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Altering Anode Thickness To Improve Power Production in Microbial Fuel Cells with Different Electrode Distances

Yongtae Ahn and Bruce E. Logan*

Department of Civil and Environmental Engineering, Pennsylvania State University, 212 Sackett Building, University Park, Pennsylvania 16802, United States

ABSTRACT: A better understanding of how anode and separator physical properties affect power production is needed to improve energy and power production by microbial fuel cells (MFCs). Oxygen crossover from the cathode can limit power production by bacteria on the anode when using closely spaced electrodes [separator electrode assembly (SEA)]. Thick graphite fiber brush anodes, as opposed to thin carbon cloth, and separators have previously been examined as methods to reduce the impact of oxygen crossover on power generation. We examined here whether the thickness of the anode could be an important factor in reducing the effect of oxygen crossover on power production, because bacteria deep in the electrode could better maintain anaerobic conditions. Carbon felt anodes with three different thicknesses were examined to see the effects of thicker anodes in two configurations: widely spaced electrodes and SEA. Power increased with anode thickness, with maximum power densities (604 mW/m^2, 0.32 cm; 764 mW/m^2, 0.64 cm; and 1048 mW/m^2, 1.27 cm), when widely spaced electrodes (4 cm) were used, where oxygen crossover does not affect power generation. Performance improved slightly using thicker anodes in the SEA configuration, but power was lower (maximum of 689 mW/m^2) than with widely spaced electrodes, despite a reduction in ohmic resistance to 10 Ω (SEA) from 51−62 Ω (widely spaced electrodes). These results show that thicker anodes can work better than thinner anodes but only when the anodes are not adversely affected by proximity to the cathode. This suggests that reducing oxygen crossover and improving SEA MFC performance will require better separators.

1. INTRODUCTION

Microbial fuel cells (MFCs) are devices that use microorganisms to covert the energy stored in chemical bonds in biodegradable organic and inorganic compounds to electrical energy.[1] Microbes release electrons to the anodes, and they are transferred through the circuit to the cathode, where they combine with protons and an electron acceptor, such as oxygen, to form water.[1,2] Several types of MFCs with different electrode arrangements have been developed, including two-chamber, single-chamber, flat-plate, and stacked electrode reactors.[3−6] Of these, the single-chamber air cathode MFC is the most commonly used configuration because of its high power output, low internal resistance, and relatively low operational cost as a result of the direct use of oxygen in air.[4,7]

Electrode materials play an important role in the performance and cost of a MFC. These materials should have good electrical conductivity, low resistance, chemical stability, corrosion resistance, and high mechanical strength. Various materials have been used, including graphite fiber brushes, graphite rods, carbon paper, carbon mesh, and carbon felt.[8−11] The modification of the surface with chemicals, metals, metal oxide, and non-metals, such as carbon nanotubes (CNTs), supported on different materials (such as textiles and sponges) are effective methods for enhancing power generation by many different types of anode materials by increasing biocompatibility and electron-transfer efficiency.[12−14] For example, the addition of carbon nanotubes to macroporous sponges improved volumetric power production by 12 times (to 182 W/m^3)[15] compared to that previously obtained with domestic wastewater.

Carbon felt has been used as an electrode material in MFCs[16,17] as well as in other electrolytic cells for ion removal.[18−20] One advantage of the carbon felt anode over other materials is that it has large porosity (~99%)[21] relative to carbon cloth or paper, allowing more surface area for bacterial growth. In addition, the cost of carbon felt and its performance (maximum power density) are similar to those of other carbon-based materials.[17,22] However, the thickness and placement of these felt materials relative to the cathode have not been well-studied. Reducing the anode−cathode distance can improve the power production by reducing ohmic (solution) resistance, but very close spacing of thin anodes can reduce power. For example, reducing the spacing between a thin carbon cloth anode (0.35 mm thick) and cathode from 3 to 2 cm increased power and decreased internal resistance from 56 to 35 Ω.[23] Although further decreases in electrode spacing reduced the internal resistance to 16 Ω, the power decreased because of oxygen crossover from the cathode to the anode, adversely affecting power generation by bacteria on the anode.

