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MINI-REVIEW

Perspective of harnessing energy from landfill leachate via microbial fuel cells: novel biofuels and electrogenic physiologies

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Abstract Organic carbon, nitrogen, and sulfur are highly concentrated in municipal solid waste (MSW) landfill leachate, which usually frustrates conventional leachate treatment technologies from the perspective of energy costs. Therefore, the possibility of converting leachate to a new energy source via microbial fuel cell (MFC) technology has been examined recently. This paper summarizes the power output and energy recovery efficiency of the leachate-fed MFCs according to different feeding patterns, cell structures, and loading rates. Also, we assess potential energy-generating chemicals in leachate like nitrogen and sulfur compounds and propose alternative pathways, which may lift strict ratios between organic carbon and nitrogen content in conventional denitrification of leachate and are expected to achieve a higher voltage than traditional organic-oxygen based cells. Although currently power output of leachate-fed MFCs is limited, it seems well possible that dynamic characteristics of MSW leachates and microbial physiologies underlying some bio-electrochemically efficient activities (e.g., direct interspecies electron transfer) could be stimulated in MFC systems to improve the present status.

Keywords Landfill leachate · MFC · Energy recovery · Direct interspecies electron transfer

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Introduction

Landfill leachate is the percolate of excessive rainwater and moisture of wastes. Although its quantity is influenced by precipitation, most (>70 %) of the liquid derives from the degradation of organics and the release of moisture (Sao Mateus Mdo et al. 2012; Zhang et al. 2010). Organic substances constitute around 60 % of municipal solid waste (MSW) in landfills and have a moisture content of 40 % or more, which means that copious amounts of leachate are generated (Zakir Hossain et al. 2014). Although composting and incineration of solid waste are preferred (EC 1999), landfill sites still receive the largest amount of MSW worldwide (350 million tons) regardless of countries' development levels (Hoornweg and Bhada-Tata 2012). It is estimated that, depending on the climate, the volume of leachate generated over the lifespan of a landfill is equivalent to 15 to 50 % of the total volume of MSWs deposited (Canziani and Cossu 1989). The concentrations of typical contaminants in MSWs landfill leachate such as biodegradable organic matter, inorganic macro-components (e.g., hydrogen sulfide, ammonia), and

xenobiotic organic compounds (XOCs) are 100 times higher than in domestic wastewater (Kjeldsen et al. 2002; Koshy et al. 2007).

Considering its high generation rates and strength, many technologies have been applied to leachate treatment including advanced oxidation processes (Deng and Englehardt 2006), membrane separation (Chan et al. 2007), passive aeration in bio-filters (Xie et al. 2010), and anaerobic digestion (Zairi et al. 2013). All these technologies, except for anaerobic digestion, consume energy (Fig. 1). Microbial fuel cell (MFC) technology is thought to be a promising treatment alternative to reduce contaminants and simultaneously recover energy from MSW landfill leachate (Ganesh and Jambeck 2013). Although MFC technology has been frequently reviewed for domestic and industrial wastewater treatment (Gil-Carrera et al. 2013a; Kim et al. 2010), its consideration towards MSW landfill leachate treatment is rarely reported. Hence, this review summarizes studies pertinent to the interface between landfill leachate characteristics and MFC technologies including energy potentials of chemicals in leachate, key factors in system efficiency enhancement, and functional microbial communities. We attempt to inspire potential researchers to explore more energy harnessing pathways from MSW leachates to convert MSW landfilling to a greener technology.

Characteristics and energy potential of MSW landfill leachates

Characteristics and composition of MSW landfill leachate

Landfills are very dynamic systems, and leachate characteristics vary as a function of landfilling procedure. MSW leachate usually falls into three categories, fresh, intermediate, and mature, according to the composition of the landfills and the degradation stages of the waste (Fig. 2). In detail, leachates drained from hydrolysis and fermentation stages are considered fresh and intermediate, respectively; organics present include volatile fatty acids (VFAs), aromatic hydrocarbons, phenols, and chlorinated aliphatic (Kjeldsen et al. 2002). Furthermore, 85 % of the organics are present as dissolved organic matter (DOM), while high molecular weight compounds account for only ~1.3 % (He et al. 2006). This readily biodegradable leachate is generated over a period of 3 to 5 years, which is relatively short compared to the overall life time of the landfill (typically 30 to 50 years). Field studies show that this organic mixture exhibits a chemical oxygen demand (COD) in the range of 4,000 to 40,000 mg/L (Tchobanoglous et al. 1993); 60~70 % of this COD is biodegradable and can be further converted into short chain VFAs in the end of the first two stages (Fig. 2). Therefore, microbiological methods can be productively applied to the treatment of fresh and intermediate leachates (Fig. 1).

