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Continuous electricity production from artificial wastewater using a mediator-less microbial fuel cell

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Abstract

A microbial fuel cell (MFC) was optimized in terms of MFC design factors and operational parameters for continuous electricity production using artificial wastewater (AW). The performance of MFC was analyzed through the polarization curve method under different conditions using a mediator-less MFC. The highest power density of 0.56 W/m^2 was achieved with AW of 300 mg/l fed at the rate of 0.53 ml/min at 35 °C. The power per unit cell working volume was 102 mW/l, which was over 60 times higher than those reported in the previous mediator-less MFCs which did not use a cathode or an anode mediator. The power could be stably generated over 2 years.

Keywords: Microbial fuel cell; Mediator; Power density; Current density; Polarization curve

1. Introduction

A microbial fuel cell (MFC) generates electricity directly from electron donors through the microbial activity (Allen and Bennetto, 1993). Most studies to date have been devoted to mediator aided MFCs. Our work has shown that an MFC can be operated using electrochemically active microorganisms without mediators (Kim et al., 1999a,b, 2002, 2004). The mediator-less MFC systems have been studied for the production of electricity from biodegradable organics in wastewater (Bond and Lovley, 2003; Chaudhuri and Lovley, 2003; Gil et al., 2003; Jang et al., 2004) and marine sediment (Reimers et al., 2001; Bond et al., 2002; Tender et al., 2002), and as a biochemical oxygen demand sensor (Chang et al., 2004; Kang et al., 2003; Kim et al., 2003a,b). Wastewater treatment using an MFC is promising since this process converts the major part of the chemical energy of the contaminants to electricity thereby reducing the generation of excess sludge (Jang et al., 2004; Kim et al., 2004). When a complex wastewater was used as the fuel, biofilm formed onto the anode in addition to microbial clumps loosely associated with the electrode. The microbial clumps are believed to ferment the complex fuel to simple fermentation products, which are oxidized by the electrochemically active microorganisms in the biofilm (Kim et al., 2004).

Studies have been made to improve MFC performance. Gil et al. (2003) have reported the effect of operational factors on the performance of the mediator-less MFC. Park and Zeikus (2000, 2002, 2003) have reported the improvement of the MFC performance through the introduction of functional mediators to the electrodes to

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increase their electron transfer efficiency. Schröder et al. (2003) obtained a power density of 6.0 W/m^2 using polyaniline-modified platinum as an anode. Although the power density is high, this MFC system needs improvements to overcome the inherent drawbacks such as the requirement of an additional reactor for fermentation, the use of a complex anode of polyaniline-modified platinum, and the need for a potential-pulse operation to maintain the catalytic activity of the anode. Bond and Lovley (2003) showed the potential of *Geobacter sulfurreducens* as a biocatalyst for an MFC. Rabaey et al. (2003) claimed that a power density of 3.6 W/m^2 could be obtained from an MFC using 100 mM ferricyanide as the cathode mediator. However this power density was not constant and several power peaks were obtained during the limited period of less than 5 days. Few studies have demonstrated long-term electricity production from MFCs. Chaudhuri and Lovley (2003) reported the operation an MFC using *Rhodoferax ferrireducens* for 900 h in repeated batch mode. However, the power density was low, viz. 8.2 mW/m^2.

In the present study, we show that a mediator-less MFC can be used for continuous and stable electricity production from an artificial wastewater. This is the first report on the continuous and long-term production of electricity using MFC. In this report, the MFC was optimized to improve the electricity production in two aspects consecutively, namely, the cathode design and the operating parameters.

2. Methods

2.1. Microbial fuel cell (MFC)

"Sensor-type" MFC, which was developed as a continuous BOD sensor (Chang et al., 2004), was used for continuous electricity production. The working volume of each compartment was 20 ml. The apparent surface area of each electrode was 48 cm^2. In some experiments, platinum-coated graphite felt was used as the cathode. Platinum powder was spray-coated onto graphite felt at the ratio of 0.4 mg/cm^2 according to the method described previously (Pham et al., 2004). The MFCs used were enriched and maintained for over 2 years. The potential between anode and cathode was measured using a multimeter (Model 2700, Keithley Instruments Inc., Cleveland, OH, USA) linked to a differential multiplexer (Model 7701, Keithley Instruments Inc.). Data were recorded digitally on a personal computer using an interface card (Model PCI-488 Keithley Instruments Inc.) every minute. The multimeter was controlled using software (TestPoint, Capital Equipment Co., Cleveland, OH, USA). The potential measured was converted to current according to the relationship of potential = current × resistance.

