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REVIEW ARTICLE

Producing electrical energy in microbial fuel cells based on sulphate reduction: a review

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Abstract

Combination of the treatment of effluents with high organic loads and the production of electricity is the driving forces stimulating the development of microbial fuel cells (MFC). The increase in electricity production in MFCs requires not only the optimization of the operational parameters but also the inhibition of the metabolic pathways, which compete with electricity production, such as methanogenesis. The presence of both sulphate and sulphide ions in conventional anaerobic reactors hampers the growth of methanogenic archaea and justifies the use of sulphate and therefore sulphate-reducing bacteria (SRB) in the anodic half-cell of MFC. Most importantly, the literature on the subject reveals that SRB are able to directly transfer electrons to solid electrodes, enabling the production of electrical energy. This technology is versatile because it associates the removal of both sulphate and the chemical oxygen demand (COD) with the production of electricity. Therefore, the current work revises the main aspects related to the inoculation of MFC with SRB focusing on (i) the microbial interactions in the anodic chamber, (ii) the electron transfer pathways to the solid anode, and also (iii) the sulphate and COD removal yields along with the electricity production efficiencies.

Keywords Bioelectrochemistry · Anaerobic digestion · Sulphate-reducing bacteria · Biofuel · Wastewater · Industrial effluents

Introduction

Following an increasing awareness of the effects of fossil fuels on global warming, new and alternative energy-producing technologies have been intensively researched. Another important concept in modern society is the circular economy, which has changed the perception of residues and effluents, now regarded as valuable resources (Goglio et al. 2019; Manzano-Agugliaro et al. 2013). Both principles have induced the development of microbial fuel cells (MFC) aiming at producing electricity from the anaerobic oxidation of organic matter (Pandey et al. 2016). A large-scale utilization of MFCs is still beyond reach, but energy recovery during wastewater treatment is most likely to happen in the near future (Logan 2009).

An MFC layout comprises two electrically connected chambers. In the anodic half-cell, a biologically mediated oxidation reaction produces electrons, which are externally transferred to the cathodic half-cell where they are used in either a chemical or biological semi-reaction of reduction. An MFC also contains a proton- or cation-exchange membrane and an external resistance (Logan et al. 2006). The growing interest in such technology does not only derive from its potential to produce electricity but also as a more environmentally-friendly technology than anaerobic digestion or any other common effluent treatment process (Pandey et al. 2016).

In addition to organic matter, industrial wastewaters may also contain a series of inorganic pollutants, such as sulphate ions, which need to be removed during effluent treatment. Sulphate-bearing effluents have been produced by several industries, such as electroplating, pulp and paper, pigments, rubber, explosives, fertilizers, and mining-metallurgy (Lens

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et al. 1998; Sarti et al. 2008). Despite its low toxicity, sulphate levels in wastewaters are regulated and a maximum contaminant level-between 250 and 500 mg L^{-1} is set by several countries. Sometimes the content of sulphate is regulated through a limit on total dissolved solids (TDS) (Nascimento 1998). Among the alternatives to remove sulphate from wastewaters, biotechnological processes applying sulphate-reducing bacteria (SRB) have been extensively investigated (Bertolino et al. 2012; Bertolino et al. 2015; Hu et al. 2020; Kaksonen et al. 2006; Kaksonen and Puhakka 2007; Luo et al. 2020). These microorganisms utilize sulphate as final electron acceptors resulting in the production of sulphide ions (Liamleam and Annachhatre 2007). Recently, it has been reported that sulphate-reducing bacteria (SRB) are electroactive bacteria (EAB), i.e. SRB are able to transfer electrons directly to an electrode rather than to a redox couple, which is an important mechanism in MFC (Kang et al. 2014; Zhou et al. 2013). This justifies the interest in coupling sulphate reduction and electricity production in these devices.

The first studies addressing sulphate reduction in the context of MFCs speculated the electroactive nature of SRB inoculated in the anodic half-cell (Agostino and Rosenbaum 2018; Hu et al. 2019; Liang et al. 2013a; Zheng et al. 2014). However, another application has also been proposed for the role of SRB. In this latter mechanism, the bacteria accept electrons directly from cathodes to convert carbon dioxide into organic substances (Agostino and Rosenbaum 2018; Luo et al. 2020). Therefore, electro-autotrophic SRB may also function as biocathodes in the production of sulphide ions, and this electrochemical device is known as microbial electrolysis cells (MEC) (Gacitúa et al. 2018; Luo et al. 2014a). Despite the several studies investigating the SRB role in MEC, the current work focuses only on anodic half-cells inoculated with SRB, which play an indisputable role on the generation of electricity (Hu et al. 2019). Specifically, the syntrophic interactions of the microbial consortium, the electron-transfer pathways in the anodic hall-cell, the oxidation of the carbon source, sulphate reduction, and the production of electricity in MFC will be revised.

