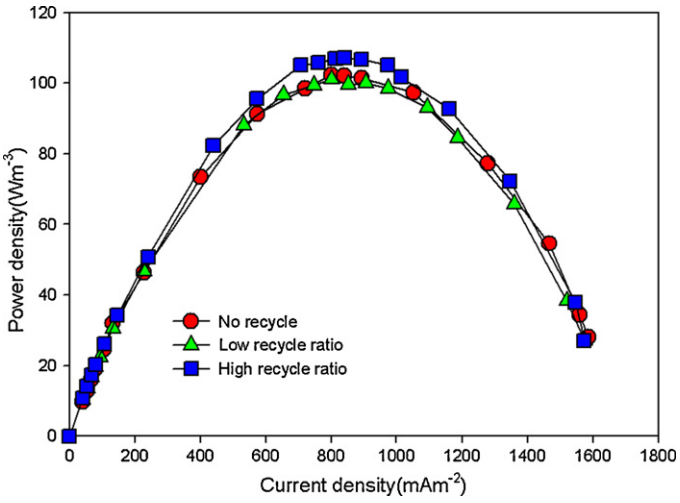

Fig. 5. Dependence of power on current in polarization curve for stainless-steel wire as current-collector, according to recycle ratios.

substrate was in high enough concentration in the anode chamber [6,28]. In this study, the OCV in the GR-2, compared with the GR-1, is increased by the reduced anode potential through are increased removal rate of bacterial oxygen in the anode chamber due to the higher substrate concentration (600 mg COD l^{-1}). It is likely that the longer current-collector in the GR-2 is mainly responsible for the small internal resistance, compared with that of GR-1.

In GR-3, in order to delineate the effects of the current-collector on internal resistance and power generation, the length of the current-collector was increased slightly to 265 cm (40% longer than in the GR-2), with the other design and operation parameters for the MFC similar to the GR-2 (Table 1). From the polarization curve (Fig. 4), at a current density of 2170 mA m^{-2}, the maximum power density is 253.6 W m^{-3}, one of the highest power densities reported from a scalable and larger MFC. The specific internal resistance obtained from the linear slope of the polarization curve is 0.187 Ω m^{-2}, i.e., just 72.5% of the GR-2. This implies that a large portion of the internal resistance consists of an ohmic resistance associated with the electrode resistances, and the longer current-collector is a very important factor in the design for a high-performance MFC.

3.2. Stainless-steel wire as a current-collector

The power density of the MFC can be increased if electron transport from the anode to the cathode is enhanced through reduction in the ohmic resistance. A highly conductive current-collector, especially for an MFC with a substantial electrode using carbon-based materials, is helpful for electron transport in the electrodes. In SSW, a stainless-steel wire with a lower resistance (10–100 μΩ cm), instead of graphite ribbon (approximately 900 μΩ cm), is used as the current-collector. After adjusting the incoming COD to 1000 mg l^{-1}, the performance of the MFC was examined at different recycle ratios, from 0 to 2.42 (Table 1). Without recycling the MFC effluent, the maximum power density is 101.1 W m^{-3}, at a current density of 860 mA m^{-2} (Fig. 5); unfortunately, it is smaller than that of graphite ribbon, which has a higher resistance as a current-collector. The specific internal resistance obtained from the polarization (0.467 Ω m^{-2}) is also higher than that of the graphite ribbon (Fig. 6). These results were not predicted because the stainless-steel wire is a more conductive material compared with the graphite ribbon of the GR-3. The internal resistance obtained from the Nyquist plot consists of 30% ohmic resistance, 23% charge-transfer resistance, and 44% diffusion resistance (Fig. 7). It has been reported that some high conductiv-

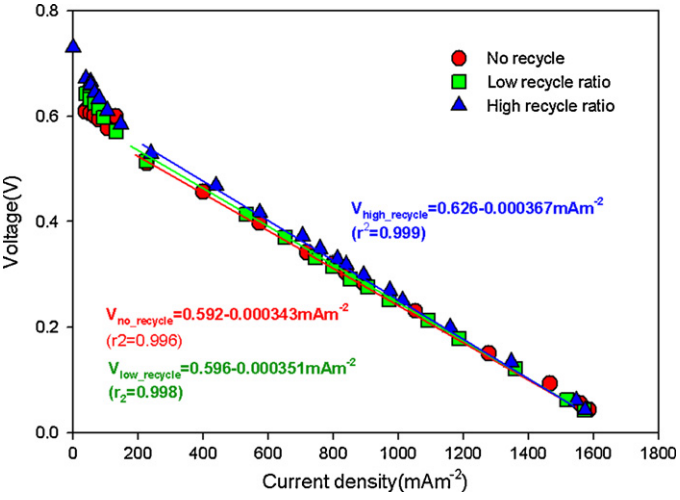

Fig. 6. Dependence of voltage on current in polarization curve for stainless-steel wire as current-collector, according to recycle ratios.

