Revue des Energies Renouvelables Vol. 17 N°2 (2014) 279 – 289
DOI: https://doi.org/10.54966/jreen.v17i2.442

Comparison of the chemical removal rates of a low cost microbial fuel cel and an aerated activated sludge bioreacteur, and evaluation of its performances

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(reçu le 10 Mai 2014 – accepté le 30 Juin 2014)

Abstract - *Microbial fuel cells can be used to reduce the organic load of a wastewater, while producing electricity at the same time. However, most tests on MFCs are done in controlled laboratory environments using synthetic wastewaters, and MFCs that contain expensive catalysts and membranes. In this work, we studied the performances of a low-cost microbial fuel cell that was made with stainless steel electrodes and no catalysts, and The COD removal in the MFC was compared to the one obtained in an aerated activated sludge bioreactor. It was found that the MFC could be used for water treatment, an 80% COD removal was reached after 15 days, compared to 90% after 10 days in the aerated reactor. However, the power densities were quite low, a maximum of 2.5 mW/m^2 could be reached, this was due to high activation losses, a high internal resistance, and the presence of few electrogenic bacteria in the biocatayst.*

Résumé - *Les piles à combustible microbiennes peuvent être utilisées pour réduire la charge organique d'un effluent, tout en produisant de l'électricité en même temps. La plupart des expériences sur les piles à combustibles microbiennes sont réalisées en conditions artificielles en laboratoire, en utilisant des effluents synthétiques. Aussi, les piles utilisées contiennent des membranes et des catalyseurs coûteux. Dans ce travail, on a étudié les performances d'une pile à combustible microbienne 'low-cost', contenant des électrodes en acier inoxydable sans catalyseurs. La vitesse de réduction de la demande chimique en oxygène de la pile a été comparée à celle d'un bioréacteur aérobie inoculé avec des boues activées. On a trouvé que la pile pouvait être utilisée dans le traitement des eaux, un taux d'abattement de la DCO de 80% a été obtenu après 15 jours, tandis qu'un taux d'abattement de 90% a été obtenu après 10 jours pour le réacteur aérobie. Les densités de puissance étaient cependant faibles, un maximum de 2.5 mW/m² a été atteint. Ces valeurs sont dues à des pertes d'activations et une résistance interne élevée.*

Mots clés: Piles à combustibles microbiennes - Traitement des eaux – Bioprocédés.

1. INTRODUCTION

The fast industrialization of the last century led to an incredible economical growth and improved life quality in most industrialized countries.

However, this also led to an increase in the use of fossil energies, which had an undeniable impact on the environment, as CO_2 emissions caused dramatic climatic changes (Lewis *et al.*, 2006). Humans also rapidly became dependent on this form of energy which is non renewable, meaning that it won't be able to indefinitely sustain the world's economy. Because of this, new and sustainable energy sources must be found.

Another problem that needs to be addressed is the water problem. Many industries

generate large amounts of wastewaters that contain numerous pollutants. These effluents must be treated to prevent the contamination of our scarce water resources, and to allow the reuse of these waters.

The microbial fuel cells, or MFCs might contribute in solving both of these problems at the same time, indeed, they can convert the organic pollutants present in the wastewaters into electricity, reducing they organic load at the same time.

The advantage of the MFCs, compared to conventional biological water treatment systems, is that they operate in anaerobic conditions, thus, there is no need to continually aerate the bioreactor, and this leads to a reduction of the energy used in the water treatment process.

Although this technology appears to be promising, many hurdles must first be overcome to make it a viable water treatment and alternative energy source. Expensive catalysts and membranes are commonly used to improve the performances of the MFC, because of this, they are mostly used in laboratory settings, and very few authors worked on MFCs being fed a real industrial effluent, in uncontrolled settings. Because of this, the behavior of the MFCs in these more realistic conditions are not known.