Microbial fuel cells

Usually, heterogenic bacteria produce energy-rich compounds, such as ATP (used to carry out biological work) via the oxidation of nutrient molecules. The electrons produced during cellular metabolism are transferred to a chemical acceptor such as O_2, NO_3^-, and SO_4^{-2} during cellular respiration (Nelson and Cox 2012; Tortora et al. 2010). Some bacteria in particular can utilize ferric iron as the final electron acceptor, which is found in nature mostly as an insoluble compound. Therefore, it was proposed that ferric iron reduction occurred outside the cell (Lovley 2006). Based on the observation of this phenomenon, the direct electron transfer to solid electrodes by microorganisms was hypothesized. Accordingly, a technology could be devised by which bacteria would transfer electrons from an electron donor to an artificial solid device, producing electricity (Madigan et al. 2015). These microorganisms are classified as either exoelectrogenic or electroactive (Logan 2009).

Electroactive microorganisms have been identified in a wide variety of ecosystems, such as soil, sediment, seawater, and freshwater and also in samples collected from several environments having a diverse microbial community (sewage, activated sludge, industrial and domestic effluents) (Logan 2009). Specifically, these bacterial strains belong to the genera *Geobacter* sp., *Rhodoferax* sp., *Shewanella* sp., *Pseudomonas* sp., *Arcobacter* sp., *Clostridium* sp., *Ochrobactrum* sp., and *Desulfovibrio* sp. (Mathuriya 2014; Santoro et al. 2017). Such microorganisms can be technologically applied in a bioelectrochemical reactor, in which electrons are transported to (and from) a solid material (electrode), such as a MFC (Zhou et al. 2013). Such devices can therefore be applied to oxidize reduced species in liquid effluents and produce electricity simultaneously (Friedman et al. 2013). The main argument supporting the development of such technology is to improve sustainability (reducing treatment costs and energy consumption) of effluent treatment operations, particularly those containing moderate to high organic loadings (Santoro et al. 2017).

In a typical MFC (Fig. 1), the anodic and cathodic chambers are separated by an ion exchange membrane. In the anodic chamber, the microbial cells in the biofilm oxidize a substrate to produce (i) protons (H^+), (ii) oxidized species, such as CO_2, and (iii) electrons, which are then transferred to the solid electrode. In addition, as the proton concentration increases in the anodic chamber, they are transferred to the cathodic compartment through the membrane. The electrons in the anode are shuttled to the negative electrode (cathode) wherein species such as oxygen, protons, ferricyanide ions, and hydrogen peroxide are reduced (He et al. 2017).

Some advantages of applying MFC in wastewater treatment are as follows (He et al. 2017; Mathuriya 2014; Santoro et al. 2017): (i) direct conversion of chemical energy in wastewaters into electricity; (ii) smaller production of biomass, even when compared with conventional anaerobic processes; (iii) aeration is eliminated; and (iv) it can be potentially used in places short of electricity supply. In addition, COD removal in MFC may attain values over 80% (Logan 2009; Santoro et al. 2017).

Extensive studies have been carried out in lab-scale in order to improve the energy efficiency of MFCs so that a large-scale use in wastewater treatment becomes feasible, in the treatment of both domestic (Lovley 2008; Tice and Kim