One way to reduce oxygen crossover is to place a separator between electrodes, forming a separator electrode assembly (SEA) configuration. Separators are effective at reducing oxygen crossover but not affecting proton transport to the cathode or increasing power densities and Coulombic efficiencies (CEs) compared to systems with larger electrode spacing because of the reduction in ohmic resistance.[24−26] In a SEA MFC, the type of anode used will affect power production and the thickness of the anode size may be a factor in improving MFC performance. A thick (2.5 cm diameter and

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1.25 cm wide, when placed against the separator) and highly porous (95%) brush anode produced much more power than that of a flat carbon mesh anode (0.2 mm thick) in a SEA MFC with wastewater as a substrate.[27] The use of a thicker anode might be an important factor in power production not only because this increases the surface area for bacterial growth but also because the thick anode could provide an environment whereby the bacteria on the inside of the anode can remain in a highly anoxic environment compared to bacteria on the outside of the electrode. Thus, the use of thick anodes could provide an alternate strategy to help mitigate the effects of oxygen contamination of the anode.

To test the importance of the anode thickness on power production, we compared flat carbon felt anodes having different thicknesses (0.32, 0.64, and 1.27 cm) in MFCs using both widely spaced and SEA electrode configurations. The greatest anode thickness of 1.27 cm was chosen to be similar to that produced by the graphite fiber brush when placed against the separator.[27] Using the two different configurations, the effect of the total electrode size could be determined separately in a spaced electrode configuration compared to the SEA configuration, where oxygen transport from the cathode would additionally affect power production.

2. MATERIALS AND METHODS

2.1. Construction of the MFCs. Single-chamber air cathode MFCs (28 mL) were constructed as previously described.[4] Carbon felt anodes of different thickness (0.32, 0.64, and 1.27 cm) with a porosity estimated by others to be 83−95%[28,29] (Alfa Aesar, Ward Hill, MA) were cut into circles having a projected surface area of 7 cm^2. Each reactor contained a single air cathode (30 wt % wet-proofed carbon cloth, type B-1B, E-TEK) with a platinum catalyst (0.5 mg/cm^2) on the water side and four diffusion layers on the air side.[7] Two layers of a textile material (Amplitude Prozorb, Contec, Inc.) were sandwiched between the anode and cathode (Figure 1A) when electrodes were used in the SEA configuration. As a control on the electrode size (total electrode surface area), MFCs were operated without a separator, with the anode and cathode spaced 4 cm apart (Figure 1B).

2.2. MFC Operation. The MFCs were inoculated with domestic wastewater from the primary clarifier of the Pennsylvania State University Wastewater Treatment Plant and operated at 30 °C. After inoculation with a 50:50 mixture of inoculum and medium, the MFCs were fed only 50 mM phosphate buffer medium containing 1 g/L sodium acetate (Na_2HPO_4, 4.58 g/L; $NaH_2PHO_4{\cdot}H_2O$, 2.45 g/L; NH_4Cl, 0.31 g/L; KCl, 0.13 g/L; trace minerals and vitamins; pH 7.1; and conductivity of 6.8 mS/cm). The reactors were operated in a fed-batch mode, where they were refilled each time when the voltage decreased to <10 mV (one fed-batch cycle of operation).

2.3. Analyses. The voltage (E) across the external resistor (1 kΩ) was measured every 20 min using a data acquisition system (model 2700, Keithley Instruments) connected to a computer. Current (I) and power ($P = IE$) were calculated as previously described[2] and normalized by the cross-sectional area of the cathode (7 cm^2). Polarization and power density curves were obtained by varying the external resistance in the circuit (20 min per resistor) over a single cycle. During each polarization test, the anode potentials were recorded using reference electrode (RE-5B; BASi, West Lafayette, IN), with the cathode potential calculated as the difference between the anode potential and cell voltage. A separate reference electrode for the cathode could not be used because of the SEA (no space between the anode and cathode for a reference electrode). CE was calculated using the ratio of the total coulombs produced during the experiment to the theoretical amount of coulombs consumed from the substrate as previously described.[2] The chemical oxygen demand (COD) was measured using a low range (0−150 mg/L) HACH COD system (Hach Co., Loveland, CO).[23] Internal resistance was characterized using electrochemical impedance spectroscopy (EIS) under working