In this work, we evaluated experimentally the performances of a MFC in which stainless steel electrodes were used, with no catalysts added, with a real industrial wastewater used as a 'fuel', and a biocatalyst that underwent a one month acclimatization. We also compared the rate of the reduction of the chemical oxygen demand in the MFC with the reduction rate in an aerated bioreactor inoculated with activated sludge.

2. MICROBIAL FUEL CELLS

2.1 Definition

Microbial fuel cells are electrochemical devices in which the chemical energy of organic compounds are converted into electricity by microorganisms directly, without the need to produce an intermediary fuel such as methane or ethanol.

It was discovered that some bacterial strains were able to generate electricity in anaerobic conditions, in microbial fuel cells (Kim *et al.*, 1999; Kim *et al.*, 1999b). the microorganisms will oxidize the organic matter present in the medium in which they are, and the electrons involved in this reaction are then transferred to an anode (Rabaey *et al.*, 2005). Protons are also produced during this reaction, and they must be allowed to migrate to the cathode, so they can combine with oxygen and the electrons to produce water (Logan, 2005).

Figure 1 shows a schematic diagram of a MFC and its main components. An anodic chamber containing the anode, microorganisms that act as biocatalysts, and the 'fuel" which is a wastewater, or more precisely the organic matter present inside it, or a culture broth whose composition is known and controlled. The cathodic chamber containing the cathode and the catholyte is separated from the anodic chamber by a proton exchange membrane that theoretically only allows protons to pass from the anodic to the cathodic chamber

Oxygen is used as an oxidant, and for economical reasons, air is usually injected in the cathodic chamber. The proton exchange membrane should limit the passage of oxygene to the anodic chamber, since it will be used as a final electron acceptor by microorganisms and in this way will reduce electricity production.

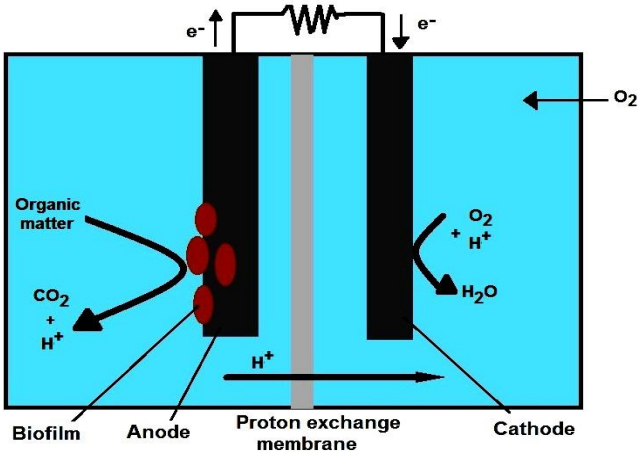

Fig. 1: Schematic of the main components of a microbial fuel cell

The typical reactions that take place in the MFC, with acetate used as a substrate, are:

$$\text{Anodic reaction-}\ CH_3COO^- + 2H_2O \Leftrightarrow 2CO_2 + 8H^+ + 8e^-$$

$$\text{Cathodic reaction-}\ O_2 + 4e^- + 4H^+ \rightarrow 4H_2O$$

The overall reaction is the breakdown of the substrate to carbon dioxide and water with a concomitant production of electricity as a by-product. Based on the electrode reaction pair above, an MFC bioreactor can generate electricity from the electron flow from the anode to cathode in the external circuit (Du *et al.*, 2007).

2.2 Electron transfer to the anode

The electron transfer mechanism is poorly understood for the time being, but two main type of electron transfers have been identified, direct electron transfer, and transfer via electron shuttles, or mediators.

2.2.1 Direct electron transfer

Microorganisms can transfer electrons directly to the anode either by direct contact between some specialized membrane proteins that are involved in the respiratory pathways and are in contact with the exterior of the cell (Lower *et al.*, 2001; Lovley *et al.*, 2004; Vargas *et al.*, 1998), or through nanowires, that are long conductive cell expansions that transport the electrons from the respiratory proteins to the electrode surface (Gorby *et al.*, 2005).