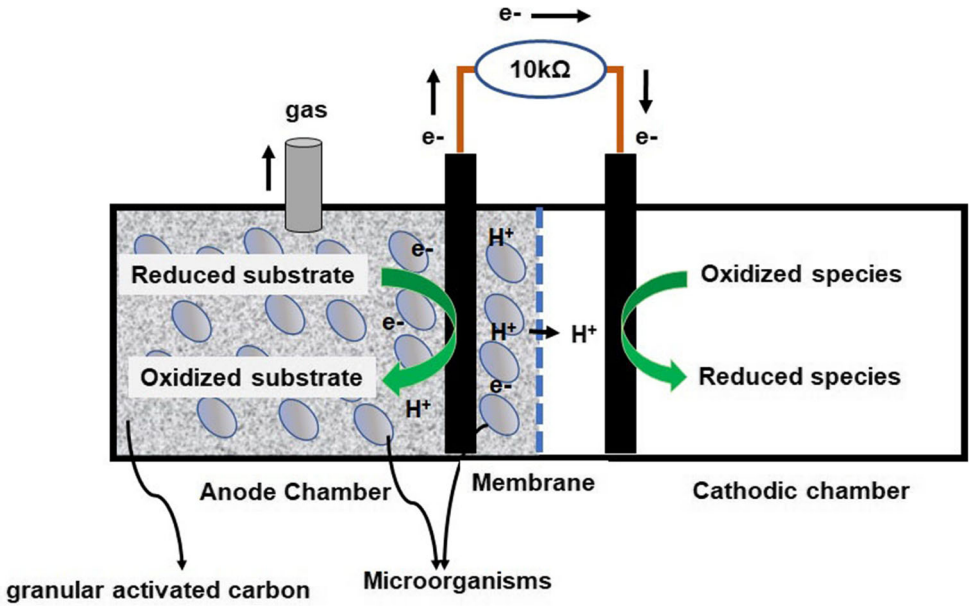

Fig. 1 Schematic representation of a typical two-chamber microbial fuel cell

2014) and industrial wastewaters, in particular those containing toxic metals (Luo et al. 2014b; Tao et al. 2014), azo dyes (Khan et al. 2015), residues produced by the oil industry (Jiang et al. 2013; Li et al. 2015), acid mine drainage (Cheng et al. 2007; Hai et al. 2016), phenol and compounds (Feng et al. 2015), and pyridine-related products (Mathuriya 2014). Its potential incorporation to existing industrial operations will require the following: (i) improvement in energetic efficiency, (ii) relative stability in energy production, and also (iii) a reduction in capital expenditure (Pant et al. 2011).

With respect to the electricity production and also the columbic efficiency (the fraction of electrons produced during oxidation, which effectively are transferred to the cathode thus producing electricity) of MFC, the microbial community plays a key role, requiring the presence of a biofilm containing exoelectrogenic species. Nevertheless, during the conversion of the organic matter, there is competition between these electroactive bacteria and methanogenic archaea (which are not electroactive) for the organic substrate, which results in a considerable reduction in electricity production and also in the columbic efficiency (Isosaari and Sillanpää 2017).

The presence of sulphide ions reduces methanogenesis in MFC because H_2S can inhibit the growth of methanogenic archaea, justifying the inoculation of the latter with SRB (Chou et al. 2014; Chou et al. 2013; Isosaari and Sillanpää 2017; Liang et al. 2013a; Sangcharoen et al. 2015; Su et al. 2012; Weng and Lee 2015; Zhao et al. 2009). Moreover, as stated, SRB are capable of transferring electrons directly to the anode (Liang et al. 2013b; Zhao et al. 2009; Zhou et al. 2013). A second hypothesis is that there is a synergic pathway involving SRB and SOB (sulphide-oxidizing bacteria), whereby SOB oxidizes the sulphide produced by SRB to elemental sulphur, transferring electrons directly to the anode material (Chou et al. 2014; Chou et al. 2013; Weng and Lee 2015; Zhao et al. 2009). Nevertheless, this latter mechanism requires sulphide diffusion (and its oxidation) in the anode, implying that the direct electron transport mechanism plays a key role in electricity production (Murugan et al. 2018). Such mechanism will be detailed in the following paragraphs.

When applied in electrochemical system aiming at effluent treatment, the participation of other microbial species is mandatory to ensure a diverse microbial community, which is required to produce the syntrophic interactions required to degrade complex organic substrates and to generate electrical energy (Kokko et al. 2016). These interactions are discussed next.

Microbiology and biochemistry of bioelectricity generation

It has been extensively demonstrated that several microbial interactions are present in anaerobic reactors (Bertolino et al. 2012; Bertolino et al. 2015; Chernicharo 1997; Hu et al. 2020; Kaksonen et al. 2006; Kaksonen and Puhakka 2007). This consortium of microorganisms is responsible for the high efficiency of wastewater treatment by anaerobic processes aiming at oxidizing organic matter. In this treatment, macromolecules are first hydrolysed by hydrolytic microorganisms producing soluble species, which will be consumed by other bacteria. Fermentation will occur through the action of acidogenic bacteria on the low molecular weight (LMW) species produced. The presence of acetogenic bacteria is also important to maintain the H_2 pressure in non-inhibitory

concentrations. Usually, these fermentation products are consumed by methanogenic archaea, but SRB can play the same role when sulphate is present (Muyzer and Stams 2008).

