Chemosphere 140 (2015) 12–17

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Electron acceptors for energy generation in microbial fuel cells fed with wastewaters: A mini-review

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HIGHLIGHTS

- This review summarized various electron acceptors adopted in microbial fuel cells.
- Merits and drawbacks of various electron acceptors were identified.
- Possible future research directions were discussed particularly from cathode aspect.

ARTICLE INFO

Article history:
Received 20 January 2014
Received in revised form 20 August 2014
Accepted 27 March 2015
Available online 21 April 2015

Keywords:
Cathode
Electron acceptor
Microbial fuel cell (MFC)
Wastewater

ABSTRACT

Microbial fuel cells (MFCs) have gained tremendous global interest over the last decades as a device that uses bacteria to oxidize organic and inorganic matters in the anode with bioelectricity generation and even for purpose of bioremediation. However, this prospective technology has not yet been carried out in field in particular because of its low power yields and target compounds removal which can be largely influenced by electron acceptors contributing to overcome the potential losses existing on the cathode. This mini review summarizes various electron acceptors used in recent years in the categories of inorganic and organic compounds, identifies their merits and drawbacks, and compares their influences on performance of MFCs, as well as briefly discusses possible future research directions particularly from cathode aspect.

1. Introduction

Microbial fuel cells (MFCs), the device that uses bacteria as the catalysts to oxidize organic and inorganic matter with current generation, has obtained tremendous global notice over the last decades (Logan et al., 2006). MFC has a number of attractive characteristics such as direct electricity generation, high efficiency, and operation in ambient temperatures (Ren et al., 2012). Particularly, MFC can accomplish wastewater treatment simultaneously as well as reduce the amount of sludge production (Pant et al., 2010b).

The electron acceptor contributes to overcome the potential losses existing on the cathode, thus it is one of the major factors influencing power generation in MFCs. The conditions of being a good electron acceptor comprise possessing high redox potential, presenting fast kinetics, being economically valuable, and preferably have sustainability and easy availability (Lu and Li, 2012). Oxygen is one of most promising electron acceptors in MFCs (Logan et al., 2006). However, with the rapid progress of MFC technology as well as a better understanding of its principle, there is a broad awareness that the cathode process is far more than just oxygen reduction reaction (ORR). Various alternative electron acceptors, such as nitrate (NO_3^-), metal ions, perchlorate, nitrobenzene, and azo dyes, have been intensively explored to achieve bioremediation in MFCs (Liu et al., 2014).

So far, several reviews have focused on the cathode processes in MFCs especially in terms of fundamentals and application (He and Angenent, 2006; Harnisch and Schroeder, 2010; Lu and Li, 2012; Liu et al., 2014). He and Angenent (2006) for the first time addressed the development and experimental progress of biocathodes in MFCs. Harnisch and Schroeder (2010) presented the first primary comparative analysis on ORR in the cathode of MFCs. More recently, Liu et al. (2014) provided a panoramic picture of various cathodic catalysts applied in MFCs and gave an insight into their catalytic characteristics, mechanisms and limitations. However, a comprehensive review on the various electron acceptors which have been used in MFCs is still lacking.

Therefore, this mini review aimed at summarizing various electron acceptors used in recent years in the categories of inorganic and organic compounds. Moreover, the merits and drawbacks of

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http://dx.doi.org/10.1016/j.chemosphere.2015.03.059
0045-6535/

different electron acceptors were identified and their influences on performance of MFCs were also compared. In addition, possible future research directions were briefly discussed in this mini review particularly from cathode aspect.

2. Fundamentals of MFCs

The operating principle of a MFC is illustrated in Fig. 1, for which a detailed description can be found elsewhere (Rozendal et al., 2008). In brief, electrons are produced by oxidizing substrates via microbial metabolism in anodic chamber of MFCs, and then transmitted to the cathode through external circuit for power generation or for further cathode applications. The overpotentials of the electrodes in an MFC can roughly be categorized as activation losses, concentration losses and bacterial metabolic losses (Logan et al., 2006). A lot of efforts have been directed to reduce these losses through decreasing electrode spacing, increasing electrode surface area and/or solution conductivity, using metal catalysts and establishment of an enriched biofilm on the electrodes (Logan et al., 2006).