2.2. Wastewater

Glucose and glutamic acid test solution for biochemical oxygen demand (BOD) was modified and used as artificial wastewater (AW) throughout the study (Chang et al., 2004). The AW was autoclaved at 121 °C for 15 min before being added with filter sterilized glucose and glutamic acid solutions to the final BOD values of 100–400 mg/l. The AW was constructed and maintained under nitrogen atmosphere in a carboy, which was connected to a nitrogen-containing gas-tight bag (SKC Inc., Valley View Road, PA, USA) to avoid air diffusion due to negative pressure.

2.3. MFC operation and analyses

The Sensor-type MFCs were operated continuously. AW was fed up-flow mode through the injection port of the anode compartment at the rate of 0.15–1.00 ml/min using a peristaltic pump (505S, Watson-Marlow, Falmouth, Cornwall, UK) equipped with Marprene II tubing (Watson-Marlow). Air-saturated tap water was fed to the cathode compartment as an oxidant at the rate of 9.5 ml/min. The MFCs were installed in a temperature-controlled chamber to 33 °C unless stated otherwise in the text. The potential was recorded at different resistances from 3 Ω to 11 kΩ to analyze the MFC performance through the polarization curve. The chemical oxygen demands (COD) of the effluent were determined using COD assay kit (Ultra-Range COD reagent, Hack, CO, USA). All assays of COD were performed in triplicate and mean values were shown.

2.4. Statistical analyses

Most the experiments were performed using at least 3 MFCs. When a single MFC was used the experiments were repeated at least 3 times. The mean values or typical results were presented. All experimental error was analyzed with an aid of statistical methodology provided by Sigma Plot® 7.1 (SPSS Inc., Chicago, IL, USA). The maximum standard error obtained through the statistical analyses was within 5%. The size of the error was similar to those of the symbols used in all figures presented so the addition of error bar was disregarded. We found that the all data presented were statistically significant.

3. Results and discussion

3.1. Effect of MFC design factors

The structure of a fuel cell device has crucial effects on the overall performance of a hydrogen fuel cell (Larminie and Dicks, 2000). The Sensor-type MFC, which had structural features of direct contact between electrodes and membrane and a rectangular cross section perpendicular to the direction of fuel-flow with the narrow width of 20 mm, was used to produce electricity continuously. The MFC was able to produce the electric power stably without any maintenance except fuel and oxidant supply for over 2 years. Under the conditions of the fuel COD of 100 mg/l, resistance of 10 Ω, and the fuel-feeding rate of 0.35 ml/min, the current density was approximately 600 mA/m^2 (data not shown), which was 3 times higher than that obtained from the batch operation of the previous design (Gil et al., 2003) at the same resistance. The effluent COD was detected lower than 2 mg/l. The Sensor-type MFC was used throughout this study.

MFCs were operated using platinum-coated graphite felt as the cathode, and the performance was compared with that of plain graphite felt through the polarization curve method (Fig. 1). The MFC was continuously operated with fuel COD of 100 mg/l at the fuel-feeding rate of 0.35 ml/min changing the external resistance to read the steady-state potential. With the increase in current density (decreasing the resistance), the potential decreased more sharply in the MFC with plain graphite than in that with platinum-coated graphite felt in the region of low current density below 0.1 A/m^2. These results indicate that the platinum-coated graphite has higher catalytic activity with regard to oxygen than plain graphite, lowering the activation overvoltage (Larminie and Dicks, 2000). The MFC using platinum-coated graphite felt as cathode material exhibited a power density of 0.15 W/m^2, which was 3 times higher than plain graphite. In the following experiments to optimize the operational parameters, the MFC with platinum-coated graphite felt as cathode was used.

