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Research review paper

A comprehensive review of microbial electrochemical systems as a platform technology

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ABSTRACT

Microbial electrochemical systems (MESs) use microorganisms to covert the chemical energy stored in biodegradable materials to direct electric current and chemicals. Compared to traditional treatment-focused, energy-intensive environmental technologies, this emerging technology offers a new and transformative solution for integrated waste treatment and energy and resource recovery, because it offers a flexible platform for both oxidation and reduction reaction oriented processes. All MESs share one common principle in the anode chamber, in which biodegradable substrates, such as waste materials, are oxidized and generate electrical current. In contrast, a great variety of applications have been developed by utilizing this *in situ* current, such as direct power generation (microbial fuel cells, MFCs), chemical production (microbial electrolysis cells, MECs; microbial electrosynthesis, MES), or water desalination (microbial desalination cells, MDCs). Different from previous reviews that either focus on one function or a specific application aspect, this article provides a comprehensive and quantitative review of all the different functions or system constructions with different acronyms developed so far from the MES platform and summarizes nearly 50 corresponding systems to date. It also provides discussions on the future development of this promising yet early-stage technology.

Contents

1. Introduction

Microbial electrochemical systems (MESs) are a rapidly growing environmental technology at the nexus of water and energy (Harnisch and Schröder, 2010; Logan and Rabaey, 2012; Rozendal et al., 2008; Torres et al., 2010). While this platform technology has only been intensively studied and developed in the past decade, it opens up a new interdisciplinary field for research and development which integrates microbiology, electrochemistry, materials science, engineering, and many related areas together. MESs not only provide a unique environment to understand the largely unexplored microbial electrochemistry, they also offer a flexible platform for many different engineering functions to be developed. While many existing environmental technologies have only one or two functions, the MES platform is so flexible that

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dozens of functions have been discovered. Almost all MESs share one common principle in the anode, in which biodegradable substrates, such as waste materials, are oxidized by microorganisms and generate electrical current. The current can be captured directly for electricity generation (microbial fuel cells, MFCs) (Fornero et al., 2010; Liu and Logan, 2004; Ren et al., 2007), or used to produce H_2 and other value-added chemicals (microbial electrolysis cells, MECs) (Cheng et al., 2009; Liu et al., 2010; Logan et al., 2008). The electrons can also be used in the cathode chamber to synthesize organic compounds (microbial electrosynthesis, MES) or remediate contaminants (microbial remediation cells, MRCs) (Aulenta et al., 2008; Butler et al., 2010; Gregory and Lovley, 2009; Lovley and Nevin, 2011; Rabaey and Rozendal, 2010). The potential across the electrodes can also drive desalination (microbial desalination cells, MDCs) (Cao et al., 2009; Jacobson et al., 2011; Luo et al., 2011; Luo et al., 2012c; Mehanna et al., 2010). The production of current associated with microbial catabolism was first reported a century ago by Potter (1911), but research interests in this concept have only blossomed in the past decade, resulting in an exponential growth in the number of journal articles (Fig. 1). There are several excellent reviews that provided information on the history and development of MESs (Borole et al., 2011; Schröder, 2011, 2012; Sleutels et al., 2012) and the substrates, materials, and microbial communities in different systems (Hamelers et al., 2010; Logan, 2009; Lovley, 2006; Pant et al., 2010; Wei et al., 2011), but there has been no comprehensive or quantitative review that directly addresses one fundamental factor: where all the known functions were originated from and all future functions will be based upon. As shown in Table 1, this article aims to provide the first complete review with the goal to summarize all the functions with different acronyms that have been developed using this platform to date, and shed light on future system development for energy and environmental science and engineering. Different groups have also used bioelectrochemical systems (BESs) or MXCs for this technology platform, but because BESs were also used in other studies to represent cell free enzyme based systems, while system acronyms have far beyond the "X" of MXCs, this review uses MESs to represent the overall technology platform (Harnisch and Schröder, 2010; Logan and Rabaey, 2012; Rozendal et al., 2008; Torres et al., 2010).

2. The shared principle in the anode chamber

Compared to traditional chemical fuel cells, the MES platform uses low-cost and self-sustaining microorganisms to oxidize organic and inorganic electron donors, mainly waste materials, and transfer electrons to the anode electrode. The electrons can be captured directly through an external circuit for electricity generation or used for chemical production. The microbial oxidation reaction in the anode chamber is a shared principle for almost all MES reactors, as shown in Table 1. However, how to use these electrons on the cathode side shows the beauty of this platform technology, because any reduction-based reaction can be realized in the cathode chamber, which creates numerous possibilities. Based on the different functions, the MES platform has been specified into many different names that some researchers name them MXCs, where X stands for different applications (Harnisch and Schröder, 2010; Torres et al., 2010). Table 1 summarizes all the reactor acronyms to date and demonstrates the shared principle on the anode and the versatile functions on the cathode.

Ideal anodic reactions in MESs generally include dynamic and effective microbial activity and community, higher substrate conversion rate and electron transfer efficiency, and lower material and system costs. MESs employ a unique group of microbes called electrochemically active bacteria (EAB), exoelectrogen, electricigen, or anode respiring bacteria (ARB) to convert the chemical energy stored in organic or inorganic substrates to electrical energy during their anaerobic respiration (Logan, 2009; Lovley, 2006; Park et al., 2001; Torres et al., 2009). Such microorganisms are able to transfer electrons out of cell membranes to the electrode either directly through membrane-bound protein structures, such as pili, *c-type* cytochrome and filaments, or using mobile electron shuttles, such as mediators for indirect electron transfer. For example, recent studies showed that *Geobacter sulfurreducens* requires conductive pili as nanowires for cell-to-cell electron conduction and *c-type* cytochrome *OmcZ* to promote electron transfer onto the electrode (Lovley, 2011; Summers et al., 2010). In contrast, *Shewanella* species were reported to make both direct electrode contact through conductive filaments and indirect electron transfer *via* mediators, such as riboflavin or flavin adenine mononucleotide (FMN) (Canstein et al., 2008; Gorby et al., 2006; Marsili et al., 2008). Many other bacteria can produce and use soluble redox mediators or electron shuttles, which transport the electrons from the cell to the electrode. For example, *Pseudomonas* species can produce phenazines as extracellular electron shuttles, and other bacteria can use externally provided mediators, such as neutral red, anthraquinone-2,6-disulfonate (AQDS), thionine, methyl viologen, methyl blue, and some humics (Aulenta et al., 2008; Milliken and May, 2007; Park and Zeikus, 2000; Rabaey et al., 2005a; Scott and Murano, 2007; Thurston et al., 1985).