Particularly, it is worthwhile to mention that biocathodes with microorganisms as catalyst have been demonstrated as one of the promising approaches to reduce activation losses occurring in the cathode of MFCs (He and Angenent, 2006). Although the biochemical mechanisms involved in microbial electron uptake from a cathode are still very unclear and thus need a further investigation, several possible extracellular electron transfer paths have been suggested, including (a) direct electron transfer using active centre of the membrane enzyme in microbe, such as cytochromes; (b) direct electron transfer via biological and fibrous protein structure nanowires; (c) mediated electron transfer using self-secreted shuttles; (d) long-distance electron transfer via a conductive biofilm matrix occupying "nanowire grid" and cytochromes related to matrix (Rosenbaum et al., 2011).

3. Inorganic electron acceptors

3.1. Oxygen

So far, the most sustainable and suitable electron acceptor known for MFCs is oxygen, because of its availability in the environment, low cost and high redox potential (Freguia et al., 2007). In order to increase the oxygen reduction kinetics and reduce cathodic activation overpotential, different kinds of catalysts have been used in the cathode (Erable et al., 2012). Platinum offers the highest catalytic performance with increased oxygen affinity and reduced activation loss, and is the most commonly used catalyst for ORR (Lu and Li, 2012). Logan et al. (2005) demonstrated that Pt-based MFC could achieve 5-fold increase in power output compared to the MFC with a plain carbon cathode. For cheaper and more sustainable, other catalysts such as lead dioxide (Morris et al., 2007), Fe/Fe_2O_3 (Zhuang et al., 2010), cobalt (Lefebvre et al., 2009), manganese dioxide (Lu and Li, 2012) or even activated charcoal (Pant et al., 2010a) have also been explored for oxygen reduction reactions at the cathode of MFCs.

Biocathodes with enzymes or microorganisms have been intensively applied for ORR in MFCs (He and Angenent, 2006). Some redox enzymes, mainly laccases and bilirubin oxidases, were applied to catalyze ORR (He and Angenent, 2006). Compared with the controlled Pt-based MFCs, the MFC with laccase generated 10-fold increase of the maximum power density (Schaetzle et al., 2009). However, enzymes have various drawbacks as the cathode catalyst for ORR, such as being sensitive to toxicants, full of complications to be immobilized on electrode surfaces, and short-life time (Erable et al., 2012). Microorganisms have been the most popular choice of biocatalyst for ORR in MFCs, due to many advantages compared to chemical and enzymatic catalysts (Lu and Li, 2012). Clauwaert et al. (2007) for the first time developed a biocathode with mixed microorganisms as biocatalysts for oxygen reduction in MFCs. Following this, mixed cultures were inoculated with various inoculums as the biocathodes for ORR in MFCs (Freguia et al., 2007; Rabaey et al., 2008). Although several pure strains were also adopted to be catalytics for ORR, the results have suggested that the MFC performance in terms of current density and power output was unable to reach to the similar levels demonstrated by the mixed population (Rabaey et al., 2008).

3.2. Nitrogen-containing compounds

Due to NO_3^- owing competitive redox potential to oxygen, denitrification process has drawn considerable attentions in MFCs with both electricity generation and wastewater treatment (Clauwaert

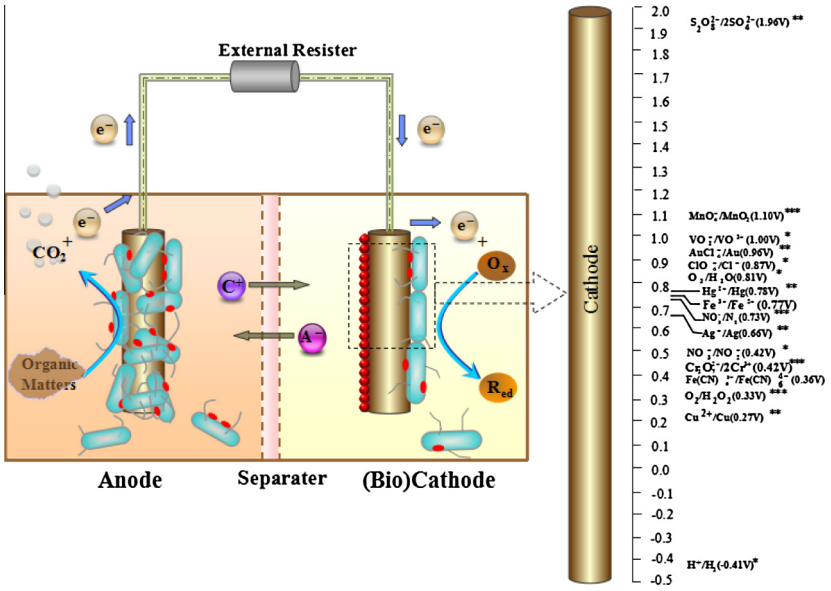

Fig. 1. Fundamental configuration of a MFC with the redox potential vs. SHE (Standard Hydrogen Electrode) of various electron acceptors (based on information from Lu and Li (2012) and He and Angenent (2006); *at pH = 7; **the molarity of ions were 5 mM; ***the molarity of ions were 5 mM at pH = 7).

