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REVIEW ARTICLE

Microbial fuel cell with high content solid wastes as substrates: a review

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HIGHLIGHTS

- Fundamentals and configuration design of MFCs fueled by HCSW were reviewed.
- HCSWs including sewage sludge, biomass and biowaste treated in MFCs were summarized.
- HCSW based MFCs technologies covered the types of sediment, soil, wetland and plant.
- Activated sludge process and composting could be coupled with HCSW-MFCs.
- HCSW-MFCs could be applied in bioremediation and biosensing.

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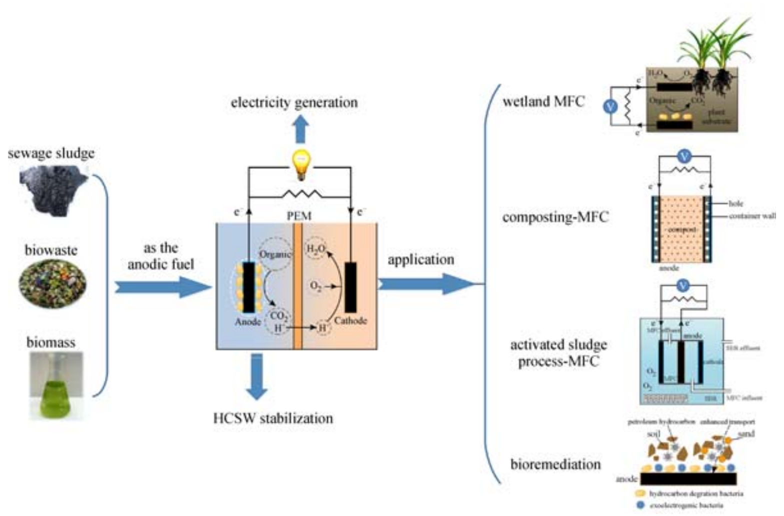
GRAPHIC ABSTRACT



ABSTRACT

With the increasing concern about the serious global energy crisis and high energy consumption during high content solid wastes (HCSWs) treatment, microbial fuel cell (MFC) has been recognized as a promising resource utilization approach for HCSW stabilization with simultaneous electrical energy recovery. In contrast to the conventional HCSW stabilization processes, MFC has its unique advantages such as direct bio-energy conversion in a single step and mild reaction conditions (viz., ambient temperature, normal pressure, and neutral pH). This review mainly introduces some important aspects of electricity generation from HCSW and its stabilization in MFC, focusing on: (1) MFCs with different fundamentals and configurations designed and constructed to produce electricity from HCSW; (2) performance of wastes degradation and electricity generation; (3) prospect and deficiency posed by MFCs with HCSW as substrates. To date, the major drawback of MFCs fueled by HCSW is the lower power output than those using simple substrates. HCSW hydrolysis and decomposition would be a major tool to improve the performance of MFCs. The optimization of parameters is needed to push the progress of MFCs with HCSW as fuel.

1 Introduction

High content solid wastes (HCSWs) mainly refer to the wastes containing high content solids (e.g. sewage sludge, biowaste, biomass, etc.). In the 21st century, a large quantity of sewage sludge is generated from wastewater treatment plants (WWTPs) due to high water consumption by an increasing population, industrialization and urbanization. The growing global urbanization of societies brings

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with it an increasing stringent sludge reuse and/or disposal regulations and raising public concerns. This is forcing sludge generators to review their sludge management strategies [1]. Biowastes, which mainly include food wastes (FWs), garden wastes and dung, are produced daily. Organic matter in the biowaste is harmful to humans and the environment. Conversely, biomass is an alternative renewable resource which can be used to produce green fuel (e.g., bioethanol or biodiesel) through sustainable approach and technological advancement. The potential exists for exploring inexpensive, renewable and sustainable biofuel due to the increasing energy demand . Many forms of biomass such as raw corn, rice and wheat stocks contain large amounts of energy, which can be a good source for power generation in an air-cathode H-type MFC [2–4]. This energy remains generally unexploited and yet to be tested. Without the appropriate treatment technic/methodology/technology, these huge amounts of this abundant organic and odor pollutants of HCSW would threaten both aquatic and the atmospheric environment.

Presently, the conventional methods for the treatment of HCSWs are facing challenges compared with the newly developed processes and concepts, which focus on cost-effectiveness and performance-efficiency. Compost, anaerobic digestion and incineration are the three main HCSW treatment technologies. Composting is a traditional HCSW treatment method, that converts HCSW to agricultural fertilizer, such that heavy metals, pathogens and micropollutants in the compost are removed because of food safety concerns [5]. In addition, anaerobic digestion can utilize HCSW to produce biogas but it requires heating and a long hydraulic retention time (more than 20 days) under mesophilic or thermophilic operation conditions. Incineration can dispose HCSW completely, but the high water content means HCSW should be mixed with high calorific value fuel, the reaction between high moist FW and low humid and high energy content wastes also may lead to the production of dioxins [6]. Overall, the deficiency and limitations of these technologies make the treatment of HCSW expensive as well. The treatment and disposal of sewage sludge account for about 25%–65% of the total expenses of a typical WWTP [7]. Now, more research is focused on the development of innovative, environmentally friendly and sustainable treatment technologies.

As an emerging energy conversion technology, microbial fuel cell (MFC) has been intensively researched in recent decade, to offer the opportunity for simultaneous pollutant removal and energy production. Recovering the rich energy in wastes makes it possible to compensate for the energy consumed in the operation of the treatment plant [8]. Recently, the available substrates of MFCs extend from micro-molecular organic to wastewater, sludge, dung and so on, which makes it possible for MFCs to treat HCSW. MFC utilizes exoelectrogenic microorganisms (EM) to convert the chemical energy stored in different substances to electric energy. The microorganisms are linked via electron donating and accepting conditions through the artificially introduced electrodes (viz., anode and cathode) that stimulates the development of a potential difference which acts as a net driving force for bioelectrogenic activity [9]. In contrast to conventional fuel cells, MFC has advantages such as direct bio-energy conversion in a single step and mild reaction conditions (viz., ambient temperature, normal pressure, and neutral pH). Due to the complexity of HCSW, MFC can provide stable external environment for EM out of frequent replacement of substrate. Steady current output with high organic matter content saves energy of transporting substrate.

In this perspective, the present review delineates HCSW as substrate of MFC pertaining to waste degradation and energy recovery. Significant developments and sustainable researches have been reported on MFC to date with the first investigation on MFC with HCSW reported by Dental, who demonstrated to harvest electrical energy from anaerobically digested sludge [10]. While a number of MFCs with HCSW as substrate have been extensively investigated, it still needs a comprehensive review paper to summarize the experimental results on bioelectrochemical oxidation and/or reduction for HCSW in MFC. This work covers important areas of MFC with HCSW, concerning fundamentals and configuration design, the wastes degradation and electricity generation, solid phase MFC, MFC coupling technology and novel applications. Furthermore, the prospect and deficiency posed by MFCs with HCSW are discussed for its practical implementation.

