Review

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Assessment of Microbial Fuel Cell Configurations and Power Densities

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Abstract

Different microbial electrochemical technologies are being developed for a many diverse applications, including wastewater treatment, biofuel production, water desalination, remote power sources, and as biosensors. Current and energy densities will always be limited relative to batteries and chemical fuel cells, but these technologies have other advantages based on the self-sustaining nature of the microorganisms that can donate or accept electrons from an electrode, the range of fuels that can be used, and versatility in the chemicals that can be produced. The high cost of membranes will likely limit applications of microbial electrochemical technologies that might require a membrane. For microbial fuel cells, which do not need a membrane, questions remain on whether larger-scale systems can produce power densities similar to those obtained in laboratory-scale systems. It is shown here that configuration and fuel (pure chemicals in laboratory media versus actual wastewaters) remain the key factors in power production, rather than the scale of the application. Systems must be scaled up through careful consideration of electrode spacing and packing per unit volume of reactor.

Introduction

Although microbial fuel cells (MFCs) have been investigated for many years, the first substantial breakthrough occurred in 1999 when it was realized that chemical mediators did not need to be added into the system to achieve power production.[1-3] Practical applications for wastewater treatment were then envisioned to be feasible based on the development of air cathodes,[4] which meant that wastewater did not need to be aerated, potentially enabling both wastewater treatment and electrical power production. However, it has been more than a decade since air cathodes and mediatorless MFCs were first proposed, and yet there are still no commercial applications of the technology. What has limited translation of laboratory-scale processes to larger scales? One main reason is the cost of the electrodes. It was estimated that the electrode materials would need to cost less than 100 € per square meter (~$110 USD) to make them economically viable.[5-7] This now seems to be possible with advances in inexpensive anodes,[8] separators,[9-11] and cathodes based on activated carbon catalysts.[12-14] Another factor that could limit the development of larger-scale MFCs is diminished power at larger scales. However, it is argued here that the main difficulty is not an intrinsic loss of power at larger scales, it is maintaining reactor geometry relative to electrode configurations and densities as larger reactors are built to handle greater water flows.

A Range of Microbial Electrochemical Technologies

MFCs can be used to produce electricity, but the use of microorganisms on the anodes or cathodes, or both electrodes, has enabled the invention of many other systems for a variety of different purposes. All of these other microbial electrochemical technologies (METs) will face similar or added challenges during scale up, and thus they are worth examining in terms of components and potential applications. METs have often been identified using variations on an MxC theme, where “x” denotes the specific application, for example x=F in the abbreviation MFC (Table 1). The first main variation on the MFC was

modifying the system to produce hydrogen gas. The omission of oxygen at the cathode, and addition of a voltage to the circuit enabled hydrogen gas production in microbial electrolysis cells (MECs) at voltages theoretically larger than ~0.2-0.3 V.[15, 16] These are much less than those used for water electrolysis of >1.2 V, although in practice applied voltages in MECs are typically 0.6 V or more.[17]

Membranes do not have to be used in MFCs or MECs, as they do in fuel cells and water electrolyzers as the water is the ion conducting medium. One important advance in improving power production in an MFC was showing that the cation exchange membrane (CEM), which was often made from expensive Nafion, could be omitted.[18] In addition, the use of anion exchange membranes (AEMs) were shown to improve power compared to CEMs due to transfer of negatively charged phosphate, carbonate ions, or hydroxide ions.[19] However, the use of membranes can create pH imbalances, limiting the extent of power generation.[20] The use of non-ion selective separators can minimize this pH problem, but all chemicals can cross between the chambers. Using a CEM or AEM enables production and recovery of commodity chemicals such as caustic solutions in MECs or MFCs, and hydrogen peroxide in MFCs.[21, 22] While membranes are needed for two-chamber MFCs, they are not used in sediment MFCs (sMFCs), where the anodes for these are immersed in organic-rich sediments which provide the fuel, and the cathodes are placed above the sediment, to allow for oxygen reduction.[23, 24]

