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Continuous Electricity Generation from Domestic Wastewater and Organic Substrates in a Flat Plate Microbial Fuel Cell

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A microbial fuel cell (MFC) is a device that converts organic matter to electricity using microorganisms as the biocatalyst. Most MFCs contain two electrodes separated into one or two chambers that are operated as a completely mixed reactor. In this study, a flat plate MFC (FPMFC) was designed to operate as a plug flow reactor (no mixing) using a combined electrode/proton exchange membrane (PEM) system. The reactor consisted of a single channel formed between two nonconductive plates that were separated into two halves by the electrode/PEM assembly. Each electrode was placed on an opposite side of the PEM, with the anode facing the chamber containing the liquid phase and the cathode facing a chamber containing only air. Electricity generation using the FPMFC was examined by continuously feeding a solution containing wastewater, or a specific substrate, into the anode chamber. The system was initially acclimated for 1 month using domestic wastewater or wastewater enriched with a specific substrate such as acetate. Average power density using only domestic wastewater was 72 ± 1 mW/m^2 at a liquid flow rate of 0.39 mL/min [42% COD (chemical oxygen demand) removal, 1.1 h HRT (hydraulic retention time)]. At a longer HRT = 4.0 h, there was 79% COD removal and an average power density of 43 ± 1 mW/m^2. Power output was found to be a function of wastewater strength according to a Monod-type relationship, with a half-saturation constant of K_s = 461 or 719 mg COD/L. Power generation was sustained at high rates with several organic substrates (all at ∼1000 mg COD/L), including glucose (212 ± 2 mW/m^2), acetate (286 ± 3 mW/m^2), butyrate (220 ± 1 mW/m^2), dextran (150 ± 1 mW/m^2), and starch (242 ± 3 mW/m^2). These results demonstrate the versatility of power generation in a MFC with a variety of organic substrates and show that power can be generated at a high rate in a continuous flow reactor system.

Introduction

In a microbial fuel cell (MFC), electricity is generated from the anaerobic oxidation of organic matter by bacteria (*1*−*4*). The MFC typically consists of two separate chambers, an anode chamber and a cathode chamber. In the anode chamber, microorganisms oxidize biodegradable organic matter and transfer electrons to the anode electrode (negative electrode). Electrons move along a circuit to the cathode chamber (positive electrode), where oxygen (*4*−*9*) or other chemicals such as ferricyanide (*10*−*12*) accept the electrons. These electrons combine with protons that diffuse from the anode chamber to the cathode chamber, forming water (from oxygen) or ferrocyanide (from ferricyanide).

There are three ways by which microorganisms can transfer electrons to the anode electrode: using exogenous mediators, such as potassium ferric cyanide, thionine, or neutral red (*13*−*18*); using mediators produced by the bacteria (*12*, *19*); or by direct transfer of electrons from respiratory enzymes (i.e., cytochromes) to the electrode (*3*−*6*, *10*, *12*, *20*, *21*). Mediators provide a method of shuttling the electrons from inside of the bacterial cell to the electrode. There are several drawbacks to using exogenous mediators, such as their expense, short lifetime, and toxicity to the microorganisms (*4*, *5*, *7*, *21*). However, when the bacteria produce their own mediators, or they transfer electrons directly to the electrode, the system operates at a high, sustained level of activity. We define this system as a mediator-less MFC based on the observation that exogenous mediators do not need to be added. Several isolates, including *Shewanella putrefaciens* (*3*, *20*, *22*), *Geobacter sulfurreducens* (*4*, *23*), *Geobacter metallireducens* (*4*, *7*), and *Rhodoferax ferrireducens* (*6*), have been shown to generate electricity in mediator-less MFC systems. The actual mechanism used by bacteria in MFCs containing a microbial assemblage using a wastewater inoculum has not yet been determined.