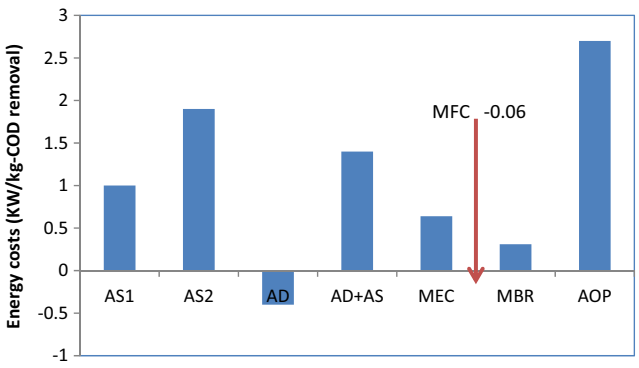

Fig. 1 Energy costs of different treatment technologies, including aerobic systems (*AS1* and *AS2* denote low and conventional aeration rates, respectively), anaerobic digestion (*AD*), and hybrid process (*AD + AS*) by Christgen et al. (2015); microbial electrolysis cells (*MEC*) by Gil-Carrera et al. 2013b); membrane bioreactor (*MBR*) by Jabornig and Podmirseg (2015); advanced oxidation process (*AOP*) by Kohler et al. (2012); and leachate-fed MFC by Zhang and He (2013)

However, the next stage is rather time consuming and may take 15 to 30 years. This lengthy degradation period can be attributed to the low activity and reproduction rates of methanogens under initial acidic conditions (Kim 2003), while another landfill specific reason is the presence of elevated levels of toxicants (heavy metals, XOCs, and ammonia) during the period of methanogenesis (Bernard et al. 1997). The content of heavy metals like cadmium (Cd^{2+}), chromium (Cr^{3+}), copper (Cu^{2+}), lead (Pb^{2+}), nickel (Ni^{2+}), and zinc (Zn^{2+}) is highly variable in leachate; their average content is typically <1 mg/L, an amount equivalent to ~0.02 % of heavy metals received in total (Flyhammar et al. 1998; Kjeldsen et al. 2002). However, the effects of these heavy metals on methanogenesis and bioelectricity generation are generally minor compared to the effects of ammonia (Ariunbaatar et al. 2015; Choi et al. 2014). Our own previous work has showed that NH_3-N reaches levels of ~1,000 mg/L in mature leachate with few volatile acids, amines, or alcohols detected (Xie et al. 2010). Among these "organic leftovers," the proportion of high molecular weight DOM increases to 32 %, and 60 % of these are detected as fulvic and humic-like compounds, which can neither be directly utilized in nitrogen reduction nor as biofuels (Puig et al. 2011; Wu et al. 2015). Indeed, their low biodegradability has largely frustrated conventional bio-treatment methods; therefore, physiochemical technologies are often used (Fig. 1).

Status quo of energy generation from organics in leachate

Acknowledging that organic carbon is abundant in leachate, particularly in fresh and intermediate ones (Fig. 2), Damiano et al. (2014) and You et al. (2006) have proven that landfill leachate can be used in MFCs, but at the same time, these authors argue that dynamic leachate should be pre-stabilized since a too low COD (<150 mg/L) concentration would be a

