MEMBRANES IN MICROBIAL FUEL CELLS: A REVIEW

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**ABSTRACT**

Currently, most of the Microbial Fuel Cells (*MFC*s) are devices designed and focused to produce electrical energy and waste treatment. We can describe a *MFC* constituted mainly by the following items: anode, cathode, microorganisms, substrates and proton exchange membrane (*PEM*). Usually, the role of a *PEM* is to divide the electrodes, the anode from the cathode in a *MFC*. Regarding the costs, the use of membranes could represent around the 40% of the *MFC* total cost.

The aim of this work was to critically review the state of the art on membranes in *MFC*s. The scope of this review includes (*i*) the different membranes used in *MFC*s, (*ii*) costs and performance of typical membranes used, (*iii*) configuration of membranes in *MFC*s, (*iv*) membraneless *MFC*s,and (*v*) perspectives on *PEM*s.

An ideal membrane separator must have some characteristics such as high proton conductivity, biocompatibility, good mechanical properties, low oxygen transfer, low permeability to substrate, good chemical and thermal stability, and low cost. The most common membrane used is Nafion®, a perflourinated membrane, due to their excellent proton conductivity, nevertheless Nafion is too expensive. Nafion belongs to the type of cation exchange membranes, also known as *PEM*s.

In order to reduce the costs, other types of proton exchange materials have been tested in *MFC*ssuch as salt bridge, anion exchange membranes, microfiltration and

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ultrafiltration membranes, bipolar membranes, among others. However, studies focused on low cost and or natural polymers for *PEM* are still scarce. Alternatively, in some works, the *MFC*s have been operated without a membrane. Yet it was found that the *CE* substantially decreased.

Because of the type of membrane affects the performance and the total cost of the *MFC*s*,* is necessary to increase efforts to develop new, more economic membranes that exhibit good properties and allow for good cell performance at the same time.

Key words: proton exchange membrane, Microbial Fuel Cell, membraneless, Nafion® 117

**Introduction**

Nowadays, the fossil fuels have been used extensively; however, it is not a renovable source of energy. Furthermore, their use has induced serious environmental problems (Das and Veziroglu 2001; Logan and Regan 2006; Yang *et al*., 2011). In order to solvent the fossil fuels problems, new efforts towards renewable alternative energy sources have been developed (Cheng-Dar *et al*., 2011; Tye *et al*., 201; Narayanaswamy *et al*., 2014).

Microbial fuel cells (*MFC*s) are proposal as a new renewable alternative energy source (Poggi-Varaldo *et al.,* 2009; Narayanaswamy *et al*., 2014). *MFCs* are an old concept. The first background was introduced by Potter, 100 years ago approximately (Potter 1911). He observed an electrical current generation by the microorganisms in presence of organic compounds (Potter 1911; Schröder 2007; Logan 2012). Recently, in the last two decades, the interest on research the *MFC*s began to grow (Liu and Logan 2004; Min *et al*., 2005; Kargi and Eker 2007; Logan 2007; Vazquez-Larios *et al.,* 2010; Modin and Gustavsson 2014).

A *MFC* is a device able to produce electrical energy by the oxidation of a variety of organic and inorganic matter present in wastewater. Obtaining energy from the oxidation of the substrates is carried out by microorganisms under anoxic conditions, these microorganisms commonly referred biocatalysts. (Li *et al*., 2011; Pandit et al., 2011; Logan 2007; Poggi-Varaldo *et al.,* 2009). Thus, A *MFC* is a bioelectrochemical system able to covert the dissolved organic or inorganic matter into electricity (Modin and Gustavsson, 2014; Poggi-Varaldo *et al.,* 2009; Vazquez-Larios *et al*., 2010). Since this overview, the *MFC*s constitute an interesting proposal for electrical energy recovery and wastewater treatment, at the same time (Logan and Rabaey, 2012; Hou *et al*., 2014).

As in other technologies, the researchers are working towards to scaling these devices (Dewan *et al.,* 2008; Hsu *et al*., 2013). They expect to improve the performance producing attractive amounts of energy and reducing the cost. However, there are some factors limiting the practical application (Li *et al*., 2011; Zhou *et al*., 2011).

In a *MFC,* a lot of technical and engineering variables are involved. All of them must be evaluated (Logan 2007). A wide variety of knowledges such as Biology, Microbiology, Physic, Chemistry, Electrochemistry, Physical Chemistry, Mathematics, wastewater, environmental engineering, Biology Molecular, among others, are necessary to design, analyzed the dates obtained and understand the operation of *MFC*s (Logan *et al*., 2006; Logan 2012).