Two or more membranes are used in other types of METs for different functions. The addition of an AEM and CEM, with salt water in the middle, can be used for water desalination in concert with electricity production in an microbial desalination cell (MDC) (Table 1).[25, 26] Using stacks of membranes can increase the energy efficiency for desalination, similar to that of water electrolyzers, although the number of pairs of membranes is limited due to the limit on the voltage produced by the MFC.[27, 28] The use of many pairs of membranes in METs can enable power production from salinity differences of solutions. Inserting a stack of paired AEM and CEM membranes, known as a reverse electrodialysis (RED) stack, with water containing high or low salt concentrations in alternating channels can produce an

electrical potential. Insertion of a RED stack between the anode and an air cathode generate more power in a microbial reverse electrodialysis fuel cell (MRFC) than an MFC,[29] or produce hydrogen gas in a microbial reverse electrodialysis electrolysis cell (MREC) without the need for electrical power source as required for an MEC.[30, 31] Inclusion of a bipolar membrane next to the anode can enable acid production in the chamber formed by the bipolar membrane and an adjacent AEM, as well as desalination of water between the bipolar membrane and AEM, with a single chamber (MEDCC) or a RED stack (MREEC).[32] These type of systems with bipolar membranes can be used to enable recovery of both acid and caustic solutions (from the cathode chamber) that have commercial value.[32] Alternatively, these solutions can be used with minerals to create a carbon sequestration technology (Table 1).[33, 34]

METs are also being explored as a method for chemical production, either through direct microbial electrosynthesis of compounds via CO_2 reduction, or through modification of organic molecules to produce higher value chemicals in a microbial electrosynthesis systems (MES).[35, 36] For example, methane can be produced by methanogens on the cathode by different routes that are thought to include hydrogen gas,[37] electrical current,[38] or via molecules excreted by microorganisms, in MESs called microbial methanogenesis cells (MMCs).[39] Current generation on the anode can be biological in origin, as in an MEC, or from water splitting. Methanogens readily grow in single chamber MECs primarily using hydrogen gas, even when there are relatively high concentrations of acetate.[40] However, there is evidence for direct electron transfer due to much higher current densities, and recent studies have shown that the supernatant from MMCs can catalyze formate reduction using current from an electrode.[39] Organic products that can be released into solution in an MES include acetate, 2-oxobutyrate, and formate.[41] Chemicals can be modified to produce more valuable products, for example conversion of acetate into caproate and caprylate.[42] However, current densities and titer (yields) for these organic products are low, and expensive membranes are used in these systems, so it is not yet clear this chemical production route can be profitable.

Analysis of these different systems that require membranes suggests that a key limitation for their implementation is the cost of the membrane(s). Thus, MFCs and MECs lacking membranes will likely need to be viable before these other systems that require expensive membranes, unless product value and titer are very high.[36, 43]

Power Based on Different Applications

MFCs have been built at many different scales, ranging from volumes of microliters to tens and hundreds of liters, with examples of different types given in Figure 1. The relative simplicity of MFCs, the lack of a need of a membrane, and the many advances in reducing the cost of materials suggest that MFCs are likely to be the most useful MET in the near term. The power produced by an MFC is a primary design factor in some applications, for example powering devices in seawater using sediment MFCs (sMFCs).[24] Micro-sized MFCs (μMFCs) are also being developed for powering small wearable devices, or lab on a chip applications.[44] In other MFC applications, for example wastewater treatment, power production is desired but a more useful aspect of using MFCs is elimination of the need to aerate wastewater and reduction in sludge production. Power production by MFCs will never be that large, although the power produced by systems based on oxygen reduction has increased over the years, from <1 mW/m^2 of projected anode area, to as much as 6.9 mW/m^2-anode by using a comparatively large cathode.[45, 46] However, the highest power densities have all been obtained under near ideal conditions by using a high concentration of a fuel (such as acetate), and well buffered and highly conductive electrolytes. It has been estimated from consideration of microbial kinetics or minimizing reactor internal resistance that power densities could reach as much as 17–19 W/m^2, but these power densities are not likely to be realized in practical designs for wastewater treatment.