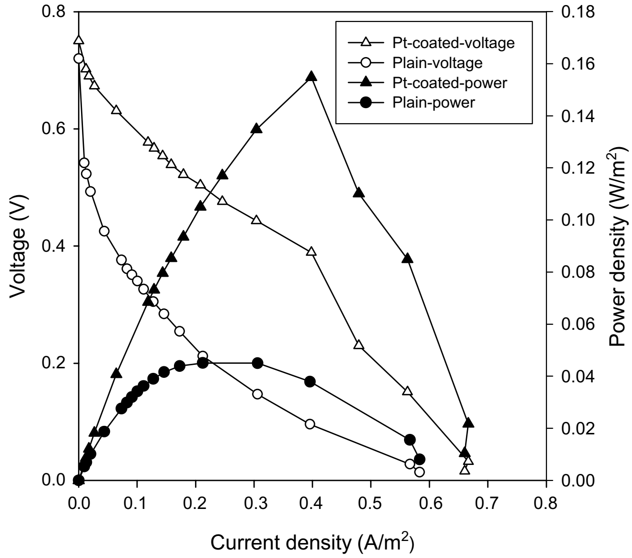

Fig. 1. Effects of platinum-coating on MFC performance. MFCs were operated continuously using artificial wastewater with COD of 100 mg/l fed at the feeding rate of 0.35 ml/min.

The Sensor-type MFC used throughout the present study has two different structural features from those used in similar studies. First of all, the electrodes were packed to occupy the entire anode and cathode compartments so as to have direct contact between the cation-specific membrane and each electrode. This configuration is believed to result in MFCs reduced ohmic overvoltage. Secondly, the cross section of the anode compartment has a rectangular shape with narrow width of 20 mm and fuel flows upward through the narrow fuel path. This design can minimize fuel channeling and flow short-circuiting problem in continuous operation. In addition the flat design of the Sensor-type MFC can be stacked easily.

The Sensor-type MFC has an inherent drawback as a packed bed reactor due to the fuel concentration gradient developed during the plug flow of fuel. Further improvement is expected through the use of direct air cathode, widely used in the direct methanol fuel cell (DMFC) (Han and Park, 2002; Chen and Yang, 2003), in the place of the cathode fed with air-saturated water used in MFC designs (Kim et al., 1999a,b, 2002, 2004; Jang et al., 2004; Gil et al., 2003; Bond and Lovley, 2003; Chaudhuri and Lovley, 2003; Schröder et al., 2003; Rabaey et al., 2003; Pham et al., 2004), DMFCs with direct air cathodes generate the power density higher by at least two orders of magnitude than those obtained using MFCs even at room temperature (Han and Park, 2002; Chen and Yang, 2003). Problems are not expected with the use of the direct air cathode in MFCs since methanol, a liquid fuel is used in DMFCs as in MFCs.

3.2. Effect of fuel concentration

The polarization curve method was employed to analyze the effects of fuel concentration on the performance of the MFC (Fig. 2A). The fuel-feeding rate was 0.35 ml/min. In the region of low current density below approximately 0.4 A/m^2, rapid voltage drops were observed in all the feeding rates tested. This means that the major overvoltage is activation overvoltage in the region of low current density. When fuel of 100 mg/l was supplied, the voltage dropped more sharply with the increase in the current density over about 0.4 A/m^2, which was not observed in the operation with a fuel concentration higher than 200 mg/l. This overvoltage might be mass transfer overvoltage due to the mass transfer loss of fuel (Larminie and Dicks, 2000), which could be improved by increasing the fuel concentration. The power density was increased with the increase in fuel concentration as shown in Fig. 2B. Fuel with a concentration of 300 mg/l gave the maximum power density of 0.36 W/m^2, and the