et al., 2007). In an abiotic cathode, Fang et al. (2011) demonstrated that NO_3^- removal rate and maximum power density of MFCs with Pt catalyst were 3.5 and 16 times higher than those without catalyst. On the other hand, biocathodes using microorganisms have been demonstrated to be a promising method for denitrification in MFCs. Clauwaert et al. (2007) firstly introduced a biocathode for denitrification process, where the MFC was able to remove up to 80 g-N m^{-3} d^{-1} and the highest power output was 4 W m^{-3}. Later, Virdis et al. (2008) developed a novel process configuration that achieved both carbon and nitrogen removal using MFCs. Removal rates up to 220 g-N m^{-3} d^{-1} were continuously achieved in the biocathode, while the MFC produced a maximum power output of 0.37 W m^{-3}. Additionally, the use of autotrophic denitrification in MFCs for waters with low ionic strengths (<1600 μS cm^{-1}) has been considered by Puig et al. (2012). In these studies, biocathodes were developed by seeding with different types of sludge and sediments as inoculums, and the microbial community included mainly denitrifying bacteria (Virdis et al., 2008, 2010; Puig et al., 2011). By employed a pure culture of *Bacillus subtilis*, electricity generation with simultaneous NO_3^- reduction in a MFC was demonstrated and a power density of 0.019 W m^{-2} was achieved (Nimje et al., 2012).

Besides NO_3^-, it has been proved that NO_2^- could also be removed as the electron acceptor in MFCs through autotrophic denitrification (Virdis et al., 2008). Puig et al. (2011) observed that the nitrogen removal rate was up to 135 ± 5 g-N m^{-3} d^{-1} and power output of 0.045 W m^{-2} when NO_2^- was fed into the biocathode of MFCs. Zhang and Angelidaki (2012) presented a new concept for in situ NO_2^- removal from eutrophic lakes by means of biocathode-based sediment MFC technology. Power density of 0.036 W m^{-2} was produced from NO_2^- synthesized lake waters, while 77% of total nitrogen removal was accomplished in this MFC.

Mitigation of N_2O emissions is necessary since it is a strong greenhouse gas with a global warming potential of about 300 times CO_2 (Desloover et al., 2011). In the abiotic or biotic cathode of MFCs, N_2O was able to be reduced into N_2 with energy generation (Desloover et al., 2011). Without the microorganism catalyst, the nitrogen removal rate was 20 g-N m^{-3} d^{-1} in 24 h with current density of 0.047 A m^{-2}. The higher N_2O removal rate ranged between 377 and 907 g-N m^{-3} d^{-1} with the maximum current density about 1.57 A m^{-2} was obtained by using an autotrophic denitrifying biocathode in MFCs.

3.3. Metal-containing ions

Due to high redox potential, convenient operation and fast kinetics, ferricyanide was a popular choice as an electron acceptor for early MFC investigations (Rabaey et al., 2004). However, ferricyanide must be chemically regenerated and thus its use is very limited in practice. Permanganate has been selected as an alternative in MFCs. By using permanganate in an MFC reactor, You et al. (2006) showed that permanganate exhibited better performances in power generation than ferricyanide, and this result was further proved by Pandit et al. (2011). Additionally, ferric iron was also used as the electron acceptor to improve cathode performance, where a maximum power density of 0.86 W m^{-2} was achieved (Ter Heijne et al., 2006).