To determine how well air-cathode MFCs are being scaled up relative to these upper limits, we examined the range of published power densities by MFCs in the following categories: μMFCs used as

small power sources; sMFCs used as remote power sources; MFCs used with laboratory media and defined substrates; and MFCs treating actual wastewaters [MFC(WW)] or using relatively complex media. The μMFC data also included systems with a ferricyanide catholyte in order to provide a more realistic picture of their range in useful power, as applications envisioned for these devices could conceivably make use of terminal electron acceptors other than oxygen.[44] Systems that produced < 1 mW/m^2-cathode were excluded from our analysis. We focused on establishing the range of values, and so we did not include studies that produced results similar to those in another study. Thus, we did not repeatedly enter into our spreadsheet values for MFCs with similar power densities and other characteristics. We also excluded from our analysis studies that did not adequately report electrode projected surface areas, volumes or test conditions.

We found that power densities normalized to the cathode projected area reported MFC(WW) data showed no real trend with reactor volume (Figure 2A). In contrast, it appeared that MFCs that used defined substrates (most with acetate) produced more power at smaller than larger scales. In addition, the power densities obtained with defined substrates clearly exceeded those reported for wastewater, although there was substantial overlap of the two domains based on other factors that affect power production such as reactor design (electrode specific surface area) and operating conditions. The μMFCs were well separated from the other systems essentially by definition, as they were defined to have volumes less than 2 mL. The sMFCs spanned a wide range, and their sizes exceeded those of the other MFCs, and overall it appeared that power densities improved with size.

A slightly different picture emerged when the comparison of area power densities was made on the basis of volumetric power density (Figure 2B). Here we see that MFCs using defined substrates had clearly outstripped MFC(WW) results on the basis of volumetric density, and that the sMFCs had the lowest volumetric power densities. MFCs for wastewater treatment are somewhat in the middle, again with no clear trends in terms of volumetric power production. A comparison of these data on the basis

of energy density would also be useful, but energy efficiency and recovery have not been well reported in MFC studies. Reviews of available data concluded that small MFCs (<100 mL) that had high power densities do not have substantially different energy recoveries than larger systems.[47, 48]

The differences in these trends based on volume and volumetric power density suggested that there were some underlying factors for differences in power production. One factor is clearly the fuel: sMFCs must use very dilute sources of organic matter in the sediment, and they are likely limited in power production by production rates of soluble substrates for power production. MFCs using single substrates (true for most µMFC studies), and therefore the limitations based on fuel availability for current generation by the anode, can be minimized. There appeared to be no trend in power generation with volumetric power density for MFCs treating wastewater. However, as we will show below, there is evidence that the critical design factor in all these systems for volumetric power is the cathode specific surface area. To better understand how cathode specific surface area might impact performance, we specifically considered how cathode configurations impacted performance.

Electrode Spacing and Cathode Specific Surface Area

The development of air-cathode MFCs resulted in much improved power densities relative to earlier designs with aqueous cathodes,[46] but electrode spacing and the use of separators were shown to affect performance.[49, 50] It was discovered that power decreased when the anode was too close to the cathode, as a result of oxygen cross over through the cathode, resulting in anode bacteria inhibited in current generation by the presence of dissolved oxygen. When the anode (carbon paper) was moved from 4 cm to 2 cm away from the air cathode (no membrane or separator), power increased in accordance with expectations based on electrochemistry as there was a reduction in solution resistance.[49] However, moving the anode to within 1 cm of the cathode decreased power production even though solution resistance was further reduced. Placing a cloth separator against the cathode can