Mixed cultures of bacteria in mediator-less MFCs have been reported to generate electricity from specific compounds (*9*, *10*, *24*) or organic matter in wastewater (*5*, *8*) and marine sediments (*7*, *25*). Power generation from domestic wastewater has recently received attention as a potential method to accomplish both wastewater treatment and electricity generation. Liu et al. (*8*) demonstrated continuous wastewater treatment and electricity generation using a single chamber MFC (SCMFC). They found that the system could generate 26 mW/m^2 of the maximum power density while removing up to 80% of the COD of wastewater. In a specially designed, smaller batch system, Liu and Logan (*9*) showed that up to 28 ± 3 mW/m^2 of power could be generated using domestic wastewater. It was further demonstrated that by removing the proton exchange membrane (PEM), they could generate a maximum of 146 ± 8 mW/m^2 of power. In these latter tests however, the wastewater was not continuously treated. In these systems, the anode was kept separated from the PEM/cathode or plain cathode in a large chamber, but the anode chamber was not mixed except by the flow of liquid into the system. In other MFCs, the anode chamber is often mixed (*4*, *14*, *15*, *20*, *22*, *26*).

In hydrogen fuel cells the electrodes are usually combined into a single strip separated by a PEM. Keeping the two electrodes in close proximity enhances proton conduction between the two electrodes. However, PEMs such as Nafion are permeable to oxygen, resulting in the transfer of small amounts of oxygen from the cathode chamber to the anode chamber. We therefore wondered if power generation would be increased in a MFC using a single electrode/PEM assembly or if transfer of oxygen from the cathode to anode chamber would prohibit successful power generation in a MFC. Bacteria known to produce electricity in MFCs range from obligate to facultative anaerobes, and thus the diffusion of oxygen to bacteria attached to the anode could restrict their

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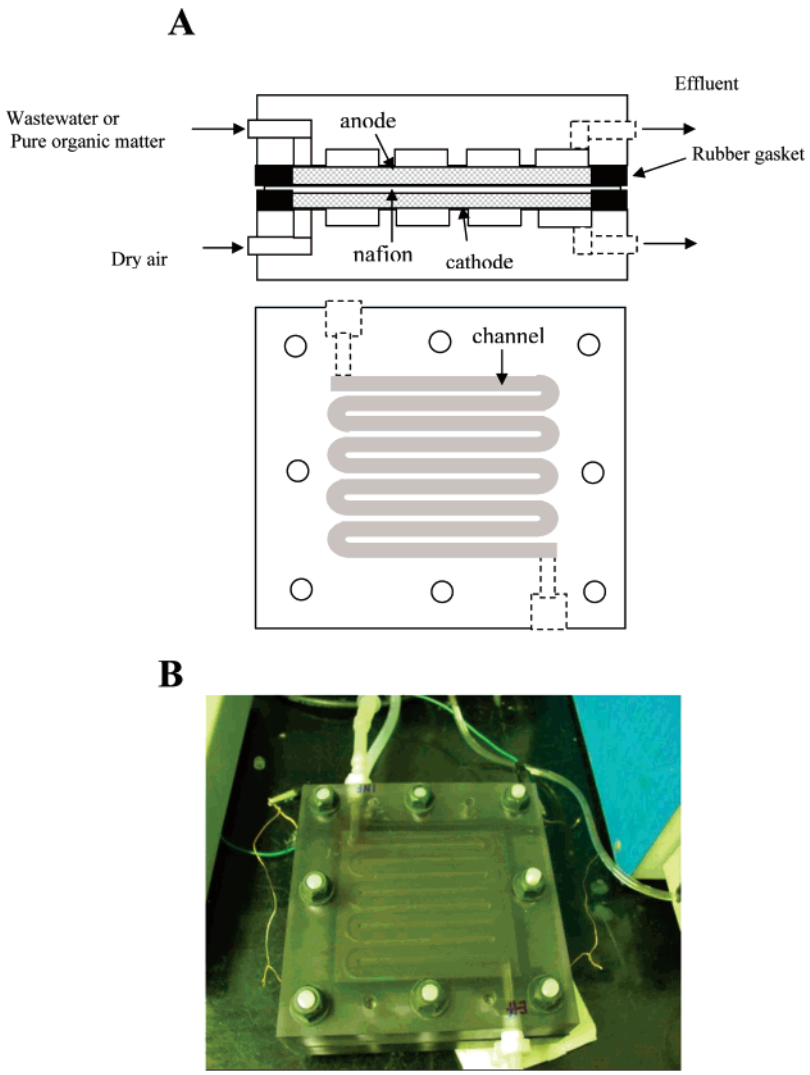