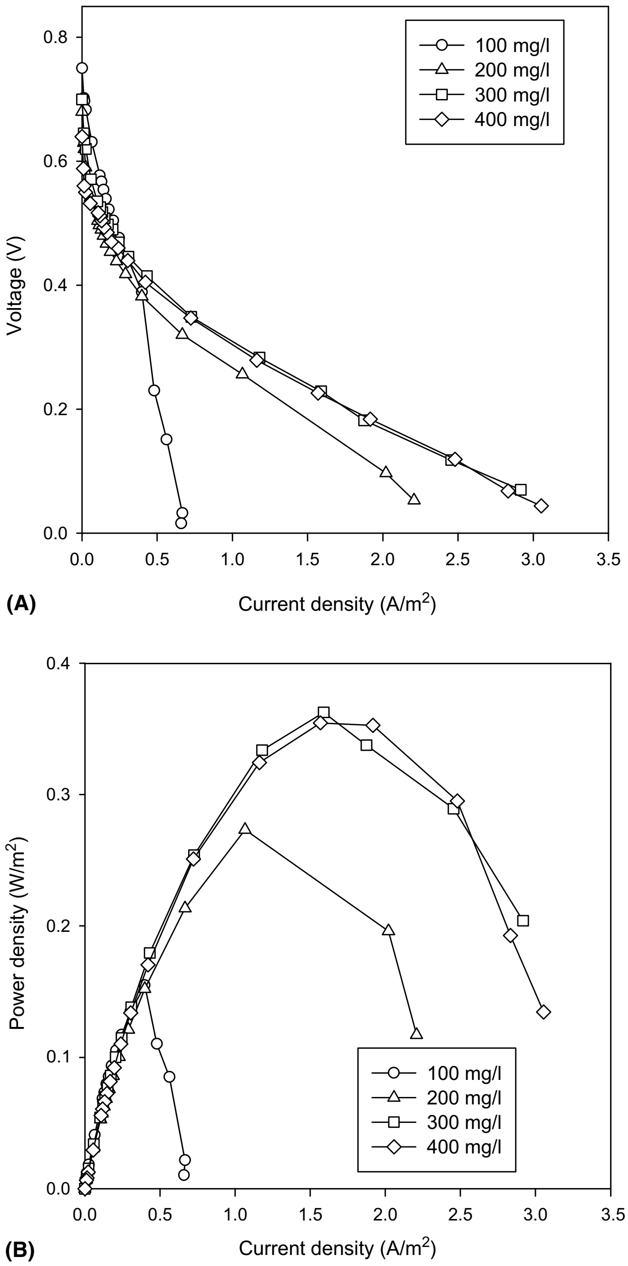

Fig. 2. Effects of fuel concentration on the MFC performance. (A) Polarization curve and (B) power density versus current density. MFCs with platinum-coated cathode were fed with artificial wastewater of different concentration at fuel-feeding rate of 0.35 ml/min. The maximum standard error obtained through the statistical analyses was within 5%. The size of the error was similar to those of the symbols used-therefore not shown.

current density of 1.6 A/m^2. This result shows that the microbial activity is saturated with a fuel concentration of 300 mg/l at the fuel-feeding rate of 0.35 ml/min. When the fuel concentration was increased, the maximum power density shifted toward higher current density. This means that the higher fuel concentration makes possible the operation of the MFC under the conditions necessary to generate higher current.

3.3. Effect of fuel-feeding rate

The effects of fuel-feeding rate on the performance of MFC were studied using a fuel concentration of 300 mg/l. As shown in Fig. 3A, current density was increased as the feeding rate was increased up to 0.65 ml/min. The increase was not substantial when the feeding rate was increased from 0.53 to 0.65 ml/min. At a feeding rate of

