PAPER

Scale up considerations for sediment microbial fuel cells†

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Scale-up of sediment microbial fuel cells (SMFCs) is important to generating practical levels of power for undersea devices. Sustained operation of many sensors and communications systems require power in the range of 0.6 mW to 20 W. Small scale SMFC systems evaluated primarily in the laboratory indicate power densities for typical graphite plate anodes on the order of 10–50 mW m^{-2}. However, previous work also suggests that SMFC power production may not scale directly with size. Here, we describe a combination of lab and field studies to evaluate scale up for carbon fabric anodes with a projected surface area ranging from 25 cm^2 to 12 m^2. The results indicate that power generation scales almost linearly with anode size up to about 1–2 m^2 of projected surface area. Our model suggests that anodes larger than this can experience significant reduction in power density, confirming laboratory observations. These results suggest that the majority of losses along the anode surface occur closest to the electronics, where the amount of current passing along an anode is the greatest. A multi-anode approach is discussed for SMFCs, suggesting that scale-up can be achieved using segmented anode arrays.

Introduction

Bioelectrochemical systems (BESs) are an emerging research field with applications in renewable energy, water treatment, product synthesis, and remediation.[1–4] One of the largest challenges facing commercialization of the technology is the design of practical solutions for large scale systems. In recent years, large scale systems have been tested in different settings with mixed results.[1,2,5] In general, most applications of commercial microbial fuel cells do not focus on energy production or harvest primarily because of the widespread availability of cheap electricity sources.[1] However, in scenarios where access to major electrical grids is limited, the cost of supplying sustained electricity can make BESs aimed at providing energy a worthwhile option.

Sediment microbial fuel cells (SMFCs) are one application where generating practical levels of power for undersea devices is a primary focus.[6] Many oceanographic sensors and communications systems require power in the range of 0.6 mW in sleep mode to 20 W in active mode.[6] Small-scale SMFC systems evaluated in the laboratory indicate power densities for typical graphite-plate anodes on the order of 10–100 mW m^{-2}.[6–10] Total power from prototype SMFC systems deployed in the field have generally resulted in power levels less than 100 mW of sustained power.[6,11–16] Scale up of SMFCs has been approached in several ways including improving power density through better electrode materials,[17–20] variations of electrode geometry,[21–24] improving efficiency through optimizing electronics or operation,[14,25–27] and by increasing the size of the system through larger electrodes or through arrays of modules.[8,22,23,28–30]

Given an electrode material with a known and reasonable level of performance such as carbon fabric, a typical two-dimensional buried panel geometry, and the assumption that large systems are likely to be deployed as arrays of smaller systems, a fundamental question for SMFCs is: what is the largest electrode that can be used for a single array module and still achieve adequate performance? This is an important question since electrode arrays constructed from large numbers of very small modules are likely to be much more complicated and expensive than arrays constructed from a small number of larger modules.

Previous work suggests that SMFC power production may not scale directly with electrode size.[31] Power density has been shown to decrease rapidly when the surface area of the current-limiting electrode is increased. Significantly higher power densities were achieved for electrodes less than 10 cm^2 and no change in power densities for electrodes greater than 50 cm^2. To evaluate this observed trend, Dewan *et al.* also conducted a scaling experiment analysis under more consistent conditions using a two-cell laboratory MFC with liquid media in the cells, and a monoculture of *Shewanella oneidensis* MR-1 grown anaerobically on the anode.[31] These results

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showed a similar trend with a reduction of maximum power density for larger anode sizes.

Other studies that focused on scale-up of MFC systems targeted at applications in wastewater treatment plants suggest that substrate concentration is important to anode performance, while solution conductivity and specific surface area are important to cathode performance.[2,5] However, to date, there have been no systematic scaling experiments performed with SMFCs under realistic natural conditions, and at scales approaching those required to power seafloor sensors and systems.