FIGURE 1. Schematic (A) (upper, side view; lower, top view) and laboratory-scale prototype (B) of the FPMFC.

ability to use the electrode assembly in the presence of dissolved oxygen. In addition, oxygen diffusion into the anode chamber can result in a loss of electron donor due to aerobic respiration by bacteria, lowering overall Coulombic efficiency.

In this study, we designed and tested the ability of a flat plate microbial fuel cell (FPMFC) containing a single electrode/PEM assembly to determine if we could continuously generate electricity using either domestic wastewater or pure compounds. The cathode electrode, consisting of a carbon cloth containing platinum as a catalyst, was hot pressed to a Nafion membrane and then placed in contact with a single sheet of carbon paper that formed the anode. Dry air was passed through the cathode chamber to provide oxygen as the electron acceptor. Bacteria present in domestic wastewater were used to inoculate the reactor.

Materials and Methods

Construction of the FPMFC. The FPMFC was comprised of two nonconductive (polycarbonate) plates bolted together (15 cm width by 15 cm length by 2 cm height), containing a channel that was separated into two halves to form the anode and cathode chambers (Figure 1). Each plate was drilled to form a rectangular channel in a serpentine path 0.7 cm wide and 0.4 cm deep, having a total surface area of 55 cm^2 and a total volume of 22 cm^3, achieving a total surface area per volume of 250 m^2/m^3. The plates were sealed using a rubber gasket and bolted together with several plastic screws. The anode electrode was a 10 cm × 10 cm piece of plain porous carbon paper, while the cathode electrode was a carbon cloth containing a platinum catalyst (0.5 mg/cm^2 catalyst containing 10% Pt) on one side of the electrode (both electrodes from De Nora North America, Inc.). The PEM (Nafion 117, Dupont, Wilmington, DE) was hot pressed to the cathode and placed on top of the anode, forming the PEM/electrode assembly sandwiched between the two polycarbonate plates. Copper wires were attached to each electrode (contact length approximately 9 cm) and sealed using electrical tape and nonconductive epoxy.

Wastewater and Organic Substrates. Domestic wastewater was collected from the primary clarifier of the Pennsylvania State University Wastewater Treatment Plant. Wastewater was used as the inoculum for the reactor and as a fuel. In some tests, the wastewater was diluted with ultrapure water (Milli-Q system; Millipore Corp., New Bedford, MA). Specific organic substrates, including glucose, acetate, dextran, starch, and butyrate, were prepared on the basis of their chemical oxygen demand (COD), at a concentration of 1000 mg COD/L, in a medium containing nutrients, minerals, vitamins, and a phosphate buffer (50 mM) (*26*). Solutions of these compounds were filter sterilized (0.45 or 0.2 μm pore diameter syringe filters; NALGENE Labware) and added to the influent storage bottle (total volume = 2.3 L).

FPMFC Operation. The reactor was operated in continuous flow mode at 30 °C in a temperature-controlled room.