The oxidation reactions of the LMW species and also hydrogen gas are represented by Eqs. (1), (2), (3), (4), (5), and (6). The electrons produced are used in the reduction of either oxygen (aerobic conditions) or other species such as NO_3^-, SO_4^{2-}, CO_2, and Fe^{3+} (anaerobic respiration), and in electrochemical systems, the electron transfer is mediated by two solid electrodes (Dong and Stams 1995; Kokko et al. 2016; Logan 2009; Lovley 2006; Madigan et al. 2015). In both anaerobic reactors and MFC, the degradation of these intermediary compounds reduces the Gibbs-free energy of the entire anaerobic organic matter oxidation process (Kokko et al. 2016).

$$CH_3COOH + 4\ H_2O \rightarrow 2\ HCO_3^- + 10\ H^+ + \mathbf{8e^-} \quad (1)$$

$$C_2H_5OCOOH + 6\ H_2O \rightarrow 3\ HCO_3^- + 15\ H^+ + \mathbf{12e^-} \quad (2)$$

$$C_3H_7COOH + 10\ H_2O \rightarrow 4\ HCO_3^- + 24\ H^+ + \mathbf{20e^-} \quad (3)$$

$$C_2H_5COOH + 7\ H_2O \rightarrow 3\ HCO_3^- + 17\ H^+ + \mathbf{14e^-} \quad (4)$$

$$C_6H_{12}O_6 + 12\ H_2O \rightarrow 6\ HCO_3^- + 30\ H^+ + \mathbf{24e^-} \quad (5)$$

$$H_2 \rightarrow 2\ H^+ + \mathbf{2e^-} \quad (6)$$

SRB and other EAB can only oxidize low molecular weight substrates such as hydrogen gas, acetate, lactate, ethanol, and glycose, which are converted to H^+ and electrons (Das and Mangwani 2010), whereas they lack the ability to degrade more complex molecules (Lovley 2006). However, the biochemical reactions occurring in the anodic cell in an MFC are similar to those observed in anaerobic reactors, as demonstrated by Kumar et al. (2017b), through molecular biology techniques. These authors found in the anode chamber of an MFC hydrolytic bacteria such as *Aminobacterium* sp. and fermentative microorganisms such as *Clostridium* sp., which convert monomers to organic acids according to Eqs. (7) and (8). The authors also detected SRB, such as *Desulfovibrio* sp. whose metabolism is represented by Eqs. (9), (10), (11), (12), (13), and (14), and also other exoelectrogenic bacteria, such as *Aeromonas taiwanensis* which use acetate as in Eq. (15). In addition, this study still found SOB, such as *Tetrathiobacter kashmirensis* and *Desulfovibrio sulfodismutans*, which oxidize sulphides to elemental sulphur (Eqs. (16), (17), and (18)). Furthermore, methanogenesis and denitrification processes compete with electricity generation, and these biochemical reactions should be inhibited so that a significant production of electricity is achieved (Das and Mangwani 2010; Kokko et al. 2018; Kumar et al. 2017a; Madigan et al. 2015). A general overview of the microbial interactions and metabolic products in an MFC is summarized in Fig. 2.

	ΔE^0 (V)	
$3\ CH_3CH(OH)COO^- \rightarrow CH_3COO^- + 2\ CH_3CH_2COO^- + HCO_3^- + H^+$	0.58	(7)
$CH_3CH_2COO^- + 3\ H_2O \rightarrow CH_3COO^- + HCO_3^- + H^+ + 3\ H_2$	− 0.79	(8)
$CH_3COO^- + SO_4^{-2} \rightarrow 2\ HCO_3^- + HS^-$	0.49	(9)
$CH_3CH_2COO^- + 0.75\ SO_4^{-2} \rightarrow CH_3COO^- + HCO_3^- + 0.75\ HS^- + 0.25\ H^+$	0.39	(10)
$CH_3CH_2CH_2COO^- + 0.5\ SO_4^{-2} \rightarrow 2\ CH_3COO^- + 0.5\ HS^- + 0.5\ H^+$	0.29	(11)
$CH_3CH(OH)COO^- + 0.5\ SO_4^{-2} \rightarrow CH_3COO^- + HCO_3^- + 0.5\ HS^- + 0.5\ H^+$	0.83	(12)
$4\ H_2 + SO_4^{-2} + H^+ \rightarrow HS^- + 4\ H_2O$	1.57	(13)
$SO_4^{-2} + 4\ HCOO^- + H^+ \rightarrow HS^- + 4\ HCO_3^-$	1.52	(14)
$CH_3COO^- + 2\ H_2O \rightarrow 2\ CO_{2\ (g)} + 7\ H^+ + 8e^-$	0.30	(15)
$H_2S \rightarrow S^0 + 2\ H^+ + 2e^-$	− 0.17	(16)
$S^{-2} \rightarrow S^0 + 2e^-$	0.45	(17)
$HS^- \rightarrow S^0 + 2e^- + H^+$	0.06	(18)

Therefore, the profile of the microbial species found in the anodic half-cell will be defined by parameters such as (i) composition of the effluent, (ii) electrode material, and (iii) operating conditions (Kokko et al. 2018).

Although the main biochemical reactions occurring in the anodic chamber are understood, the syntrophic and competitive interactions between different microbial strains should be investigated further on. In addition, new ways of inhibiting the metabolic pathways competing with the production of electricity are yet to be discovered (Bratkova et al. 2019).