Specific challenges related to SMFC systems need to be considered when attempting to increase size and power output. Unlike other reactor systems, just increasing electrode or reactor size is insufficient for benthic sub-marine systems. Both deployment (how the system will be implanted in the sediment) and waterproofing for electronics systems need to be considered in addition to electrode size. These factors increase the complexity of making SMFC systems functional in the ocean environment and require a balance of several engineering considerations.

Here we describe a combination of lab and field studies to identify scale-up considerations for carbon fabric anodes for systems with geometries compatible with deployment without divers.[23] Initial lab studies with anode surface areas between 25 cm^2–6,400 cm^2 were performed before follow-on studies to evaluate large scale single anode or multiple anode systems for in the lab and field. These results are discussed in the context of scale-up design considerations for large scale SMFC systems.

Methods

Laboratory SMFCs

Five sets of SMFCs were set up with projected surface areas of the anodes at 25, 100, 400, 1600, and 6400 cm^2. The 25, 100, and 400 cm^2 anodes included triplicate replication, while the two larger anodes had duplicates. The anodes all had the same 1 : 8 aspect ratio and were constructed of carbon fabric (CC6; Fuel Cell Earth LLC; Stoneham, MA). This relatively long and narrow geometry was selected so that these laboratory experiment would be consistent with the field systems that required this shape for deployment.[23] Short lengths of insulated titanium wire were sewn onto the end of each anode for secure connections to be used as electrical leads. All SMFCs used a common cathode consisting of a 150 cm long carbon brush with a titanium core wire (The Mill-Rose Company; Mentor, OH).

The fuel cells were all set up in a large tank (183 cm × 93 cm × 61 cm). Sediment from a field site in San Diego Bay was collected and homogenized before being used to fill the tub to a depth of about 15 cm. The anodes were laid flat onto this sediment (Fig. 1) and an additional 10 cm layer of sediment was placed on top to uniformly bury the anodes. Each of the titanium wires from the anodes were connected to insulated copper wire that terminated at individual screw terminal

Fig. 1 Lab SMFC scaling experimental layout in the large tank prior to burial of the anodes. The anodes are sitting on top of the underlying sediment layer in the tank. The smallest anodes are just visible at the bottom of the photo. The insulated titanium wires can be seen leading from the anodes out of the tank. A yardstick is also seen on the right-most electrode for scale.

junctions. The titanium to copper connection was made outside of the tub to prevent oxidation of the copper wire.

The remainder of tank was then filled with seawater from the laboratory flow-through system that provided water directly from San Diego Bay. The cathode was suspended in the water approximately 10 cm above the sediment bed and connected at each end to an insulated copper conductor running to the screw terminal strip. To maintain constant aerobic conditions in the overlying water, an air bubbler was installed in the overlying water and fresh seawater was continually exchanged with the overlying water at a rate of approximately 10 L min^{-1}. Environmental conditions were monitored with handheld meters during the experiment and average observed values are given in Table 1.

Anode and cathode potentials were measured against a Ag/AgCl reference electrode (Model 13-620-45; Fisher Scientific; Hampton, NH) suspended in the overlying water. Potentials were measured and recorded every 5 min utilizing a custom program created with Labview (NI Labview 2011; National Instruments Corporation; Austin, TX) interfacing with a data acquisition unit (Model USB-1616FS; Measurement Computing Corporation; Norton, MA). The SMFCs were monitored under open circuit conditions until a stable open-circuit potential (OCP) of about 0.7–0.8 V was reached. Over the next month, the individual SMFCs were subjected to increasingly higher loads by installing progressively smaller passive resistors until all of the systems had stabilized at

Table 1 Average environmental conditions measured in laboratory studies. The reported range indicates one standard deviation

Environmental Parameters:

Seawater
pH	7.83 ± 0.02
Conductivity	51.9 ± 0.7 mS
Temperature	20.6 ± 0.9 °C
Total dissolved solids (as NaCl)	31.8 ± 0.5 ppt
Oxidation-Reduction Potential (ORP)	$+176 \pm 23$ mV