reduce oxygen transfer and increase power production.[9] In general, however, there has been a trade off in power densities and performance using separators, as the material can reduce oxygen crossover into the anolyte, but it can also impair ion movement to and from the cathode.[11] While some researchers have found it possible to greatly increase power with a very small electrode spacing using solutions with a high concentration of substrate (acetate),[50] others have found MFCs with closely-spaced flat electrodes to have unstable performance over time with more dilute solutions such as domestic wastewater.[51] It is likely that high substrate concentrations enable bacteria to quickly remove the oxygen and reduce the oxygen mass transfer into the anode water, or some exoelectrogenic strains can develop an ability to generate current in the presence of oxygen, but these factors have not been systematically investigated.

The use of thicker anodes, such as graphite fiber brush anodes and thick carbon felt, seems to result in more stable power over time than thin, flat anodes, even with low substrate concentrations.[8, 52, 53] Even when the edge of a brush anode is placed very close to the edge of the cathode, power production may not be affected,[51, 52] as it is with a thin, flat anode. It is possible that stable anoxic zones can develop within the brush and generate current while other bacteria on the exterior of the brush (or those in a separator, if present) consume dissolved oxygen. The tolerance of a brush anode to oxygen from the cathode relative to its size was examined by gradually trimming brush fibers farthest from the cathode over time.[52] Power was relatively unchanged until more than 65% of the brush most distant from the cathode was removed, leaving a brush that remained only 0.88 cm long, with an edge 0.4 cm from the cathode. The use of small brushes (0.8 cm) can reduce power compared to larger brushes (2.5 cm) when compared on the basis of the centerline of the brush relative to the cathode, as the distance of the edge of the brush to the cathode increases as brush size decreases. However, when smaller brushes were moved closer to the cathodes, so they had the same edge-to-cathode distance as the larger brushes, power was found to increase.[54] In both the brush trimming and brush size experiments, however, high

concentrations of acetate were used in a well buffered and high conductivity medium. Stable power production might not be obtained with very small brushes in solutions with lower substrate concentrations, such a domestic wastewater.

Based on consideration of the above results on electrode spacing, it seems logical that a critical factor in impacting power production when scaling up MFCs are electrode spacing, in terms of power production per area of electrode, and electrode packing density relative to volumetric power density as the size of the reactor is increased. If we examine the data for the different types of MFCs on the basis of cathode specific surface area, defined as the area of the cathode per volume of the reactor, we can see that in general the packing density shows a general decreasing trend with larger reactor size (Figure 2C). The trend is less clear for just MFCs using single substrates and wastewater. When those combined data are plotted versus size, the correlation is not significant, but the slope of the line is significantly less than unity ($p=0.01$) (Figure 2D). Thus, our survey of the literature suggests that larger reactors are not being designed with sufficient cathode area appropriate for the relative increase in reactor volume. Consistent with laboratory tests, if the distance between the electrodes increases (beyond the point that oxygen contamination is an issue), then the areal power density will decrease. If the volume is increased, and the areas of the anodes and cathodes are not sufficiently increased, volumetric performance will decrease due to insufficient electrode surface area to capture the substrate as electrical current.

Modular Designs. Commercialization of MFCs will require mass manufacturing of the materials in a modular format. The success of the design will depend on many factors, but we argue that the potential success of the reactor can be anticipated from laboratory tests with the same materials and wastewaters only if the cathode specific surface area is maintained with reactor size, for scales at which the conductivity of the materials (e.g. the current collectors) could limit performance. The cathode specific surface area must therefore be defined for each module design.