Inside of *MFC,* at least two electrodes are presents: anode and cathode. The anaerobic anodic chamber and aerobic cathodic chamber are commonly divided by a separator. The typical separator used in *MFC*s is a proton exchange membrane (*PEM*), Nafion® 117 (Logan 2007; Narayanaswamy *et al*., 2014; Kim *et al*., 2014).

In the anodic compartment, the biocatalysts anaerobically oxidize the substrate and release electrons and protons. By the one hand, the electrons are collected by the anode to transport to the cathode by and external circuit (Poggi-Varaldo *et al*., 2009; Vazquez-Larios *et al*., 2010). On the other hand, the protons diffuse through the liquor and *PEM* of the *MFC* to reach the cathodic section. At the cathode, the protons react with electrons and molecular oxygen from the air producing water in what is known as the oxygen reduction reaction (*ORR*) (Logan *et al*., 2006; Yang *et al*., 2010). It is illustrated in the figure 1.

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**FIGURE 1. The membrane separator and the reduction reaction at cathode.**

In the operation of the *MFC*s, there are a lot of factors affecting the *MFC* performance. The material and surface area of electrodes, the catalyst used at the cathode and the biocatalysts at the anode, the internal resistance (*Rint*) of the *MFC* and the external resistance used along the operation, electrolytic conductivity of the solution, distance between the anode and cathode, the membrane, among others (Logan 2007; Dewan *et al*., 2008; Wei *et al*., 2011; Cho *et al*., 2006; Shahgaldi *et al*., 2014). However, the membrane is one of the most important factors in *MFC* configurations and it depicts by itself, around the 40% of the *MFC* total cost (Ghasemi *et al*., 2013; Shahgaldi *et al*., 2014).

The presence or absence of these pieces is reflected directly in the performance and the cost.

Nafion membrane is the membrane choosen by the researches to use in fuel cells, includen *MFC*s (Logan *et al*., 2006; Rabaey and Verstraete 2005). Nowadays, Nafion has the best characteristics to use in *MFC*s. However, notwithstanding Nafion membranes are used as the best membranes for *MFC*s, its high price is discouraging to use in a big scale and also regarding the *MFC* performance it is affected by the increasing of the *Rint* (Rozendal *et al*., 2008; Sivasankaran and Sangeetha, 2011). Both are two big challenges to overcome.

The studies on *MFC*s as a new technology have awoken a big interest in the researchers. New alternatives membranes to Nafion, needs to be developed. Some considerations must be taken in account for propose new alternatives membranes or separators. To develop new alternative membranes to apply in *MFC*s is a difficult task, there are limited works related to new alternative membranes tested in *MFC*s (Grzebyk and Poźniak 2005; Li *et al*., 2011; Choi *et al.,* 2012; Kim *et al*., 2014).

The aim of this work was to critically review the state of the art on membranes in *MFC*s. The scope of this review includes (*i*) the different membranes used in *MFC*s, (*ii*) costs and performance of typical membranes used, (*iii*) configuration of membranes in *MFC*s, (*iv*) membraneless *MFC*s,and (*v*) perspectives on *PEM*s.

**Membrane separator functions in MFCs**

A membrane separator has some important functions in *MFC*s. One of them is to prevent the short circuiting between the electrodes (Figure 2).

By the other hand, the membrane acts as a channel to conduct the protons to the cathode and avoid the oxygen diffusion into the anodic compartment from the cathode. The substrate flux from the anode to cathode is also reduced by the membrane separator (Figure 2) (Logan 2007).



**FIGURE 2. A *MFC* divided by a membrane separator.**

The oxygen in the anodic chamber affects negatively the *MFC*s performance(Watson *et al*., 2011). The oxygen is an alternative electron acceptor for the biocatalyst at the anode. It plays versus anode and is toxic to them.

In the case of the facultative bacteria (also present in the anodic chamber), the oxygen is used as the first option as terminal electron acceptor instead of the anodic material, then the Coulombic efficiency (*CE*) decrease. In the case of the anode potential, it become more positive, decreasing the current density (Harnisch and Schröder 2009; Watson *et al*., 2011).

The *EC* is one of the dates that must be included in the data reported as *MFC* performace. The *CE* (*ɳcoul*) is defined as the transfer efficiency of the available electrons to the anode. It is the total Coulombs actually transferred to the anode from the substrate, to maximum possible Coulombs if all substrate removal were converted in current (Logan *et al*., 2006; Logan 2007; Li *et al*., 2011; Liu and Logan 2004).

