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Microbial Fuel Cells in Waste Treatment: Recent Advances

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Abstract: The increasing awareness that there are limits to the availability of nonrenewable resources, as well as that there are limits to the biosphere's ability to absorb wastes, are at the basis of the growing interest about Microbial Fuel Cells (MFCs) technology, with particular regards to their application to wastes treatment. MFCs, in fact, couple the direct electric power production to the degradation of organic compounds, liquid and solid wastes included. From municipal wastewater, to landfill leachate and solid waste, the application of MFCs to waste treatment achieved important results both in COD removal and power output. Unlike traditional Fuel Cells (FC), MFCs don't require chemical catalysts neither high working temperatures. Moreover, there is no net production of CO_2. In the course of 20 years, the performances of MFC increased significantly and scaled prototypes were realized. The constant progresses achieved make a wide-scale application of MFC to waste treatment more reliable.

Keywords: *Microbial fuel cells, wastes treatment, energy recovery, biomass valorization.*

1. Inroduction

The increasing necessity of more sustainable sources of energy drives the constant research about new and more environmental friendly technologies [1]. In recent years, an increasing attention to biomass valorization as sustainable source of chemicals and fuels [2] as well as to the necessity to find fuels alternative to H_2 for traditional Fuel Cells (FCs) [3] is spreading among scientific community. Biomass valorization and FC technology met in the so-called Bio Electrochemical Systems (BESs). BESs take advantage of biological capacities (microbes, enzymes, plants) for the catalysis of electrochemical reactions [4]. They mainly include Micro-Electrolysis Cell (MECs) and Microbial Fuel Cells (MFCs). While MECs can produce valuable products (like H_2) providing a suitable potential at the electrodes, MFCs don't need any energetic input to convert chemical energy (stored in organic compounds) into electric power.

BESs are essentially based on the ability of "electrogenic" or "electroactive" bacteria to exchange electrons with an anode in an electrochemical cell. Such microorganisms are able to form biofilm at the anode (bioanode) as well as at the cathode (biocathode). Different extracellular electronic transfer pathways are possible at both electrodes, accordingly to the physiology of biofilm-forming-bacteria and to the terminal electronic acceptors. In general, microorganisms can exchange electrons directly (by nanowires, for example) or by means of chemical intermediates, redox compounds acting as shuttles [4]. As to their high metabolic versatility, bacteria can virtually use any kind of chemicals (both inorganic and organic) for their metabolism. For this reason, MFCs can be fed by a great number of substrates: from acetate and glucose, to municipal and industrial wastewaters, and polluted soils [4][5][6][7]. Recently, a possible application of MFCs to composite food waste had been proved by Mohan and Chandrasekar [8], thus extending the application field of BES from liquid to solid wastes. A special mention is due to the

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Photosynthetic Microbial Fuel Cells (PMFCs), which are based on photosynthetic organisms (bacteria, algae and plants) and, thus, able to use sunlight as source of energy. The most promising PMFCs are the Bio-Electrochemical Systems (BESs), that convert hydrogen from photosynthesis, and Sediment-based BESs (SBESs), able to convert excreted organics from cyanobacteria or plants into electric power. These systems are quite complex and the scientific interest about them is more and more increasing **[**9**].**

It is a fact that BES technology advances had really improved in the last 20 years. MFCs Power Densities (PDs) increased from 0.001 to 0.01 milliWatts per square meter (mWm^{-2}) of anodic projected surface area in 1999 to 787 mWm^{-2} in 2003 and, finally, to levels of 2,770 mW m^{-2} in 2008 [4]. More recently, a PD of 661 W m^{-3} of anodic volume in an innovative MFC with flexible graphene-nickel 3D foam had been detected [10]. Different studies proved the reliability of low-strength wastewater treatment with MFCs [11][12][13], with some encouraging outcomes [14][15][16][17]. Nevertheless, the realization of scaled up prototypes, in addition to traditional wastewater treatments, is still limited [18] mainly because of different technological constraints still to overcome [15]. Recent progresses in the application of MFCs technology to liquid and solid wastes treatment are discussed in this paper.

2. MFC Principles and Configurations

Organic matter in the anodic compartment represents the fuel of MFCs. While bacteria carry out the degradation of organic compounds, they transfer electrons on an anode. Electrons are then transferred to a cathode, placed in an aerobic environment. At the same time, protons produced in the anodic compartment pass to the cathodic compartment, thus reacting with the electronic acceptor (usually oxygen). The flow of electrons through an external electric circuit is responsible for the electric power generation [19]. In figure 1, few examples of the possible MFCs configurations and electrochemical reactions occurring in them are reported [20]. Essential components of MFC are: an anode placed in an anoxic/anaerobic environment, a cathode placed in an aerobic environment or in contact with an electronic acceptor (ferrocyanide or potassium permanganate, for example) and the cell itself, often made by Plexiglas. Different materials are used for large prototypes [15]. A MFC can be made by one or two compartments. In this last case, a Cationic Exchanging Membrane (CEM) regulating the protonic flow from the anodic to the cathodic semi-cell is always present (Fig.1). A CEM can be also used in single chamber MFCs, usually adhering to the cathode by means of a hydrogel [21], or applied as ion-exchange resin [22].