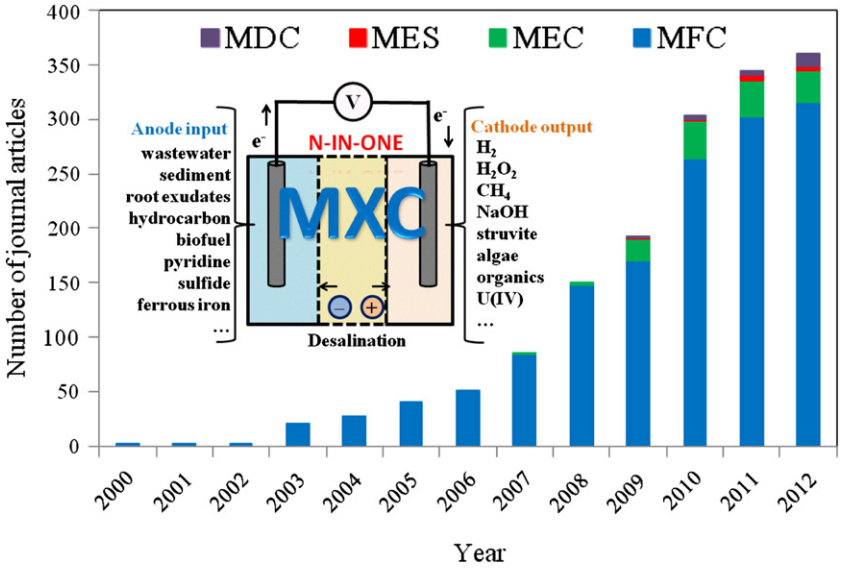

Fig. 1. Number of published journal articles on MESs containing the phrases "microbial fuel cell", "microbial electrolysis cell", "microbial electrosynthesis" or "microbial desalination cell". Source: Scopus on 7/1/2013; document type: Journal; Language: English; duplicates were removed from searching results.

Table 1
Summary of all types of MESs with different acronyms.

#	Types of MESs	Electron donor for anode oxidization	Electron acceptor for cathode reduction	Main products	Ref.
	MFC-based systems for electricity generation				
	Microbial fuel cells (MFCs) — in general	*Any biodegradable material*	*Oxygen, potassium ferricyanide, or other oxidants*	*Electricity*	Kim et al. (1999), Tanaka et al. (1983)
1	Tubular microbial fuel cell (tubular MFC)	Acetate, glucose, domestic wastewater, hospital wastewater, digester effluent from a potato processing plant	Potassium ferricyanide	Electricity	Rabaey et al. (2005b)
2	Upflow microbial fuel cell (UMFC)	Sucrose	Potassium ferricyanide, oxygen	Electricity	He et al. (2005), He et al. (2006)
3	Baffled air-cathode microbial fuel cell (BAFMFC)	Glucose, liquid from corn stover steam explosion process	Oxygen	Electricity	Feng et al. (2010)
4	Up-flow anaerobic sludge blanket reactor–microbial fuel cell (UASB–MFC)	Glucose, sulfide	Oxygen, sulfer	Electricity	Zhang et al. (2012)
5	Slalom-flow cassette-electrode microbial fuel cell (sCE-MFC)	Starch, yeast extract, peptone, plant oil, detergent	Oxygen	Electricity	Miyahara et al. (2013)
6	Plug flow microbial fuel cell (PF-MFC)	Wastewater, sodium acetate	Oxygen	Electricity	Karra et al. (2013)
7	Complete mixing microbial fuel cell (CM-MFC)	Wastewater, sodium acetate	Oxygen	Electricity	Karra et al. (2013)
8	Stacked microbial fuel cell (stacked MFC)	Sodium acetate	Potassium ferricyanide	Electricity	Aelterman et al. (2006)
9	Submersible microbial fuel cell (SBMFC)	Domestic wastewater	Oxygen	Electricity	Zhang and Angelidaki (2012c)
10	Benthic microbial fuel cell (BMFC)	Sediment	Oxygen	Electricity	Gong et al. (2011), Nielsen et al. (2007), Tender et al. (2008)
11	Sediment microbial fuel cell (AKA benthic unattended generator or BUG)	Acetate and other fermentation products in the sediment	Oxygen	Electricity	Lovley (2006)
12	Self-stacked submersible microbial fuel cell (SSMFC)	Sediment, acetate	Oxygen	Electricity	Zhang and Angelidaki (2012b)
13	Microbial remediation cell (MRC)	Diesel, ethanol, 1,2-dichloroethane, pyridine, phenol	Chlorinated solvents, perchlorate, chromium, and uranium	Reduced/non-toxic chemicals	Aulenta et al. (2008), Butler et al. (2010), Gregory and Lovley (2009), Kim et al. (2007), Luo et al. (2009), Morris et al. (2009), Pham et al. (2009), Zhang et al. (2009), T. Zhang et al. (2010)
14	Photo-microbial fuel cell (p-MFC)	Water	Potassium ferricyanide	Electricity	Thorne et al. (2011)
15	Microbial photoelectrochemical solar cell	Marine sediment	Oxygen	Electricity, glucose, oxygen	Malik et al. (2009)
16	Solar-powered microbial fuel cell	Succinate, propionate	Oxygen	Electricity, hydrogen	Cho et al. (2008), Strik et al. (2010)
17	Photobioelectrochemical fuel cell	Organic acids, alcohols	Potassium ferricyanide	Electricity, hydrogen	Rosenbaum et al. (2005)
18	Photosynthetic microbial fuel cells (PMFCs)	Water	Oxygen	Electricity	Zou et al. (2009)
19	Photosynthetic electrochemical cell	Water, glucose	Potassium ferricyanide	Electricity	Yagishita et al. (1997)
20	Solar-driven microbial photoelectrochemical cell (solar MPC)	Trypticase soy broth (TSB)	Proton	Electricity	Qian et al. (2010)
21	Plant microbial fuel cell (PMFC)	Plant-derived organics (root exudates)	Oxygen, potassium ferricyanide	Electricity	Deng et al. (2012)
22	Phototrophic microbial fuel cells (phototrophic MFCs)	Sediment	Oxygen	Electricity	He et al. (2009)
23	Photosynthetic algal microbial fuel cell (PAMFC)	Algae	Potassium ferricyanide	Electricity	Strik et al. (2008b)
24	Microbial electrochemical snorkel (MES, AKA short-circuited microbial fuel cell)	Wastewater	Oxygen	Treated wastewater, no electricity	Erable et al. (2011)
25	Acid-mine drainage fuel cell (AMD-FC)	Ferrous ion	Oxygen	Electricity, removing iron	Cheng et al. (2007)