In recent years, the intensive studies of recovery or removal of heavy and noble metals were not only because of extreme toxicity to environment, but even more on account of their rarity. Using metal-containing ions as electron acceptors, MFC can not only generate electricity but also remove or recover them from wastewaters, as summarized in Table 1. Li et al. (2008) employed a MFC with graphite paper cathode for Cr(VI) reduction, where 99.5% of Cr(VI) was removed after 25 h treatment with a maximum power density of 1.60 W m^{-2}. A novel rutile-coated graphite cathode was designed to accomplish cathodic reduction of Cr(VI) with or without visible light irradiation (Li et al., 2009). Moreover, biocathodes have shown potential application in the reduction of Cr(VI) to form insoluble Cr(III) precipitates in MFCs. For instance, Tandukar et al. (2009) operated a biocathode-based MFC to achieve a maximum Cr(VI) reduction rate of 0.46 mg Cr(VI) g-VSS^{-1} h^{-1} and a power density of 0.056 W m^{-2}. By setting a biocathode potential at −300 mV vs SHE, the reduction of Cr(VI) was improved to 19.7 g m^{-3} d^{-1} and the maximum power density was increased to 0.034 W m^{-2} compared to the control without set potential (14.0 g m^{-3} d^{-1} and 0.022 W m^{-2}) (Huang et al., 2011). Recently, Xafenias et al. (2013) used *Shewanella oneidensis* MR-1 as biocatalyst in the cathode and the Cr(VI) removal rate of 54 g m^{-3} d^{-1} was obtained with a maximum power density of 1.72 W m^{-2}. Ter Heijne et al. (2010) studied the recovery of copper in a MFC and the maximum power density was up to 0.43 W m^{-2} with higher than 99.98% of copper removal efficiency. Through X-ray diffraction (XRD) analysis, Tao et al. (2011) indicated that Cu^{2+} was reduced to Cu_2O or Cu_2O plus Cu depositing on the cathode of

Table 1
Metal-containing electron acceptors in MFCs.

Electron acceptors	Cathode/catalyst	Anode substrate	Current density (A m^{-2})[a]	MPD (W m^{-2})[a]	Reference
$Fe(CN)_6^{3-}$	Graphite	Glucose	61.8	4.31	Rabaey et al. (2004)
$Fe(CN)_6^{3-}$	Carbon paper	Acetate	N.A.	190	Oh and Logan (2006)
MnO_4^-	Carbon cloth	Glucose	5.9	3.99	You et al. (2006)
MnO_4^-	Graphite block	LB agar	N.A.	0.12	Pandit et al. (2011)
Fe^{3+}	Graphite felt	Acetate	4.5	0.86	Ter Heijne et al. (2006)
$Cr_2O_7^{2-}$	Graphite block	LB agar	N.A.	0.046	Pandit et al. (2011)
$Cr_2O_7^{2-}$	Carbon fiber	Glucose	2.5	1.03	Zhang et al. (2012a)
$Cr_2O_7^{2-}$	Graphite paper	Acetate	0.40	1.60	Li et al. (2008)
$Cr_2O_7^{2-}$	Graphite plate/biofilm	Acetate	0.12	0.056	Tandukar et al. (2009)
$Cr_2O_7^{2-}$	Granular graphite/biofilm	Acetate	0.11	0.034	Huang et al. (2011)
$Cr_2O_7^{2-}$	Graphite felt/biofilm	Acetate	0.033	1.72	Xafenias et al. (2013)
Cu^{2+}	Graphite plate	Acetate	0.74	0.2	Zhang et al. (2012b)
Cu^{2+}	Graphite plate	Glucose	0.42	0.13	Tao et al. (2011)
Cu^{2+}	Graphite foil	Acetate	1.7	0.43	Ter Heijne et al. (2010)
VO_3^-	Carbon fiber	Glucose	1.1	0.57	Zhang et al. (2009)
SeO_3^{2-}	Carbon cloth	Acetate	N.A.	2.90	Catal et al. (2009)
Hg^+	Carbon paper	Acetate	1.44	0.43	Wang et al. (2011b)
$AuCl_4^-$	Carbon cloth/titanium	Acetate	10.16	9.08	Choi and Hu (2013)
Ag^+	Carbon cloth/titanium	Acetate	5.67	4.25	Choi and Cui (2012)

Note: MPD is maximum power density.
[a] Normalized by the projected surface area of cathode.

MFCs. Zhang et al. (2012b) also obtained the same reduction pathways using $Cu(NH_3)_4^{2+}$ as the electron acceptor in MFCs.