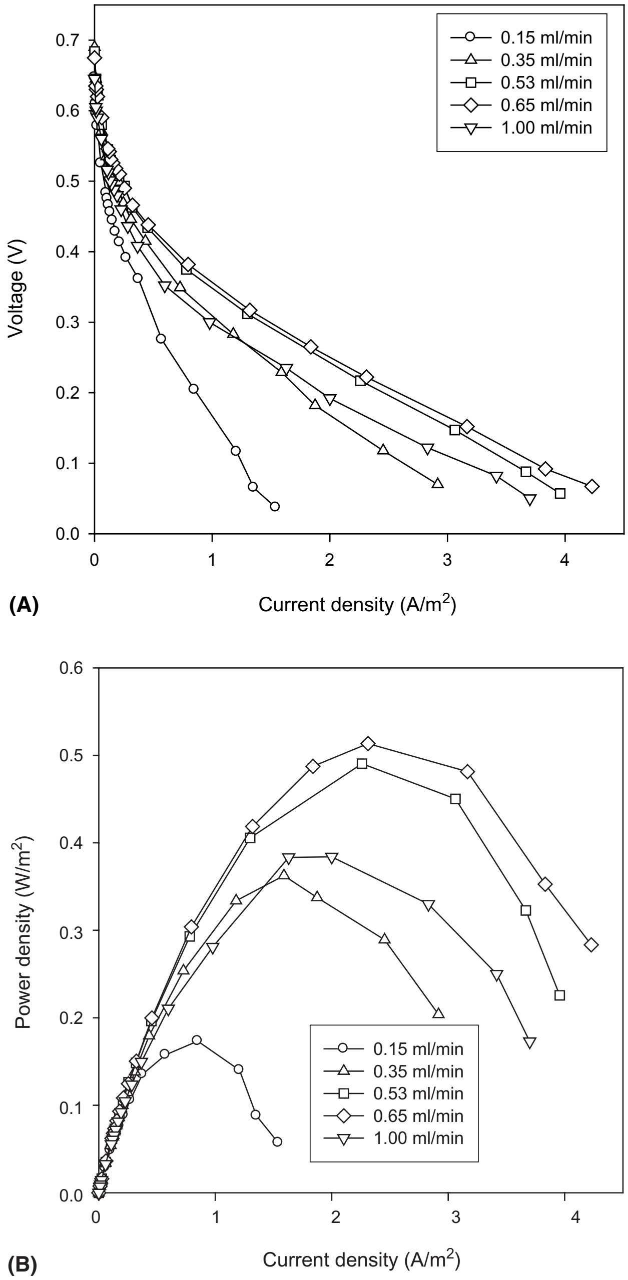

Fig. 3. Effects of fuel-feeding rate on the MFC performance. (A) Polarization curve and (B) power density versus current density. Artificial wastewater with COD of 300 mg/l was fed to the MFCs with platinum-coated cathode at the feeding-rate shown in the figure. The maximum standard error obtained through the statistical analyses was within 5%. The size of the error was similar to those of the symbols used-therefore not shown.

1.00 ml/min, the MFC showed lower current density than at 0.65 ml/min. A similar trend was observed in the power density (Fig. 3B). The increase in the current density with a feeding rate up to 0.65 ml/min shows that the microbial activity is saturated with fuel under these conditions. In Fig. 2 it was apparent that the microbial activity was saturated with 300 mg/l fuel at the feeding rate of 0.35 ml/min without any further increase in current with the increase in concentration to 400 mg/l. This comparison shows that some AW component(s) other than glucose and glutamate might be limiting.

The current and power densities were lower when MFCs were fed at the rate of 1.00 ml/min than at lower feeding rates. This might be due to the faster growth of the fermentative bacteria than of the electrochemically active bacteria acidifying the anode at this feeding rate. A similar phenomenon was observed previously (Kim et al., 2004; Rabaey et al., 2003). In the region of low current density below approximately 0.5 A/m^2, rapid voltage drops were observed in all the feeding rates tested, and in the high current density above 0.5 A/m^2, the voltage decreased linearly at the lower rate. Rapid falls of voltage in the region of the high current density were not observed. In all the regions of current density, the slopes of voltage drops were less marked as the feeding rate was increased to 0.65 ml/min. These results mean that the increase in fuel-feeding rate from 0.15 to 0.65 ml/min reduces activation and ohmic overvoltages.

The maximum power density of 0.51 W/m^2 was obtained at the feeding rate of 0.65 ml/min as shown in Fig. 3B. The COD values were decreased from 300 mg/l to 2.3 ± 0.47, 10.2 ± 0.58, 9.6 ± 0.10, and 9.7 ± 0.10 mg/l at the feeding rates of 0.15, 0.35, 0.53 and 0.65 ml/min, respectively, when the MFCs were operated to obtain the maximum power density at each feeding rate. This operational range of feeding rates satisfies the domestic regulation (below 10 mg/l) on the effluent COD in wastewater treatment facilities.