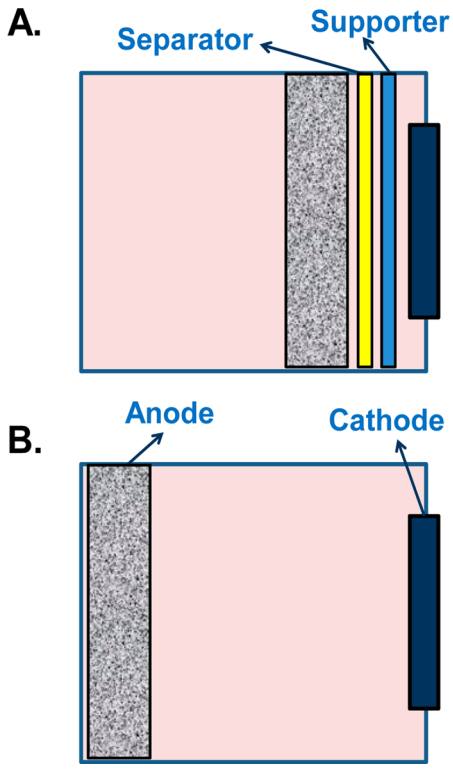

Figure 1. Schematic of the MFCs with (A) SEA configuration and (B) spaced electrode configuration.

cell conditions (R_{ext} = 1000 Ω). The impedance measurements were taken from 100 kHz to 10 mHz by applying a sine wave (10 mV) on top of the bias potentials with a potentiostat (BioLogic, VMP3). The ohmic resistance was determined by reading the real axis value at the high-frequency intercept, and the charge transfer and diffusion resistances were determined by fitting the EIS spectra with semicircles.[30,31]

3. RESULTS

3.1. Current Production. The SEA MFCs with three different anode thicknesses operated for more than 2 months (1 kΩ fixed resistance) demonstrated stable and reproducible performance during each fed-batch cycle. Representative current output curves are shown in Figure 2A. Even at this high fixed resistance, there were current differences among the reactors operated in this SEA configuration. The current was higher for the thickest anode material with 0.46 mA (1.27 cm thick carbon felt), followed by 0.39 mA (0.64 cm) and 0.32 mA (0.32 cm). The higher current densities for the thicker anode material appeared to be due to the sustained highly negative anode potentials (Figure 2B). In contrast, the thinnest anode started out with a highly negative potential, but this potential quickly became more positive, resulting in a lower maximum current. It appeared that the MFC with the thinnest anode (0.32 cm) had a more negative cathode potential (−244 mV) compared to those for the other two anodes (−217 mV, 0.68 cm; −214 mV, 1.27 cm). However, this difference in potential could be due to the placement of the reference electrode near the outer edge of these very thick anodes. The rapid increase in the anode potential was likely the critical factor in the different performance over time.

When these three different anodes were tested using a more typical configuration, with the anode placed on the other side of the chamber from the cathode, all three anodes produced

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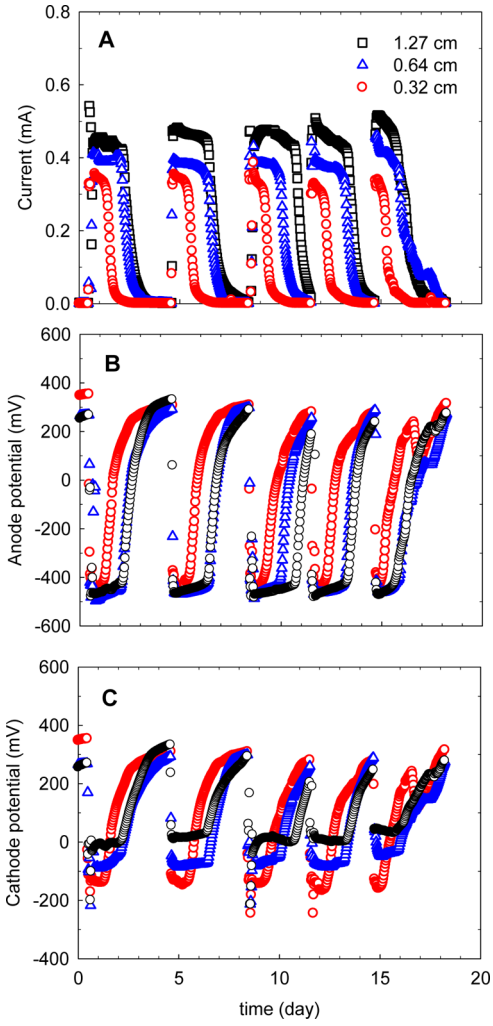

Figure 2. Performance of the MFCs with a SEA configuration in terms of (A) current production, (B) anode potential, and (C) cathode potentials over time.