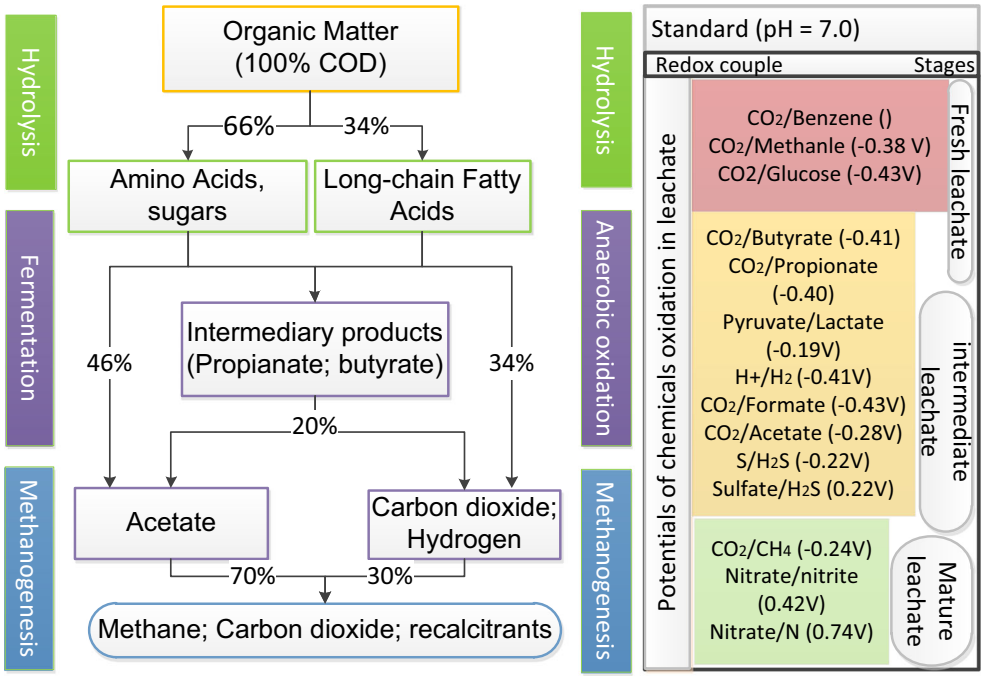

Fig. 2 COD balance of the organic fraction (redrawn from Lema et al. (1988)) and chemicals' redox potential in landfill leachate at different stages

limiting factor for bioelectrochemical reactions at the anode, while on the other hand, excessively high COD concentrations (>1,000 mg/L) may bring down coulombic efficiency (CE), especially in membraneless systems. Likewise, Ozkaya et al. (2013) found that a continuous increase in influent COD concentration was companied by an initial rise and subsequent sharp decrease in power density (Table 1); but in contrast, the CE value of consumed leachate showed a constant decline (57 to 1 %). Therefore, dilution of the carbon-rich stream to a proper COD and/or loading range is strongly recommended. Interesting in this context is also a study by Teng et al. (2010), who working with simulated leachate pointed out that at increasing proportions of butyrate and propionate power density and CE decreased from 1.9 to 1.0 W/m^3 and from 34 to 20 %, respectively. This decline is partially consistent with the observation of Puig et al. (2011) that even though power density increased from 0.15 to 0.3 W/m^3 with the addition of more raw leachate, CE finally dropped to ~2 %, whereas this value generally ranges from 20 to 30 % in pure culture (alcohol-fed) MFCs (Kim et al. 2007). It appears that high heterogeneity of carbon sources in landfill leachate could potentially put a dent in the energy and treatment efficiency of MFCs. This implies that more (bio)engineering is needed before this highly complex and carbon abundant mixture can serve as a suitable substrate for current generation in MFCs.

Apart from substrates, the effectiveness of a leachate-fed MFCs could also be affected by the reactor configuration and the operational conditions of the reactor. Table 1 shows that the dual chamber configuration slightly increases the systems' CE but that this has no positive effect on power density, probably due to the correspondingly increased internal resistance. In single chamber MFCs, molecular oxygen can easily diffuse across the membrane to the anode and bring down the CE. In membraneless fuel cells, which are characterized by low internal resistances and high power densities, the system uses only 1.5 % of its consumed electrons for electricity generation (Zhang et al. 2008). In addition, compared with batch feeding, the continuous pattern is clearly at a disadvantage with respect to energy generation (Table 1), probably because the microbially produced mediators (as secondary metabolites) involved in shuttling electrons to the anode (Lovley 2006; Rabaey et al. 2004) are continuously removed from the system. Indeed, these setup constrains on electricity generation are not specific to leachate-fed systems. What characterizes these systems is their high susceptibility to electron losses given the high concentrations of potential electron acceptors (nitrate, nitrite, sulfate, etc.) and amenable substrates for methanogens in the anode compartment; commonly identified competing pathways therefore include denitrification and methanogenesis (You et al. 2006; Zhang and He 2013).