26	Integrated photobioelectrochemical system (IPB)	Wastewater	Oxygen	Electricity, algal biomass	Xiao et al. (2012)
27	Osmotic microbial fuel cell (OsMFC)	Sodium acetate	Oxygen	Diluted draw solution, electricity	Zhang et al. (2011)
28	Microbial reverse electrodialysis cell (MRC)	Sodium acetate	Oxygen	Electricity	Cusick et al. (2012), Kim and Logan (2011b)
29	Microbial reverse-electrodialysis chemical-production cell (MRCC)	Sodium acetate	Oxygen	Electricity, acid, alkali	Zhu et al. (2013)
	MEC-based systems for chemical production				
	Microbial electrolysis cells (MECs) — in general	*Any biodegradable material*	*Proton,*	*Hydrogen, hydrogen peroxide, methane, sodium hydroxide*	Cheng et al. (2009), Liu et al. (2005b), Rabaey et al. (2010), Rozendal et al. (2009)
30	Bioelectro-chemically assisted microbial reactor (BEAMR)	Wastewater	Proton	Hydrogen	Ditzig et al. (2007)
31	Solar-powered microbial electrolysis fuel (solar MEC)	Acetate	Proton	Hydrogen	Chae et al. (2009)
32	Microbial reverse-electrodialysis electrolysis cell (MREC)	Acetate	Proton	Hydrogen	Kim and Logan (2011a)
33	Microbial electrolysis struvite-precipitation cell (MESC)	Sodium acetate	Proton	Hydrogen, struvite	Cusick and Logan (2012)
34	Submersible microbial electrolysis cell (SMEC)	Acetate	Proton	Hydrogen	Zhang and Angelidaki (2012a)
	MES-based systems for chemical production				
	Microbial electrosynthesis (MES) — in general	*Organic, hydrogen sulfide, water*	*Acetic acid or other organics, carbon dioxide*	*Ethanol, acetate, 2-oxobutyrate, formate*	Gong et al. (2013), Nevin et al. (2010), Nevin et al. (2011), Rabaey and Rozendal (2010), Rabaey et al. (2011), Steinbusch et al. (2010)
35	Microbial carbon capture cell (MCC)	Glucose	Carbon dioxide	Algal biomass, electricity	Wang et al. (2010)
	MDC-based systems for water desalination and beneficial reuse				
	Microbial desalination cells (MDCs) — in general	*Any biodegradable material*	*Oxygen, potassium ferricyanide, organics, or other oxidants*	*Desalinated water*	Cao et al. (2009)
36	Microbial saline-wastewater electrolysis cell (MSC)	Sodium acetate	Hydrogen	Treated saline wastewater, electricity	Kim and Logan (2013b)
37	Osmotic MDC (OsMDC, MODC)	Sodium acetate, xylose, wastewater	Oxygen, potassium ferricyanide, proton	Desalinated water, electricity	Kim and Logan (2013a), Zhang and He (2012)
38	Microbial desalination cell with capacitive adsorption capability (cMDC)	Sodium acetate	Potassium ferricyanide	Desalinated water	Forrestal et al. (2012a)
39	Microbial desalination cell packed with ion-exchange resin (R-MDC)	Sodium acetate	Oxygen	Desalinated water, electricity	Morel et al. (2012)
40	Microbial electrolysis desalination cell (MEDC)	Sodium acetate	Proton	Hydrogen, desalinated water	Luo et al. (2011)
41	Microbial electrolysis desalination and chemical-production cell (MEDCC)	Sodium acetate	Oxygen	Desalinated water, sodium hydroxide, hydrochloric acid	Chen et al. (2012)
42	Microbial capacitive desalination cell (MCDC)	Sodium acetate	Oxygen	Desalinated water	Forrestal et al. (2012b)
43	Capacitive deionization coupled with microbial fuel cell (CDI-MFC)	Sodium acetate	Potassium ferricyanide	Desalinated water	Yuan et al. (2012)
44	Upflow microbial desalination cell (UMDC)	Sodium acetate	Oxygen	Desalinated water, electricity	Jacobson et al. (2011)
45	Stacked microbial desalination cell (SMDC)	Sodium acetate	Oxygen	Desalinated water, electricity	Chen et al. (2011)
46	Recirculation microbial desalination cell (rMDC)	Xylose	Oxygen	Desalinated water, electricity	Qu et al. (2012)
47	Submerged microbial desalination–denitrification cell (SMDDC)	Sodium acetate	Nitrate	Electricity, nitrogen	Zhang and Angelidaki (2013)

Using microorganisms as biocatalysts, MESs can theoretically convert any biodegradable substrate into energy and chemicals. Besides simple sugars and derivatives used in most lab scale studies, many complex waste materials have also been utilized, such as different wastewaters from municipal and industrial sources, biomass wastes, and inorganic substrates such as ammonia, sulfide, and acid mine drainage (Cheng et al., 2007; Kuntke et al., 2012; Pant et al., 2010; Rabaey et al., 2006; Velasquez-Orta et al., 2009). The utilization of complex waste materials generally requires the cooperation of polymer-degrading bacteria and electrochemically active bacteria, with the first group breaking down the complex polymers, such as cellulose or protein, into simple organic matter, such as volatile fatty acids, alcohol, or amino acids, and then the second group oxidizes these simple organic products with the anode serving as the electron acceptor (Freguia et al., 2008; Parameswaran et al., 2009; Ren et al., 2007, 2008). In terms of waste treatment in the anode chamber, MESs represent a new generation of technology, because they carry the potential to transform traditional energy-intensive, treatment-focused processes into integrated systems that recover energy, nutrient, water, and other value-added products.

3. The diverse application possibilities in the cathode chamber

As shown in Table 1, there have been 47 systems presented so far with different functions or system constructions that were developed using the MES platform, and people used different acronyms to represent the various functions and systems. Though no specific rules have been established to name the different reactors, this article attempts for the first time to summarize and categorize all the systems that have been reported so far and provides some insights on future technology development.

In many cases, the different MESs can be summarized as MXCs, in which the X simply presents the main function and benefit of a specific cell. For example, a microbial fuel cell (MFC) is the very original type of MES, whose main function is direct electricity generation (Fig. 2A) (Logan et al., 2006). When an external power source is added in an MFC reactor to reduce cathode potential, the system becomes a microbial electrolysis cell (MEC), where hydrogen gas and other products can be generated (Fig. 2B) (Cheng et al., 2009; Ditzig et al., 2007; Logan et al., 2008; Rabaey et al., 2010; Rozendal et al., 2009). If the main function of the system is to use the cathode to reduce oxidized contaminants, such as uranium, perchlorate or chlorinated solvents, the cell can be named a microbial remediation cell (MRC) (Aulenta et al., 2008; Butler et al., 2010; Gregory and Lovley, 2009), and if the main goal of the system is to synthesize value-added chemicals through microbially catalyzed cathodic reductions, the system can be named microbial electrosynthesis (MES), which can be a little confusing with the general microbial electrochemical system acronym (Fig. 2C) (Lovley and Nevin, 2011; Rabaey and Rozendal, 2010). Another system called a microbial desalination cell (MDC) (Fig. 2D) (Cao et al., 2009; Kim and Logan, 2011a) includes additional chambers between the anode and cathode and uses the internal potential to drive water desalination.