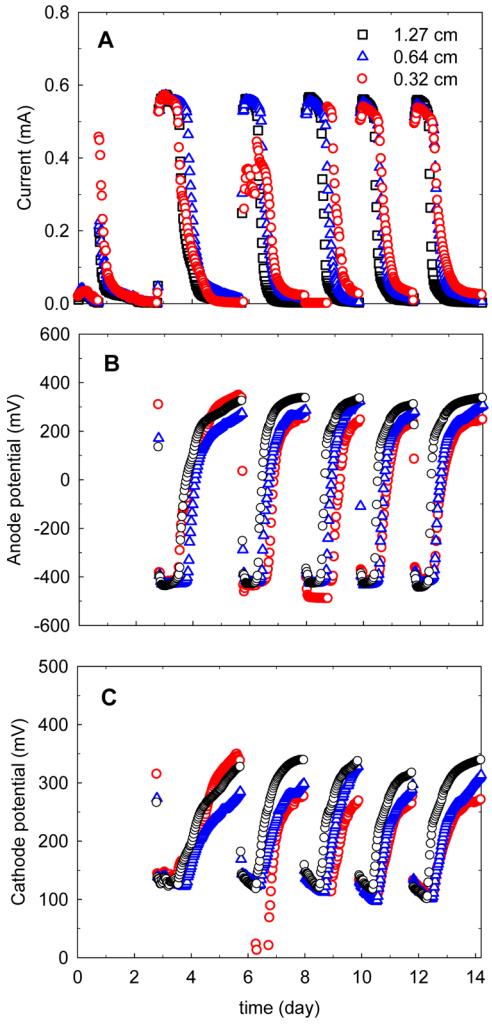

Figure 3. Performance of the MFCs with a spaced electrode configuration in terms of (A) current production, (B) anode potential, and (C) cathode potentials over time.

similar currents (Figure 3B). The lowest anode potential (−454 ± 31 mV; Figure 3B) was similar to that obtained with the best anodes with the SEA configuration (Figure 2B). In all MFCs, the anode potentials remained similar to each other and highly negative, until the end of the fed-batch cycle. These results show that the use of thicker anodes at a high external resistance was only beneficial when the anode was placed directly against the cathode in the SEA configuration. Thus, the adverse effect of oxygen leakage through the cathode was more apparent for the thinner anodes but only when the anode was placed next to the cathode.

COD removals were good (>85%) for all configurations (Figure 4). These COD removals were only slightly less than that previously reported using the same medium with brush anodes (>90%).[32] CEs ranged from 19 to 23% in the normal configuration, widely spaced electrodes, but higher CEs of 37−50% were obtained with a SEA type of configuration. The highest CE was obtained using the thickest anode (1.27 cm) in the SEA configuration. These results with higher CEs for the MFCs with the SEA configuration than widely spaced electrodes are consistent with previous results that using a separator increases CEs.[33] The CE was slightly lower with 1.27 cm anode than with other anode thicknesses for SEA configuration, while the COD removal efficiency was highest.

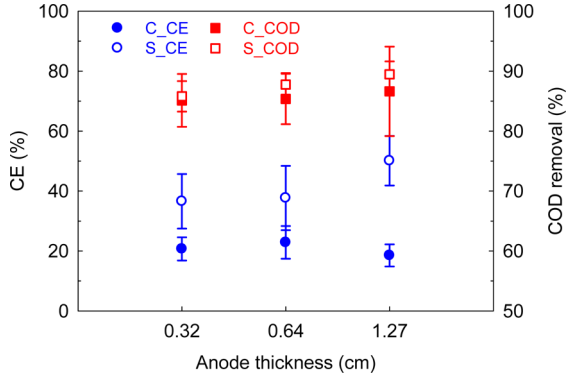

Figure 4. CEs and COD removals with different anode thicknesses for SEA configuration and control reactors, where C represents "spaced electrode configuration" and S represents "separator electrode assembly configuration".