In summary, even though the high organic content in landfill leachate could presumably reduce the "side effects" of carbon sources and reactor setup on power density, systems mentioned above all additionally suffered from low CE issues. Thus, transforming carbon-diverse leachates to substrates (e.g., acetate) favored by exoelectrogens should be given priority in operation. This would suggest a multi-stage approach. Indeed, it has been well established that running systems in series (stacks) or implementing anaerobic pretreatment can both increase electrogenic potential of leachates and lower the competitiveness of competing electron acceptors like nitrate (Galvez et al. 2009; Tugtas et al. 2013). The reason why

Table 1 Operational conditions and performance of waste leachate-fed MFCs (using carbon electrodes)

Leachate[a] (BOD/COD)	Org-loading (kg COD m^{-3} day^{-1}) or influent COD	Configurations	Operational conditions (pretreatment)	Max. stable output power density (CE %)	References
Mature	1.5–21.9	Single chamber	Continuous	0.06–0.35 W/m^3 (2)	Puig et al. (2011)
Intermediate	4.17	3 single chamber	Continuous (in series)	Reactor 1; 1.8 mW/m^2 Reactor 2; 5.6 mW/m^2 Reactor 3; 1.7 mW/m^2	Galvez et al. (2009)
Fresh	5.2	Single chamber	Continuous (no membrane)	12.8 W/m^3 (1.2)	Zhang et al. (2008)
Mature	2.4	Single chamber Single chamber	Semi-continuous Batch	14 mW/m^2 (0.5) 40 mW/m^2 (9.3)	Ganesh and Jambeck (2013)
Intermediate	<100 mg/L COD 4,900 mg/L COD	Dual chamber Dual chamber	Batch Batch	2.1 W/m^3 (22.1) 6.8 W/m^3 (3.4)	You et al. (2006)
Intermediate	0.3–2.9	Dual chamber	Continuous	0.07–0.26 mW/m^2	Greenman et al. (2009)
Intermediate	~250 mg/L	Dual chamber	Continuous (NO_x acceptor)	12 mW/m^2	Lee et al. (2013)
Fresh	4.3 0.4	Dual chamber	Continuous (pre-digestion) Batch[b] (pre-digestion)	158 mW/m^2 109 mW/m^2	Tugtas et al. (2013)
Fresh	50 100 200	Dual chamber	Batch[c] (Ti-TiO_2)	400 mW/m^2 (57) 800 mW/m^2 (7.0) 200 mW/m^2 (1.0)	Ozkaya et al. (2013)

[a] Determined by (Kjeldsen et al. (2002)), BOD_5/COD ratio of 0.5 and 0.2 for fresh and mature leachates, respectively

[b] Leachate pretreatment

[c] Electrode modification

the use of metal-modified electrodes, which are routinely recommended as enhancing CE, is not advised is that young leachate is very corrosive while its high hydrogen sulfide content could lead to metal poisoning or sulfur precipitation problems. Even though Ozkaya et al. (2013) stated that their Ti-TiO_2 electrodes enhanced power by 15 times and sustained for 20 days, the CE problem in the high leachate loading rate scenario is yet to be resolved (Table 1).

Energy generation from inorganics in leachate and prospected pathways

Using nitrate as electron acceptor in MFCs is promising since (i) its redox potential is comparable to that of oxygen (nitrate to N_2, Fig. 2) and (ii) it alleviates the concern about oxygen diffusion into the anode compartment. The first published designs based the use of bio-cathodes on the argument that exoelectrogens can reduce nitrate/nitrite with an electrode as electron donor (Gregory et al. 2004). However, this concept had always been realized in a potentiostat-poised half cell (external current supply is necessary), until Clauwaert et al. (2007) developed a system where a biocathode catalyzed denitrification by utilizing electrons from microorganisms oxidizing acetate in the anode.

As shown in Fig. 2, the inorganic nitrogen content starts to increase with age in intermediate and mature leachates, mainly in the form of NH_3-N, and typically reaches values of 700 mg/L (Kjeldsen et al. 2002). Harnessing this energy source necessitates a pre-nitrification step. For example, Lee et al. (2013) used an external aeration column to oxidize leachates and then fed the highly concentrated N-ions (mostly nitrate) to the cathode as potential electron acceptors. Their system achieved a peak power density of 12 mW/m^2 (Table 1), lower than obtained with oxygen biocathode (Zhang and He 2013). This is not surprising because the redox potential of nitrate/nitrogen is theoretically inferior to the O_2/H_2O's (0.7 vs. 0.8 V). However, Zhang and He (2013) further showed that the CE based on leachate-organic/nitrate (8.4 %) is ten times higher than on leachate-organic/oxygen (0.6 %). Thus, we may infer from these studies that the presence of highly concentrated inorganic N can turn the leachate into a suitable MFC substrate.