The optimum feeding rate was determined to be 0.53 ml/min considering operational stability as well as power density. When the MFC was fed with artificial wastewater of 300 mg/l COD at 0.53 ml/min, the COD loading rate was 9.2 kg/m^3 day, which is the highest among those ever obtained using MFC and over 10 times higher than those obtained using the conventional activated sludge processes (Tchobanoglous et al., 2003). and comparable with those obtained using anaerobic digestion, where loading rates are in the range of 5–25 kg/m^3 day (Rabaey et al., 2003). This shows the high potential of the MFC system as an alternative biological wastewater treatment process.

3.4. Effect of temperature

The MFC was operated at different temperatures, and the results were analyzed through the polarization curve method (Fig. 4A). The fuel concentration and feeding rates were 300 mg/l and 0.53 ml/min, respectively. The performance of MFC was slightly improved with the increase of temperature from 24 to 35 °C and decreased at over 38 °C. Temperature affected mainly ohmic overvoltage in all experiments. The maximum power density of 0.56 W/m^2 was obtained at 35 °C, as shown in Fig. 4B, and could be stably generated over

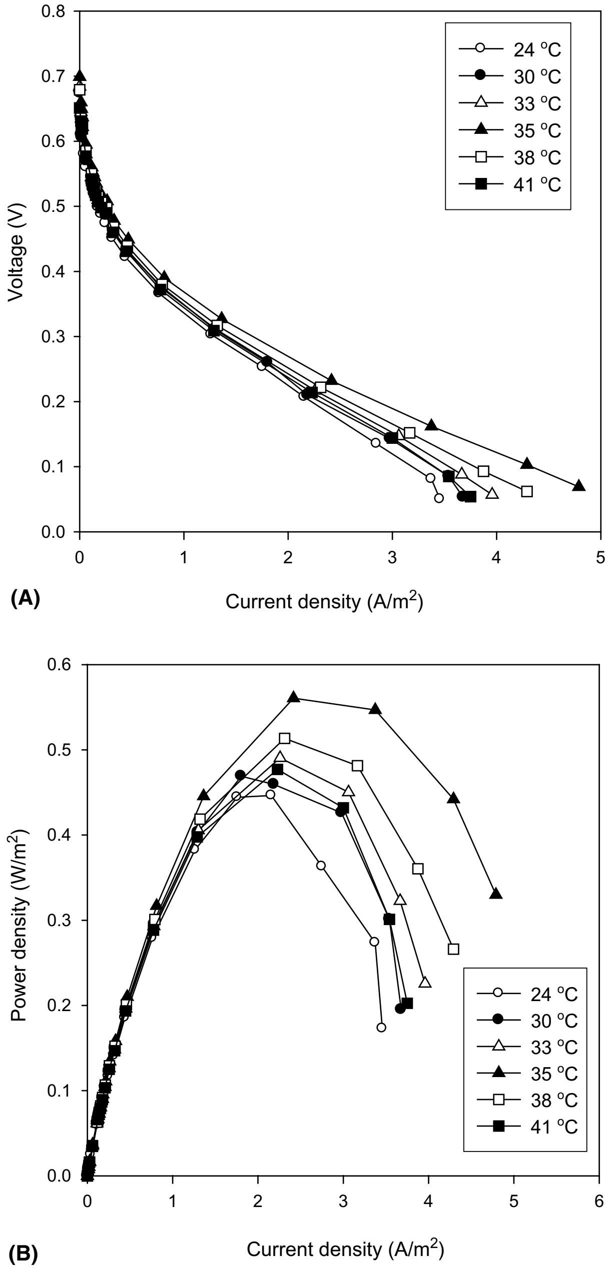

Fig. 4. Effects of temperature on MFC performance. (A) Polarization curve (B) power density versus current density. MFCs platinum-coated cathode were run at the temperature given in the figure with artificial wastewater of 300 mg/l COD at the feeding rate of 0.53 ml/min. The maximum standard error obtained through the statistical analyses was within 5%. The size of the error was similar to those of the symbols used-therefore not shown.