This low CE might be due to a loss of substrate to microorganisms through aerobic respiration.

3.2. Maximum Power Densities. The maximum power densities of the MFCs operated in the SEA configuration were only slightly improved using thicker anodes (Figure 5). The maximum power density was 689 ± 16 mW/m^2 (17 W/m^3) for

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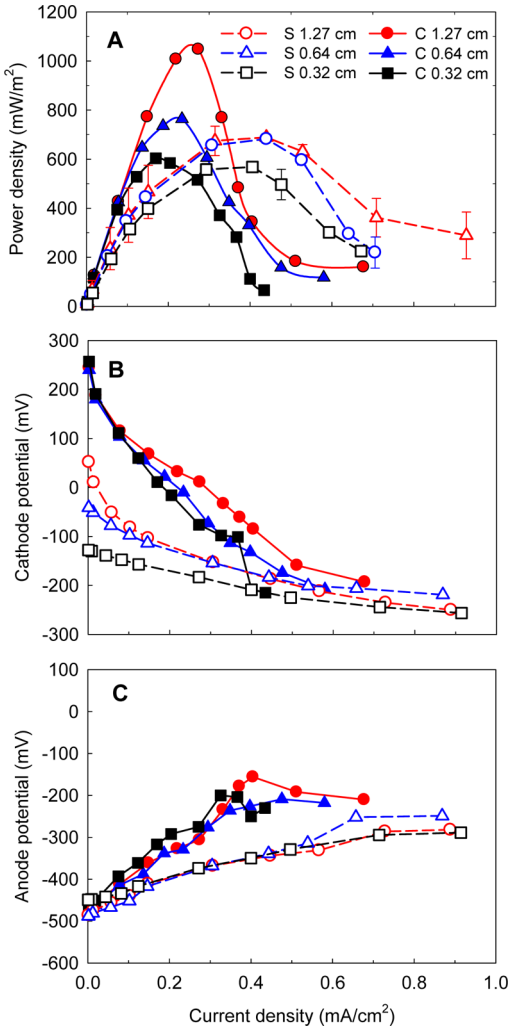

Figure 5. (A) Power density curves and (B and C) electrode potentials with different anode thicknesses and reactor configuration, where S represents "separator electrode assembly configuration" and C represents "spaced electrode configuration".

the thickest (1.32 cm) anode compared to 656 ± 6 mW/m^2 (16 W/m^3) for the 0.64 cm anode and 558 ± 60 mW/m^2 (14 W/m^3) for the 0.32 cm anode.

The maximum power densities obtained with the MFCs using the widely spaced anodes demonstrated improvement with using thicker anodes, and the two thickest anodes produced higher maximum power densities than those with the SEA configuration (Figure 5). The maximum power density of 1048 mW/m^2 (26 W/m^3) for the 1.27 cm anode was 52% higher than that obtained using the SEA MFCs with the same anode thickness. The MFC with the 0.64 cm anode produced 764 mW/m^2 (19 W/m^3), and the MFC with the 0.32 cm anode produced 604 mW/m^2 (15 W/m^3).

The results of the individual electrode potential assemblies suggested that the decreased performance of the SEA configuration compared to that of the widely spaced electrode configuration resulted from more negative cathode potentials (Figure 5B). At similar current densities, the cathode potentials for the SEA configuration were all calculated to be more negative than those in the MFCs with the spaced electrodes. However, it cannot be determined with any certainty that the cathode potentials changed because of the position of the reference electrode and the thick anodes, as will be discussed below. The anode potentials were measured to be the same at the different current densities for the SEA configuration, with values that are all more negative than those of the widely spaced electrodes (Figure 5C).