Nevertheless, bio-cathodes as mentioned above still need external energy to maintain a negative potential and implement nitrification (Gregory et al. 2004; Lee et al. 2013). Therefore, it would be intriguing to know if energy can be harnessed from the ammonia oxidation aiming to offset, if any, the aeration costs. Kim et al. (2008) detected no electricity generated with the addition of ammonia at first, but Fig. 3a shows that both aerobic and anaerobic ammonia oxidation (anammox) are exergonic and may theoretically indicate a spontaneous electron flow. Thus, the pending issue is whether the corresponding bio-catalysts (electrogenic species/enzymes) exist. Fortunately, recent studies concerning N-based MFCs suggested that not only ammonia but also intermediate products (e.g., nitrate and hydroxylamine) could all be utilized

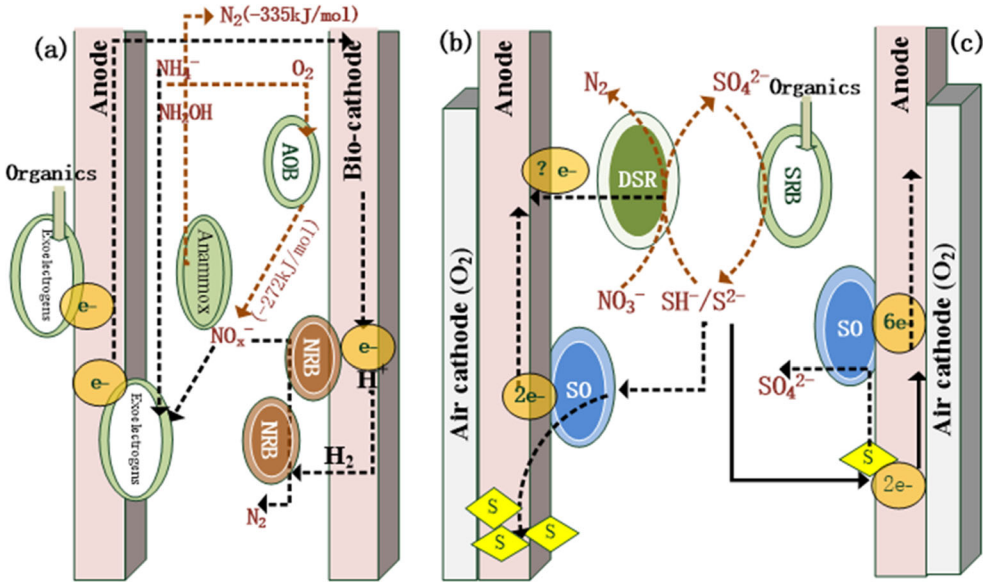

Fig. 3 Mechanisms by which nitrogen (**a**; −335 and −272 kJ/mol indicate energy released of anaerobic and aerobic ammonia oxidation, respectively, under standard conditions) and sulfur metabolites (**b**, **c**) contribute to electricity generation in microbial fuel cells. *Dashed line* and *solid line* represent biotic and abiotic pathways, respectively. *SO*, *SRB*, *AOB*, and *NRB* denote sulfide-oxidizing bacteria (e.g., *Desulfobulbus propionicus*), sulfate-reducing bacteria, ammonia-oxidizing bacteria, and nitrate-reducing bacteria, respectively. *DSR* represents the denitrification sulfide removal process, where one of the currently identified functional microorganism is *Pseudomonas* sp. C27 (Cai et al. 2013). Those electrons released by sulfide oxidation but not accepted by nitrate were finally transferred to the anode electrode; the exact electron numbers cannot be confirmed at our current knowledge

as fuels at the anode by typical nitrifying bacteria *Nitrosomonas* and *Nitrospira* (Chen et al. 2014). Lee et al. (2013) observed that an anammox biocathode generated slightly higher power density than a denitrifying biocathode (~30 vs. ~8 W/m^2) in a leachate-fed system, although the exact pathways were not unequivocally confirmed. Initiatives to integrate anammox in leachate-fed MFCs are based on the premises that the configuration of bioelectrogenic systems (a combination of anoxic and aerobic physiologies) and the low carbon/nitrogen ratio in mature leachate can both facilitate this autotrophic process. However, these drivers may lead to the misuse or overgeneralization of anammox-MFC concept. For example, Li et al. (2014a) just took advantage of anoxic condition in anodic chamber for implementing anammox but did not explain its role in electricity generation. From the perspective of energy generation, future research hence may need to explore the possibility of anammox (or similar functional) bacteria in electrode respiration, which in principle has been proven possible by Jadhav and Ghangrekar (2015).