The reactor was inoculated with wastewater and operated in continuous flow mode for approximately 1 month using either wastewater or wastewater containing acetate (20 mM). Wastewater in the influent tank was continuously sparged with N_2 gas to maintain anoxic conditions and prevent aerobic oxidation of the organic matter in the wastewater. Dry air was passed into the cathode chamber at a flow rate of 2 mL/min (unless stated otherwise) and monitored using a volumetric flowmeter (Cole-Parmer Instrument Co., Vernon Hills, IL). The hydraulic retention time was measured as previously described (*27*) using a spike injection of a salt tracer (4.7 M) for a flow velocity of 2.4 cm/min. Results indicated a reactor volume of 26.5 mL, dispersion coefficient of 1.84 cm^2/min, and a Peclet number of 108.

Analyses. The COD of the wastewater and other organic compounds was measured using Standard Methods (method 5220; HACH COD system, HACH Company, Loveland, CO.) (*28*). All samples except the wastewater samples were filtered to remove bacteria (0.2 μm pore-diameter, syringe membrane filter, Pall Corp., Ann Arbor, MI) prior to measuring the COD concentration. The concentrations of organic acids (acetate, butyrate, and propionate) and two alcohols (ethanol, methanol) in the effluent from the reactor fed dextran, glucose, or starch were analyzed (triplicate samples) using a gas chromatograph (*26*; Agilent, 6890) and a 30 m × 0.32 mm × 0.5 μm fused-silica capillary column. Reactor concentrations were expressed on the basis of the log mean COD concentration, C_{lm} (*29*−*30*), calculated using the influent (C_{in}) and effluent (C_{out}) COD concentrations as

$$C_{lm} = \frac{C_{in} - C_{out}}{\ln(C_{in}/C_{out})} \quad (1)$$

Current was determined from measuring the voltage (V) across a resistor in the circuit between the two electrodes using a multimeter and a data acquisition system (2700, Keithly, Cleveland, OH), with the data automatically recorded on a personal computer. Power density ($P = VI/A$) was calculated from the measured voltage (V), current ($I = V/R$), and surface area of the anode electrode (A). The Coulombic Efficiency of the system was calculated by integrating the measured current relative to the maximum current possible based on the observed COD removal (*24*). The maximum power output was determined by varying the load on the system using a series of resistors.

Results and Discussion

Power Generation Using Domestic Wastewater. The flat plate system developed here was able to continuously generate electricity from the organic matter in the wastewater while accomplishing wastewater treatment. Following an acclimation period of approximately 1 month, constant power generation from wastewater was obtained with the FPMFC over a period of 5 months. For wastewater containing 246 ± 3 mg COD/L, an average power density of 56 ± 0 mW/m^2 was obtained with a hydraulic retention time (HRT) of 2.0 h (0.22 mL/min flow rate; 164 mg/L log mean COD) and an air flow rate of 2 mL/min with a 470 Ω resistor (Figure 2A). Under these operation conditions, the COD removal rate was 1.2 mg/L min (58% COD removal). The maximum power density at a flow rate of 0.22 mL/min, obtained by varying the circuit resistance from 47 to 22 000 Ω (Figure 2B), was 63 mW/m^2 with a current of 1.03 mA (326 Ω). This power density is about 10% higher than that obtained under typical operating conditions with a 470 Ω resistor.

Increasing the HRT to 4.0 h decreased the power density to 43 ± 1 mW/m^2 and the COD removal rate to 1.0 mg/L min (79% removal; influent COD = 280 ± 19 mg/L; log mean COD = 141 mg/L; data not shown). When the HRT was decreased to 1.1 h (0.39 mL/min), the average power generation was 72 ± 1 mW/m^2 with a COD removal rate of 2.4 mg/L min (42% removal; influent COD = 379 ± 9 mg/L; log mean COD = 292 mg/L; data not shown). This power density was 2.8 times larger than that obtained in previous tests using domestic wastewater and a MFC with a PEM (26 mW/m^2; ref *8*).