3.3. Internal Resistance. EIS spectra obtained were much different for the reactors with different anode thicknesses and configurations (Figure 6). Total internal resistances were much

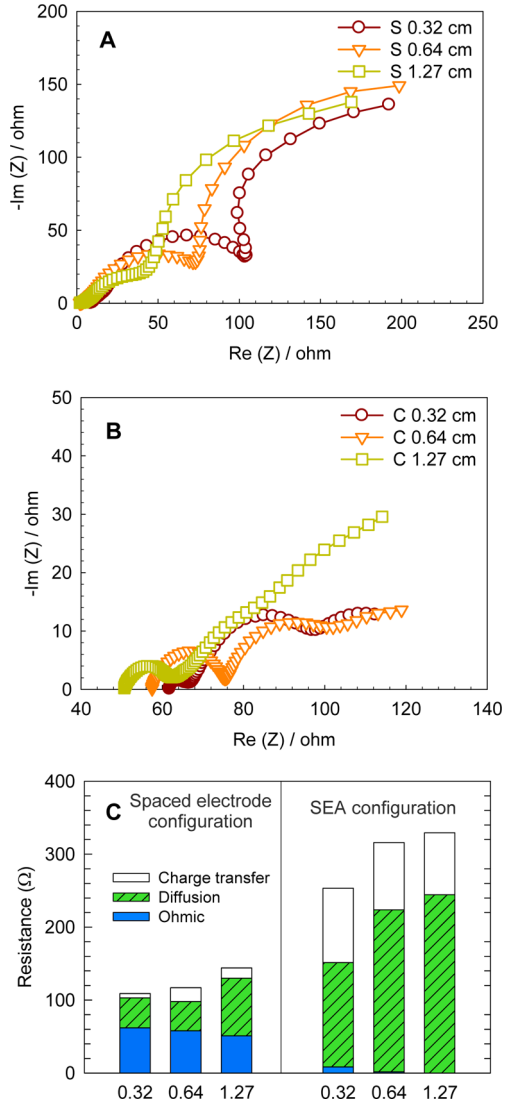

Figure 6. (A and B) Whole cell impedance spectra under working cell conditions and (C) internal resistance distribution for the MFCs with separator electrode assembly configuration and spaced electrode configuration reactors.

higher with the SEA type of configuration, despite a reduction in the solution resistance because of the decrease in the electrode spacing (Figure 6C). The ohmic resistances ranged from 51 to 62 Ω for the MFCs with the spaced electrodes compared to <8.4 Ω for MFCs with the SEA configuration. The SEA configuration resulted in much larger charge transfer and diffusion resistances of 85−102 Ω, resulting in total internal resistances that ranged from 253 to 329 Ω for the SEA configuration compared to 109−144 Ω for the spaced electrode configuration. For both configurations, the solution resistance slightly decreased with an increasing anode thickness and the

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diffusion resistances were much larger than charge-transfer resistances in all conditions. These results are consistent with the maximum power density results, demonstrating that the spaced electrode configuration was superior to the SEA configuration.

4. DISCUSSION

Increasing the thickness of the anodes in MFCs with space between the electrodes increased power from 604 mW/m^2 (0.32 cm) to 1048 mW/m^2 (1.27 cm). This result was not due to a slightly reduced distance that occurred with the thicker anode, because the solution resistance changed by only 18% (from 62 to 51 Ω) using the thicker anode, which was only a 10% change in the total internal resistance. These results for anode thickness show a trend similar to effects on power generation between thick graphite fiber brush anodes and thin anodes.[14,27] The brush configuration produces a greater anode volume and more surface area for bacterial growth than much thinner and flat anodes, resulting in higher power densities.[14] Here, a similar outcome was achieved using a relatively thick carbon felt anode.

The maximum power density obtained here with the very thick (1.27 cm) carbon felt anode was quite similar to that reported for a 2.5 cm diameter (2.5 cm long) brush anode (940 ± 100 mW/m^2), where both solutions had similar ionic conductivities.[31] However, it was found for the brush anode that removing up to 65% of the brush (the part most distant from the cathode), producing a 0.88 cm thick brush, did not affect power generation.[31] Similarly, the brush could be decreased in thickness from 2.5 to 1.25 cm by pressing it against a separator without affecting power.[27] However, here a decrease in the carbon felt thickness from 1.27 to 0.64 cm clearly reduced power generation. The carbon felt has a relatively high porosity (~99%) compared to other types of flat anodes[21] (i.e., carbon paper > 70%),[34] suggesting that both electrode porosity and size are important for power generation in MFCs.