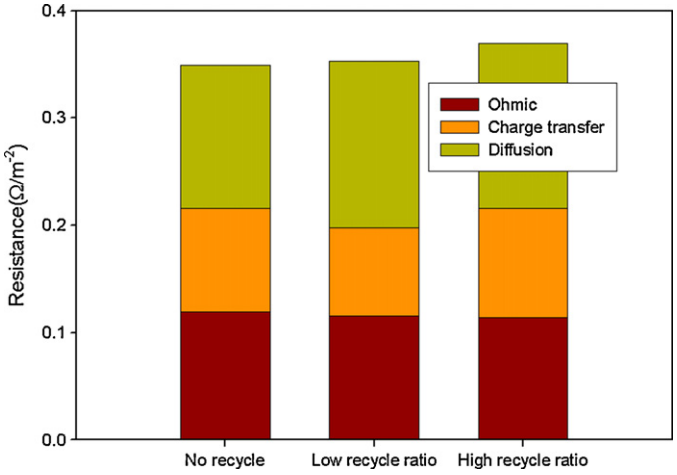

Fig. 7. Internal resistances of MFC with stainless-steel wire as current-collector, according to recycle ratios.

ity metals including stainless-steel, nickel, and aluminum, behave like a polarized electrode with a high electrode|solution interfacial impedance [29], indicating that this impedance, as well as conductivity, biocompatibility, high surface area and porosity, corrosion resistance, and price, is an important factor in the material selection for the electrode and the current-collector [24,30]. When the recycle ratio of the effluent is increased from 0 to 2.42, the OCV values increase from 0.62 to 0.72 V. The maximum power densities, however, are not much different from that obtained without recycling of the effluent (Fig. 5). It seems that the solid|liquid interfacial impedance for the charge-transfer between the anode and current-collector is not affected by flow turbulence. Therefore, in order to improve MFC performance using a highly conductive current-collector, it is important to use a current-collector with low solid|liquid interfacial impedance or to avoid interfacial impedance through integration of the current-collector and the electrode.

3.3. Implication for design, operation, and maintenance of MFC

Operation of the MFC devised in this study is very simple and straightforward as both feed supply and recirculation of the effluent (using pumps) are at sufficient levels. In particular, during continuous operation of the MFC, issues with leakage through the cathode or the wetted surface of the cathode, common for a vertical type of air cathode MFC, do not occur. For application in genuine wastewater, the electrode size of the MFC could be enlarged without additional internal resistance through use of a highly conductive current-collector, and the subsequent space is easily saved by stacking the cells, as shown in Fig. 1e. In a stacked MFC system, the influent wastewater and effluent for recirculation are only supplied to the inlet zone at the top of the MFC cells. Therefore, the running costs of the stacked system will be reduced. Overall, the surface floating, air cathode, MFC with a horizontal flow is a scalable MFC for wastewater treatment. Nevertheless, during continuous operation in this study, some capricious problems concerning maintenance of the MFC were encountered. First, after lengthy periods of operation, some chemical deposits and microbial slime were observed on the water-side surface of the cathode. Both are suspected to affect catalytic oxygen reduction on the cathode surface [5,31] and cause problems concerning the floating of the cathode as the specific gravity of the cathode can be increased by both deposits and slime. On the other hand, the slime on the cathode surface may be useful in terms of preventing oxygen diffusion from the cathode side into the anodic compartment [6,22,28]. In order to avoid the floating problem of the cathode, a frame supporting the cathode floating on the water surface proves helpful. Second, in the inlet zone of the MFC, aerobic microorganisms using dissolved oxygen contained within the feeding wastewater can grow and cause clogging of the path for water flow. Clogging in the inlet zone can hinder even distribution of the wastewater flow in the MFC. An artificial flow channel in the anode chamber would be helpful for distribution of wastewater flow. Third, there exists a certain degree of bubble formation due to gaseous products from the anodic reaction, and this can hinder proton transport to the cathode. Removal of these bubbles by placing the electrode at a slope to the horizontal plane might prove helpful. The bubbles could be also removed through a porous cathode or a vent in the cathode. In the case of a sloped MFC, it is expected that the concentration overpotential is reduced by increasing the flow velocity of the wastewater. Although the glucose-based synthetic wastewater is benign, compared with the complex genuine wastewater, the results obtained from the study provide some valuable information concerning design and operation of the treatment process for genuine wastewater.

4. Conclusions

A new surface floating, air cathode, MFC with a horizontal flow has been devised, and the factors affecting the performance examined. The following conclusions are drawn. Some considerations in design and operation of the MFC, including distance between electrodes, length and type of current-collector, surface area to volume ratio, substrate concentration and horizontal flow velocity, influence the maximum power density of the MFC. Graphite ribbon is a good current-collector for both the anode and the cathode with an internal resistance that is a function of the current-collector length. The maximum power density obtained from the MFC is 253.6 $W\,m^{-3}$ at a current density of 2170 $mA\,m^{-2}$ when using a graphite ribbon current-collector. By contrast stainless-steel wire is interior as a material for the current-collector due to its high liquid|solid interfacial impedance. The MFC is simple to scale-up without additional ohmic resistance and operation and maintenance is straightforward. It is concluded that the surface floating, air cathode, MFC with horizontal flow is a scalable configuration for wastewater treatment.

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