There are also many different sub-systems within each main category. Take MFCs as an example, based on different substrates used in MFC reactors, there are wastewater MFCs, sediment or benthic MFCs, *etc.* (Liu et al., 2004; Reimers et al., 2001). By utilizing different photosynthetic organisms for solar energy capturing, people have developed plant-MFCs, phototrophic-MFCs, and algae-MFCs (Deng et al., 2012; He et al., 2009; Strik et al., 2011). By integrating other technologies with the MES platform, new systems with superior performance can be developed. For instance, by incorporating reverse-electrodialysis (RED) with an MEC, the microbial reverse-electrodialysis electrolysis cell (MREC) can produce H_2 without any external power supply (Kim and Logan, 2011a). By integrating capacitive deionization (CDI) with an MDC, the microbial capacitive desalination cell (MCDC) could improve desalination efficiency by 7–25 times compared to traditional CDI processes (Forrestal et al., 2012b). Other names may come from the combination of multiple functions in one system, and they are generally straightforward, such as microbial electrolysis desalination cell (MEDC) (Luo et al., 2011), microbial electrolysis desalination and chemical-production cell (MEDCC) (Chen et al., 2012), osmotic microbial fuel cell (OsMFC) (Zhang et al., 2011), and microbial electrolysis struvite-precipitation cell (MESC) (Cusick and Logan, 2012), *etc.*

4. MFC-based systems for electricity generation

4.1. Wastewater microbial fuel cells (wastewater MFCs)

MFCs refer to the reactor systems that focus on electricity production from biodegradable materials. Table 1 provides a complete list of different MFCs to date by our best count. Early lab scale MFC studies mostly used acetate, glucose, or other simple substrates to characterize the performance of materials, reactor configurations, or microbial activities (Liu et al., 2005a; Rabaey et al., 2003). The first MFC study that used real wastewater as the substrate was reported in 2004 (Liu et al., 2004), and since then hundreds of studies have been published to report power production from different substrates, including both organic and inorganic waste streams using various electrode or separator materials and reactor configurations. Several review articles have provided comprehensive information on the substrates (Pant et al., 2010), electrode materials (Wei et al., 2011), separator materials (X. Zhang et al., 2010), and reactor configurations (Logan et al., 2006) used in different MFC studies.

Classic MFC designs include the single-chamber air-cathode MFCs (SCMFCs) developed by Liu and Logan, which for the first time eliminated the membrane and therefore significantly reduced system internal resistance and cost (Fig. 3A) (Liu and Logan, 2004; Liu et al., 2005a). Tubular designs (Tubular MFCs) with different flow patterns simplified construction processes and optimized systems with increased electrode surface area and reduced system resistance (He et al., 2005; Rabaey et al., 2005b). A baffled air-cathode microbial fuel cell (BAFMFC) was designed to increase organic loading rate (Feng et al., 2010), and stacked MFCs were able to increase direct voltage or current output while also enhance substrate oxidation (Aelterman et al., 2006). Other MFC systems used in wastewater applications include submersible MFCs (SBMFCs) (Zhang and Angelidaki, 2012c), which may convert the information of substrate concentration, toxicity, or dissolved oxygen concentration into electronic signals as MFC sensors.

The main advantages of using MFCs in wastewater treatment come from the savings of aeration energy and sludge disposal (Oh et al., 2010; Ren, 2013; Xiao et al., 2012). For traditional activated sludge systems, aeration can amount to 45–75% of plant energy costs, so the conversion of aeration tank to MFC units is very beneficial because it not only eliminates aeration energy consumption, studies also showed that the MFC can produce 10–20% more energy that can be used for other processes (Huggins et al., 2013; Pant et al., 2010). The reported maximum power density from lab scale air-cathode MFCs has reached 2.87 kW/m^3, making it promising for commercialization development (Fan et al., 2012), even though the system scale up remains a major challenge. Another main benefit of MFC systems is the low biomass production. The MFC is a biofilm based system, and the cell yield of electrochemically active bacteria (0.07–0.16 gVSS/gCOD) is much less than the activated sludge (0.35–0.45 gVSS/gCOD), so it can reduce sludge production by 50–70% (Fan et al., 2012; Huggins et al.), which in turn may reduce 20–30% of the plant operation cost. Other benefits may include nutrient removal and the production of value-added products, such as caustic solutions for disinfection, or H_2 and biogas for energy, which will be discussed more extensively in the following sections.

4.2. Benthic microbial fuel cells (benthic MFCs)

Benthic MFCs (BMFCs), also known as sediment MFCs (SMFCs) are systems that utilize the naturally occurring potential difference

between the anoxic sediment and oxic seawater to produce electricity (Lovley, 2006). Microorganisms oxidize the substrates in the sediment and transfer electrons to the anode either embedded in or rested on top of the sediment, and then the electrons are transferred to the cathode suspended in the overlying seawater, where dissolved oxygen is reduced to water (Fig. 3B) (Donovan et al., 2011). The abundant availability of substrates in the sediment makes BMFC a very promising power source for autonomous marine sensors and underwater vehicles, because they provide consistent and maintenance-free power supply for a long period of time without using batteries. This is a huge advantage compared to batteries, because batteries are limited in service life for about 2–4 years, and the replacement can be very expensive, especially in deep water. It was estimated that the initial organic matter in 1 L marine sediment could generate an average current of 0.3 mA continuously for 22 years (Malik et al., 2009). While the concept of BMFC was only introduced in 2001 by Reimers et al. (2001), it is a type of MES device that is closest toward commercialization. The first demonstration of BMFC as a viable power source was reported by Tender et al. in 2008, where an 18 mW meteorological buoy was powered for nearly 7 months (Tender et al., 2008). Another study showed a chambered BMFC was used to power an acoustic modem interfaced with an oceanographic sensor for over 50 days with an average power density of 44 mW/m^2 (Gong et al., 2011). So far, the longest field demonstration of BMFCs has been reported continually operated for at least 2 years without depletion in power (Tender et al., 2008). Different configurations of BMFCs have been developed and deployed. Initial designs include simple graphite plates buried in the sediment with suspended cathode in water, but such designs are fragile and the power output is very low (Tender et al., 2002). Nielsen et al. developed a chamber-based BMFC that incorporates a suspended and semi-enclosed anode, which reduced system footprint and increased power output to a range of 380 mW/m^2 (3.8 W/m^3) (Nielsen et al., 2007). A self-stacked submersible microbial fuel cell (SSMFC) showed an open circuit voltage (OCV) of 1.12 V and a maximum power density of 294 mW/m^2 (Zhang and Angelidaki, 2012b).

4.3. Microbial remediation cells (MRCs)

Another emerging application of the MES platform is using the electrodes to serve as inexhaustible electron acceptors (anode) or donors (cathode) for underground contaminant remediation (Huang et al., 2011; Morris and Jin, 2008; Yuan et al., 2010). Like sediment MFCs, MRCs used in groundwater or soil remediation can be a single or an array of electrodes without using enclosed containers. Such bioelectrochemically enhanced approach can stimulate microbes to concurrently degrade underground pollutants and produce additional electricity. Such process is considered sustainable because it eliminates the injection of expensive chemicals and reduces operational energy cost as compared to other technologies.