The use of thicker anodes had much less impact on power generation when they were used in MFCs with the SEA type of configuration. The maximum power density using the thickest anode (689 mW/m^2, 1.27 cm) was only 23% more than that obtained using a 0.32 cm thick anode (558 mW/m^2), which was lower than maximum power densities obtained with the widely spaced electrodes. The ohmic resistance decreased as the electrodes were moved closer to each other, but it is well-known that this does not necessarily increase power output by MFCs. Oxygen transfer through the cathode adversely affects power production by the anode,[23] and therefore, power can decrease with a reduced electrode spacing, despite the improvement in ohmic resistance. Using a separator between the electrodes can help to mitigate this effect of oxygen, because the separator decreases oxygen crossover to the anode.[4,23] However, the separator used here, in conjunction with the thicker felt anodes, did not improve power generation in the SEA configuration. This suggests that improved power will require improved separators and not just thicker anodes.

The decrease in the power generation using the SEA configuration was indicated to be a result of a decrease in the cathode potentials based on reference electrode measurements. However, this conclusion is likely an artifact of only being able to make anode potential measurements on the side of the anode distant from the cathode. The reference electrode was placed close to the anode, but the anode is a highly porous structure. The cathode potential could not be directly measured in the SEA configuration, and therefore, it was calculated from the anode and whole cell potentials. While a separate reference electrode can be used to measure the cathode potential when there is space between the electrodes, it cannot be inserted next to the cathode with a SEA configuration because there is no room between the separator and the cathode. Thus, any changes in performance were attributed to the cathode based on the measured change in the anode potential. When using highly porous and very thick anodes, the potential measured relative to the reference electrode near the outer edge of the anode in the SEA configuration may not reflect the whole anode potential. Thus, it is possible that the anode potentials measured here did not adequately reflect the overall anode potentials.

There is good evidence in the results obtained with the SEA configuration compared to the widely spaced electrode results to support this hypothesis that the whole anode potentials were not properly evaluated for the thick and highly porous anodes. The anode potentials measured in the SEA configuration were indicated to be uniformly low and essentially identical for the anodes with three different thicknesses (Figure 5). However, if oxygen leaks through the cathode, we would expect the anode potential to become more positive in the SEA configuration, consistent with previous results using thin anodes that anode potential increases as the power density decreases in SEA configurations. Here, the power did decrease in the SEA configuration relative to the spaced electrode configuration, but the anode potentials apparently remained relatively negative. This suggests that the potential of the anode surface relative to the reference electrode remained relatively negative (because oxygen was consumed inside the anode) but that the anode surface near the cathode likely had a more positive potential. Because the power was lower with the SEA configuration and the anode potential was lower, then the calculated result is that the cathode potential was reduced in the SEA configuration. This appears unlikely, because it is counter to results obtained in many other studies. In addition, we see a much faster rise in the anode potential for the spaced electrode results than that for the SEA anode results. Thus, we conclude that there was an adverse effect of oxygen on the anode and not the cathode, and the derived results suggested that the changes in cathode potentials were not accurately portraying the effect of the SEA configuration on the individual electrode potentials.

5. CONCLUSION

The use of thicker anodes improved power production in MFCs when there was sufficient spacing between the anode and cathode. Under these conditions, increasing the anode thickness from 0.32 to 1.27 cm improved power densities by 23% (from 558 to 689 mW/m^2). When the anodes were used in a SEA configuration, maximum power densities were reduced in comparison to those produced with the spaced electrodes. In addition, there was less of an effect of anode thickness on performance. The SEA configuration, however, did improve CEs to 37−50% compared to those of 19−23% with the widely spaced configuration. These results show that the use of thick anodes alone cannot overcome the deleterious effect of oxygen crossover from the cathode to the anode. However, if this oxygen crossover can be better controlled and reduced using effective separators, then it is likely that the use of thicker anodes will improve performance.

AUTHOR INFORMATION

Corresponding Author

*Telephone: 814-863-7908. E-mail: blogan@psu.edu.

Notes

The authors declare no competing financial interest.

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