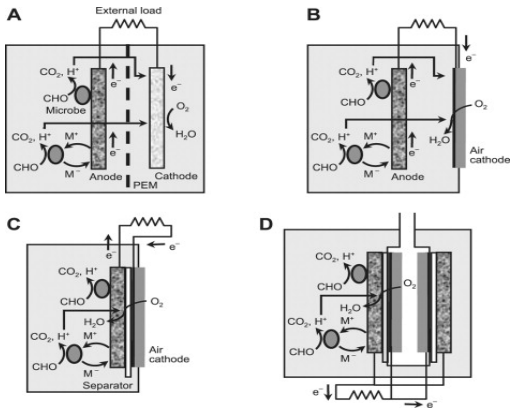

Figure 1: MFC configurations and electrochemical reactions. (A) A double-chamber MFC using oxygen as the cathode electron acceptor. (B) A single-chamber air-cathode MFC. (C) A single-chamber air cathode MFC with cloth electrode assembly separator. (D) A cassette-electrode MFC. M, Mediator; CHO, organics; PEM, proton exchange membrane. [20].

Many authors report a membraneless, single-chamber, air-cathode MFC as the most effective configuration in wastewater as well as in solid wastes treatment, even if some higher power output can be achieved in presence of a CEM at the cathode [9]. As to electrodes, different materials can be used but, the most commonly adopted in wastes treatments are carbon-based electrodes (graphite plates and carbon clothes for example) for their low cost and good performances [19][20][21]. Biocathodes (biofilm-forming bacteria at the cathode) are in most part of cases advised for any MFC practical application for both efficiency and low cost [22][23]. Examples of different configurations proposed for solid wastes treatment are reported in figures 2 and 3.

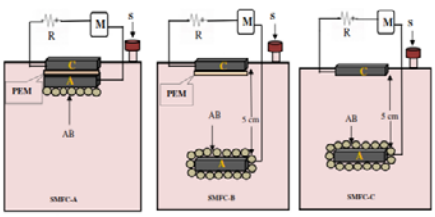

Figure 2: Schematic details of various configurations of Solid MFC used in [8] [A: anode; C: cathode; PEM: proton exchange membrane; AB: anodophilic bacteria; M: multi meters (digital display); R: resistance (100 Ω), S: sampling port [8].

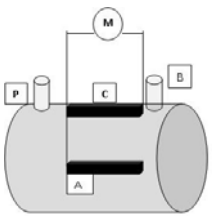

Figure 3: Solid MFC fed with Organic Fraction of Solid Municipal Wastes. A: anode; C: cathode; P: pHmeter probe; B: biogas sampling opening; M: digital multimeter.[35].

3. Energy Recovery

The power production of MFCs is calculated as P(W)=I(A)*E_{cell}(V), where I is the current and E_{cell} the cell voltage at current I. P value is usually normalized to the anodic surface (Power Density, PD) and expressed as W/m^2. In order to compare the performances of MFCs having different volumes, PD is also expressed as W/m^3. This last definition is of particular importance at the light of a possible industrial application of MFCs. As in traditional fuel cell, maximum power value is calculated by means of a polarization curve, plotting cell voltage (E_{cell}) against current (I_{cell}) at the variation of an external resistance (R_{ext}). Current intensity is also reported to the anodic surface and defined as Current Density (CD). Columbic Efficiency (CE), instead, is calculated on the base of the amount of electrons really transferred to the anode in relation to the amount of electrons theoretically delivered by the substrate [20]. The CE for a MFC run in fed-batch mode for a period of time is calculated according to (1)[21].

$$\epsilon_{Cb} = \frac{M \int_0^{t_b} I \, dt}{Fbv_{An}\Delta COD} \tag{1}$$

where M is 32 (molecular weight of oxygen), F is the Faraday constant, b is 4 the number of electrons exchanged per mole of oxygen v_{An} is the volume of liquid in the anodic compartment, and ΔCOD is the change in Chemical Oxygen Demand (COD) over time t. CE for continuous flow systems is calculated on the basis of current generated under steady conditions. When bacteria use a terminal electron acceptor different from the oxygen (at the cathode) like nitrate or sulphate, CE decreases and, in general, it diminishes because of competitive processes and bacterial growth [21]. As a consequence, only a fraction of the organic compounds are converted into electricity, limiting MFCs performances (Columbic losses). It is possible to determine the COD converted to electricity by means of a simple formula [24]:

$$\begin{aligned} COD_{ini} = {} & COD_{current} + COD_{biomass,\,sus} + COD_{biomass,\,att} \\ & + COD_{gases} + COD_{fin} + COD_{unknown} \end{aligned} \tag{2}$$