The sulfate content in leachate generally ranges from 500 to 2,000 mg/L and is usually readily reduced by sulfate-reducing bacteria (SRB) to sulfide in stage 1 (~1 year after landfilling) (El-Fadela et al. 2002; Kjeldsen et al. 2002). Sulfur-based MFCs have revealed thatSO_4^{2-}, S^{2-} and S^0 can act as electron mediators rather than as substrates (Ieropoulos et al. 2013; Rabaey et al. 2006). The high organics and sulfur content in landfill leachate therefore can be viewed as wastewater mixed with electron mediators. Ieropoulos et al. (2013) and Lee et al. (2014) reported that oxidation of this mixture achieved a higher specific power of 80 and 60 mW/m^2, respectively, in comparison to other leachate-fed MFCs (Table 1), and that the oxidation of sulfide involved two electrons transferring to the anode with the formation of S^0 nanoparticles (Fig. 3b). However, Holmes et al. (2004) and Gong et al. (2013) have reported that these two electrons were harvested abiotically (at a potential no less than −0.27 V) and another six were harvested biotically, possibly by *Desulfobulbus propionicus*, on an electrode poised at a potential above 0.3 V (Fig. 3c). Interestingly, a higher anode voltage is thus required for complete sulfur oxidation and consequently for harnessing all electrons and eliminating S^0 deposition induced electrode fouling issues, while on the other hand, an elevated anode potential lowers the output energy, which is a function of the difference between anode and cathode potential. Tailored research will be needed to determine the optimum for specific leachates.

Stepping away from these existing organic/sulfur systems, Cai et al. (2013) have constructed a new system that was expected to use sulfide and nitrate as electron donor and acceptor, respectively. This system achieved a constant current density of ~150 mA/m^2, where >90 % sulfide and nitrate was removed. This research is inspiring considering the large amounts of sulfide and inorganic N coexisting in mature leachate (Fig 2). Also, sulfide/nitrate-based cells could

theoretically gain 1.87 V (Eq. 1, by Cai et al. (2013); this value is considerably higher than the theoretical gain for the traditional organic C (acetate)/oxygen redox couple (~1.2 V).

$$5H + 2 \rightarrow 5S + N_2 + 6H_2O + 7e^- \left(E^0 = 1.87V\right) \quad (1)$$

It is unknown which portion of the energy/voltage electrogenic microorganisms retain for their own metabolism. More research is needed to explore in which pathways and with what efficiencies functional microorganism transfer electrons to or gain electrons in bioelectrochemical systems treating mature leachate with high S and N content.

Specific microbial accesses in MSW leachate to energy in the MFC

As part of the stepwise degradation process depicted in Fig. 1, many anaerobes in landfill leachate indeed are capable of intracellular electron transfer to nitrate and sulfate for anaerobic respiration (Wang et al. 2014). What distinguishes electrogenic bacteria is their ability to transport electrons outside their membrane. In their special respiration chains, electrodes are used as terminal electron acceptors, while the protons generated are transferred to and consumed at the cathode. Commonly detected electrogenic bacteria belong to three genera, *Shewanella* (*Shewanella oneidensis*, *Shewanella putrefaciens*), *Geobacter* (*Geobacter sulfurreducens*, *Geobacter metallireducens*), and *Rhodoferax* (*Rhodoferax ferrireducens*), which can all gain energy from the use of insoluble metal as an electron acceptor in nature (Chaudhuri and Lovley 2003; Szollosi et al. 2015). It is notable that these microorganisms are all classified as *Proteobacteria*, organisms that are abundant in both fresh and mature MSW landfill leachate (Kobayashi et al. 2013; Liu et al. 2011). This may imply that landfill leachate possesses a wealth of organisms that can gain energy in MFCs. In effect, rarely does one single species or genus dominate in the bacterial communities attached on the anode, especially when fed with complex substrates such as landfill leachate (Logan and Regan 2006).