Sediment (near anode)
pH (before SMFC operation)	7.13 ± 0.07
pH (during steady state SMFC operation)	6.05 ± 0.36

working potentials (WPs) of about 0.4 V. The known resistances (R), the measured WPs (V), and the constructed surface areas (A) were then used to calculate current, power, and power density as described previously.[3]

Field microbial fuel cell

A preliminary effort to extend knowledge gained from the lab experiments was done by deploying a larger MFC system in the same site that sediments were gathered. Because of the large size, only one system was deployed in the same area due to the prohibitive cost and effort associated with deploying multiple large scale systems.

The large SMFC was deployed using an unmanned sled deployment device[23] at the same site in San Diego Bay where the sediments were collected for the laboratory experiments. This single SMFC system had an anode surface area of 12,000 cm² with a similar aspect ratio as the lab systems (1 : 6) and was constructed from the same carbon fabric (note that the deployed anode had a 1 : 8 aspect ratio, but the last two meters inadvertently tore off during deployment resulting in the 1 : 6 aspect ratio). The fabric was terminated at one end with an aluminum flat-bar clamp that was secured along one of the narrow ends of the fabric. An underwater cable (Teledyne Impulse; San Diego, CA) was connected to screw terminals on the aluminum flat-bar. The entire aluminum clamp and underwater cable connection was placed inside a 25 cm diameter PVC pipe and sealed in waterproof epoxy to ensure protection against oxidation and seawater.

The cathode consisted of an identical titanium-carbon fiber brush used in the lab, except the length was 600 cm. An underwater cable was connected to the titanium stem of the brush, and the entire connection was then sealed in polyurethane. The anode and cathode were connected to an underwater electronics housing that contained a custom energy harvesting and voltage conversion system (Northwest Metasystems; Bainbridge, WA) and a data logging system (QuadVolt; Madgetech, Inc.; Contoocook, NH). This system contained a potentiostat that maintained open circuit conditions for the SMFC until the OCP rose to 0.4 V. Once sufficient potential was available, the system regulated the WP to 0.4 V. Outputs from the system including WP and SMFC current were monitored and recorded every 5 min. The field SMFC was installed and buried in San Diego Bay using a sled device as described previously.[23] The system was left in place and monitored for about 4 months from August 25, 2011 through December 12, 2011. The measured current, WP, and the surface area were then used to calculate power and power density.

Validation for a multi-anode SMFC

Operational SMFCs from the initial laboratory testing were used to examine whether any additional loss in performance would result from a multi-anode SMFC approach to scale up. Anodes that were the same size were connected to a single insulated copper wire to make multi-anode systems. Monitoring of the WP was performed using the Labview data acquisition setup described previously. Passive resistors were used to load each of these systems to a WP of 0.4 V. Current, power, and power density calculations were made once WPs stabilized for each system.

Linear sweep voltametry was then used on these systems to generate polarization and power density curves. The systems were allowed to equilibrate under open circuit conditions (about 12 h) before a potential sweep was performed with a potentiostat (Model EA163; eDAQ Inc.; Colorado Springs, CO) using the anode as the working electrode, cathode as the counter electrode, and a Ag/AgCl reference. The potential sweep was set to begin at the anode potential under open circuit conditions (-0.4 V vs. Ag/AgCl) and was raised to the potential of the cathode ($+0.4$ V vs. Ag/AgCl) under open circuit conditions with a scan rate of 0.05 mV s^{-1}.