2 Fundamentals and configuration design of MFC with HCSW as substrates

2.1 Fundamentals of MFC

Appropriate configuration is an important characteristics of MFC, and researchers have developed several configurations over the years with improved performance. Fig. 1 shows the detail components of a typical two-chamber MFC. In MFC, organic matter in HCSW is oxidized by EM under anaerobic conditions of the anode chamber with electrons and protons production. The electrons are transferred through the external circuit to the cathode chamber, where electrons, protons and oxygen (electron acceptor) combine to produce water.

The insoluble macromolecules in HCSW should be hydrolyzed into simple molecules before the microbes utilize them in the anode chamber. Conventionally, four major steps can be distinguished (Fig. 2). Both biological decomposition of organic polymers and solubilization of insoluble particulate matter take place in the first hydrolysis step. Acidogenesis and acetogenesis follow in the second and the third step, where a wide variety of fermentation end products are formed. In the final step, methanogenic community transforms these products into

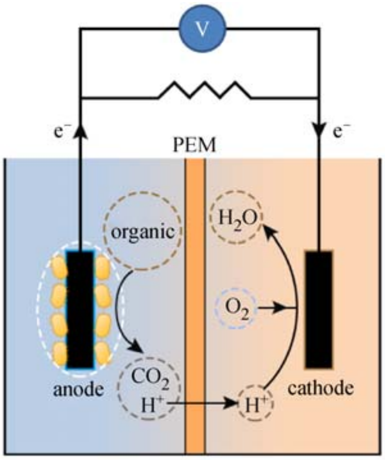

Fig. 1 Schematic illustration of MFC components and the principle of operation

methane. Biofilm attached on the anode surface of MFC hydrolyzes the complex organic matter in HCSW into simple molecules, which are further oxidized by EM. The electrons that the anode accepted flow through an external circuit to the cathode, where an oxidant (viz., electron acceptor) such as oxygen or metal ions are reduced resulting in the production of bioelectricity.

The electrode reactions in MFC fueled by HCSW are also illustrated in Fig. 1. At the cathode, Fe^{3+} ($K_3Fe(CN)_6$, $FePO_4$), Cu^{2+} ($CuSO_4$ wastewater), oxygen (air, dissolved oxygen), etc. are used as electron acceptors [11]. The electrons flowing from the anode to the cathode through the external resistor and circuit result in the production of electric current. A series of proton exchange pathways are also employed to make the proton transfer from the anode chamber to the cathode chamber possible, such as proton exchange membranes (PEM), sediment-water interface, baffle plate and cation exchange membrane (CEM).

2.2 Configuration design of MFC

MFC fueled by HCSW contains two main configurations (viz., single-chamber and two-chamber), and research on MFC configuration is based on the improvement of the single-chamber and two-chamber. PEMs are widely employed in two-chamber MFC that are the most popular configuration in MFC fueled by HCSW (Fig. 1).

In two-chamber MFC, the anode chamber is fueled with HCSW, the cathode chamber is filled with electrolyte or dissolved oxygen, and PEM is placed between these two chambers. Single-chamber MFC fueled by HCSW can be constructed using waste-oxygen interface as proton exchange pathway (Fig. 3). Single-chamber MFC has some advantages over two-chamber MFC for practical application: (1) the operation is simplified with no recycle or chemical regeneration of catholyte; (2) higher volumetric power density due to smaller cell volume; (3) less investment in membrane. However, the coulombic efficiency of single-chamber MFC is lower than that of two-chamber MFC with membrane as separators due to oxygen diffusion through the cathode. Another drawback is that the distance between anode and cathode in the single-chamber MFC is limited to a certain range, due to the potential negative effect of oxygen on the activity of the anaerobic bacteria and the risk of short circuit. The single-chamber MFC fueled by HCSW gradually presents the trend of semi-open style (e.g., soil MFC and compost MFC) [12,13]. The cell does not need to have a strictly anaerobic environment [14], and the semi-open style chamber decreases the cost of MFC with increased oxygen involved in the reaction and hence improved electricity generation performance.

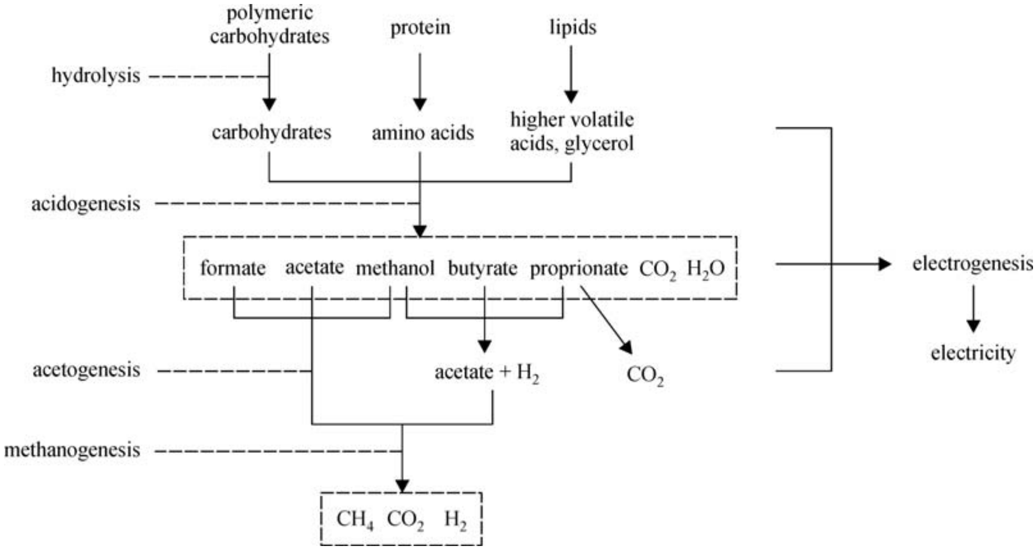

Fig. 2 Process of organic matter degradation and available fuel for MFC within HCSW

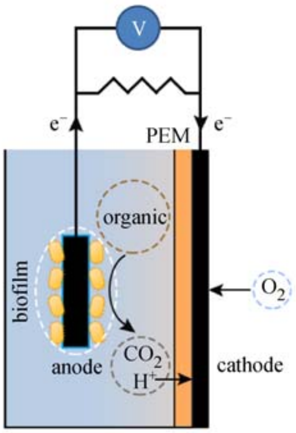

Fig. 3 Single-chamber MFC with open air cathode

Recently, a few multiple-chamber MFCs have emerged to satisfy the needs of multifunction especially aiming at improving electricity generation performance. Zhang et al. constructed a three-compartment MFC (one anode and two cathode chambers joined with a pipe) for sewage sludge treatment (Fig. 4(a)) [15]. Two PEMs with the same cross-sectional area of 4900 mm^2 were used to separate the anode and the cathode chambers. By using the three-chamber MFC configuration in this study, electricity yielded from sewage sludge at maximum power output of 13.2±1.7 $W\cdot m^{-3}$ during polarization. In addition, the stable current in MFC system leads to the flow of multiple ions. Based on this feature, Meng et al. developed a biocathode microbial desalination cell (MDC) with dewatered sludge as fuel for synergistic desalination, electricity generation and sludge stabilization [16]. The MDC reactor consisted of five plexiglas chambers with a symmetric construction, i.e. two cathode chambers, two desalination chambers and one anode chamber. Each of the two chambers was connected with square window to facilitate ionic migration. Anion-exchange membranes and CEMs were installed on both side of desalination chambers, the permselectivity of these membranes achieved the enrichment of salinity (Fig. 4(b)). In the stable operation period, the desalination rates were 46.37±1.14% and 40.74±0.89% with the initial salt concentrations of 5 $g\cdot L^{-1}$ and 10 $g\cdot L^{-1}$ in 24 h. With the different number of chambers and setting of ion-exchange membranes, improvement of electricity generation and specific ion removal were achieved. It is worth noting that the high cost of complicated reactor design/components and the ion-exchange membranes constrain the application of this MDC.