Besides above mentioned cases, many other metals distributed in wastewater have the potentials to be acting as the electron acceptors for both electricity generation and their removal or recovery in MFCs, as shown in Table 1. Zhang et al. (2009) demonstrated that the MFC could reduce 25.3% of V(V) in 72 h with a maximum power density of 0.57 W m^{-2}. About 99% of 0.2 g-Se L^{-1} selenite was reduced at the cathode in 72 h for MFCs and the power density was higher than 1.00 W m^{-2} (Catal et al., 2009). Wang et al. (2011b) reported that Hg^+ removal efficiency of 98.22–99.54% was achieved in MFCs with a maximum power density of 0.43 W m^{-2}. Choi and Hu (2013) operated a cubic MFC containing $AuCl_4^-$ catholyte to accomplish 99.89% recovery of Au and meanwhile acquire the maximum power density of 9.08 W m^{-2}. The same research group successfully reduced Ag^+ into Ag in a MFC with the recovery efficiency as high as 99% and a maximum power density of 4.25 W m^{-2} (Choi and Cui, 2012).

3.4. Others

Perchlorate is known to be an emerging drinking water contaminant, and dissimilatory perchlorate-reducing bacteria have been demonstrated to reduce perchlorate to chloride in MFCs (Butler et al., 2010). Pandit et al. (2011) evaluated persulfate ($S_2O_8^{2-}$) with three other acceptors and the results shown that the maximum power density of $S_2O_8^{2-}$ was higher than that of dichromate and ferricyanide but slightly lower than that of potassium permanganate. An MFC with triiodide as the electron acceptor was carried out and the power density of MFCs was significantly higher compared to ferricyanide (Fu et al., 2010).

4. Organic electron acceptors

4.1. Azo dyes

Recently, MFCs have shown the great potential for treating wastewater containing azo dyes for decolorization and simultaneous production of electricity (Solis et al., 2012; Solanki et al., 2013). While some studies have investigated azo dye decolorization through co-mechanism in the anode or electro-Fenton process in the cathode (Feng et al., 2010; Hou et al., 2012), this review is majorly focusing on azo dye as the electron acceptor for decolorization in the cathode of MFCs.

Using Acid Orange 7 as a model azo dye, Mu et al. (2009a) investigated its decolorization as the electron acceptor in MFCs. Effective decolorization of Acid Orange 7 at rates up to 501 ± 6 g m^{-3} d^{-1} was achieved at the cathode with concomitant energy recovery. Liu et al. (2009) found that different types of azo dyes including Methyl Orange, Orange I, and Acid Orange 7 could also be successfully degraded in the cathode of dual-chamber MFCs. In order to reduce the high internal resistance of the traditional dual-chamber MFC, Kong et al. (2013) modified the MFC configuration to be sleeve-type with an inner anode chamber and an outer cathode chamber to decolorize Acid Orange 7. This sleeve-type MFC with large area and small distance between anode and cathode could lead to a lower internal resistance and therefore a higher decolorization performance.

Electrode modification is also a pathway to improve azo dye degradation in MFCs. Ding et al. (2010) observed that replacement of the graphite electrode with a rutile-coated graphite electrode results in an increase in the decolorization efficiency of Methyl Orange from 37.8% to 47.4%. In addition, the output current density increases more significantly when the rutile-coated cathode is irradiated by visible light. Liu et al. (2011) modified the cathode electrode with redox mediator thionine and anthraquinone-2, 6-disulfonate to enhance decolorization in MFCs. Consequently, the decolorization rate of Methyl Orange increased by over 20% and the power density was enhanced by over three times.

Enzyme laccase has been used as the biocatalyst in the cathode of MFCs for enhanced azo dyes decolorization and simultaneous electricity generation. Suspended laccase could catalyze the decolorization of reactive blue 221 in the cathode and maximum power density of the MFC increased about 30% (Bakhshian et al., 2011). Furthermore, the same research group immobilized laccase on the surface of a methylene blue-modified graphite electrode in the cathode compartment to decolorize the azo dye (Savizi et al., 2012). With this modification on the cathode, the power density of MFCs had a 65% improvement with a 74% increase in decolorization efficiency compared to the simple graphite cathode.