For both plate and frame and tubular MFC designs, the cathode specific surface area is easily assessed based on geometry used for these configurations. For a plate and frame arrangement, assuming electrodes with equal projected surface area, the cathode specific surface area is calculated from the "repeating width" of the anode-cathode pair. For example, if the anode chamber is AC = 4 cm wide and the cathode chamber is CC = 1 cm wide (including support materials), the repeating width is AC+CC = 5 cm, and a 1 m^3 MFC would have a cathode specific surface area of $(AC+CC)^{-1}$, or A_c = 20 m^2/m^3 after conversion of cm to m.[55] If the reactor contains a lot of space for the wastewater, for example 20 cm between the cathodes, then the cathode specific surface area is reduced to only A_c = 5 m^2/m^3, and volumetric power production will decrease. For tubular designs, with a cathode wrapped around the anode, the cathode specific surface area is simply calculated as $2r^{-1}$, where r is the tube radius. Thus, a tube 2.5 cm in radius has a specific surface area of A_c = 80 m^2/m^3 for just the tube, or A_c = 57 m^2/m^3 if we assume an additional 1 cm for air flow around the tube. If the tube radius is increased to 20 cm, with 10 cm around the tube allowed for air flow, the cathode specific surface area becomes only A_c = 7 m^2/m^3, and thus the larger reactor will not function as efficiently as the smaller reactor.

Few comparisons can be made of the same reactor configuration at different scales, but we can consider situations based on similar materials and electrode packing. For example, a small MFC (28 mL, 23 mL working volume) with a single carbon fiber brush anode and air cathode (A_c = 25 m^2/m^3) (Figure 1B) produced 130–240 mW/m^2 (7-13 W/m^3) using domestic wastewater.[51] In studies with a larger MFC (130 mL) containing 3 larger brush anodes and a single cathode, with a similar cathode specific surface area (A_c = 27 m^2/m^3) (Figure 1D), power densities of 120 mW/m^2 (3.2 W/m^3) and 282 mW/m^2 (7.6 W/m^3) were obtained using wastewater from the same plant. [56,57] Variations in wastewater strength and composition make it difficult to provide exact comparisons between studies. In general power densities might be expected to increase when using a higher strength wastewater, but electrode packing density is more important relative to volumetric power density for the studies included in this analysis. In tests

using different reactor configurations, but all with high strength wastewaters (brewery and piggery), 4.1 W/m^3 was produced using a 5.7 L reactor with 62 m^2/m^3,[58] compared to 1.1 W/m^3 using a 5-L MFC with 5 m^2/m^3,[59] and 1 W/m^3 in a 90-L reactor with 6 m^2/m^3 (Figure 1F).[60] Thus if cathode specific surface areas are not maintained for larger reactor sizes, volumetric power densities will be low due to the lack of sufficient cathode specific surface area.

Discussion

In the near term, the most promising MET applications will likely be the use of MFCs for wastewater treatment, as these systems lack expensive membranes and can accomplish removal of organic matter without aeration, and sMFCs as remote power sources. Successful scale up will require maintaining high cathode specific surface areas in order to maximize volumetric power production and organic matter utilization rates. With successful commercialization of MFCs, and in the future the availability of inexpensive membranes, it may be possible to further develop other METs. The development of low cost membranes could aid the development of other renewable energy technologies, such as those based on waste heat and salinity gradient energy.[29, 61-66]

MFCs can be used by inserting an MFC reactor filled with modules into a conventional treatment plant train to replace the biological treatment unit, such as an activated sludge reactor, thereby accomplishing COD removal, electricity production, and a reduction in sludge production relative to an aerobic process such as activated sludge. However, a secondary process would be needed to fully remove COD to levels suitable for discharge, as current generation is minimal once the COD is less than ~100-150 mg/L.[67] The use of another developing technology, an anaerobic fluidized bed membrane bioreactor (AFMBR), was shown to successfully reduce COD to <20 mg/L and total suspended solids to <1 mg/L, following treatment of a domestic wastewater using MFCs.[68] Alternatively, other processes could be used including activated sludge and a secondary clarifier as a polishing step, with greatly

reduced aeration requirements compared to treatment of the wastewater without MFC pretreatment. Nutrient removal, however, still needs to be addressed as there is minimal removal of ammonia or phosphorus in the systems.