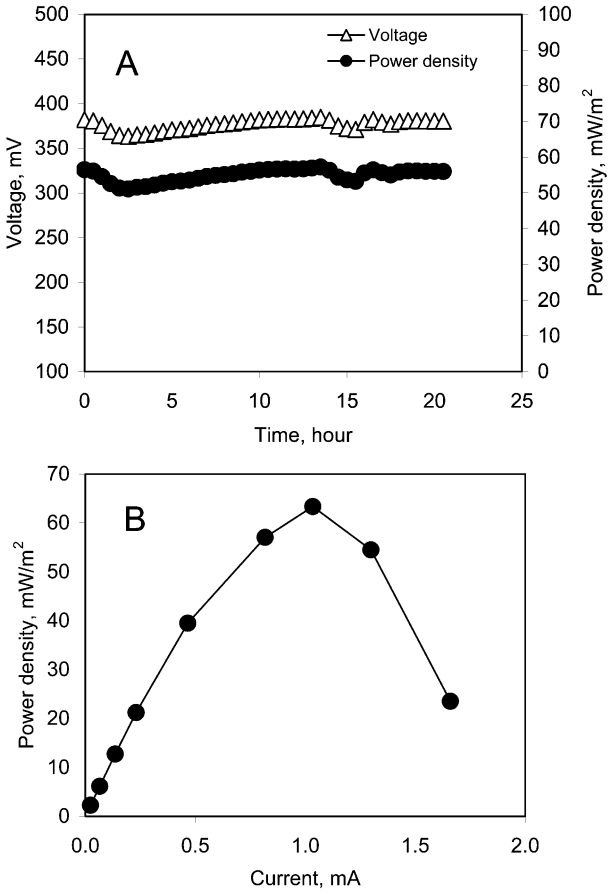

FIGURE 2. Voltage (△) and power generation (●) with 470 Ω (A) and changes in power with different currents (different resistors) (B) in a FPMFC using wastewater at a 2.0 h HRT.

Power generation was examined as a function of wastewater strength at fixed HRTs of 36 min (0.73 mL/min) and 33 min (0.81 mL/min) in two separate experiments conducted 3 months apart. Power density showed a Monod-type trend as a function of the wastewater strength over a range of 38−324 mg COD/L (Figure 3). In the first test (HRT = 36 min), the maximum power generation constant was P_{max} = 166 ±56 mW/m^2 with a half-saturation coefficient of K_s = 461 ±236 mg/L. In the second set of experiments, the maximum power was P_{max} = 218 ±142 mW/m^2 with a half-saturation coefficient of K_s = 719 ± 621 mg/L (HRT = 33 min). COD removal in the above two tests was found to be a linear function of the log-mean COD concentration (Figure 4). This indicated that while electricity production was reaching saturation conditions, COD removal rates were not. The loss of COD could be due to gas production (hydrogen or methane) or to the use of alternate electron acceptors by bacteria such as oxygen diffusing through the PEM (*4*, *5*, *8*−*10*, *16*, *26*). The fact that the two rates were different likely reflects the different compositions of the wastewater samples collected at different times.

It is possible that by diluting the wastewater with a low conductivity water (deionized water) we altered power generation due to the change in the conductivity of the sample. Therefore, we compared wastewater diluted 50%

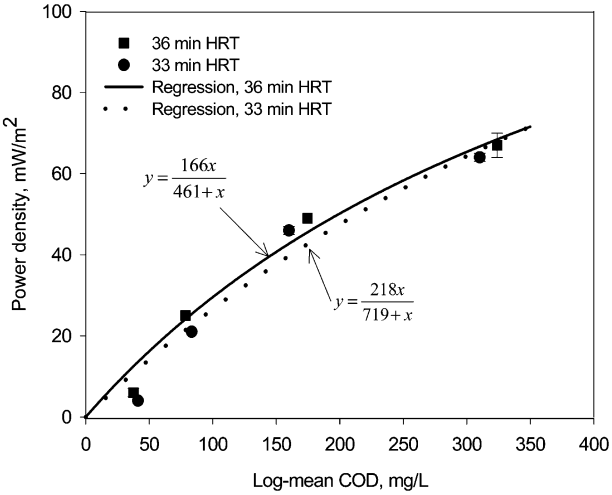

FIGURE 3. Power production in the FPMFC as a function of log-mean COD at different wastewater strengths (470 Ω, 36 min HRT for the first run, 33 min HRT for the second run).