Table 1
Comparison of MFC performances

Biocatalyst	Mode of operation	Anode mediator	Cathode mediator	Power density (mW/m^2)	Volumetric power (mW/l)	References
Enriched microbial consortium	Continuous	None	None	560	102	This work
Enriched microbial consortium	Batch	None	None	8.3	1.6	Kim et al. (2004)
Shewanella putrefaciens	Batch	None	None	0.32	0.067	Kim et al. (2002)
Geobacter sulfurreducens	Batch	None	Yes	16	0.4	Bond and Lovley (2003)
Rhodoferax ferrireducens	Batch	None	Yes	8.2	0.15	Chaudhuri and Lovley (2003)
Enriched microbial consortium	Batch	None	Yes	3600	388	Rabaey et al. (2003)
Sewage sludge	Batch	Yes	Yes	788	16	Park and Zeikus (2003)

2 years (data not shown). The power density was maximum throughout the present study and compared favorably with all those ever reported using other MFC systems (Table 1).

3.5. Characteristics of MFC performance

The optimal operational conditions were a fuel concentration of 300 mg/l, a fuel-feeding rate of 0.53 ml/min, and a temperature of 35 °C through the present study. The polarization curve plotted at different conditions shows the effect of these parameters on the overall MFC performance. Except in the case of a fuel concentration of 100 mg/l, activation overvoltage and ohmic overvoltage limited overall performance. Activation overvoltage is caused by the slow reactions on the electrode surface, and ohmic overvoltage is straightforward resistance to the flow of electrons and ions (Larminie and Dicks, 2000). It is possible to improve the performance of MFC by reducing the overvoltages, adopting a method which takes into account the origin of the overvoltages. For example, using microorganisms with greater microbial activity and incorporating cathode material with greater catalytic activity can decrease activation overvoltage. In the case of ohmic overvoltage, an optimal design of membrane-electrode assembly and the screening of membrane giving a higher proton transfer rate would be considered.

The maximum power density obtained in the present study was lower than those obtained in batch systems by Park and Zeikus (2003) and Rabaey et al. (2003) as shown in Table 1. However it should be stressed that the power could be stably produced over 2 years. In addition to this, our system did not use any cathode mediators such as ferricyanide, which are not adequate for practical application (Schröder et al., 2003). The volumetric power, that is, power per unit cell working volume of our MFC, was 102 mW/l, which was over 60 times higher than those obtained in other mediator-less systems not using anode and cathode mediators. Up to now, power density has been used as a representative performance index for MFCs. But a value of power density can not clearly indicate the performance because it can be substantially varied according to the determination method of electrode area. In MFCs, only a part of the electrode surface would take part in the electrode reaction rather than total surface including internal surface as in typical chemical fuel cells such as proton exchange membrane fuel cells. For electrode material with internal structure, the total area of electrodes including the internal surface area (Park and Zeikus, 2003) or apparent area (Chaudhuri and Lovley, 2003; Reimers et al., 2001; Park and Zeikus, 2003) has been adopted for the computing of power density in MFCs. In this sense, volumetric power can be used as an alternative performance index to power density in a large volume process such as wastewater treatment. Volumetric productivity is one of the most important, if not the most important parameter.

4. Conclusions

The performance of a continuous microbial fuel cell could be considered in terms of microbiology and electrochemistry. In this study the polarization curve method was employed to optimize the performance of MFCs electrochemically assuming that microorganisms in the MFCs, enriched and maintained for over 2 years, were well adapted to the conditions studied. The optimal operational conditions for the MFC with the anode working volume of 20 ml were a fuel concentration of 300 mg/l, a fuel-feeding rate of 0.53 ml/min, and a temperature of 35 °C. The polarization curve plotted at different conditions could elucidate the effect of these parameters on the overall MFC performance. Except in the case of fuel concentration of 100 mg/l, activation overvoltage and ohmic overvoltage limited overall performance. The maximum power per unit cell working volume was 102 mW/l, which was over 60 times higher than those reported in the previous mediator-less MFCs that did not adopt cathode mediator.

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