To be able to transfer electrons to the anode in this way, the microorganisms must first adhere to the electrode's surface and form a biofilm.

2.2.2 Electron shuttles

In the fist experiments carried out on microbial fuel cells, external electron shuttles like neutral red (Park *et al.*, 1999), anthraquinone-2-6,disulfonate (AQDS), thionin, potassium ferricyanide (Bond *et al.*, 2002), were added to the culture medium in the anodic chamber, although the results obtained were promising, systems in which external mediators are added cannot find any application.

It was recently discovered that some microorganisms are able to produce their own electron mediators, the most notable example being *Peudomonas aeruginosa* who produces pyocyanins that can effectively transfer electrons to the anode (Rabaey *et al.*, 2004).

2.3 Microorganisms used in MFCs

Very few microorganisms that are able to generate electricity in a MFC have been identified. There are few isolates that have been obtained from an MFC, with most of these obtained by plating using iron as an electron acceptor. So far, there does not appear to be an isolate capable of producing power densities that are as large as those produced by mixed cultures in the same MFC (Logan, 2008).

3. MATERIALS AND METHODS

3.1 Biocatalyst

Activated sludge was collected from the aeration tank in the Baraki wastewater treatment plant, and underwent a one month acclimatization period, during which increasing concentrations of the industrial wastewater used in this study were introduced in the culture medium in anaerobic conditions.

The acclimated anaerobic sludge was then introduced in the anodic chamber of the MFC as a biocatalyst.

Another sample of the same activated sludge collected in the Baraki wastewater treatment plant was left in aerobic conditions and introduced in an aerated bioreactor. This bioreactor was used as a point of comparison to evaluate the COD reduction properties of the MFC

3.2 Electrodes

Stainless steel electrodes were used in this study. Stainless steel was chosen because of its good mechanical properties and resistance to corrosion.

Moreover, these electrodes were biocompatible, and didn't have any inhibitory effect on the metabolic activity of the microogranisms.

They were used without any type of pretreatment, and to reduce the cost of the MFC, no catalyst coatings were added to them. Insulated copper wires were fastened to the electrodes using epoxy.

3.3 Wastewater

A real industrial effluent was used for this study, it was collected in plastic bottles in a dairy processing plant in Algiers. The wastewaters didn't receive any kind of pretreatment before being introduced into the MFC.

The characteristics of the effluent are given in **Table 1**:

Table 1: Characteristics of the effluent

DCO (mg/l O_2)	pH	Conductivity (mS/cm^2)	Nitrites (mg/l)	Nitrates (mg/l)	Ammonium (mg/l)	Phosphate (mg/l)
400-800	7.8-9	0.2	3.5	5.7	67	17

3.4 Chemical oxygen demand

To evaluate the oxidation of organic matter in the wastewater, the COD was measured. Although this method is used to measure the amount of matter that could be oxidized and not only biodegradable organic matter, it is faster and more convenient than other tests such as the measurement of the biochemical oxygen demand.

Also, it is a good approximation if we consider that all of the oxidation reactions that take place in the MFC are catalyzed by the microorganisms.

A colorimetric method was used to measure the COD using potassium dichromate as an oxidizing agent under acidic conditions.

3.5 Microbial fuel cell

For this study, a two chamber MFC was used in batch mode, the anodic and cathodic chambers both had a volume of 15 ml.

The anodic chamber contained the biocatalyst (i.e., the acclimated anaerobic sludge) and the wastewater in a 1:10 ratio. It was tightly sealed after being filled to maintain an anaerobic atmosphere. A sampling port, closed using a rubber plug when the MFC is operating and when no sampling is done, was used to collect samples at regular intervals using syringes.

The samples were then centrifuged, and the COD of the supernatants was measured.

Also, a tube going out of the top of the anodic chamber and going to a beaker filled with water was used to allow gases produced during the biological activity to be evacuated while preventing air, and oxygen, to enter.