Microbial electrochemical remediation of petroleum contaminants was demonstrated by using electrode as a channel linking underground hydrocarbon oxidation and upground O_2 reduction. One study showed that the active MRC increased the degradation of diesel range organics (DRO) by 164% as compared to open circuit potential (Morris et al., 2009), and another study using a U-tube MFC showed crude oil degradation can be increased by 120% at the location near the electrode (X. Wang et al., 2012). The dramatic increase in contaminant oxidation rate is hypothesized due to the faster electron transfer by more conductive electrode as compared with electron shuttles. It is also possible that the competition between microbes to access and deliver electrons to the electrodes triggered higher metabolic activities, and the immediate removal of electrons *via* the electrode eliminated the potential feedback inhabitation. Similar remediation studies on other reduced pollutants including diesel, ethanol, 1,2-dichloroethane, pyridine, and other contaminants were also reported (Luo et al., 2009; Pham et al., 2009; Zhang et al., 2009). Conversely, oxidized contaminants, such as

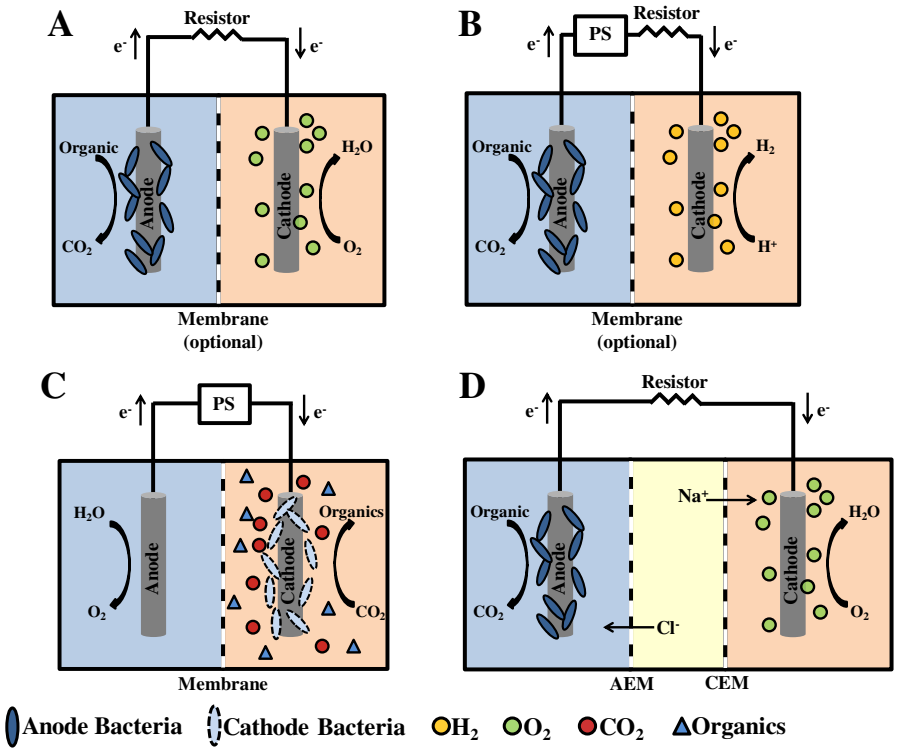

Fig. 2. Basic principles in four typical MESs (left chamber: anode; right chamber: cathode). (A) Electricity generation in air-cathode microbial fuel cells (MFCs); (B) hydrogen generation with external power supply in microbial electrolysis cells (MECs); (C) chemical production by microbial electrosynthesis (MES); (D) middle chamber desalination in microbial desalination cells (MDCs).

chlorinated solvents, perchlorate, chromium, and uranium, can be reduced using the electrode as the electron donor (Aulenta et al., 2008; Butler et al., 2010; Gregory and Lovley, 2009; Wang et al., 2008). For instance, studies showed that a negatively polarized electrode could act as an electron donor for the dechlorination of trichloroethene (TCE) to ethene by a mixed culture of microorganisms (Aulenta et al., 2008). The similar approach was also used in both lab and field tests for U(VI) reduction, where the horizontally distributed anodes and cathodes enabled direct correlation between acetate injection and uranium reduction, and current production may be an effective proxy for monitoring *in situ* microbial activity and remediation performance (Fig. 3C) (Williams et al., 2010).

4.4. Microbial solar cells (MSCs)

Microbial solar cells are collective names for different MESs that integrate the photosynthetic reaction with microbial electricity (or chemical) production using synergistic relationships between photosynthetic organisms and EAB (Strik et al., 2011). While EAB are generally the same bacterial groups in other MESs, the organisms that are responsible for converting solar energy to organic matter may include higher plants, photoautotrophic bacteria, and algae. A very wide variety of names and systems related to MSCs have appeared in literature, such as photo-microbial fuel cell (p-MFC) (Thorne et al., 2011), microbial photoelectrochemical solar cell (Malik et al., 2009), solar-powered