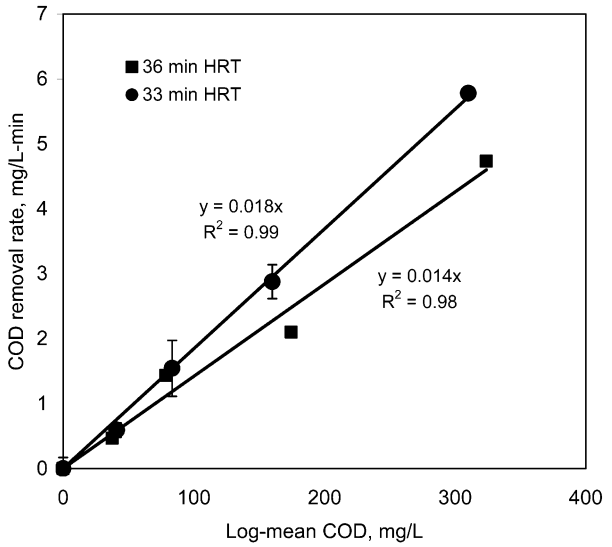

FIGURE 4. Removal rate of organic matter in wastewater by the FPMFC as a function of log mean COD (470 Ω, 36 min HRT for the first run, 33 min HRT for the second run).

with deionized water to the same sample containing sufficient NaCl to achieve the original conductivity. There was no significant difference in the power density measured for these two samples (data not shown), indicating that changes in conductivity due to dilution of the wastewater did not adversely affect power generation under these conditions.

Air Flow Rate. Previous studies have shown that the flow rate of air to the cathode can affect power generation in air−cathode types of microbial fuel cells (*8*, *9*). The effect of increasing the air flow rate to the cathode from 2 to 200 mL/min was therefore examined in relation to power generation (Figure 5). The power density was found to decrease as the air flow rate increased. However, the COD removal rate increased slightly with air flow rate. These results suggested that the increase in air flow resulted in an increase in oxygen transfer into the anode chamber, resulting in a decrease in the power density but an increase in the COD removal rate. To test this hypothesis, we calculated the Coulombic Efficiencies of the system under these conditions. As shown in Figure 5, there was a slight decrease in Coulombic efficiency with an increase in air flow rate, although the range in values was large.

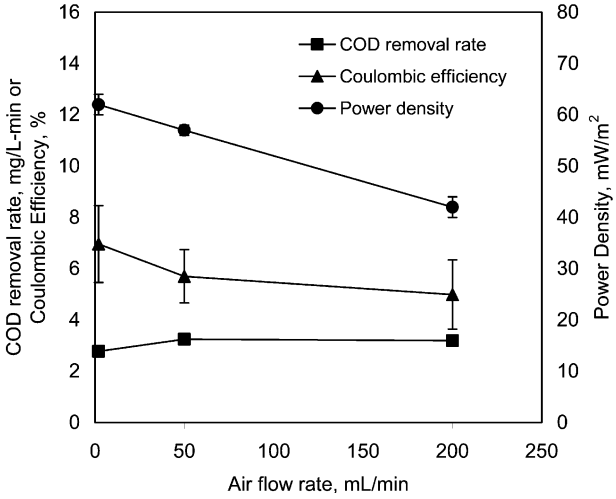

FIGURE 5. Effect of oxygen flow to the cathode chamber on electricity generation, organic matter removal rate in wastewater, and Coulombic efficiency (470 Ω, HRT = 1.4 h).