Validation for a SMFC module

Following the scaling experiment, a conceptual design was developed for a segmented anode where the segments were of a scale large enough to be practical, but not so large as to incur significant reductions in performance. Duplicate anode segments each consisted of a 100 cm × 100 cm carbon fabric sheet with a projected surface area of 20,000 cm². At locations 25 cm from each end, a titanium collector wire was sewn across the entire width of the sheet. These collector wires were connected to a pair of two conductors, 12 gauge insulated copper flat wires (9688T16; McMaster-Carr Supply Company; Robbinsville, NJ) that ran the length of the sheet. The flat wires were fixed to the sheet with an adhesive backing. The connection between the titanium wire and the flat wire was made by piercing the titanium wire through the insulation of the flat wire, injecting methyl 2-cyanoacrylate adhesive into the connection region, and clamping the two wires together while the adhesive dried. At one end of the segment, the flat wires were connected to 12 gage insulated copper wire for connection to a terminal block. Any potentially exposed flat wire (unconnected ends or connections with titanium) was then covered with flexible marine sealant (Type 5200; 3M Company; St. Paul, MN) to prevent degradation of the copper wire. The segment was installed in the large laboratory tank and covered with about 15 cm of sediment as described previously for the scaling experiment.

The tank was then filled with seawater from the laboratory flow-through system that provided water directly from San Diego Bay. A cathode consisting of a 365 cm long by 10 cm wide carbon fabric strip sewn onto a titanium wire was suspended in the overlying water approximately 10 cm above the sediment bed. Each end of the titanium wire was

connected to a terminal block by 12 gage insulated copper wire. Monitoring of anode and cathode potentials during operation was performed as described for the previous laboratory SMFCs.

The segment was monitored at open circuit until it stabilized at an OCP of about 0.8 V. Over the next month, the SMFC was conditioned for power output by installing progressively smaller resistors until the system had stabilized at a WP of about 0.4 V. Current, power output, and power density were calculated as described previously.

Results and discussion

Lab SMFC scaling results

Power output from the laboratory SMFCs increased as a linear function of anode size, ranging from 0.04 mW for the smallest anodes to 13.3 mW for the largest anodes (Fig. 2). Although power did increase with size, it did not increase in direct proportion, and thus larger anodes generally exhibited lower power densities than small anodes (Fig. 3).

Observed water quality parameters of incoming seawater to the SMFC system were not observed to vary significantly over the course of the experiment. The pH of the sediment did appear to decrease by slightly more than one pH unit. This can be attributed to the oxidation of carbon compounds and mass transfer limitations for proton transport out of the sediment to the water column. While Gardel *et al.* indicated almost no change in pH with SMFC operation,[28] our studies showed a

Fig. 3 Lab SMFC (black diamonds) and field SMFC (red square) maximum power density as a function of anode surface area. Lab SMFC data was generated from constant load experiments and field data was obtained from potentiostatic load. Error bars are one standard deviation on the lab replicates. The dashed lines represent estimated power density from the model results for SMFCs with anode material resistivities in the ranges shown.

decrease in pH by a full pH unit which is consistent with a freshwater SMFC system reported by Hong *et al.*[32]

A general trend that was observed and that has been seen previously was that the smaller anodes tended to have higher variability (Data not shown). We attribute this variability to the heterogeneity of natural sediments and of biofilm development at the electrode surface, *i.e.* the larger anodes could tend to average out variations sediment composition and biofilm development. It was also observed that the power produced by the 400 cm^2 anodes was about 80% of the expected value (Fig. 3). A potential explanation of this performance could be linked to the conditioning of these SMFCs, which were unintentionally under-loaded (allowing WP to drop below 0.4 V) during the startup phase. Although the WP recovered under different resistive loading, these systems never reached the same power density levels as the other similarly sized anodes indicating the importance of anode potential during biofilm formation reported by others.[33]