2.3 Typical exoelectrogenic microorganism in MFC with HCSW as substrates

Catalytic reaction of EM plays a key role in electricity generation of MFCs. *Shewanella putrefaciens*, *Geobacteraceae sulfurreducens*, and *Geobacter metallireducens* are well-known EM. Besides these, a variety of other species have also been reported (Table 1). No specific ubiquitous microorganism has been found in different MFCs with HCSW as substrates, indicating that a great amount of bacterial species can adapt to the situation of electricity generation under HCSW substrates. Most bacteria species present in raw activated sludge (AS) are potential EM, which can explain why AS and anaerobic sludge are utilized widely as inoculums in MFCs. It can be observed from Table 1 that *Proteobacteria*, *Bacteroidetes*, and *Frimicutes* are dominant in MFCs inoculated with AS or anaerobic sludge. The addition of bioelectrochemical system (BES) can affect the dominant archaea (especially methanogens) and slightly influence the dominant bacteria (including EM), but cannot kill them.

The inoculum source can significantly affect the formation of the anode bacteria community and other bacteria can be involved in the generation of electricity according to inoculum type. Different bacteria were developed in SMFC-I with anion exchange membrane-seperator-electrode assemblies (AEM-SEA) (δ-*proteobacteria* and *Firmicutes*) and SMFC-II with Nafion-SEA (γ-*proteobacteri*a, β-*proteobacteria* and *Bacteroidetes*) [17]. Different inocula [AS, garden soil (GS), wastewater (WW) and river sediment (RS)] resulted in different

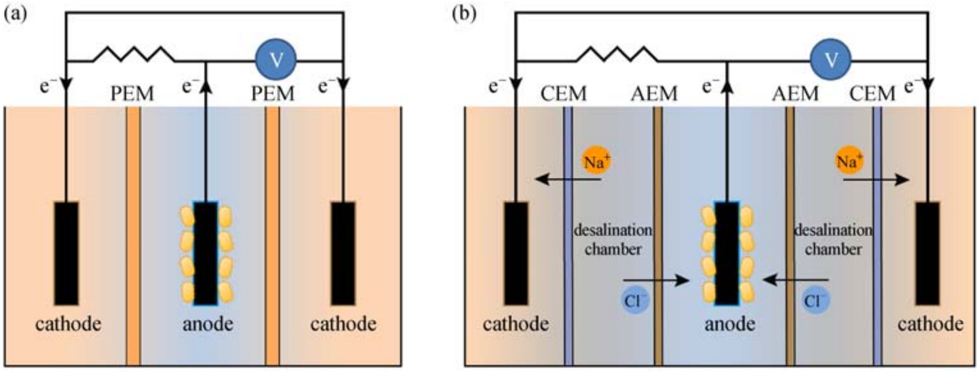

Fig. 4 (a) Schematic drawing of the three-chamber biocathode MFC; (b) Schematic diagram of the MDC reactor

Table 1 Microbial species involved in MFCs for electricity generation

inoculum	substrate	predominant species	reference
activated sludge from municipal wastewater treatment plant	anaerobic sludge	*Proteobacteria*, *Bacteroidetes*, uncultured bacteria, *Actinobacteria*, *Firmicutes*, *Chloroflex*.	Zhang et al. [15]
anaerobic sludge	anaerobic sludge	Unclassified_*Clostridiales*, *Clostridium XI*, unclassified_*Comamonadaceae*, *Arcobacter*, *Desulfobulbus*, *Desulfovibrio*, *Geobacter*.	Wang et al. [29]
anaerobic sludge	sewage sludge	*Clostridium* sp., *Lactobacillus* sp., *Flavobacterium* sp, *Methanolinea* sp., *Methanospirillum* sp., *Methanosarcina* sp., *Methanosphaera* sp.	Xiao et al. [121]
anaerobic sludge	anaerobic and aerobic activated sludge	δ-*proteobacteria*, γ-*proteobacteria*, *Firmicutes*.	Gao et al. [122]
anaerobic sludge	anaerobic sludge	*Geothrix*, *Geobacter*, *Desulfuromonas*.	Yoshizawa et al. [123]
aerobic sludge	aerobic sludge	*Bacteroidetes*, *Nitrospirae*, β-*proteobacteria*, *Chloroflexi*, *Chlorobi*, *Gammaproteobacteria*.	Gao et al. [122]
anaerobic sludge	food waste leachate	*Proteobacteria*, *Acidobacteria*.	Li et al. [124]
food wastes	food wastes	*Proteobacteria*, *Bacteroidetes*, *Firmicutes*.	Jia et al. [125]
activated sludge	food waste	*Proteobacteria*, *Bacteroidetes*.	Blanchet et al. [46]
anaerobic activated sludge	dairy manure	*Clostridium*, *Ochrobactrum*, *Pseudomonas*, *Comamonas, Desulfobulbus*.	Zhang et al. [126]
activated sludge	swine manure	*Clostridium*.	Vilajeliu-Pons et al. [127]
anaerobic consortia	powder orange peel waste	*Proteobacteria*, *Bacteroidetes*, *Chloroflex*.	Miran et al. [128]
lake water	sediments with cyanobacterial bloom biomass amendment	*Proteobacteria*, *Bacteroidetes*, *Chloroflexi*, *Acidobacteria*, *Actinobacteria*, *Firmicutes*, *Nitrospirae*, *Planctomyceltes*, *Chlorobi*, *Spirochaetes*.	Zhou et al. [64]
anaerobic sludge from anaerobic digester process	Microalga *Chlorella Vulgaris*	*Proteobacteria*, *Bacteroidetes*, *Synergistertes*, *Chlorophyta*, *Spirochaetes*.	Lakaniemi et al. [129]
anaerobic sludge from anaerobic digester process	Microalga *Dunallella Tertiolecta*	*Proteobacteria*, *Bacteroidetes*, Deferribacteres, *Firmicutes*.	Lakaniemi et al. [129]

community composition at the anode biofilms. MFC-AS and MFC-GS were closely clustered and were separated from MFC-WW and MFC-RS [18]. There was substantial difference in community composition in MFCs fed with different pretreated (ozonation and microwave digestion) sludge. *Bacteroidetes* was abundant bacterial phylum which dominated on anodes of higher productivity MFC [19]. High-performing MFC's fed with paddy soils of high dissolved organic carbon (DOC) and NH_4^+ concentrations in porewater selected for an active, highly electrogenic bacterial community (dominated by δ-*proteobacteria*), while the dominant bacterial community for the low-performing MFCs from soils of low DOC and NH_4^+ was β-*proteobacteria* [20].