The *ɳcoul* is calculated as follow (Poggi-Varaldo *et al*., 2010):

(1)

where *ACS* is the actual charge due the substrate obtained by the Eq. 2 and *CTS* is the theoretical charge due the substrate calculate by the Eq. 3.

(2)

(3)

*IMFC* current intensity over time delivered by the cell

*F* is Faraday’s constant, 96 485.33 Coulombs/mol *e-*

*bCOD* is the number of electrons exchanged per mole of oxygen generated by chemical oxygen demand (COD), 4

COD*i* initial COD (g/L)

*CODf* final COD (g/L)

*V* the volume of liquid in the anode compartment (L)

*MCOD* molecular weight of oxygen (32 gO2/mol COD)

However, the *ɳcoul* is adversely affected by others factors such as the own bacterial growth, competitive processes and the utilization of alternate electron acceptors in the influent by the microorganisms (Logan *et al*., 2006)

Regarding to the *Rint* of the *MFC*s, the presence of a membrane separator brings by itself the increasing of *Rint* affecting adversely the power generation (*Kim et al., 2014*)*.*

Also the membrane, along the operation generates pH gradients (Rozendal *et al*. 2006; Kim *et al*., 2007).

One alternative to improve the *MFC* performance is removing the membrane from the system and decreasing the distance, between the electrodes. Both, reduce the ohmic losses (Liu and Logan 2004; Logan *et al.,* 2006; Vazquez-Larios *et al*., 2010).

**Ideal membranes**

Amembrane must perform a lot of characteristics to be considerate as an ideal membrane. Some researchers characterize their membranes out of the cell before to apply to the *MFC* (Hernández-Flores 2014; Narayanaswamy *et al*., 2014; Kim *et al*., 2014). The most of them synthesize and apply the membrane separators directly in *MFC*s, evaluating the membrane *in situ.*

A membrane could be considerate as an ideal membrane to use in a *MFC* whether has the follow properties: a high proton conductivity, waterproof, low thickness, impermeability to the oxygen and cations such as Na+, K+, Ca2+, Mg2+ and NH4+, good mechanical properties, chemical stability, low values of *Rint*, impermeability to gases such as H2, N2 and definitely a low price. Regarding using the membrane directly in *MFC* we can add the following properties: low average *Rint* of the device, long stability or durability along the operation time under different operating conditions, biofouling resistance, nonbiodegradable, high *CE* and an attractive overall power density(Peighambardoust *et al*., 2010; Li *et al*., 2011; Watson *et al*., 2011; Rismani-Yazdi *et al.,* 2011; Brunetti *et al*., 2012; Logan 2010; Kim *et al*., 2014).

Unfortunately, there is not an ideal membrane. There are membranes that depict some of the most important properties and this allows the membranes can be applied (used) directly in *MFC*s.

In the case of Nafion membranes (perflourinated membranes), these *PEM*s shows the best characteristics of the main properties membrane. It is the model membrane worked in a *MFC*. The main properties of Nafion**®** membrane are relative high proton conductivity, chemical stability, and excellent mechanical properties; nevertheless the high cost of this membrane makes it not viable (Logan 2007; Rismani-Yazdi *et al.,* 2011; Hernández-Flores, 2014; Lou and Pu 2011).

The table 1, shows the Nafion**®** specific characteristics.

**TABLE 1. Nafion® 117 characteristics.**

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| --- | --- |
| **Membrane specifications** | **Nafion® 117** |
| Proton conductivity (S/cm) | 2.0×10-3 |
| Thermal stability (°C) | 90 |
| IEC (meq/g) | 1.23 |
| Water swelling (%) | 22 |
| Thickness (cm) | 0.019 |
| K0 | 1.6×10-5 cm/s |
| Cost ($/m2) | 1733 |

Modified from Sivasankaran and Sangeetha, 2011.

We have mentionated a*PEM* is an essential component in the configuration of the *MFC*s. Below we highlight the advantages using membranes as separators in *MFC*s (Liu and Logan, 2004; Logan, 2008; Li *et al*., 2011):

* Increasing the selective passage of protons
* Separating the anodic from the cathodic chamber
* Increasing the *CE*, reducing the oxygen flux from the cathode chamber to the solution in the anode chamber
* Keeping the anoxic conditions for the biocatalysts in the anode chamber
* Avoiding the transfer of other ions between the chambers
* Prohibiting the substrate flux from the anode to cathode
* Ensuring an efficient and sustainable operation along the time
* Isolating the catalyst from the cathode in single-chamber *MFC*s

However, also there are technical and economical disadvantages related to the *PEM* use directly in *MFC*s. The main one is the high cost of standard membranes (Liu and Logan, 2004; Logan, 2008; Li *et al*., 2011). Logan (2007) reported that Nafion® can cost $1400/m2. Currently the cost has increasing up to $1733/m2 (Hernández-Flores, 2013). Furthermore, its use affects negatively the power generated due to the increasing of *Rint* (Logan, 2007; Li *et al*., 2011; Xia *et al*., 2013).