Field SMFC results

Starting from the installation date on 8/25/11, the WP for the field SMFC rose until it reached the regulated level of 0.4 V on 8/27/11 (Figure S1). The SMFC then began to generate power and increased until power output stabilized at about 80–90 mW on about 9/18/11 (22 days). The SMFC maintained these
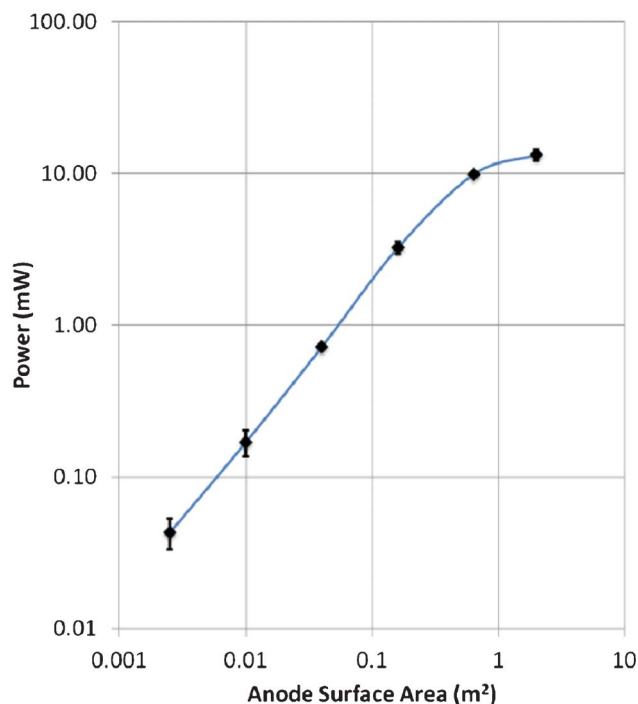
Fig. 2 Lab SMFC scaling results showing the increase in maximum power output as a function of anode surface area. Error bars represent one standard deviation.
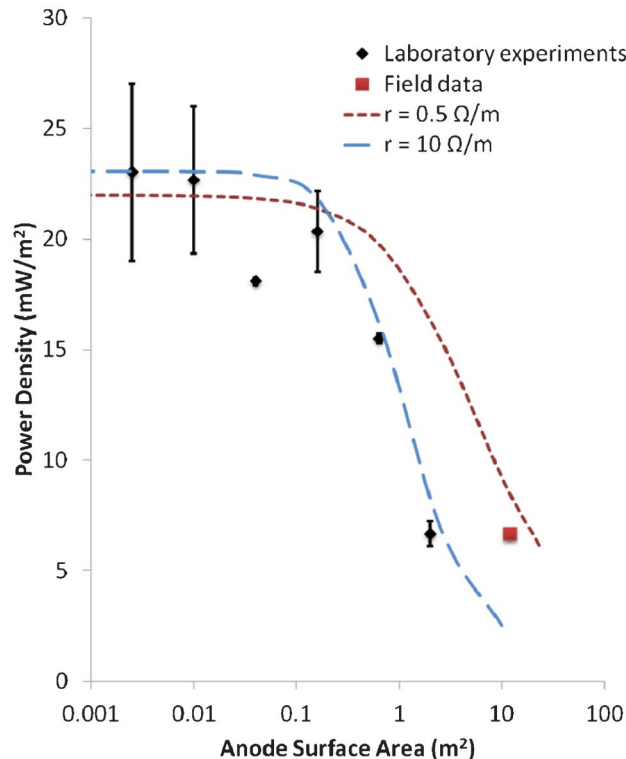
power levels with only minor deviations until the system was shut down on 12/12/11. The average power produced during the period following stabilization was about 84 mW, resulting in a power density of about 7 mW m^{-2}. This result is included in Fig. 3, extending the relationship between size and power density to this larger scale. Although this is only a single measurement, the result is consistent with our observation that power density drops off substantially as anode size increases above 1 m^2.

Altogether, the scaling results suggest a trend in which fabric anode sizes with surface areas greater than about 1 m^2 may suffer significant reductions in performance. Mass transport limitations of organics may also be a factor in establishing maximum power density of a system, especially at smaller anode sizes. However, the potentiostat is able to minimize operation in the mass transfer limited regime by keeping the WP at an appropriate level. Because these systems were all deployed in the same sediment, using the same materials, and comparable connections and measurement methodologies, the differences in performance seem to be specifically related to the anode size.