Different biocathode materials had important effect on the microbial species in the biocathode MFC. *Bacteroidetes* and *Proteobacteria* were the dominant phyla on the materials packed biocathode MFCs. *Comamonas* of β-*proteobacteria* played significant roles in the electron transfer process of granular activated carbon, granular semicoke and carbon felt cube packed biocathode MFCs, while in granular graphite packed MFC *Acidovorax* correlated with power generation [21]. The microbial communities on anodic surface of MFCs varied with cathode types and operational procedure [22].

3 MFCs with high content solid substrates

MFCs can utilize substrates ranged from wastewater, sludge to polluted soil, dung or even gas. The severity of environmental hazards of high content solid substrates has made MFCs operated with high content solid substrates to gain more attention. Researches have proven that different kinds of high content solid substrates are compatible with MFCs technology. Studies on high content solid substrates used in MFCs are listed in Table 2. These progresses indicate that MFCs with high content solid substrates have the potential to be applied in multiple aspects.

3.1 High content solid substrates treatment in MFCs

3.1.1 Sewage sludge

During the early stage of MFCs study, marine sediments were commonly used as anodic substrates for harvesting

Table 2 MFCs operated with different HCSW substrates

section	MFCs configuration	inoculum source	substrate	pretreatment	maxium power density "()" means max power density wasn't given	electron acceptor	reference
sewage sludge	membraneless MFCs	–	digester sludge	–	–	oxygen	Dentel et al. [10]
	two-chamber MFCs	sewage sludge	sewage sludge	–	$8.5 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Jiang et al. [23]
	two-chamber MFCs	–	saline domestic sewage sludge	–	$41 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Karthikeyan and Selvam [24]
	two-chamber MFCs	anaerobic sludge	sewage sludge	–	$11.04 W \cdot m^{-3}$	$KMnO_4$	Begera and Ghangrekar [25]
	single chamber air cathode MFCs	anaerobic mesophilic sludge	anaerobic mesophilic sludge	–	$53.3 W \cdot m^{-3}$	oxygen	Martin et al. [26]
	two-chamber MFCs	anaerobic digestion sludge	digested sludge	ultrasound	$12.67 W \cdot m^{-2}$	oxygen	Oh et al. [28]
				heat/alkaline	$12.53 W \cdot m^{-2}$		
	two-chamber MFCs	sewage sludge	anaerobic sewage sludge	–	$38.1 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Wang et al. [29]
	two-chamber MFCs	sewage sludge	sewage sludge	ultrasonic and alkaline	$12.5 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Jiang et al. [30]
	two-chamber MFCs	activated sludge	activated sludge	microwave	$(42 \pm 3 mW \cdot m^{-2})$	oxygen	Yusoff et al. [31]
	two-chamber MFCs	–	dairy waste activated sludge	low temperature thermo-chemical	$0.715 W \cdot m^{-3}$	oxygen	Jayashree et al. [32]
	single chamber air cathode MFCs	MFCs effluent	fermented primary sludge	–	$320 \pm 10 W \cdot m^{-2}$	oxygen	Yang et al. [33]
	two-chamber MFCs	sewage sludge	sewage sludge	freezing/thawing	$10.2 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Chen et al. [34]
	two-chamber MFCs	anaerobic sludge	anaerobic sludge	–	$(36.8\sim40.1 mW \cdot m^{-2})$	oxygen	Xiao et al. [35]
	two-chamber MFCs	*Escherichia coli*	digested sewage sludge	–	$3.1 W \cdot m^{-3}$	Fe^{3+}	Fischer et al. [36]
	triple-chamber MFCs (dual anode)	*Escherichia coli*	digested and dewatered sewage sludge	–	–	Fe^{3+}	Happe et al. [37]
	two-chamber MFCs	activated sludge	activated sludge	–	$8.7 W \cdot m^{-3}$	NaOCl	Ghadge et al. [38]
	two-chamber MFCs	sewage sludge	sewage sludge	ultrasound	$11.8 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Jiang et al. [39]
	two-chamber MFCs	sewage sludge	sewage sludge	–	$9.1 \pm 0.1 W \cdot m^{-3}$	$K_3Fe(CN)_6$	Jiang et al. [40]

(*Continued*)

section	MFCs configuration	inoculum source	substrate	pretreatment	maxium power density "()" means max power density wasn't given	electron acceptor	reference
biowaste	membraneless MFCs	–	dried blended farm manure	–	$5\mathrm{mW\cdot m^{-2}}$	oxygen	Scott and Murano [14]
	two-chamber MFCs	biogas slurry	cattle dung	–	$220\mathrm{W\cdot m^{-3}}$	$KMnO_4$	Zhao et al. [41]
	single chamber air cathode MFCs	anaerobic sludge	food wastes	oil removal	$5.6\mathrm{W\cdot m^{-3}}$	oxygen	Li et al. [130]
	two-chamber MFCs	digestate	livestock manure and agricultural wastes	–	$73\mathrm{mW\cdot m^{-2}}$	oxygen	Di et al. [43]
	triple-chamber MFCs (dual anode)	*Escherichia coli* and manure leachate	cattle wastes	–	$215\mathrm{mW\cdot m^{-2}}$	oxygen	Zheng and Nirmalakhanadan [44]
	single compartment combined membrane-electrodes	digested slurry	livestock organic solid waste	–	$36.6\mathrm{mW\cdot m^{-2}}$	oxygen	Lee and Nirmalakhanadan [45]
	twin-compartment brush-type anode electrodes				$67\mathrm{mW\cdot m^{-2}}$		
	membraneless MFCs	–	composite food waste	oil removal	$170.81\mathrm{mW\cdot m^{-2}}$	oxygen	Mohan and Chandrasekhar [131]
	two-chamber MFCs	garden compost leachate	dairy wastes	–	$91\mathrm{mW\cdot m^{-2}}$	oxygen	Cercado-Quezada et al. [132]
	triple-chamber MFCs (dual anode)	activated sludge	food wastes	dilution	$(21.9\pm2.1\mathrm{A\cdot m^{-2}})$	oxygen	Blanchet et al. [49]
	triple-chamber MFCs (dual cathode)	topsoil	dairy manure	–	$14.11\pm0.20\mathrm{W\cdot m^{-3}}$	oxygen	Zhang et al. [47]
	two-chamber MFCs	activated sludge	food wastes	–	$(14.8\mathrm{A\cdot m^{-2}})$	oxygen	Bridier et al. [48]
biomass	two-chamber MFCs	wastewater	*Scenedesmus obliquusas*	acid-thermal	$951\mathrm{mW\cdot m^{-3}}$	$K_3Fe(CN)_6$	Kondaveeti et al. [19]
	two-chamber MFCs	anaerobic consortium	*Chlorella vulgaris*	–	$5.3\mathrm{mW\cdot m^{-2}}$	oxygen	Lakaniemi et al. [49]
			Dunaliella tertiolecta		$15.0\mathrm{mW\cdot m^{-2}}$		
	two-chamber MFCs	polluted water	*Microcystis aeruginosa*	ultrasound	$4.2\pm0.1\mathrm{W\cdot m^{-3}}$	oxygen	Wang et al. [50]
	two-chamber MFCs	domestic wastewater	algae sludge	alkaline	$2.8\mathrm{W\cdot m^{-3}}$	oxygen	Wang et al. [51]
	membraneless MFCs	sewage sludge	cyanobacteria	acidic fermentation	$72\mathrm{mW\cdot m^{-2}}$	oxygen	Zhao et al. [52]