TABLE 1. Power Generation, COD Removal, and Coulombic Efficiencies Measured for Different Compounds with the Reactor Acclimated to Wastewater (except as noted)

		continuous flow tests (33 Ω resistor)		
substrates	max power density (mW/m^2)	power density (mW/m^2)	COD removal (%)	Coulombic efficiency (%)
acetate	309	286 ± 3	8 ± 1	65
starch	261	242 ± 5	26 ± 2	21
glucose	220	212 ± 2	32 ± 2	14
dextran	189	150 ± 1	22 ± 4	17
butyrate	nm[a]	5 ± 0	3 ± 1	28
butyrate[b]	274	220 ± 1	10 ± 0	50

[a] Not measured. [b] Sample acclimated to wastewater and 1000 mg/L of butyrate.

The air flow rate was decreased from 2 to 0.2 mL/min in order to determine if lowering the air flow rate would affect power generation. There was no difference in either the power density or COD removal rate at these lower air flow rates (data now shown).

Power Generation with Specific Chemicals. Following 5 months of constant operation of the FPMFC with domestic wastewater, the feed to the reactor was changed to a medium containing specific chemicals at concentrations of 1000 mg COD/L. The reactor was run at a HRT = 0.68 h (41 min) with each substrate until the power density was constant (36−59 HRTs). The power density of 286 ± 3 mW/m^2 was the largest value with acetate, a compound that is a fermentation end product (Table 1). Acetate also had the largest Coulombic efficiency of 65%. Lower power densities were achieved with carbohydrates, with 242 ± 5 mW/m^2 for starch, 212 ± 2 mW/m^2 for glucose, and 150 ±1 mW/m^2 for dextran. The Coulombic Efficiencies for these chemicals were substantially lower, ranging from 14% to 21% (Table 1).

Power production using butyrate was initially found to be quite low (5 ± 0 mW/m^2) relative to other organic substrates. To determine if the low power output was due to the acclimation procedure, the system was reacclimated to domestic wastewater containing 1000 mg COD/L of butyrate. The system was then switched after 10 d to a feed containing only butyrate as a carbon source. Stable power generation was obtained after 17 d. The system then produced 220 mW/m^2 of power with a Coulombic Efficiency of 50% (Table 1).

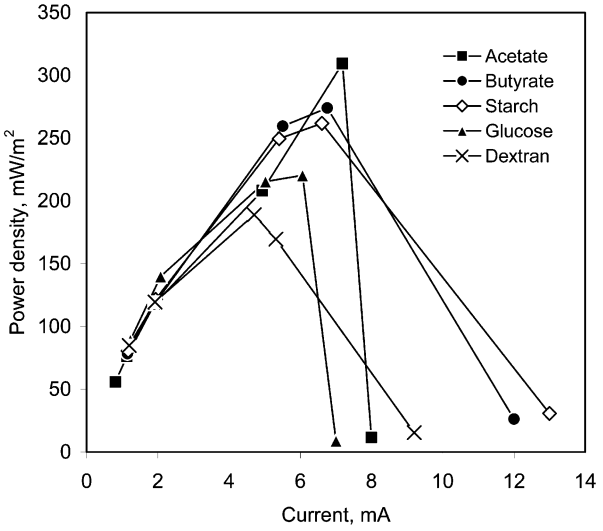

FIGURE 6. Power densities generated in the FPMFC from different substrates as a function of current (1000 mg-COD/L for each substrate; HRT = 41 min).

TABLE 2. Organic Acids and Alcohols Measured in the Effluent of the Reactor Fed Fermentable Substrates

	influent substrate		
condition	glucose	dextran	starch
influent COD (mg/L)	1066 ± 33	1003 ± 1	925 ± 5
effluent			
COD (mg/L)	728 ± 14	782 ± 41	686 ± 18
acetate (mg COD/L)	142 ± 1	68 ± 12	200 ± 9
butyrate (mg COD/L)	0 ± 0	4 ± 2	4 ± 0
propionate (mg COD/L)	39 ± 1	83 ± 13	51 ± 4
ethanol (mg COD/L)	42 ± 3	2 ± 1	173 ± 27
methanol (mg COD/L)	16 ± 1	18 ± 1	18 ± 1
total COD removal (%)	32 ± 3	22 ± 4	26 ± 2