Recent metagenomic analysis has revealed a clear disparity in microbial communities between acetate-fed and leachate-fed scenarios, where the relative abundance of *Geobacter* declined tenfold upon addition of leachate (Zhang et al. 2014). One possible reason is that complex organics cannot be efficiently or directly used by the electrogenic microorganisms (detailed in next section). Another reason may be that heavy metals present in leachate may adversely affect the viability of exoelectrogens. Abourached et al. (2014) reported that maximum tolerable concentrations (MTCs) of the ions Cd (VI) and Zn (II) for the electrochemically active microorganisms are 0.2 and 0.4 mg/L; a higher concentration (>0.5 mg/L) could significantly inhibit this reaction, as indicated by a 70 % reduction in voltage. As summarized by Kjeldsen et al. (2002), total zinc in MSW landfill leachate (0.2 to 5.3 mg/L) exceeded the MTC in most cases. However, heavy metal ions can readily form complexes with inorganic and organic ligands, which would safeguard MFCs against "heavy metal poisoning." Thus, further study on heavy metals' toxicity in leachate to electrogenic microorganisms is urgently needed.

Fermentation and interspecies electron transfer in leachate

Landfill leachate degradation processes are distinct from those studied in single substrate incubations (e.g., studies with acetate in microbial fuel cells) or typical wastewater treatment pathways: they feature an intricate combination of sequential and parallel processing of substrates, dictated by distinct and stepwise microbial activities (Renou et al. 2008). Therefore, enhancing energy recovery efficiency from leachate is difficult to achieve, if the abundance of exoelectrogens is truly the key factor in electricity production. However, the "food chain" mentioned above may imply alternative potentialities for electricity generation (Dolfing 2014). Firstly, fermentation is a prerequisite for the effective oxidation of complex organic matter, such as aromatic compounds and long-chain fatty acids (McInerney et al. 2009). Pre-fermentation of leachate (acetate and succinate as major products) increased the electron recovery rates in a fuel cell by 20 % (Mahmoud et al. 2014). This is because the currently known exoelectrogens cannot metabolize complex substrates (Chaudhuri and Lovley 2003). Thus, MFCs fed with complex substrates, which generally have a lower redox potential than acetate (e.g., glucose vs. acetate, Fig. 2), only recovered 2–6 % of the theoretical voltage (Lee et al. 2008; Logan 2004). It appears that abundant energy, stored in complex substrates, for example carbohydrates, in waste streams is harnessed limitedly in MFCs as a substantial part of this energy is released during fermentation to the substrates that are "edible to exoelectrogens."

However, there is evidence that electrons could be shuttled to electrodes directly from fermentative microorganisms (Fig. 4a), such as *Geothrix fermentans*, *Clostridium butyricum*, and *Pseudomonas chlororaphis* via self-produced electron mediators, Fe (III) reduction, or abiotic oxidation of their products (e.g., hydrogen) at the anode (Hernandez et al. 2004; Lovley 2006; Park et al. 2001). Nevertheless, the role and function of fermentative microorganisms in leachate-fed MFCs are still controversial. Hydrogen-oxidizing methanogens (responsible for hydrogen transfer in leachate fermentation) can outcompete exoelectrogens (Lee et al. 2008), and pragmatically the methanogens are sometimes allowed to take control (in anode) to enhance proton recover rates (Chae et al. 2010). But, importantly, coexistence of *Hydrogenophaga* (hydrogen-gas consuming exoelectrogens) and fermentative bacteria significantly increased energy output (Kimura and Okabe 2013). Thus, further research is

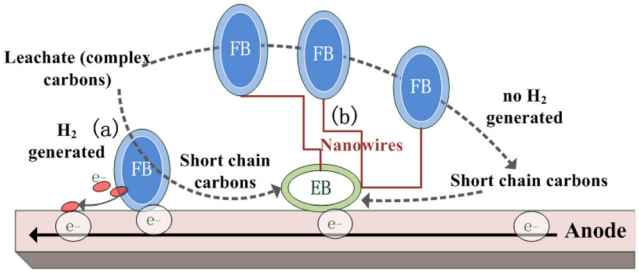

Fig. 4 Mechanisms of electrons transferred to anode by fermentative bacteria (**a**) and proposed pathways involving exoelectrogens and fermentative bacteria (**b**). *Dashed line* and *solid line* represent substrate and electron pathways, respectively, and *FB* and *EB* denote fermentative bacteria and exoelectrogenic bacteria, respectively. *Red circles* denote self-produced electron mediators

warranted to evaluate whether fermentation should be combined with bioelectricity generation.