The main obstacle at this point in time for commercial production of MFCs is manufacturing capacity to produce the reactor cathodes. While brush anode manufacturing could easily be implemented, large scale cathode production does not yet exist. Reactors will require hundreds to thousands of square meters of cathode, and currently there is no commercial provider of cathodes larger than a few hundred square centimeters. Thus, commercialization of MFCs represents both a business opportunity and a production challenge.

■ Associated Content

Supporting Information. A copy of the spreadsheet used for calculations is included, in the form of 4 tables and associated references. This material is available free of charge via the Internet at http://pubs.acs.org.

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569 **Table 1: Examples of different microbial electrochemical technologies (METs).**

MxC	Full name	Comments	References
MDC	Microbial desalination cells	Can use reverse electrodialysis (RED) stacks (MEDC, microbial electrodialysis cell); forward osmosis (MOFC, microbial osmotic fuel cell); pressure retarded osmosis	25-27, 69-74
MEC	Microbial electrolysis cell	Typically used for hydrogen gas production from the cathode, but also used for metals reduction	15-17, 75-77
MEDCC	Microbial electrolysis desalination and chemical production cell (MEDCC)	Includes a bipolar membrane, so energy must be input for chemical production	32, 78
MES	Microbial electrosynthesis cell	An MEC that is designed to produce soluble organics such as acetate	35, 79-81
MFC	Microbial fuel cell	Electrical power production	2, 4, 8, 46, 55, 82-88
MxC-MBR	MFC with a cathode membrane	The cathode serves a dual function reduction and filtration of the water using either MFCs or MECs	89-91
MMC	Microbial methanogenesis cell	Methane production from the cathode	38, 92-97
MREEC	Microbial reverse-electrodialysis electrolysis and chemical-production cell (MREEC)	An MEDCC that includes a RED stack, and is used for production of acid and bases; can be used for carbon capture; can produce hydrogen gas; can also be used for desalination	33, 34, 98
MREC	Microbial reverse electrodialysis electrolysis cell	RED stack inserted into an MEC	30, 99, 100
MRFC	Microbial reverse electrodialysis fuel cell	RED stack inserted into an MFC	29, 31, 101
MSC	Microbial struvite production cell	Designed to precipitate struvite on the cathode	102-106
sMFC	Sediment microbial fuel cell	Also known as a benthic MFC	23, 24, 107, 108