energy. Based on the concept of sediment MFCs, Dentel et al. first proved that electricity could be generated directly from digested sludge [10]. Afterwards, Jiang et al. studied the characteristics of MFCs operated with sludge systematically and pointed out that sludge MFCs had a stable electricity production performance under a wide range of operation conditions [23]. In hostile environment, MFCs show a strong tolerant capacity, the ability to treat high salinity sewage sludge is an example [24]. However, adjusting suitable operation conditions such as sludge loading rate, concentration, pH and temperature is equally important for optimizing system performance [25–27].

The feasibility of MFCs fed with different type of sewage sludge has been widely studied [28,29]. Presently, most research focuses on the enhancement of the MFC system. The bottleneck of sludge treatment technology is the degradation of the recalcitrant organic matter, thus for the complex components, various pretreatment methods have been adopted to improve the sludge MFCs performance. Jiang et al. tested ultrasonic and alkaline pretreatments of sludge, which favored organic matter removal but had a relatively small impact on power output [30]. However, microwave pretreatment significantly improved MFC electricity productivity while ozonation pretreatment had less effect in Yusoff's study [31]. In addition, fermentation process, low temperature thermo-chemical and freezing/thawing treatment are all found to enhance both organic matter removal and electricity generation [32–34].

Other than feeding the anode chamber, sewage sludge can be added into the cathode chamber, for instance, processing aerobic sludge digestion in the cathode chamber had a similar promotion effect as sludge pretreatment [35] and the potential to recover phosphate from sludge [36,37]. Above researches prove that traditional sludge pretreatment methods can accelerate the degradation of sludge but it does not lead to the improvement of MFC's electricity generation performance. Pretreatment facilitates the release of organic substance, sufficient organic substrate that is indispensable for electricity generation process, however, the electron transfer is the rate-limiting step of this process. Ghadage et al. used NaClO as the catholyte to improve cathode electron transfer capacity in a sludge MFC, obtaining a twofold power density compared to the aeration catholyte [38]. Although many strengthening methods have been proposed, the electric energy recovered from sludge MFC today is only a small percentage of the total sludge latent energy. How to enhance electricity generation according to the principle of sustainability requires further study.

Overall, combining sewage sludge treatment and bioelectrochemical processes, Jiang et al. identified MFCs as an enhanced sludge digester, which provides extra hydrolysis and degradation pathways with components such as aliphatic substrate and carbohydrate being hydrolyzed easily in sludge MFC system [39,40].

3.1.2 Biowaste

Biowaste includes FW from households and restaurants, garden waste and dung etc. Organic matter in the biowaste is harmful to the environment but rich in bioenergy. MFCs with HCSW provides a new way to utilize biowaste. Scott and Murano proposed early the use of biowaste in MFCs, and recent studies have practiced all kinds of biowastes in MFCs as substrate [14]. Similar to sludge MFCs, the rate-limiting of hydrolysis–acidification process along with the competition of methanogenesis is the key challenge of biowaste MFC. It was found that with the processing of acidogenesis, there was a change of dominant bacterial community. The *Methanobacterium* genus disappeared gradually, which was beneficial for the robust performance of MFCs [41].

Aiming at improving biowaste degradation efficiency, it is useful to know the fate of the organic matter in MFCs, the fractionation method using XAD-8/XAD-4 resins are thus broadly used in MFCs studies [42]. For FW MFCs, aromatic compounds were degraded first, this result is instructive for choosing appropriate pretreatment or posttreatment methods [43]. It is worth mentioning that in a three-stage MFC reactor, nitrogen removal (60% nitrogen removal rate) of livestock manure and agriculture wastes was observed [44]. By developing particular reactor configuration, dung fed MFCs showed a comparative power density as traditional wastewater MFCs [45,46]. Electrode distance, PEM, substrate buffering capacity and temperature show significant influence on FW electrogenesis performance [47,48]. Using water-based additive (viz., wastewater) to dilute the concentrated waste is another possible strengthening process [49]. Considering the long-term operation of the system, dairy manure fed MFC was able to keep a stable high performance over 110 days [50]. A new procedure of the use of bioanode can lead to the stability and recovery of electricity generation performance of MFC [51].

3.1.3 Biomass

The high growth rate of algae leads to many emerging environmental problems. Algal contains protein, carbohydrate and other organic matter recyclable (algal organic matter, AOM). Algae have been applied as biofuel in hydrogen and methane production. As an emerging renewable energy technology, researchers have put forward and evaluated algae feedstock MFC in the past few years. Kondaveeti et al. used *Scenedesmus obliquus* as the anode substrate in a two chamber MFC, the degradation process released acetate and lactate byproducts, which led to a higher power density compared to similar MFC reactor with different substrate [19]. Lakaniemi et al. comparatively studied the different electrogenesis characteristics of two different microalgae, *Chlorella vulgaris* showed a higher power output while *Dunaliella tertiolecta* had a more sustained power output potential [52].

MFC had a better COD removal rate (82%) of *Microcystis aeruginosa* ultrasound broken solution than anaerobic fermentation (24%) [53]. But for untreated algae sludge from drinking water treatment plant, MFCs could not run efficiently, and alkaline pretreatment might solve this problem [54]. Zhao et al. combined alkaline and acidic fermentation pretreatment for a stable electricity generation performance of cyanobacteria solution [55]. Pretreatment methods played a vital role in the high content solid substrates used in MFCs, but algae and activated sludge contained diversified organism aggregations. The acclimatization and resistance ability of these organisms makes the degradation of algae and the reduction of activated sludge quite difficult for all biotreatment technologies.

3.2 High content solid substrates based MFCs technology (Solid phase MFCs)

MFC fed with high content solid substrate changes the anode phase from liquid to solid or semisolid. According to the characteristics of the solid phase, multiple components or particular environment combined MFC reactors were designed and used as described below (Fig. 5).