The occurrence of syntrophy in landfill sites based on hydrogen-mediated interspecies electrons transfer is well established (Jakobsen et al. 1998). Direct interspecies electron transfer (DIET) on the other hand is favored thermodynamically because it is more energy efficient: (i) hydrogen inhibits fermentation of organics and (ii) the hydrogen gradients needed for interspecies hydrogen transfer dissipate energy (Dolfing 1992; Fukuzaki et al. 1990; Summers et al. 2010). One prerequisite for DIET is physical contact. The existence of DIET was first observed by Gorby et al. (2006), who observed that fermentative microbes (*Pelotomaculum thermopropionicum*) were wired to methanogens (*Methanothermobacter thermoautotropicus*) by conductive filaments, which were previously thought to be exclusive to exoelectrogens transferring electrons to metals and electrodes. Subsequently, Summers et al. (2010) observed that biomass aggregates are another form of physical contact where DIET can occur.

It has been suggested by Lovley (2006) that transfer of electrons to natural extracellular material is an adaptive evolution process selecting for the most effective strategies for energy production. We speculate that large organic polymers could trigger similar processes at leachate-fed anodes. It therefore would be interesting to know whether electrons can be directly transferred from fermentative organisms (complex organics) via exoelectrogens (presumably via NAD^+/NADH) to an anode (Fig. 4b). These physical contacts allowing DIET would overcome the energy-harvesting barriers of non-degradable substrate (by exoelectrogens) in fresh and intermediate leachate. Recently, Li et al. (2014b) have successfully used nanoFe_3O_4, analogous to nanowires, to connect syntrophic microorganisms in an engineered form of DIET. It is illuminating that the addition of nanomaterial may facilitate this energy efficient electron pathway. Apart from direct electron transfer, mediated electron transfer (MET) in bioelectrochemical systems is also frequently observed (Schröder 2007). In MET, electron mediators wire microbial metabolism to a fuel cell anode via the shifts of their redox potentials. Actually, the high content of accumulated humic substances in mature leachate (60 % of the DOM) compared to <6 % in most other types of wastewater, specifically favor MET. Regular addition of exogenous mediators would be technologically unfeasible and economically questionable (Kjeldsen et al. 2002). For this reason, mature leachate has been mixed with waste and/or wastewater in order to streamline the electron transfer chain in substrate oxidation and fermentation. For example, Capodici et al. (2014) reported that the electron transfer efficiency (respiration rate) increased by 40 % with addition mature leachate. Also, research by Ferraz et al. (2014) has shown that humic substances at a low leachate mix ratio (~2 %) enhanced the systems' bioactivity and indicated that this refractory organic material underwent self-degradation. From these observations, we conclude that mature leachate can be theoretically converted to energy harnessing facilitators.

Conclusions

Landfill leachate-based MFCs provide us a new research platform, which straddles engineering and the underpinning science. This review shows that fresh and intermediate leachate can in principle be used for bioelectricity generation, but that efficient energy recovery will require integrated decomposition of a highly diverse waste stream, for example by operating fuel cells in series and/or coupled to anaerobic pre-digestion. Metal modification of electrodes can be applied to enhance output energy if the material is cost-efficient, and Ti-TiO_2 is a good option compared to platinum. But this approach with respect to leachate treatment is clouded by issues like H_2S-induced metal catalyst poisoning and sulfur precipitation. Most MFCs therefore opt for the metal-modified electrode as air cathode. The current application of leachate-fed bioelectrochemical systems is mainly aimed at contaminant removal and has circumvented the tension between organic carbon and nitrogen content, which plagues conventional denitrification in mature leachate, through anammox and simultaneous N and S removal. These designs do however suggest that the high concentrations of inorganic N and S metabolites in leachate can also be used as novel biofuels; the energy "appropriation" by electrogenic microorganisms will be the key limit in their application. Also, it appears that the relationship between fermentative microorganisms and electrogenic bacteria is critical in energy output enhancement, particularly via the energy efficient DIET. This potential can be realized by the addition of nanomaterials and/or humic substance-rich mature leachate. The current generation of "off-the-shelf" MFC technology has already been shown to be applicable to the treatment of landfill leachate, but economics will eventually decide whether this technology is appropriate.

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