**Membranes used in MFCs**

A great variety of different membranes and separators have been used by the researchers. The aim of them is lower costs; reduce *Rint* and increase power output and *CE,* improving the membrane separator as key component.

At the beginning of the *MFC*s development technology, a salt bridge was applied instead of a membrane separator. In another cases the *MFC*s were operated without a separator (Karube *et al*., 1977; Kim *et al*., 2014).

In search of new membrane or separators able to reach the properties Nafion and reduce cost, many membranes or separator have been studied. Here there are some of them: Zirfon® membrane, sulphonated polyether ether ketone membrane (SPEEK), ultrafiltration and microfiltration membranes, anion and cation exchange membranes, bipolar membrane, forward osmosis membrane, Cloth (J-cloth) separator, glass fiber separator, cation exchange layer made of purified kaolin, porous porcelain coated with Nafion-117 solution, polystyrene and divinylbenzene with sulfonic acid groups, dialysis membranes, organic polymers, spray coated to thin layers of hydrophilic cation exchange, anion exchange and neutral polymers, porous fabrics and course-pore filter material, selemion HSF, polytetrafluoroethylene membrane, isoporo membrane filter, biomax ultrafiltration disc, glass wool, nylon membranes, polycarbonate membranes, cellulose nitrate membranes, kaolin, porcelain and polyethylene membranes interpolymer, forward osmosis membrane (Grzebyk and Pozniak, 2005; Min *et al*., 2005; Logan *et al*., 2006; Logan and Regan 2006; Logan, 2007; [Zhang](http://pubs.acs.org/action/doSearch?action=search&author=Zhang%2C+X&qsSearchArea=author) *et al*., 2009; Yang *et al*., 2010; Pant *et al*., 2010; Lefebvre *et al*., 2011; Li *et al*., 2011; Sivasankaran and Sangeetha, 2011; Wei *et al*., 2011; Zhang *et al*., 2011; Watson *et al.*, 2011; Hernández-Flores, 2014).

**Membraneless MFCs**

*MFC*s without a separator or membrane were one of the first configurations used in the principle of *MFC*s (Min *et al*., 2005; Kim *et al*., 2014).

The basis to propose *MFC*s operating without membranes or separators is due to the water has the property of conducting the protons by itself (Logan 2007). Then,the membrane is not strictly necessary in a *MFC* configuration.

There are some disadvantages using Membraneless *MFC*s. The absence of a separator allow the substrate and oxygen diffusion, it affects reducing the *CE* and microorganism activity bioelectrocatalytic (Liu *et al*., 2005; Yang *et al*., 2010; Liu and Logan, 2004; Logan, 2007; Li *et al*., 2011; Logan *et al*., 2006).

Regarding the substrate diffusion, this causes a rapid formation of biofilm at the cathode, it is called biofouling. This causes deactivation of the cathode and reduce the *MFC* performace (Tartakovsky and Guiot, 2006).

Liu and Logan (2004) used a membrane-less *MFC* to the bioelectricity generation. Their objective was to increase the energy output and reduce the cost. The results obtained by them were a power density of 146 ± 8 mW/m2 and 20% of *CE* for their membrane-less *MFC*. In contrast, their *MFC* fitted with Nafion membrane displayed a power density of 28 ± 3 mW/m2 and 28% of *CE*.

In another case, Liu *et al*., 2005 and Cheng *et al*., 2006 proposed a configuration to reduce the oxygen diffusion. They separated the electrodes in the *MFC,* however it increase the *Rint*.

The researches have concluded a separator or membrane is necessary to ensure an efficient and sustainable MFC operation (Harnisch and ShrÖder, 2009).

**Perspectives on PEMs**

The challenge of developing alternative membranes to use in*MFC*sis tasktaken only by a few researchers(Kim *et al*., 2014). However, to find a new membrane able to reach attractive *MFC* performance with a low cost is necessary for a cost-effective scaling upof the *MFC*s.

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**Notation**

CE coulombic efficiency

MFC microbial fuel cell

ORR oxygen reduction reaction

PEM proton exchange membrane

Rint internal resistance

SPEEK sulphonated polyether ether ketone

*Greek characters*

ηcoul  coulombic efficiency

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