The maximum power that could be generated with each of these substrates, at a fixed HRT = 0.68 h, was examined for each substrate by varying the circuit load (Figure 6). The maximum power densities varied in a manner similar to that observed in continuous power tests. Acetate produced the largest maximum power density of 309 mW/m^2 with a circuit load of 33 Ω, versus 76 mW/m^2 with a 320 Ω resistor. With glucose, a maximum current of 6 μA was obtained with a 33 Ω resistor, producing 220 mW/m^2. This value is larger by 147% (131 mW/m^2) than that obtained with a 320 Ω resistor (89 mW/m^2).

The observation that Coulombic Efficiencies were much larger for the nonfermentable substrates (acetate and butyrate) than the fermentable substrates (glucose, dextran, and starch) suggested that there was substantial loss in terms of COD of the materials into the biofilm that did not result in power generation. COD loss linked to fermentation would require a loss of the material as a gas (i.e. hydrogen or methane) or as biomass. As there was no gas phase in the system, gases were not monitored. However, there was substantial production of alcohols (ethanol and methanol) and organic acids (acetate, butyrate, and propionate), indicating significant fermentation of the substrates by the biofilms (Table 2). The composition of the effluent in terms of these acids and alcohols varied for each substrate (glucose, starch, and dextran), but acetate was usually the main fermentation product. There was little butyrate production relative to acetate production, and when the reactor was subsequently fed butyrate as the central substrate, there was little power generation. Butyrate is a primary fermentation end product during hydrogen production in mixed culture bioreactors (*31*), suggesting that there was little biohydrogen production in the reactor.

Implications for Wastewater Treatment. The reactor that was used here had the anode and cathode on either side of the PEM, and therefore there was oxygen diffusion to the anode because the PEM is permeable to oxygen (*4*, *5*, *8*−*10*, *16*, *26*). The diffusion of oxygen into the anode chamber did not prevent electricity generation in the MFC, but it may have affected reactor performance. There are many possible reasons for electricity generation under these conditions that cannot be answered without a better understanding of the characteristics of the bacteria that generate electricity in MFCs. Bacteria attached directly on the electrode may be able to produce electricity even in the presence of small concentrations of oxygen. Alternatively, the uneven surface of the carbon electrode might permit some bacteria in the deeper parts of the electrode (closer to the cathode) to sufficiently scavenge oxygen before it could reach other bacteria on the outer surface of the anode producing electricity. It is also possible that some bacteria, not directly attached to the surface, produced electricity due to mediators produced by the bacteria (*19*). While the specific reason for continued power generation is not known, the ability to have these electrodes spaced so closely to each other is an important finding, as it increases the potential options for finding more efficient designs of MFC reactors.

The maximum power density obtained here of 72 mW/m^2 for domestic wastewater represents an increase by a factor of 2.8 times compared to that previously obtained under continuous flow conditions with a different type of reactor (single chambered MFC; ref *8*). While this power density is relatively low compared to hydrogen fuel cells, it should be recognized that fixed film bioreactors used for wastewater treatment already provide very large surface areas for biofilm growth. For example, plastic media trickling filters typically have approximately 100 m^2/m^3 of reactor volume and are 6 m high, providing 600 m^2 of surface for each square meter of cross section for wastewater application. While the surface area of the FPMFC (250 m^2/m^3) was larger than that typical of a trickling filter, it was substantially larger than the 20 m^2/m^3 used in a single chamber MFC reactor in a previous study (*8*). This increase in specific surface area was likely a contributor to the higher power density achieved here versus that obtained in the single chamber MFC. As we learn more about how to optimize MFC designs for wastewater treatment, we should be able to design reactors that have more optimized specific surface areas and geometries of the electrodes and the reactor.

Acknowledgments

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