On the other hand, the cathodic chamber was left open and was aerated passively.

An external load of 1 kΩ was used to close the circuit when the MFC was under normal operating conditions.

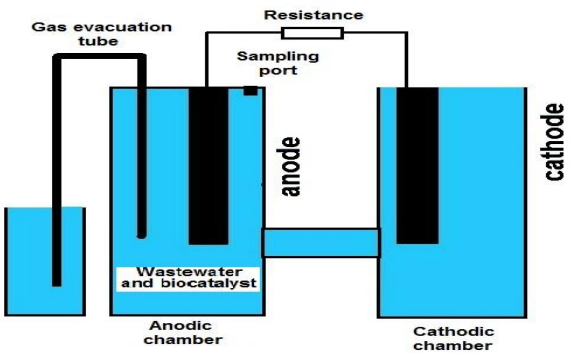

Fig. 2: Diagram of the MFC used for the experiments

3.6 Aerated bioreactor

Activated sludge and wastewaters were added to a beaker in a 1:10 ratio. The mixture was constantly stirred, and a small air pump was used to inject aerate the mixture.

Samples were collected at regular interval from the beaker

3.7 MFC performance tests

Polarization curves were obtained by recording the stable cell voltage produced across different external resistances. And since no consensus exists in the field of

microbial fuel cells concerning external loads used to evaluate electrical power production, a 1 kΩ resistor was used as a load since it is the most used one in the literature, and the cell voltage was measured at regular intervals to obtain the power *vs.* time curve.

4. RESULTS AND DISCUSSION

4.1 Chemical oxygen demand reduction

The evolution of the chemical oxygen demand was followed in both the MFC and the aerated bioreactor. Since the COD reduction was much slower in the MFC the sampling intervals were bigger, and the period during which this experiment was carried out was longer.

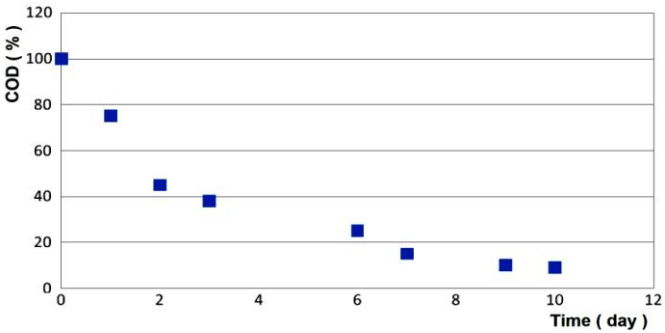

Fig. 3: COD evolution in aerated bioreactor

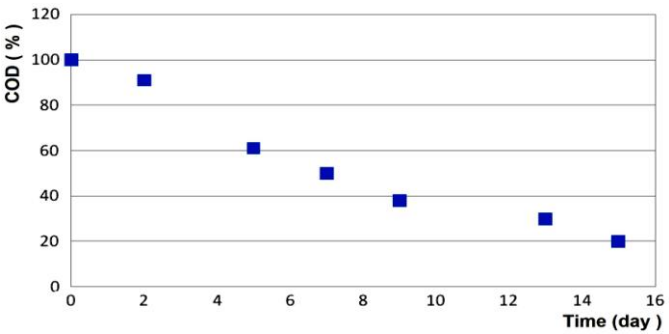

Fig. 4: COD evolution in MFC

Unsurprisingly, the COD reduction in the aerated bioreactor was good, as the efficiency of this type of bioreactor in the treatment of industrial wastewaters was already shown (Moletta *et al.*, 1999) as it can be seen in figure 3, the reduction is fast during the first 3 days after which 60 % of the COD was reduced, after that, the COD removal rate decreases. After 10 days, a COD reduction of 90 % is achieved.

This COD reduction could be considered slow when compared to the results of Castillo de Campins, (2005) who reached a maximum COD removal of 90 % after only three days using an aerated bioreactor containing activated sludge and a synthetic wastewater prepared using whole milk.