We expect that the performance of these SMFCs is generally constrained by diffusion-limited mass transport within the sediment. However, we don't expect that organic carbon mass transfer limitations have a major impact in SMFCs utilizing our anode sizes. Instead, we have concluded that the reduction in performance may be related to two factors. For progressively larger carbon fabric anodes, ohmic losses grow as current levels rise. This leads to a non-uniform operating potential (and deviation from our operational setpoint) along the anode and leads to underperforming anode sections. Thus, the overall reduction in performance is due to the both the ohmic losses associated with conduction of electrons through the anode material and the impact of underutilizing sections of larger anodes. Secondly, the proton transfer limitations result in low sediment pH around the anode during long term SMFC operation in the lab, potentially limiting power output.

The microbial communities in the field and lab systems are expected to be similar since sediment for the lab systems was collected from the same location and depth as the field system. Operational conditions in the field and lab were designed to mirror each other. In addition, good agreement between the different systems in terms of electrochemical performance was observed and the systems are comparable from a performance point-of-view. Determining the exact nature of the microbial community may be helpful to determine fundamental microbial interactions in the MFC environment, and a more detailed microbial ecology study will be performed in the future.

SMFC performance modeling

To evaluate the influence of these factors on SMFC power production, we constructed a simple finite-element model that incorporated the ohmic losses (described by the characteristic I–V drop along the anode due to the resistance of the anode material. This potential drop along the anode results variations in anode operating potential, which have a direct impact on the amount of current generated by a particular section of anode. In this model, anode potential is governed by the

current (determined from current-power relationships) passing through each element of the model and the resistance of the anode material (eqn (1)–(3)).The resistance associated with anode-cathode separation were calculated for observed seawater conductivities and did not appear to be a significant contributor to losses.

$$E_{anode}(x) = \max\left\{ E_{pot} - \int_D^x I_{anode}r\,dx, E_{oc} \right\} \qquad (1)$$

$$I_{anode} = \int_l^{l-x} iw\,dx \qquad (2)$$

$$i = \frac{(mE_{anode} + b)}{A_{anode}} \qquad (3)$$

(over the linear portion of the I − V curve)

Here, E_{anode} is the anode potential as a function of distance (x) along the anode, E_{pot} is the WP set by the potentiostat (or the load) at the root of the anode ($x = 0$), I_{anode} is the current flowing through the anode at location x, r is the specific resistance of the anode material, E_{oc} is the open circuit potential, i is the current density, w is the width of the anode, l is the length of the anode, m and b are the slope and intercept of the I–V curve, and A_{anode} is the surface area of the anode.

Environmental characteristics (pH, temperature, substrate concentration) affect current production in eqn (3). Changes in these parameters that impact the redox chemistry and biofilm behavior could be incorporated into the model through proposed bioanode models.[34,35] Hamelers et al. described a Butler–Volmer–Monod model to describe MFC polarization curves from fundamental reaction kinetics with good agreement to experimental data.[34] Since our operation conditions were considered at steady state, we modeled our SMFC system using graphical approximation (for "m" and "b") taken from experimental SMFC polarization curves (Figure S2).

The model was first used to evaluate the effect of anode material resistance for the field anode configuration (1 m × 6 m) using several different material resistivities (Fig. 3 and 4). The results show minimal impact of resistivity on anode potential for values between 0.01–0.001 Ω m^{-1}. On the other hand, the potential begins to drop significantly along the length of the anode at resistivities of 0.1 Ω m^{-1} and larger. This potential drop has direct impacts on current generation and power performance and suggests that anode size begins to limit power production at sizes similar to the deployed system. The model was then used to predict power density for anodes with the same geometries that were used in the lab and field experiments. The results for two model runs that bracket the range of measured specific resistance for the anode material are shown plotted with the experimental data in Fig. 3. Results from the model are also consistent with observed power output from the field SMFC (Figure S1) given the range of resistivities measured for the carbon fabric (Fig. 5).