Some authors report that in the anodic chamber of an MFC, sulphate ions may compete with the anode as the final electron acceptor during the metabolism of SRB (Habermann and Pommer 1991; Kokko et al. 2016). However, these microorganisms can still contribute to the generation of electricity by transferring electrons via the oxidation of organic matter to the electrode to the detriment of sulphate (Hu et al. 2019; Kang et al. 2014; Loukanov et al. 2019; Miran et al. 2018). Moreover, in terms of electricity production, sulphate reduction does not represent the main role of SRB in the anodic half-cell. Uppermost is the SRB role in (i) the oxidation of the organic substrate and (ii) contribution to biofilm formation as exopolysaccharides and filamentous proteins are excreted by them, in addition to the several electron transfer pathways already proposed for this group of microorganisms (Bratkova et al. 2019; Kokko et al. 2018; Lee et al. 2012; Zhao et al. 2008). Understanding the mechanisms related to electron transfer by SRB to anodes will improve electricity production in MFC and also expand its applications (Murugan et al. 2018).

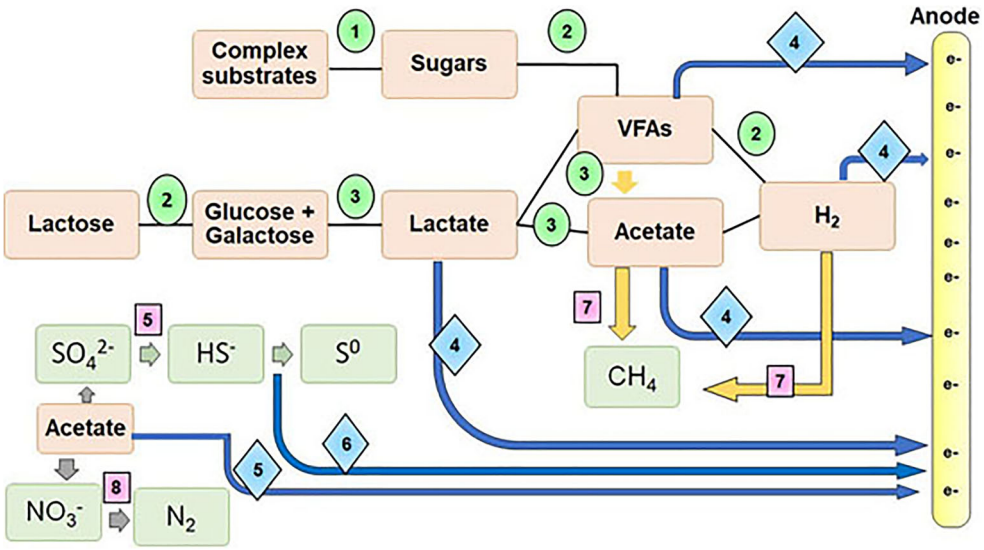

Fig. 2 Microbial consortium in the anodic chamber of a microbial fuel cell. 1, hydrolytic bacteria; 2, fermentative bacteria; 3, syntrophic bacteria; 4, exoelectrogens; 5, sulphate-reducing bacteria (SRB); 6, sulphide-oxidizing bacteria (SOB); 7, methanogens; 8, denitrifies. Circle indicates reactions that precede electricity production; diamond indicates electricity production reactions; square indicates reactions that compete with electricity production (This figure was devised based on references Madigan et al. 2015; Kumar et al. 2017b)

Electron transfer pathways by SRB

Most studies investigating the role of SRB in MFC proposed somewhat simplified mechanisms for the electron transfer to anodes (Murugan et al. 2018). According to these mechanisms, SRB are generally able to transfer electrons throughout four possible pathways (Blázquez et al. 2019; Kumar et al. 2017b; Miran et al. 2018; Zhou et al. 2013), which are (i) syntrophic interaction with SOB; (ii) via either the outer membrane or periplasmic cytochromes when there is direct contact of the cell with the electrode; (iii) synthesis of nanowires, i.e. electron-conducting pilli produced by bacteria attached to the electrode surface; and (iv) nanoparticles of metal sulphides, such as FeS, transfer electrons via the external membrane of the microbial cells (Chou et al. 2013; Hu et al. 2019; Kang et al. 2014; Lee et al. 2012; Logan 2009; Miran et al. 2017; Murugan et al. 2018; Sangcharoen et al. 2015; Santoro et al. 2017; Zhao et al. 2008). Figure 3 describes schematically electron transfers to the anode in an SRB-containing MFC.