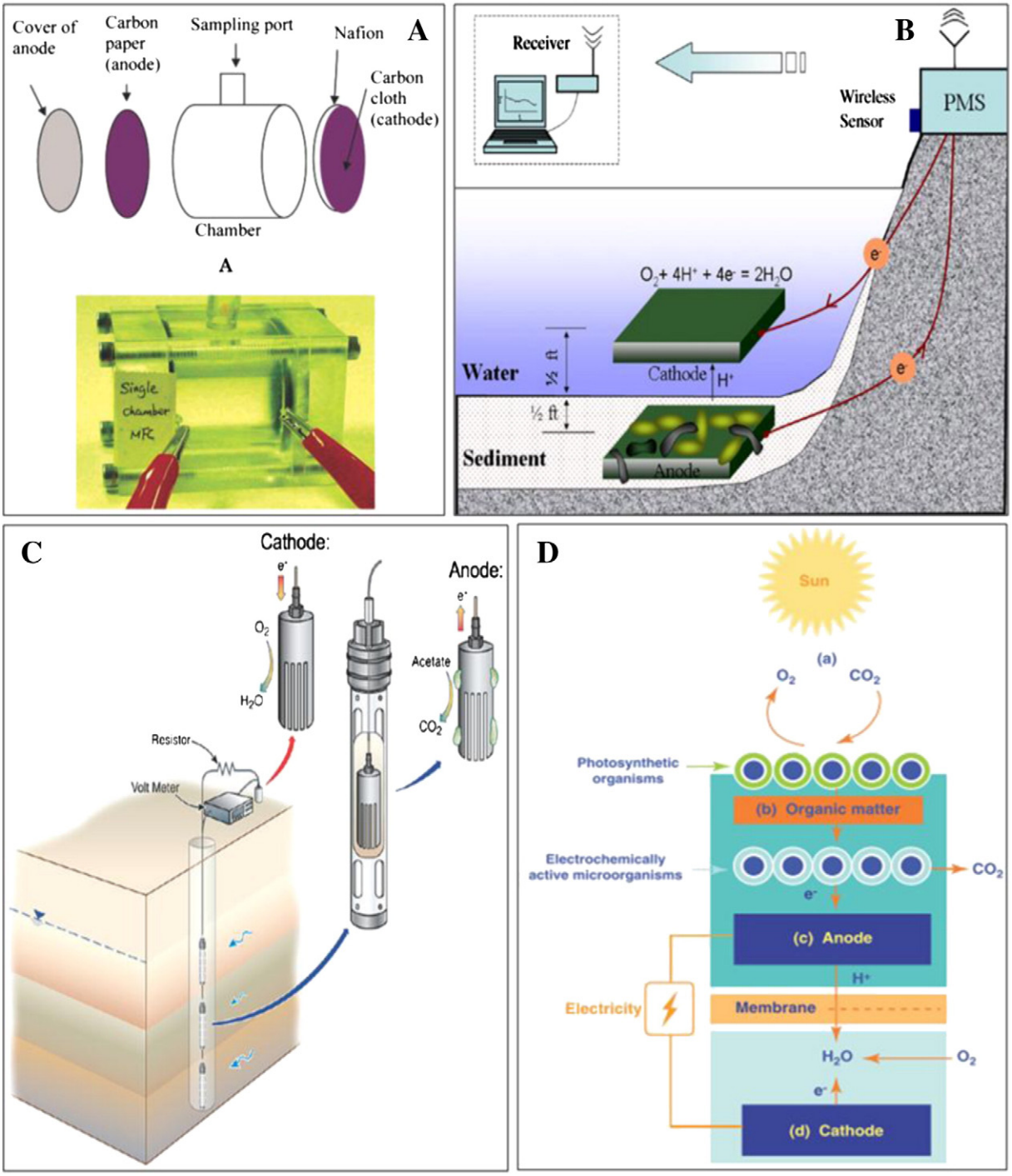

Fig. 3. MFC-based systems for electricity generation: (A) wastewater microbial fuel cells, (B) benthic microbial fuel cells, (C) microbial remediation cells, and (D) microbial solar cells. Reproduced with permission from refs. Donovan et al. (2011), Liu and Logan (2004), Strik et al. (2011), Williams et al. (2010).

microbial fuel cell (Strik et al., 2010), photobioelectrochemical fuel cell (Rosenbaum et al., 2005), photosynthetic microbial fuel cells (PMFCs) (Zou et al., 2009), photosynthetic electrochemical cell (Yagishita et al., 1997), and solar-driven microbial photoelectrochemical cell (solar MPC) (Qian et al., 2010). Despite the variations in system designs, the basic principle of MSCs usually include 4 steps, as described by Strik et al. (2011), and illustrated in Fig. 3D, (i) photosynthesis of organic matter; (ii) transport of organic matter to the anode compartment; (iii) anodic oxidation of organic matter by EAB; and (iv) cathodic reduction of oxygen or other electron acceptors. Here we categorize the MSCs into 3 groups based on the organisms responsible for photosynthesis — plant MSCs, phototrophic MSCs, and algae MSCs. More detailed information can be found in other reviews (Deng et al., 2012; He et al., 2009; Strik et al., 2011).

The most popular MSCs are plant MSCs, which use the organic rhizodeposits excreted from living higher plants to feed EAB for electricity production. Reed mannagrass and rice plants were used first to demonstrate the syntrophic relations, with maximal power outputs of 67 mW/m^2 and 26 mW/m^2, respectively (Schamphelaire et al., 2008; Strik et al., 2008a). Other plants such as *Spartina anglica*, *Arundinella anomala* and *Arundo donax* were also investigated for concurrent electricity and biomass production. *A. donax* failed (Helder et al., 2010), but *S. anglica* was able to generate current for up to 119 days (Timmers et al., 2010). Despite the low power output at the current stage, a European research consortium estimated that the power production from plant-MSCs could reach 1000 GJ/ha/year (3.2 W/m^2) (Strik et al., 2011). Unlike plant MSCs, the phototrophic MSCs do not require the cooperation between the two groups of microbes, because studies showed that strains of photosynthetic bacteria such as *Rhodobacter sphaeroides* can generate electricity through the metabolic activity of *in situ* oxidation of photobiological hydrogen (Rosenbaum et al., 2005), and the power density can be comparable with nonphotosynthetic MFCs (Cao et al., 2008). A self-assembling self-repairing marine sediment system with photosynthetic microbes was reported to generate electricity from sunlight without the need of providing constant flux of glucose and oxygen (Malik et al., 2009). The algae MSC is an emerging system, because the functions of algae and EAB are complementary. The consortium not only can convert solar energy to electric energy, it can also remove nutrients and produce value-added chemicals, such as protein and biodiesel. Both microalgae (*e.g.*, *Chlorella vulgaris*) and macroalgae (*e.g.*, *Ulva lactuca*) have been used in algae MSCs to provide substrates for EAB (Velasquez-Orta et al., 2009). In addition to traditional batch reactors, Strik et al. developed a flow through photosynthetic algal microbial fuel cell (PAMFC) to automatically feed algae to MFCs (Strik et al., 2008b). Another study integrated photobioreactor, anaerobic digester, and MFC reactors together to recover both biogas and electricity (Schamphelaire and Verstraete, 2009). Other systems include recycling anode off gas (CO_2) into an algae grown cathode for additional carbon capture (Wang et al., 2010), and an integrated photobioelectrochemical system with an MFC enclosed inside an algal bioreactor (Xiao et al., 2012). Utilizing the algae, cyanobacteria and protozoa, Strik et al. reported an MSC with a reversible bioelectrode, which can function as a biocathode during illumination for photosynthesis reaction and can then switch to the anode in the dark for organic degradation (Strik et al., 2010). MSCs are the only MESs that do not rely on external electron donors but convert inexhaustible solar energy into electrical energy and chemicals, so they carry great potential if current challenges such as low power output are addressed.

5. MEC-based systems for chemical production

The concept of microbial electrolysis cell was originated in 2005, with the key feature of using an external voltage on top of the MFC potential to enable hydrogen gas evolution at the cathode through the reduction of protons (Liu et al., 2005b; Rozendal et al., 2006). Early studies used external power supplies ranged from 0.6 to 1.0 V to catalyze H_2 evolution, which was much lower than the 1.8–2.0 V used in traditional water electrolysis (Liu et al., 2005b; Logan et al., 2008). Another advantage was that the substrates can be from renewable and waste materials rather than fossil fuels, and the H_2 production rate can be more than 1 m^3/day/m^3 reactor with a yield up to 11 mol H_2/mol glucose, which is more than 3 times higher than dark fermentation (Liu et al., 2010; Logan et al., 2008). Several excellent reviews summarized the material and system development of the MECs for H_2 production (Lee et al., 2010; Liu et al., 2010; Logan et al., 2008).

The elimination of membranes or separators converted dual chamber MECs to single chamber reactors and significantly increased H_2 generation rate, but the produced H_2 was more likely consumed by methanogenesis to generate CH_4 (Liu et al., 2010; Logan et al., 2008). Researchers have tried different inhibition approaches, such as adding expensive methanogen inhibitors, periodically expose solution in aerobic environment, and control the pH and redox potentials, but the CH_4 contamination of H_2 in single chamber MECs still remains a major obstacle (Hu et al., 2008; Liu et al., 2010; Logan et al., 2008). The small external voltage can be supplied by MFC stacks or other renewable power sources such as solar and wind (Chae et al., 2009; Sun et al., 2008). Recently, reverse electrodialysis (RED) was added into MECs generating a new system called microbial reverse-electrodialysis electrolysis cells (MRECs) with spontaneous H_2 production by combining together the driving forces from anode organic oxidation and salinity gradient energy (Fig. 4A), and salt solutions could be continuously regenerated with waste heat (≥40 °C) (Cusick et al., 2012; Kim and Logan, 2011a).