3.2.1 Sediment MFCs (SMFC)

Reimers et al. connected marine sediment and seawater with electrodes, wires and resistance constructing the first SMFC [56]. In 2001, their study only obtained a power of 0.01 $W \cdot m^{-2}$. Fifteen years later, MFC power output has been enhanced for more than one hundred times, the stack operation strategy and high performance electrode materials developed lately made the application of MFC one step closer to reality [57,58].Through the adsorption effect, sediment concentrates a variety of pollutants including various organic matters, which is available to the bioanodes. Sediment organic matters (SOM) was promoted to be humified, aromatic, polydispersed and obtained a high average molecular weight in SMFC [59]. Applying SMFC in a petroleum hydrocarbon contaminated sediments increased the natural biodegradation rate by about 12-folds [60]. SMFC could stabilize the anode sediment, but the limitation of electricigens to utilize simple substrate made macromolecular organic compounds residual in the sediments require further studies.

Pretreatment such as autoclaving (30 or 60 min) and heating (150°C, 3h) accelerated the dissolution of dissolved organic matter (DOM), which contributed to the better electricity generation performance, and the molecular size got smaller after pretreatment [61]. However, the underwater position and large quantity of sediments ex situ pretreatment is not required in most cases. In situ addition of appropriate biodegradable substrate in the anode chamber is an easy way to improve the power generation [62,63]. Xia et al. studied the degradation rate of 68 organic matters in SMFCs and summarized that high polarity organic matter was preferentially degraded [64]. This result is a significant guide for researchers to effectively constructed SMFC and a minute quantity of surfactant would be available for purifying the contaminated sediment and overlying water [65].

The membraneless design makes SMFC realize some unique coupling processes compared to the membraned SMFC. Jeon et al. inoculated *Chlorella vulgaris* in the cathode to investigate the relevance between biomass production and SMFCs current generation [66]. Algae consumed CO_2 produced through organic matter degradation in the anode to grow and reproduce. On the other hand, the large algae biomass provided more oxygen to the cathode, enhancing the oxygen reduction reaction (ORR) and the power output of SMFCs. In a similar system, the preponderance of electrode bacterium inhibited the growth of fermentative bacterium and methanogen, sediment bulking and black water agglomerate were prevented successfully when embedded with MFCs [67].

3.2.2 Soil MFCs

Soil is a common inoculating source of many biological treatment technologies; the rich microorganism in soil suggests its potential as MFCs anode substrate [68].

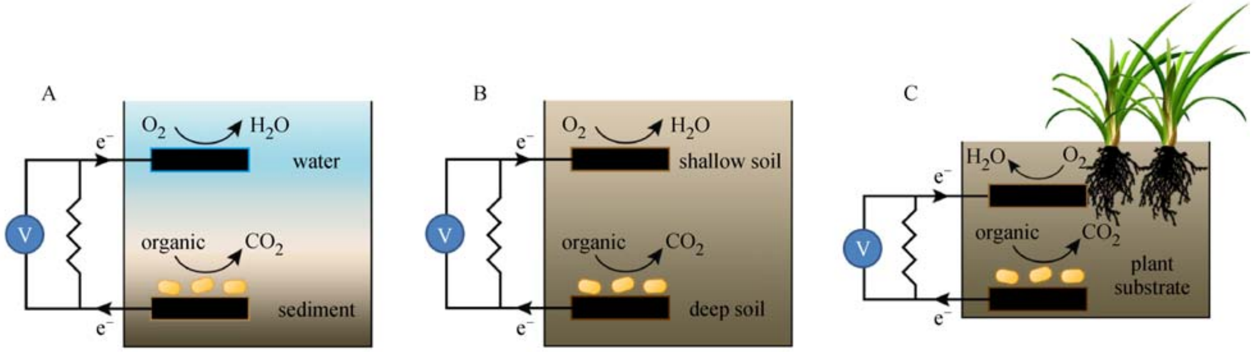

Fig. 5 Schematic illustration of (A) Sediment MFC (B) Soil MFC (C) Wetland and plant MFC

Temperature, soil depth, structure, moisture and salt content are all important factors affecting power generation [69,70]. Especially, proportional mixtures of soil and FW lead to a remarkable voltage output [71]. Because of the seriousness of soil organic pollution, most soil MFC studies are focused on the in situ bioremediation of refractory organic pollutants. In the subsequent section, MFC bioremediation technology will be discussed in detail.

3.2.3 Wetland and plant MFCs

Wetland or plant wastewater treatment processes are widely used, which are considered as green technologies with low impact on the environment. Wetland or plant treatment and MFCs processes are inseparable from microorganism, and the uneven distributing of oxygen concentration along the wetland or plant matrix depth makes MFCs compatible with wetland or plant treatment facility [72]. Furthermore, based on the abundant studies of soil or sediment MFCs, wetland and plant MFCs have become burgeoning treatment technologies.

Zhao et al. conducted a preliminary investigation of constructed wetland microbial fuel cells (CW-MFCs), a continuous, upward flow system was constructed using alum sludge and gravel as the matrix and reed as the wetland plant [73]. In a similar system, they also obtained a high nutrient (ammonium and total phosphorus) and organics removal rate from swine slurry [74]. This result supports MFCs to be a biological nutrient removal (BNR) technology. The subsequent studies tested the influence of various operational factors including electrode separation and flow regime [75]. Distinctively, as a natural treatment combined system, water level variation through evaporation was one main parameter that controlled the voltage output [76]. Diverse plants such as *Oryza sativa*, *Glyceria maxima*, *Spartina anglica* have been well applied in MFCs system, intensifying the solid growing substrate through modifying plant-growth medium and adding organic wastes to facilitate both the growth of plants and MFCs power generation [77–81]. Multiple processes happen in CW or plant MFCs simultaneously. A better understanding of these processes could guide the construction of MFCs. For example, rhizodeposition is the transportation of organic matter to the rhizosphere of the plants, as a result, positioning the anode in the rhizosphere will improve power generation of the system [82]. A latest study also reported that MFC could enhance the survival of a macrophyte (*Potamogeton malaianus*), demonstrating the advantage of plant-MFC combined systems from the angle of phytophysiology [83]. If more mechanisms between plant and MFCs processes are revealed, methods to optimize and strengthen the plant-MFC combined system will be clearer.

3.3 High content solid substrates MFCs coupling technology

3.3.1 Activated sludge process-MFC

Activated sludge process based technology is still the most widely used technology in practice [84]. The high content solid MFCs technology reveals the feasibility of MFC-activated sludge coupling technology, providing the possibility of upgrading and reconstruction of the traditional technology (Fig. 6) [85]. Research has observed the improvement in nitrogen removal and reduction of sludge production [86]. Tomoya converted an AS reactor to cassette-electrode MFC with a scalable configuration which is attractive for future application [87]. Other high concentration sludge based wastewater treatment technologies such as AAO and UASB all get boosted when combined with MFCs [88,89]. Yang et al. found that aerobic granular sludge was able to stabilize in biocathode chamber to treat synthetic and pharmaceutical wastewater [90]. All the above researchers expanded the application of MFC and attested to the possibility of coupling MFC–sludge based treatment technologies.