The first pathway proposed for to the production of electricity in MFCs was the autotrophic oxidation (by SOB) of the sulphide produced during sulphate reduction (Blázquez et al. 2019; Chou et al. 2013; Kang et al. 2014; Kumar et al. 2017b; Lee et al. 2012; Sangcharoen et al. 2015; Zhao et al. 2008). An alternative pathway is abiotic sulphide oxidation to elemental sulphur (Sangcharoen et al. 2015).

The direct transfer of electrons to an extracellular solid acceptor (electrode) by SRB is reported in several studies (Eaktasang et al. 2016; Eaktasang et al. 2013; Hu et al. 2019; Loukanov et al. 2019; Miran et al. 2017; Miran et al. 2018). Eaktasang et al. (2016) revealed evidences of electrically conductive nanoscale filaments produced by SRB in MFC using scanning electron microscopy (SEM) and atomic force microscopy (AFM) techniques. Loukanov et al. (2019) reported the formation of nanowires in the solid electrode using SEM-EDS techniques, and Hu et al. (2019) determined the conductive properties of these nanostructures by AFM. In addition, Kang et al. (2014) demonstrated that *Desulfovibrio desulfuricans* is able to transfer electrons to the anode directly via cytochrome C-type proteins. This was accomplished by producing and isolating this recombinant protein and by using the latter to effectively produce electricity in an MFC. Fourier-transform infrared spectroscopy (FTIR) analyses have proved that the microorganisms bind to the anode surface via hydrogen and peptide bonds. The latter are bonds between amino groups belonging to the cytochrome C, located on the outer membrane of the microorganism, with carboxylic groups present on the carbon-anode surface (Kang et al. 2014). Therefore, the direct transfer of electrons from the periplasmatic region to the extracellular solid acceptor is enabled.

More recently, it was proposed that other conductive metabolites such as iron sulphides (shown in Fig. 3 as Me_yS_x) also mediate the transfer of electrons to the anode (Hu et al. 2018; Murugan et al. 2018). Specifically, Murugan et al. (2018) proposed that the presence of iron in the growth medium doubled the values of the anodic current in electrochemical studies, which was justified by the contribution of iron sulphides to the formation of cell aggregates on the electrode surface. In addition, FeS nanoparticles on the cell surface enabled electron transfers through the bacteria outer membrane to the extracellular medium, which is a more effective and faster pathway than the diffusion of sulphide ions and their oxidation on the anode surface (Murugan et al. 2018). It must be emphasized that sulphides are semi-conductors and may contribute to transfer of charges in the system (Eaktasang et al. 2013).

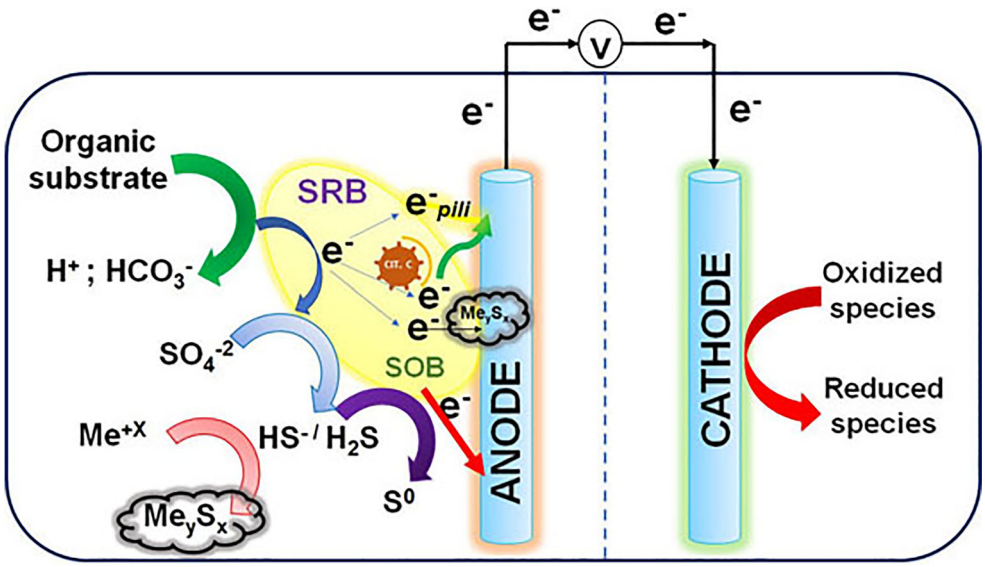

Fig. 3 Schematic representation of electron transfer in microbial fuel cells based on sulphate-reducing bacteria. SRB, sulphate-reducing bacteria; SOB, sulphide-oxidizing bacteria; Me, metal (This figure was devised based on reference Miran et al. 2017)

In addition to these studies discussing the electroactive features of SRB, several studies covering sulphate removal associated to electricity generation in MFC have been published (Bratkova et al. 2019; Chou et al. 2013; Cooney et al. 1996; Habermann and Pommer 1991; Kang et al. 2014; Kumar et al. 2017a; Kumar et al. 2019; Lee et al. 2012; Lee et al. 2014; Liang et al. 2013b; Miran et al. 2017; Niyom et al. 2018; Sangcharoen et al. 2015; Zhao et al. 2008). Some of these works are revised next.