By using similar strategies in MECs, other inorganic chemicals have been produced in the cathode chamber. Cusick and Logan discovered that phosphate can be recovered as struvite ($MgNH_4PO_4 \cdot 6H_2O$) in a modified microbial electrolysis struvite-precipitation cell (MESC) (Cusick and Logan, 2012). Rozendal et al. reported that hydrogen peroxide can be produced by reducing oxygen through the two electron reduction, and the proof-of-concept study showed that at an applied voltage of 0.5 V, H_2O_2 can be generated at a rate of 1.9 ± 0.2 kg H_2O_2/m^3/day with a concentration of 0.13 ± 0.01 wt.% and an overall efficiency of 83.1 ± 4.8% (Rozendal et al., 2009). The same group later used a similar approach to produce alkaline solutions, as they found that by using acetate as the electron donor in the anode, the MEC generated up to 1.05A in current at an applied voltage of 1.77 V, which allowed for the production of caustic to 3.4 wt.% (Rabaey et al., 2010). Such chemicals can be produced during wastewater treatment process and then used as low-cost disinfectants for many industries.

6. MES-based systems for chemical production

Microbial electrosynthesis, also shortened as MES in literature, is an emerging area in microbial electrochemical research and development, and it uses the electrons derived from the cathode to reduce carbon dioxide and other chemicals into a variety of organic compounds, especially those with multiple carbons that are precursors for desirable value-added chemicals or liquid transportation fuels (Lovley and Nevin, 2011; Rabaey and Rozendal, 2010; Rabaey et al., 2011). The potential of MES not only comes from the double benefits of carbon sequestration and organic production, but may also address the harvesting, storage, and distribution problems associated with energy crops, solar and wind farms, and natural gas exploration, because the electrons can be from any renewable source, and microbes may harvest solar energy in a 100-fold higher efficiency than biomass-based chemical production (Lovley and Nevin, 2011).

The concept of microbial electrosynthesis was only introduced in 2009–2010, with the initial findings associated with methane generation from a reactor with an abiotic anode and a biocathode acclimated with *Methanobacterium palustre* (Cheng et al., 2009). Another early study demonstrated that biofilms of *Sporomusa ovata* could use the electrons supplied by the cathode to reduce carbon dioxide into acetate and small amounts of 2-oxobutyrate. Electrons appearing in these products

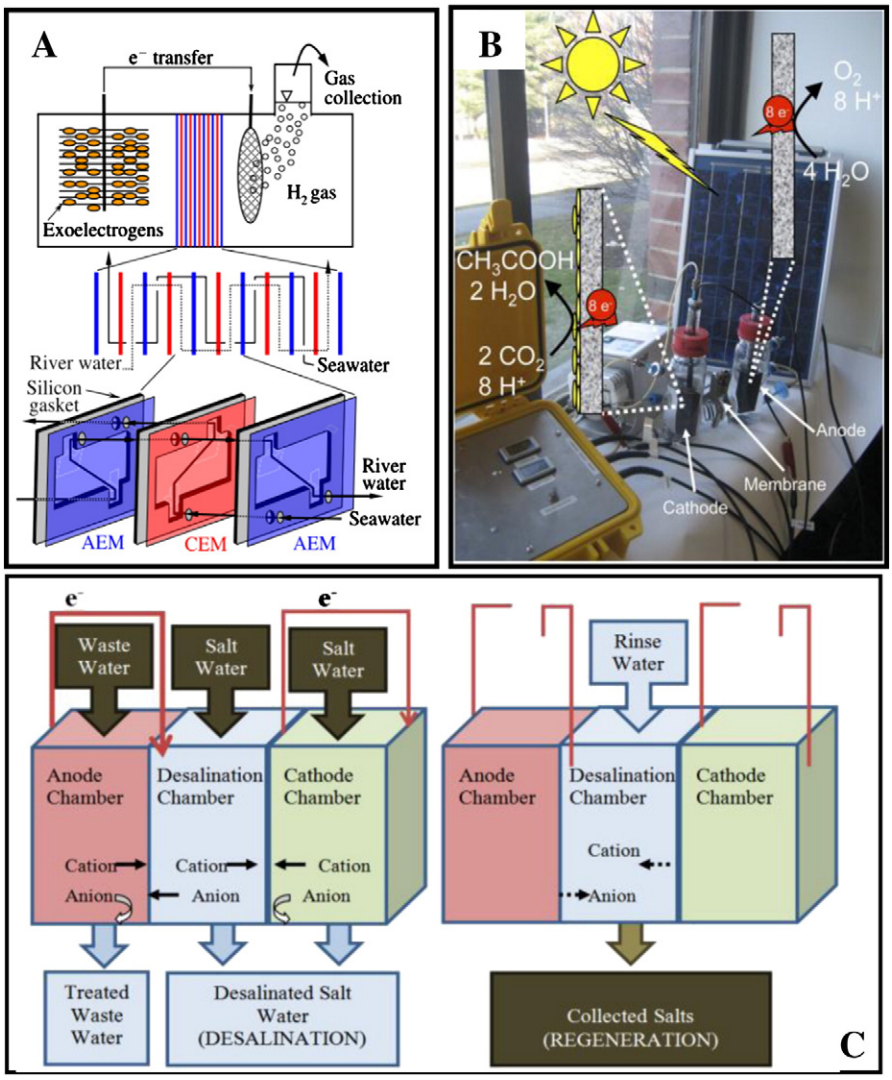

Fig. 4. Some advanced MESs: (A) a microbial reverse-electrodialysis electrolysis cell (MREC), (B) a microbial electrosynthesis (MES), and (C) a microbial capacitive desalination cell (MCDC).
Reproduced with permission from refs. Forrestal et al. (2012b), Kim and Logan (2011a), Nevin et al. (2010).

accounted for over 85% of the electrons consumed (Fig. 4B) (Nevin et al., 2010). In general, acetogenic bacteria use hydrogen as the electron donor for carbon dioxide reduction, but it was found that many acetogenic bacteria, such as *Clostridium ljungdahlii*, *Clostridium aceticum*, *Sporomusa sphaeroides*, and *Moorella thermoacetica*, were all able to consume electrical current and produce organic acids (Nevin et al., 2011). Studies also showed that ethanol can be produced by reducing acetate at the cathode, but some processes required addition of mediators, such as methyl viologen (MV) (Steinbusch et al., 2010). The mixed culture originated from brewery wastewater was reported to generate methane, acetate, and hydrogen gas from a biocathode poised at −590 mV (vs SHE) with CO_2 as the only carbon source (Marshall et al., 2012), and research on genetically modified microorganisms may significantly facilitate electron uptake and organic synthesis. As discussed in several conceptual review articles, the microbial electrosynthesis carries great potential, but there are also many technological and economic challenges to be solved before it can be implemented in large scale (Lovley and Nevin, 2011; Rabaey and Rozendal, 2010; Rabaey et al., 2011).