3.3.2 Composting-MFC

Composting process is widely used to treat organic wastes with the advantage of recovering nutrients for further utilization. Oxygen influences microbial growth and gas emission during composting processes [91]. Based on different substrates and objectives, a compost reactor can be operated under anaerobic and/or aerobic conditions. As a requirement of MFCs in electricity generation process, keeping the anode substrate in anaerobic conditions is appropriate. Most researches focused on combining anaerobic compost with MFC, but Takayuki's study of aerobic compost MFCs showed a higher power output than those under anaerobic conditions (Fig. 7) [92].Oxygen can inhibit the anode reaction but contributes to the degradation of anode substrates. The balance between these two different conditions need further study. In addition, long retention time and poor maturity effect are two main limiting factors [93]. Yu's study also revealed the practicality of applying MFCs to enhance sludge compost maturity, implying the effects of MFCs process to accelerate compost process [94]. Just like conventional compost processes, the performance of MFCs-compost system is highly controlled by operation condition. The impact of carbon/nitrogen ratio and moisture content are the most significant factors [95]. As a result of the inhibitory effect to microorganism, a high level of salinity deteriorated the performance of the compost MFCs fed with fruit and vegetable wastes [13]. Despite the promotion effect of MFCs, the slow hydrolysis process of complex

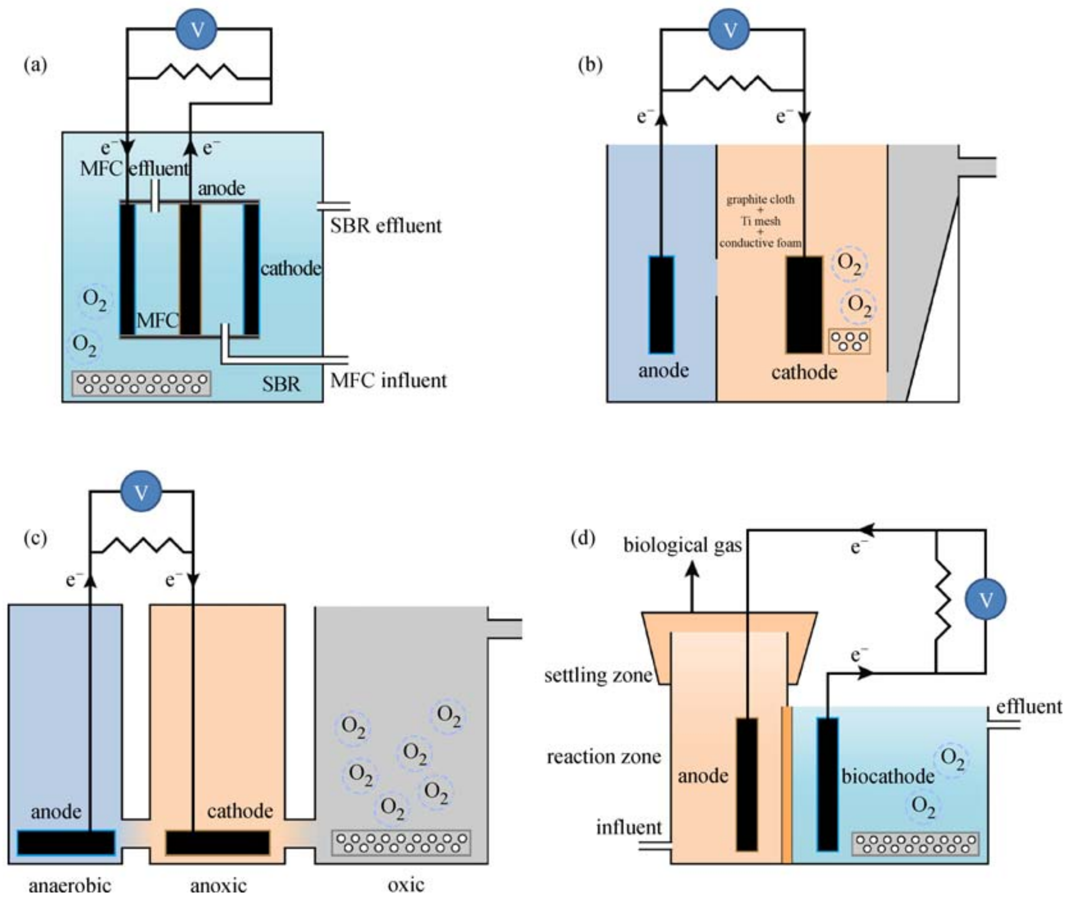

Fig. 6 Schematic illustration of MFC with activated sludge process: (a) Sequencing Batch Reactor; (b) anaerobic-anoxic-oxic wastewater treatment process; (c) activated sludge wastewater treatment processes to improve nitrogen removal and reduce sludge production; (d) continue flow microbial fuel cell system

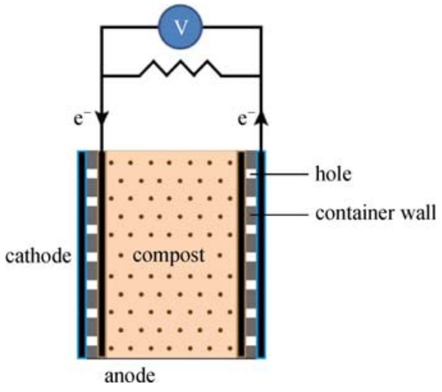

Fig. 7 Schematic illustration a composting MFC with blower

organic compounds limits the performance of MFCs. However, through the addition of acetate in a garden compost MFCs, Parot et al. got a high current of 545 $mA \cdot m^{-2}$ [96]. Other methods such as the addition of bio-enzymes had effects to reduce the internal resistance and enhance the electricity generation performance [97].

3.4 Novel applications of high content solid substrates MFCs

Many researches have proven that MFC system is favorable for HCSWs based treatment technology, and this concept of MFCs can be applied in diversified domains of environment technology.

3.4.1 Bioremediation

The discussion on sediment (and soil) MFCs indicates that sediment (and soil) organic matter is degraded in the anode domain and some specific pollutants can be effectively removed, showing MFCs have the potential to act as a novel sediment or soil bioremediation technology (Fig. 8). Especially, as a remediation technology MFCs show many potential benefits, such as accelerated decontamination, self-sustained operation, and environmental friendliness [98,99]. Recent studies have demonstrated its feasibility in the remediation of refractory organics and heavy metal pollution. Mohan et al. placed petroleum sludge in MFCs anode chamber to achieve efficient organic contaminants