Sulphate reduction in microbial fuel cells

The first reports of the use of SRB in MFCs appeared in the 1990s (Habermann and Pommer 1991; Lee et al. 2012). However, sulphate removal was a controversial hypothesis because it would compete with the anode surface for the electrons available in the anodic half-cell. A decade later, a new theory proposed that the sulphide produced during sulphate reduction was oxidized to elemental sulphur in the anode (Rabaey et al. 2006). As a consequence of this second theory, the number of papers addressing sulphate reduction in the anodic half-cell started to increase, as listed in Table 1.

It can be seen in Table 1 that the current density values are low. Habermann and Pommer (1991) stated that low currents and an unstable production of energy would limit the application of this technology in an industrial scale. However, the main advantage of the use of MFCs would be a significant COD removal associated with the production of electricity, which was proposed for the first time by these authors, who reported 75% COD removal from treating a landfill leachate in an MFC.

Habermann and Pommer (1991) also verified sulphate reduction to sulphide by SRB in the anodic chamber, but, according to the authors, the species must have been re-oxidized to sulphate, justifying the low sulphate removal observed when glucose was used as an energy source. However, this hypothesis is unlikely because elemental sulphur oxidation to sulphate is kinetically slow in the absence of sulphur-oxidation bacteria. The highest current density, as compared with other studies, could have been related to the electrode impregnation with Ni, Co, and Fe (Cooney et al. 1996) or even due to the long acclimatization periods, as these experiments took over 5 years to conclude.

A comparison of the power output values of these studies is challenging, due to the different MFC configurations and the differences in operating conditions. Logan (2012) states that a complete description of the layout and operating conditions of the different MFCs is required to properly assess their performance.

Carbonaceous materials are the main types of electrodes, which include carbon felts, cloths, sheets, and bars. Such low-cost materials are biocompatible, good electrical conductors, corrosion resistant, besides having a high surface area (Santoro et al. 2017). Current density values ranged from 0.002 to 0.09 mA cm^{-2} in studies using activated carbon as the anode (Table 1). On the other hand, Cooney et al. (1996) and Zhao et al. (2008) reported significantly higher current densities (1.7 mA cm^{-2} and 1.3 mA cm^{-2}), respectively, when platinum impregnated cathodes and lactate (carbon source) were selected. Thus, the electrode material is the key component in the production of electricity in MFCs using electroactive SRB. Therefore, low-cost and recyclable alternatives must be investigated for this parameter in order to devise a feasible technology (Goglio et al. 2019).

In all the studies listed in Table 1, sulphate and COD removals were greater than 60% irrespective of the carbon source, suggesting therefore that this is not a limiting factor in MFC inoculated with SRB. Nevertheless, it must be added

Table 1 Review of studies with microbial fuel cell for removal of COD and SO_4^{-2}

Carbon source	MFC type	SO_4^{-2} removal	DQO removal	Current density (mA cm^{-2})	Reference
Glucose	Single chamber; cation exclusion membrane (CEM); graphite electrodes and metal impregnated anode	8.3%	Without mention	32	(Habermann and Pommer 1991)
Lactate	Single chamber; proton exclusion membrane (Nafion®); activated carbon anode	75%	Without mention	1.7	(Cooney et al. 1996)
Lactate	Single chamber; Nafion®; carbon electrodes; cathode with Pt	99%	Without mention	1.3	(Zhao et al. 2008)
Lactate	Double chamber; CEM (CMI-7000); carbon electrodes[a]	84%	24%	0.07	(Lee et al. 2012)
Lactate	Double chamber; Nafion®; carbon electrodes[a]	61%	94%	0.03	(Chou et al. 2013)
Ethanol	Electrochemical system of three electrodes (single chamber, without membrane); cloth activated carbon electrodes	87%	76%	0.05	(Liang et al. 2013b)
Lactate	Double chamber; Nafion®; carbon electrodes[a]	99%	Without mention	0.02	(Lee et al. 2014)
Lactate	Double chamber; Nafion®; graphite electrodes[a]	Not evaluate	98%	0.02	(Kang et al. 2014)
Lactate	Single chamber; Nafion®; carbon anode and Ag screen cathode	18%	57%	0.02	(Sangcharoen et al. 2015)
Landfill leachate + Lactate	Double chamber; Nafion®; graphite electrodes[b]	Without mention	100%	0,02	(Kumar et al. 2017b)
Glucose	Single chamber; Nafion®; carbon electrodes, cathode with Pt	43%	56%	0.003	(Niyom et al. 2018)
Lactate	Double chamber; Nafion®; graphite electrodes and cathode with microalgae	Without mention	Without mention	0.002	(Kumar et al. 2019)
Lactate; glucose	Single chamber; (CMI-7000); graphite bar electrodes	93% (lactate); 90% (glucose)	78% (lactate); 68% (glucose)	0.09 (lactate); < 0.01 (glucose)	(Bratkova et al. 2019)
Lactate + simulated textile effluent	Double chamber; Nafion®; graphite electrodes[c]	> 95%	> 85%	0.06	(Miran et al. 2018)