Consistent with the experimental data, the model suggests that performance drops off for anode areas larger than 0.1 m^2 and significant performance impacts for anodes larger than 1
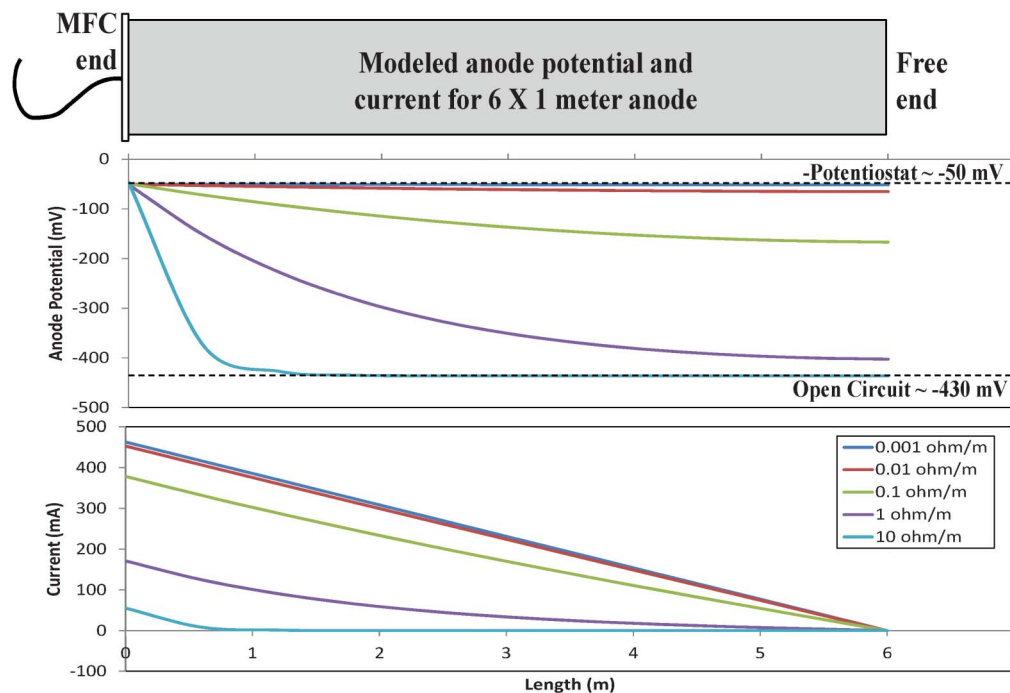
Fig. 4 Modeled anode potential and current behavior for the 6 m × 1 m anode geometry deployed in the field.
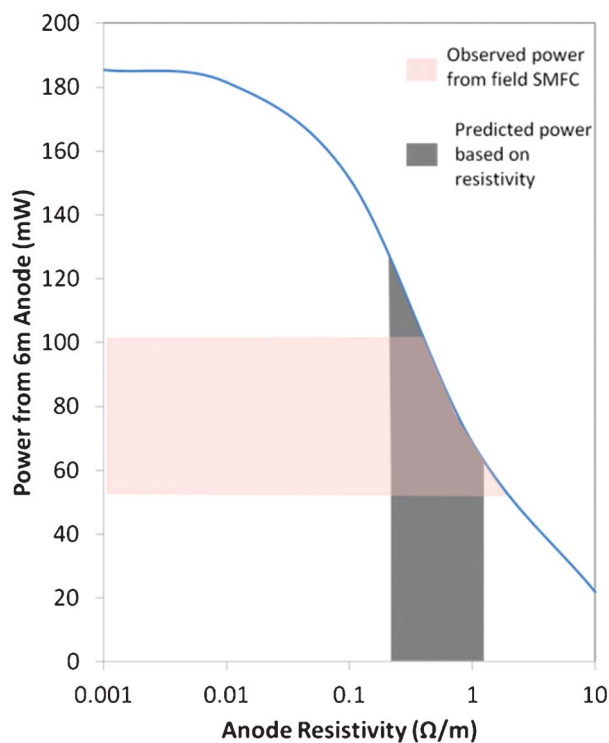
m². Model results also suggest that only about 30% of the power degradation is related to internal losses, and the majority is related to the potential drop across the anode, resulting in operation of those anode portions at suboptimal electrode potentials. Interestingly, the majority of the ohmic losses occur near the root of the anode (connected end) where the cumulative current becomes very high, while the majority of the suboptimal cell voltage losses occur toward the free end of the anode where the cell voltage approaches the open circuit potential.