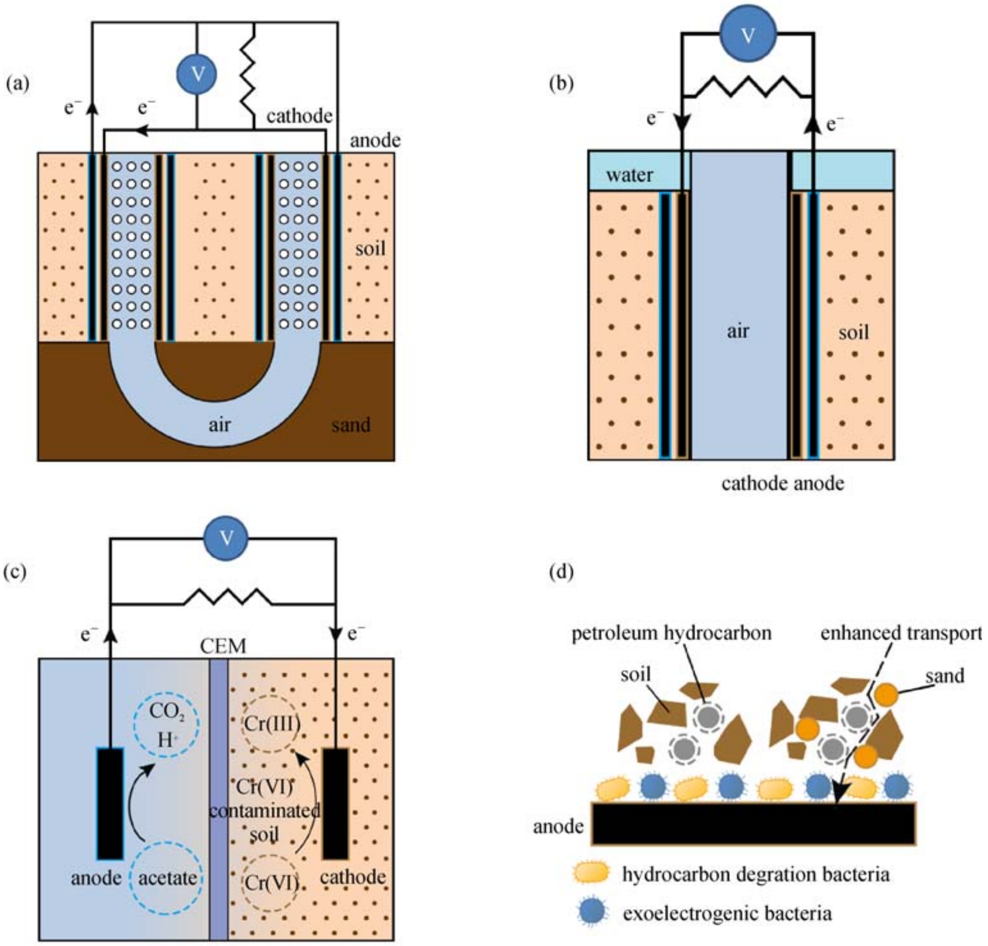

Fig. 8 MFC for bioremediation: (a) U-tube MFC; (b) soil MFC; (c) remediation of Chromium (VI)-contaminated soils; (d) remediation of petroleum hydrocarbon contaminated soil

removal, and Sherafatmand et al. used sediment MFCs to treat three kinds of PAHs including naphthalene, acenaphthene and phenanthrene showing the high resistant activity to PAHs of MFCs [100–101]. Other organic pollutants such as phenol and pesticide have proven to be degraded efficiently in MFCs [102–103]. Furthermore, applying MFCs to remediate heavy metal such as chromium-contaminated soils is realizable [104]. Ryu et al. also reported that Cr(VI)-reducing bacteria could enrich on the anode with Cr(VI)-containing sludge [105]. The high internal resistance and complicated soil/sediment content are the two limiting factors of MFCs bioremediation systems. Li et al. added sand to soil, with the increasing of porosity and the decreasing of ohmic resistance, both the contaminant removal rate and power output were enhanced [106]. Also, aiming at in situ remediation of petroleum hydrocarbon-contaminated soil, Zhang et al. designed a horizontally arranged anodes which had a high remediation efficiency [107]. Another study considered the application of plant MFC for the remediation of heavy metal contaminated water in soils. The Cr(VI) removal efficiency reached 99% under various conditions. Only a small amount of soluble Cr(III) remained and that most Cr(III) precipitated in the form of the $Cr(OH)_3(s)$ or was adsorbed onto the electrodes [108].

3.4.2 Biosensoring

Owing to the complexity of HCSW, some reaction processes are difficult to detect using traditional methods and through the current intensity of MFCs, multiple biochemical indexes and toxic substance concentration can be determined as a result of the different biological activity of EM [109]. This technology is a branch of biosensor where transducers are not required. MFCs biosensor has an expansive development prospect to act as a disposable and portable device [110]. Especially, high solid content MFCs make it possible for monitoring the state of sludge in real time. As the substrate degradation is positively related to electricity generation, Dena et al. evaluated the active sludge and metabolic pathway activity using MFCs [111].

In addition, MFCs can serve multiple functions including gas flow meter and pH meter in a anaerobic fermentation reactor as a result of the good linear correlation between MFCs potential and proton concentration, gas flow rate and gas volume [112].

4 Prospect and challenges

Organic matter in HCSW usually exists in the insoluble and large particulate forms, whereas microbes in MFCs tend to utilize soluble and biodegradable organic matter (e. g., proteins and carbohydrates) as substrates. Hence, providing appropriate substrates for EM is one of the challenges of MFC, which also regulates the performance of electricity generation. To date, MFCs with HCSW as substrates possess a relatively higher internal resistance and lower power density compared to those with pure compounds substrates (e.g., glucose and acetate).

The challenges of MFCs with HCSW as substrates can be summarized as follows: (1) low coulombic efficiency seriously restricts the energy recovery of HCSW substrate [113]; (2) MFCs have an accelerated effect on the degradation of organic matter in HCSW compared to the natural biodegradation process, but the electricity generation performance is severely restricted by the low mass transfer rate in solids [114]; (3) relatively slow HCSW biodegradation rate; and (4) high mass transfer resistance and internal resistance.

The performance of MFC's fueled by HCSW needs improvement in reactor design and optimum operation parameters before remarkable progress can be made. One major measure to improve the performance of MFCs fueled by HCSW is the improvement in EM's function and efficiency during electricity generation. Isolation of bacterial strains with strong HCSW degradation ability and efficient electricity generation is in high demand. Metagenomics is another kind of technology that can be used to enhance MFCs' energy-conversion efficiency and performance. Another major measure to improve the performance is pretreatment prior to using HCSW as fuel. Current researches have attempted ultrasonication, alkalination, and microwave to pretreat sewage sludge so as to decompose the insoluble and large particulate organics, and they are effective in improving the performance of MFCs [39,115]. In the future, methods such as ozonation, enzymatic hydrolysis, and other advanced oxidation pathways that have been explored in conventional treatment, should be further investigated [116–121]. Improvement of mass transfer at the anode may be another pathway to enhance the performance of MFCs fueled by HCSW. The HCSW generally have high mixed liquor suspended solids, which could restrain the mass transfer at the anode. The enhancement of mixing conditions in the anode chamber contributes to high MFC performance.

Besides, it can also be applicable to MFCs powered by HCSW through other pathways (viz., electrode modification, configuration improvement, operation optimization, nutrient additive), which have been investigated in MFCs fed with other substrates. Researches in the future are needed to explain the behaviors of toxic substances in MFCs and the subsequent treatment of the used HCSW. Furthermore, HCSW can be practiced in other electrochemical technologies (e.g. microbial electrolysis cell) in the future, which promotes the multiple utilization of HCSW [122,123].

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