7. MDC-based systems for water desalination and beneficial reuse

Water desalination using the MDC process was first introduced in 2009 by Cao et al., and the proof-of-concept study was selected as the top technology paper by *Environmental Science & Technology* (Cao et al., 2009). The basic principle of MDC is to utilize the electric potential generated across the anode and cathode to drive desalination *in situ*. Compare to other MESs, MDCs have a third chamber for desalination by inserting an anion exchange membrane (AEM) and a cation exchange membrane (CEM) in between the anode and cathode chambers. When bacteria in the anode chamber oxidize biodegradable substrates and produce current and protons, the anions (*e.g.*, Cl^-) in the middle chamber migrate to the anode and the cations (*e.g.*, Na^+) are drawn to the cathode for charge balance, thus the middle chamber solution is desalinated (Cao et al., 2009; Luo et al., 2012c). Recently, other approaches were developed to achieve desalination as well. For example, by switching the CEM to the anode side and AEM to the cathode side, a microbial saline–wastewater electrolysis cell (MSC) desalinates anolyte and catholyte by driving salts into the middle chamber (Kim and Logan, 2013b). Osmotic microbial fuel cells (OsMFCs) or osmotic MDCs (OsMDCs, MODCs) use a forward osmosis membrane to replace the AEM and withdraw pure water from wastewater to the draw solution, and then water can be recovered during draw solution regeneration (Kim and Logan, 2013a; Zhang et al., 2011). A capacitive microbial desalination cell (cMDC) incorporates capacitive deionization into an MDC to improve desalination efficiency (Forrestal et al., 2012a, 2012b; Yuan et al., 2012). In addition to desalination, acid (HCl) and

base (NaOH) solutions can be produced if a bipolar membrane is placed into the MDC next to the anode chamber, creating a four-chamber system called a microbial electrolysis desalination and chemical-production cell (MEDCC) (Chen et al., 2012).

The MDC can be used as either a stand-alone for simultaneous organic and salt removal with energy production or a pretreatment for conventional desalination processes such as reverse osmosis (RO) to reduce the salt concentration in feed solution, and minimize energy consumption and membrane fouling. Compared with current technologies that use 6–68 kWh to desalinate 1 m^3 of seawater, MDC studies showed that 180–231% more energy can be recovered as H_2 than the reactor energy input when desalinating 5–20 g/L NaCl solutions (Luo et al., 2011; Mehanna et al., 2010), and it was estimated that an MDC may produce up to 58% of the electrical energy required by downstream RO systems (Jacobson et al., 2011). Higher desalination efficiency and current output can be achieved through membrane stacks (Chen et al., 2011; Kim and Logan, 2011c), and electrolyte recirculation was shown effective in stabilizing electrolyte pH (Luo et al., 2012a; Qu et al., 2012). Traditional MDC designs accomplish desalination by transporting ions from the middle chamber to the anode and cathode chambers, which increases the conductivity of the anolyte and catholyte. This change has been shown beneficial to electricity generation due to improved mass transfer, but the increased salinity may also affect effluent water quality and prevent subsequent beneficial use of treated wastewater (Luo et al., 2012c). One solution for complete salt removal from all the liquids may involve the physical and electrical adsorption of ions onto high surface area membrane electrode assemblies, such as microbial capacitive desalination cells (MCDCs), which showed up to 25 times of increase in salt removal and complete salt recovery (Fig. 4C) (Forrestal et al., 2012b). Similar as many membrane based technologies, one challenge for MDCs may come from membrane fouling due to biofilm growth and scaling due to the deposition of hardness-causing cations, but studies on understanding and addressing such problems are just getting started, and solutions remain to be found (Luo et al., 2012a, 2012b).

8. Outlook

In about one decade of research and development, the functionality of MESs has expanded dramatically and the performance has improved exponentially. However, despite the many different functions discovered, there are many remaining challenges before this technology can be implemented in larger scale. Taking MFCs as an example, the power density has increased by orders of magnitude, from less than 1 mW/m^3 to 2.87 kW/m^3 (or 10.9 kA/m^3) (Fan et al., 2012), primarily due to the advancements in reactor architecture, material, and operation, which relieves the physical and chemical constraints of the system. The projected wastewater treatment capacity of MFCs can reach 7.1 kg chemical oxygen demand (COD)/m^3 reactor volume/day, which is even higher than conventional activated sludge systems (~0.5–2 kg COD/m^3 reactor volume/day) (Rozendal et al., 2008). However, there are still many challenges that need to be addressed before the technology can be applied in commercial scale. The replacement of expensive metal catalysts and membranes with cheaper alternatives has dramatically reduced the reactor costs, but the overall cost of MESs is still considered expensive for wastewater treatment, unless an estimated threshold of internal resistance < 40 mΩ m^2 in combination with a current density around 25 A/m^2 can be reached (Sleutels et al., 2012). Most studies are still limited in lab scale, and several pilot scale plants with capacities between 20 and 1000 l have not yet shown stable and high enough performance due to the problems of water leaking, low power output, influent fluctuation, and unfavorable products (Cusick et al., 2011; Keller and Rabaey, 2008; Logan, 2010). To achieve practical implementation, MESs will need to be scaled-up to at least in cubic meter scale, the reactor configurations have to be easily integrated with current infrastructure, and effectively harvesting systems instead of resistors have to be developed to deliver usable power (Park and Ren, 2012; H. Wang et al., 2012). Multiple reviews have summarized the progresses of MFC system development and provided insights in further directions (Logan, 2010; Lovley, 2011; Rozendal et al., 2008; Wei et al., 2011).

Compared to electricity generation in MFCs, chemical production and desalination from MESs have been considered technically and economically more feasible due to the higher price of chemicals and relatively simple collection process. But such processes are relatively new and mainly in lab scale, and there have been few reports in scale-ups (Cusick et al., 2011; Logan, 2010). Among the many different functions developed using this MES platform technology, as discussed across this article, it is not clear where the MES can contribute the most to the current environmental infrastructure and chemical industries. There have been very limited evaluations of different systems regarding to their life cycles in terms of function selections or comparisons with established technologies, which they may complement (Foley et al., 2010; Pant et al., 2011). It has been assumed that the most environmental benefits from MESs come from the displacement of fossil fuel dependent resources (*i.e.* grid electricity, or chemical manufacture) through co-product production (*i.e.* electricity, chemicals) from renewable sources, but the energy and environmental footprints of different systems have to be clearly quantified before implementing large scale applications. In addition, fundamental understandings on the unique electron transfer mechanisms between bacterial cells and electrodes as well as among different microbial species are crucial for further system development. Such characterizations should be performed on both pure cultures at different growth stages as well as microbial consortiums that are present in the environment. Overall, despite the remaining challenges, if MES keeps its pace in research and development, it is reasonable to believe that in the near future this platform technology will provide viable solutions to address many energy and environmental problems.

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