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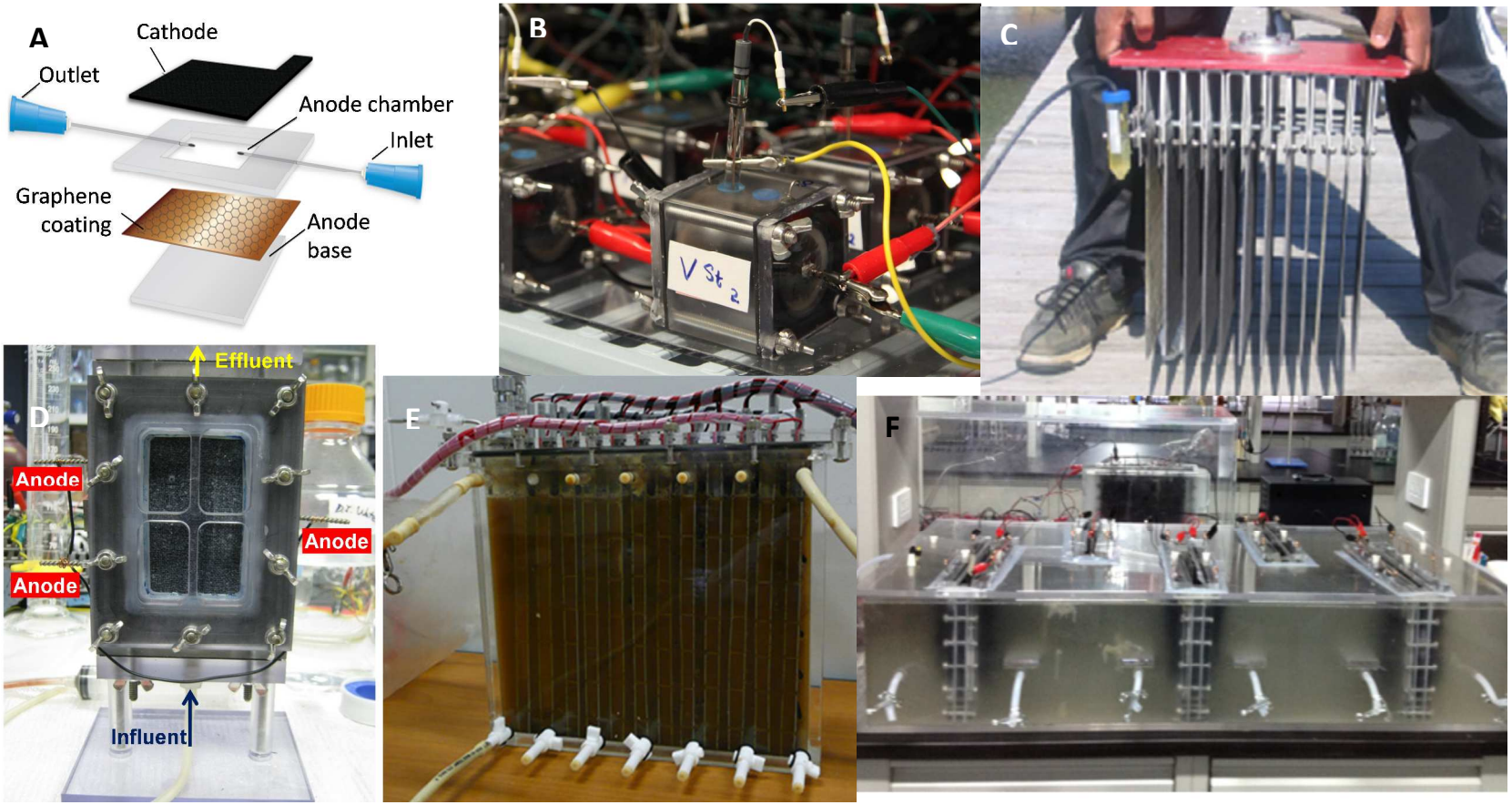


Figure 1. Photographs or schematics of different types of MFCs: (A) µMFC (25 µL anode volume);[109] (B) MFC with air cathode and reference electrode used in many different laboratory studies, shown here with a brush anode;[8, 18] (C) sMFC anode array;[24] (D) a 3-brush electrode MFC (130 mL) designed for continuous flow;[110] (E) larger-scale MFC containing 12 cassettes;[111] (F) baffled MFC with multiple cassettes.[60] (A, C, D, E and F, reprinted with permission; B, credit B.E. Logan.)