Validation for a multi-anode SMFC

Power density calculations from creating a multi-anode SMFC from operational SMFC anodes showed no decrease in power output when anodes of similar size were ganged together. Power density curves of these combined anode systems from the polarization curve data (Figure S2) indicated that the maximum power density for multi-anode larger surface area systems remained relatively unchanged from their operation as single anode systems shown in Fig. 2. These results suggest little to no impact on performance would result from using a multi-anode approach to create higher power SMFC systems.

Based on these experimental and modeling results, we approached the issue of minimizing scale up losses from anode size by adopting a multi-electrode approach. For this method, we constructed a single segment of what could be part of a multi-anode SMFC as described in the methods section above. Results from the module testing resulted in stable power generation of about 42 mW from a total surface area of 2 m² (a power density of about 21 mW m⁻²). This power density is consistent with the maximum power density achieved from the small anodes in the original scaling

Fig. 5 Predicted maximum power production for the 6 m × 1 m anode geometry over a range of anode resistance levels. The dark shaded range corresponds to the range of measured resistance levels for the carbon fabric, and the lightly shaded area indicates the power levels seen in the field.

experiment, suggesting that an SMFC constructed from several of these modules may perform significantly better than an SMFC based on a single large anode configuration, and that these modules can be constructed at a practical size scales that can minimize the complexity and logistics in creating large numbers of very small anodes.

We can extrapolate, from this data here, the design of a multi-anode SMFC with a maximum single anode size of between 1–2 m^2. Above this size, the resistive losses in the materials dominate losses and limit the efficiency of the system. In future systems, we will use this information as design criteria for large scale SMFCs.

Conclusions

These results provide a basis for scale-up of SMFCs using fabric anodes that can operate in the desired range for many seafloor applications. Inexpensive installation of SMFCs capable of sustaining meaningful power could provide infrastructure for sensor networks or power stations for remote vehicles at the seafloor. The studies reported here identify several design considerations that will need to be addressed while engineering large-scale SMFC systems.

We have identified specific design criteria for large SMFC systems in addition to identifying key system losses that should be avoided by utilizing a multi-anode approach. This multiple anode approach has several additional benefits beyond just avoiding resistive losses. For example, each of the anodes in a system can be stacked (using appropriate electronic circuits) to yield output voltages greater than the typical low voltages observed with MFC systems. Furthermore, the multiple anode system gives some robustness to SMFC systems by allowing non-performing or ineffective sections of anode (e.g. from oxygen intrusion or unburying) to be isolated. This prevents the whole SMFC system from being affected by localized unfavorable environmental conditions.

Future work on these systems will focus on designing and testing of a multi-anode SMFC utilizing a diver-less deployment method. Efforts to identify effects of sediment quality on power output at the seafloor (e.g. TOC, grain size, temperature) are also under way. Work to follow up on the full effects of sediment permeability on mass transport limitations is planned. Additional efforts to minimize power requirements for a payload (sensors and/or communications) as well as efficient power management techniques are being explored as well.

Acknowledgements

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