4.2. Nitrogenous aromatic compounds

Nitrogenous aromatic compounds are priority pollutants listed by many countries due to their mutagenicity and recalcitrance. Mu et al. (2009b) firstly investigated the use of an MFC to remove nitrobenzene in an abiotic cathode and found that effective removal of nitrobenzene at rates up to 4.92 g m^{-3} d^{-1} was achieved with concomitant energy recovery. Using a Pt-based cathode, Li et al. (2010) obtained an average nitrobenzene removal rate of 1751 g m^{-3} d^{-1} and the maximum power density reached 0.069 W m^{-2} in MFCs. However, the reductive intermediates nitrosobenzene and phenylhydroxylamine could be accumulated in the abiotic cathode during nitrobenzene reduction especially at high current densities (Mu et al., 2009b; Li et al., 2010). Nitrosoaromatics are more toxic than parent nitroaromatics in many instances, and can chemically condense with hydroxylamino aromatics to produce azo compounds which are difficult to mineralize (Wang et al., 2011a). In order to solve this problem, Wang et al. (2011a) successfully developed a microbially catalyzed cathode for selective transformation of nitrobenzene to aniline in MFCs. Additionally, the effect of nitro-substituent on mononitrophenol reduction in MFCs was also explored and the reduction of the three mononitrophenols followed in the order of *o*-nitrophenol > *m*-nitrophenol > *p*-nitrophenol (Shen et al., 2013).

4.3. Chlorophenols

Chlorophenols are commonly found in ground waters, sediment and surface soils, industrial wastewater effluents and treatment lagoons. It has been shown that pentachlorophenol can be reductively degraded in the biocathode of MFCs (Huang et al., 2012a,b). By using a mediator-less biocathode, the pentachlorophenol could be reductively dechlorinated at a rate of 6.3 ± 1.2 g m^{-3} d^{-1} coupled with power generation of 0.083 W m^{-2}. Both pentachlorophenol degradation rate and power production were enhanced to 12.6 ± 1.9 g m^{-3} d^{-1} and 0.173 W m^{-2} respectively when operating the MFC at 50 °C. Recently, 4-chlorophenol was also successfully adopted as the electron acceptor in an abiotically cathode of MFCs for dechlorination and simultaneously electricity generation with the maximum power output of 0.033 W m^{-2} (Wen et al., 2013).

5. Conclusions and perspectives

Electron acceptor is one of the indispensable and highly important components in MFCs. This mini review summarizes the various inorganic and organic electron acceptors that have been used in MFCs for not only bio-energy generation but also bioremediation in the cathode. Only from electricity generation point of view, oxygen will still be a promising electron acceptor in MFCs

especially for the scaling up due to its sustainability, easy availability and cost-effective.

Besides for electricity generation, MFCs have shown the great bioremediation potentials for the removal or recovery of various inorganic and organic pollutants from wastewater in the cathode. Therefore, in the near future it is reasonable to further extend this novel bioremediation technology to treat various important waste streams such as acid mine drainage from mining industry (Johnson and Hallberg, 2005). In addition, most of studies so far only investigated the conversion of single electron acceptor in the cathode of MFCs, the interaction and competition between different electron acceptors in the cathode should be further evaluated since most of wastewaters have a very complex composition. On the other hand, although biocathode with microorganism has shown its promise, fewer researchers focused on this area for treating organic pollutants and metals probably because of their toxicity to the microbes (Wang et al., 2011a). Thus development of suitable biocathode with microorganism for treating such kinds of pollutants might be another research area in the future. Most importantly, the deep understanding of electron transfer mechanisms between electrode, microbe and even electron acceptors in the cathode is necessary for the optimization of functional biocathodes. Moreover, no matter what kind of electron acceptors are used in the cathode, we believe a comprehensive life cycle assessment of MFCs should be conducted for energy generation or bioremediation in order to upscale this technology to convince the end-users of its potential (Pant et al., 2011).

Although MFCs were intensively investigated over the past decade, so far very limited success in the practical application has been reported due to the limitations and challenges of MFCs. According to our analysis, there are several promising niches where MFCs might be heading toward practical application. First, MFCs can be adopted as power supplies for low-power devices used especially in remote, rural and environmentally unfriendly areas where it is difficult to replace batteries of the system (Tender et al., 2008). Second, by virtue of the signal variation of electric current as exoelectrogen is sensitive to the dynamic changes of substrate degradation or/and microbial activities, it is highly possible to establish a MFC-based miniaturized biosensor for monitoring of various biodegradation processes in the wastewater treatment plant (Li and Yu, 2014). Lastly, rather than electricity generation, the MFC can be hopefully used as a wastewater refinery to produce or recover high valuable chemicals such as biofuels and hydrogen gas (Logan and Rabaey, 2012).

Acknowledgements

The authors wish to thank the Natural Science Foundation of China (51222812), the Recruitment Program of Global Experts, the Program for Changjiang Scholars and Innovative Research Team in University of the Ministry of Education of China, and the Jiangsu Key Laboratory of Chemical Pollution Control and Resources Reuse in NJUST (Grant No. 30920140122008) for the financial support of this study.

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