This difference in performance could be explained by the fact that the real effluent

used in our study contained detergents and disinfectants that are commonly used to inhibit bacterial proliferation in industrial installations, and it is well known that these substances reduce the performances of biological water treatment systems (Nadais *et al.*, 2010).

In the MFC, although acceptable, the COD reduction is much slower, after 3 days, only 20 % of the organic matter is oxidized, and after 10 days 60 % of it is oxidized. We also notice that the digestion of the organic matter slows down a little after day 8.

Finally, it took 15 days to reach an 80 % reduction of the COD, 10 % lower that the one observed in the aerated bioreactor.

Although the COD removal is slower and less efficient in the MFC, it must be noted that the reaction took place in anaerobic conditions, so no aeration device was used, in other words, no energy was used to promote the oxidation of organic matter present in the wastewater by the microorganisms.

In a laboratory scale experiment, the energy used for the aeration of the bioreactor is low, however, in large scale applications, such as in water treatment plants, more energy is needed. According to a study in which the energy consumption of almost 1,000 wastewater treatment plants in Japan was evaluated, 1.0 to 7.5 MJ/m^3 are needed, and in most of the cases, biological treatment represented half of the total operating costs (LA kennedy et Rtsuchashi, 2005; Elías-Maxil, 2013).

MFCs could be used to replace, or complement conventional biological treatments and reduce considerably the energy consumption in water treatment plants.

4.2 Power generation in the MFC

The recording of the power started as soon as the anodic chamber was filled with wastewater and inoculated.

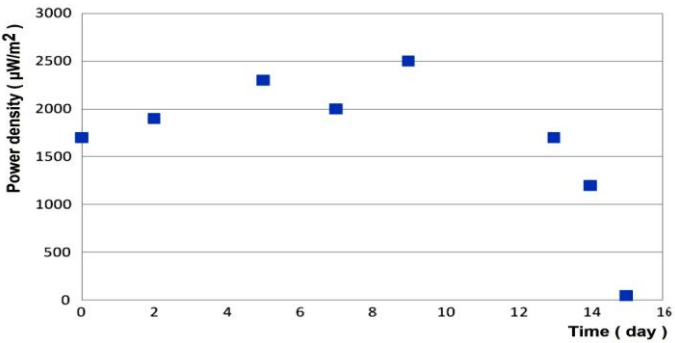

Fig. 5: Evolution of the power densities in the MFC

We observed one cycle of power generation in batch mode, it can be seen that power started being generated immediately after inoculation, while this startup period was longer for other authors (Wen *et al.,* 2011). Since, the biocatalyst underwent a one month acclimatization, this could suggest the presence of mediators, or electron shuttles produced by the microorganisms in the medium. The microorganisms couldn't have colonized the electrode instantaneously after inoculation.

This mode of electron transfer might make the MFC unusable in continuous mode, since the microorganisms must first synthesize the mediators that must be accumulated in the anodic chamber.

We can see that the power density rises steadily, with a slight drop on the seventh day, until it reached a maximum of 2.5mW/m^2 on day 8.

After that, it gradually decreased until it became negligible on the fifteenth day.

Our power density curve lools similar to the ones obtained by other authors (Xia *et al.*, 2010), it seems like this evolution of power densities could be linked to the digestion of organic matter present in the medium, indeed, it was shown that the power output ou a MFC was dependent on the substrate concentrations (Zhang *et al.*, 1995), when the COD became sufficientlly low, the power outputs decreased(Velasquez-Orta *et al.*, 2009), this result is in accordance with the hypothesis that electricity production in the MFC is the result of the microbial metabolic activity.