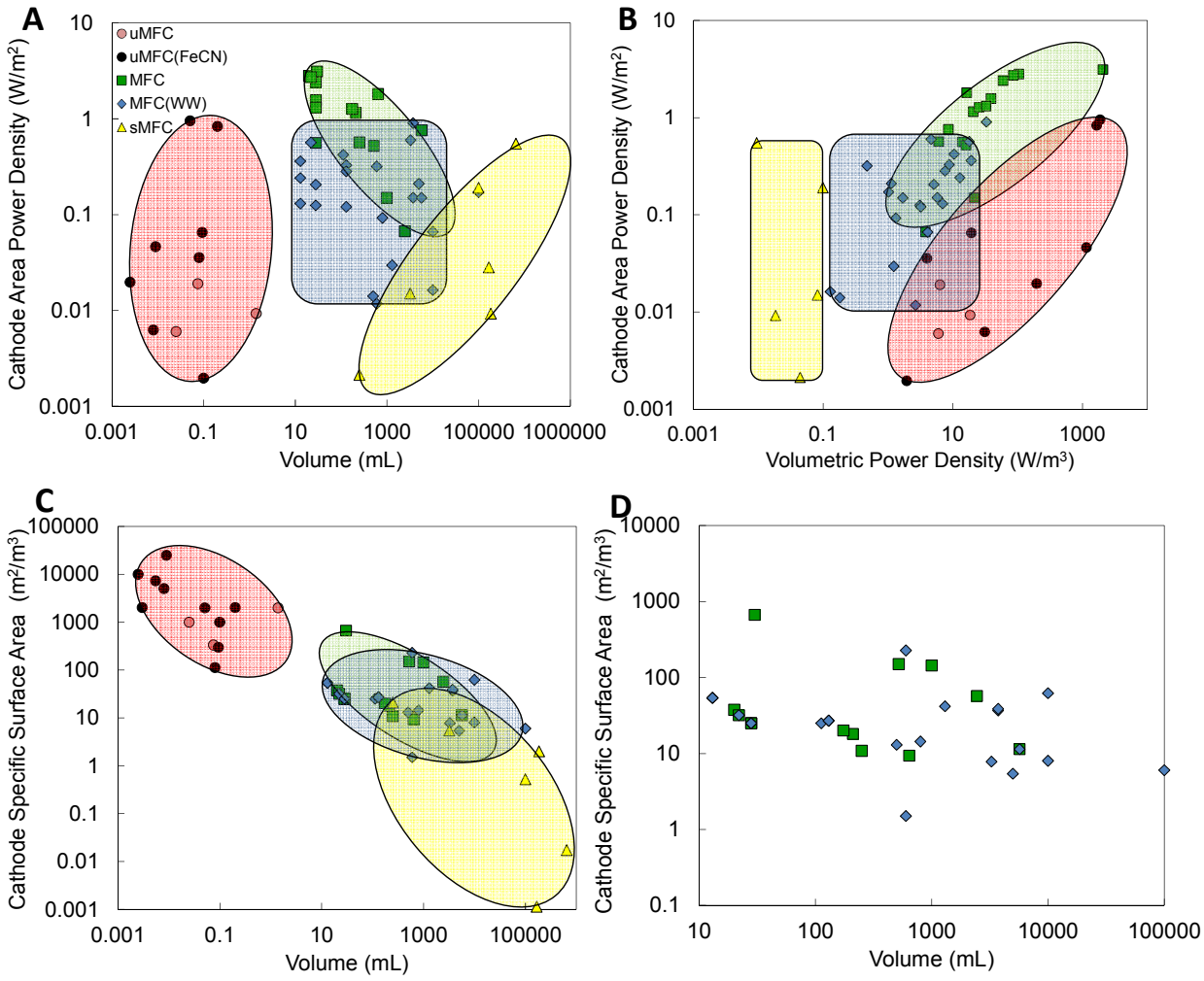
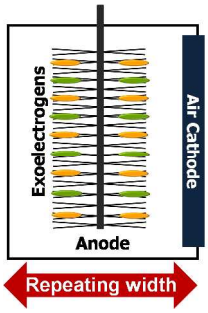


Figure 2. Power production per cathode area based on (A) the volume of the MFC and (B) the volumetric power density. (C) cathode specific surface area as a function of MFC volume for all data, and (D) for only MFCs and MFCs using wastewater. No data were included from the literature for MFCs that produced <1 mW/m^2. All data are for MFCs with air cathodes, except the µMFCs which include ferricyanide (FeCN) catholytes as indicated.

TOC Art



Repeating width	Cathode specific area
4 cm	25 m^2/m^3
8 cm	12.5 m^2/m^3
10 cm	10 m^2/m^3