[a] Catholyte: ferricyanide + phosphate buffer

[b] Catholyte: $KMnO_4$ + phosphate buffer

[c] Catholyte: phosphate buffer

that both the pH and COD/SO_4^{-2} ratio are key parameters in the performance of both sulphate and COD removals in anaerobic reactors (Bertolino et al. 2012; Bertolino et al. 2015; Isosaari and Sillanpää 2017; Kaksonen and Puhakka 2007). Theoretically, a COD/SO_4^{-2} ratio of 0.67 would enable SRB growth and, consequently, sulphate removal (Isosaari and Sillanpää 2017). However, quite a few studies have proposed that COD/SO_4^{-2} ratios around 2 are required if the sulphate reduction is to improve (Bertolino et al. 2012; Bertolino et al. 2015; Bratkova et al. 2019; Miran et al. 2018). Working with COD/SO_4^{-2} ratios below 2 may justify the results presented in Table 1, in which the sulphate removal yields were low (Niyom et al. 2018; Sangcharoen et al. 2015). Similar finding was observed when the COD removal was also low (Lee et al. 2012; Niyom et al. 2018; Sangcharoen et al. 2015). On the other hand, a correlation between sulphate and COD removal with electricity production was not established so far, and this is one of the challenges to be faced by the scientific community.

An important parameter in sulphate reduction in MFC is pH because it defines the concentration of H_2S in the reactor, which inhibits sulphate reduction and bacterial growth. The maximum sulphide S^{2-} or H_2S concentration tolerated by SRB was proposed to be 230 mg L^{-1} at pH 7 (Cooney et al. 1996). Liang et al. (2013b) determined the effect of pH on the performance of a MFC inoculated with SRB and reported the best results when the pH was in the 6.5–8.5 range.

The configuration of the MFC, single or double chamber, could not be related to either the production of energy or to sulphate removal, similarly to what was observed with carbonic substrate.

Recently, studies in which a MFC is used for sulphate reduction associated to other technological important process are being published (Eaktasang et al. 2013; Kumar et al. 2019; Miran et al. 2018; Zhao et al. 2008). For instance, Miran et al. (2018) reported significant sulphate removal in a MFC treating azo compounds. Another promising application is to associate sulphate reduction in the anode to algae growth in the cathode, as Kumar et al. (2019) observed that increasing the lipid content in the cathodic half-cell improved the production of electricity. This is an interesting proposition because the algal biomass can be converted subsequently to biodiesel. Furthermore, an advantage of bio-electrochemical systems over conventional biological sulphate removal methods is the oxidation of sulphide ions to elemental sulphur because the accumulation of sulphide in conventional systems inhibits SRB growth, in addition to causing corrosion and malodour issues (Eaktasang et al. 2013; Zhao et al. 2008).

New studies should be focused on the selection of the most appropriate electrode material, and on the COD/SO_4^{-2} ratio so that the maximum COD and SO_4^{-2} removals and also energy production are achieved.

Conclusions and perspectives

The pathways of electron transfer to a solid electrode by SRB are well established. These microorganisms transfer electrons directly to the anode surface either by using cytochrome C-type periplasmic proteins or by producing conducting nanowires. In addition, electron transfer may also occur via active metabolites such as FeS. Sulphide oxidation by SOB is also widely regarded as an electricity producing pathway in these systems. In any case, a microbial consortium is essential for sulphate and COD removal, as in any anaerobic system, and also for electricity production.

Regarding the operating conditions: electrode type and COD/SO_4^{-2} ratio are the main parameters controlling sulphate reduction and the simultaneous production of electricity. The success of the technology will be achieved when these parameters were optimized

Summarizing, the development of the MFC technology requires a multidisciplinary approach to find alternatives sources of electrical energy. It symbolizes the confluence of chemical, physical, and life sciences and is a meeting point for basic and applied research.

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