Even though the power generation is linked to the substrate concentration, the outputs cannot be predicted, the biocatalyst contained different bacterial strains, many of which are unable to generate electricity in an MFC, and the interactions between them are not totally understood, and a detailed screening of bacterial biofilms may therefore be needed to identify important strains capable of high-power generation (Kiely *et al.*, 2010)

The power, and current densities obtained in our work are quite low when compared to the ones obtained by other authors. We had a maximum power density of 2.5 mW/m^2 while others reached power densities that exceeded 487mW/m^2 using artificial wastewaters containg acetate as a carbon source (Nien *et al.*, 2011)

This can be explained by the fact that no catalysts were used, and also, a real industrial effluent was used as fuel, as mentioned before, the presence of disinfectants could have inhibited the microbial metabolism.

4.3 Polarization curve of the MFC

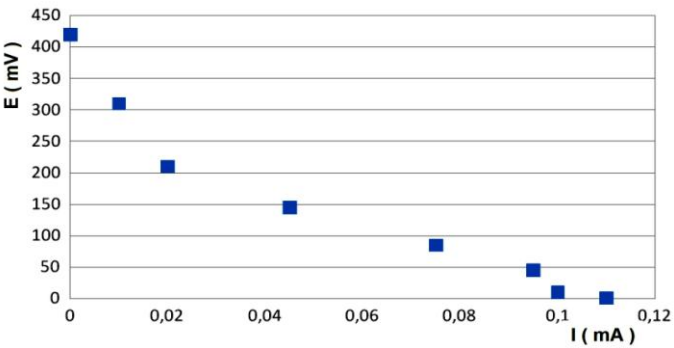

Fig. 6: Polarization curve of the MFC

The rapid voltage losses at low currents, are due to activation losses, they are due to energy lost for initiating the oxidation and reduction reactions, and the energy lost through the transfer of an electron from the cell terminal protein or electron mediator to the anode surface (Logan, 2008).

These losses are high, meaning we must find new bacterial strains that are able to transfer their electrons more easily, or investigate ways that might improve this transfer in known strains.

Voltage losses at higher currents are low, meaning that mass transfer aren't mainly

responsible for the low power outputs.

The region of constant voltage drop corresponds to ohmic losses. These losses are due to internal resistance of the MFC.

The slope of this region was calculated, and the internal resistance was found to be approximately 2200Ω, meaning the ionic transfer between the two compartments is slow.

These losses must be overcome to improve the MFC performances.

4.4 Coulombic efficiency

The coulombic efficiency was calculated using the following formula:

$$C_E = \frac{M_s \times \int_0^{t_b} I \times dt}{F \times b_{es} \times v_{An} \times \Delta c}$$

C_E : Coulombic efficiency; M_S : Molecular weight of the substrate; I : Current; F : Faraday's constant; b_{es} : Number of electrons involved in the reaction; v_{an} : Volume of liquid inside the anodic chamber; Δc : Variation of the substrate concentration.

Since the concentration of the substrate was evaluated using the COD, this formula becomes:

$$C_E = \frac{8 \times \int_0^{t_b} I \times dt}{F \times v_{An} \times \Delta COD}$$

After calculation, it was found that the efficiency was 2.8 %. This value is extremely low, it means that most of the organic matter was digested without being converted into electricity, this is because most of the strains found in the biocatalyst used to inoculate the anodic chamber weren't electrogenic bacteria.

A selection of the strains prior to inoculation might improve the efficiency, but little is known about this type of microorganisms.

5. CONCLUSION

Although the COD removal is slower and slightly lower in MFCs than in conventional aerated bioreactors, the fact that they do not need aeration to operate, which means a lower energy consumption, makes them an interesting alternative in treating industrial wastewaters.

However, the power generation was quite low, a low coulombic efficiency, high activation losses, and a high internal resistance limited the power outputs. It also seems that the use of catalysts is necessary, and an architecture that can be upscaled while reducing internal resistance must be found.

The results also suggest that the electrons are mainly transferred by external mediators. This might hinder the use of the MFC in continuous mode. A more in depth study of the microorganisms that develop in the anodic chamber must be made to

understand the interactions between different strains, and how these strains interact with the anode.

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