where COD_{ini} is the initial COD in the anode chamber, $COD_{current}$ is the COD for current generation over the incubation time, $COD_{biomass,sus}$ relates to the net accumulation of suspended biomass in the anode chamber, $COD_{biomass,\,att}$ relates to the net accumulation of the attached biomass at the anode, COD_{gases} is the COD for cumulative gas production in the anode chamber, $COD_{fin\ is}$ the soluble COD in anode chamber at the end of current generation, and finally $COD_{unknown}$ is the COD unaccounted for in the liquid of the anode chamber. COD values are expressed as mgO_2. The same authors put electron equivalents as COD (1 e^-eq = 8 g COD) to establish a mass balance [24]. In case of SMFCs, the COD of biomass attached to solid particles (fuel) in the anodic compartment should be also considered in equation 2.

As one of the advantages of MFC is to couple waste treatment with power production, the efficiency of a MFC is also referred to COD removal. Some of the most recent findings are reported in Table 1.

Table 1: Performances of MFC fed with Different Wastes.

Author	Waste	MFC type	Maximum power output	Coulombic efficiency	COD removal
Zhang B. *et al.*, 2009 [33].	Molasses wastewater	UASB-MFC-BAF reactor*	1410.2 mWm^{-2}	n.r.	53,2%
Aeltermann *et al.*, 2006 [19].	Potato-processing factory wastewater	Single-chambered with silver cathode	58±2 Wm^{-3}	20%	20-60%
Feng Y. *et al.*, 2008 [34].	Brewery wastewater	Single chambered	5.1 Wm^{-3}	Variable according to operational conditions	Variable according to operational conditions
Cha J. *et al.*, 2010 [14].	Raw municipal wastewater	Single-chambered	16.7 Wm^{-3}	14.9% to 39.6%	From 1.4 to 1.1 Kg $m^{-3}d^{-1}$
Cha J. *et al.*, 2010 [14].	Raw municipal wastewater		5,77 Wm^{-3}		
Gálvez A. *et al.*, 2009[6].	Landfill leachate	Single-chambered stack	18.04 Wm^{-2}	79.4%	
Mohan S. V. and Chandrasekhar K., 2011 [8].	Composite food waste	Single chambered with and without membrane	8,8mWm^{-2} Kg**	n.r.	76% (maximum value achieved)
Nastro R.A. *et al.*, 2013 [35]	Organic Fraction of Municipal Solid Waste	Single chambered	10 mWm^{-2} Kg**	n.r.	n.r.

* UASB- MFC-BAF: up-flow anaerobic sludge blanket reactor–microbial fuel cell–biological aerated filter; ** values are reported to the anodic surface and mass fuel; n.r.: not reported.

3.1 Factors Affecting MFCs Performance

The performances of MFCs are highly affected by chemical and operational factors. Fuel composition, ionic strength, buffer capacity, and pH as well as operational temperature and flow mode (continuous or in-batch feed mode) are important parameters and must be taken into account while projecting an MFC. The distance between the electrodes, as well as their composition, the shape of the cell and the chemical nature of the electronic acceptor are of fundamental importance as well [19][21][25][26]. Moreover, the composition of the microbial community on both the anode (bioanode) and the cathode (biocathode) plays a crucial role in determining MFCs performances [27][28][29]. Even if

most part of laboratory-scale MFCs are colonized by one or few microbial strains, a different approach must be taken into account in a scaled, practical application. It is a fact that microbial consortia made by environmental species present in the fuel (wastewater for instance) would colonize the electrodes, overcoming any electroactive microorganism previously inoculated in a MFC or, at least, competing with it.

Wastewater treatment sludge is usually considered as a good source of microorganisms for MFCs treating liquid wastes, while endogenous microflora had been used in MFCs fed with solid organic wastes [11][8][36]. Recently, a chemical modification of anodic surface with nitrogen-containing amines resulted in higher and more rapid production of current, compared to those without this modification, when placed in microbial cultures [30]. Moreover, some attempts to use photo-reactive semiconductors at the cathode (the so called "photocathode") are in progress [31]. If confirmed, the encouraging results of these new experiments will open up new prospects for practical application of MFCs and, in general, that of BESs.

4. MFC and Anaerobic Digestion

Anaerobic Digestion (AD) is one of the most commonly used treatment of both solid and liquid wastes. The process itself is based on the ability of anaerobic microorganisms to degrade organic compounds, with the consequent decrease in COD content and production of biogas rich in methane [36]. Acidogenesis and methanogenesis are the main processes occurring during AD. "Digestate" is solid waste after AD treatment which is firstly stabilized by an aerobic microflora and, then, applied to open fields soil or for field improvement purposes [37].

MFC technology can find a similar application in waste treatments, offering different advantages. Unlike AD, a MFC can work at relatively low temperature (10-20°C) and electric energy is produced directly from the organic matter, without any combustion. The AD energetic outcome derives, in fact, from the utilization of biogas for heating and electric power production. So, a possible niche application of MFC could be as substitute of AD in low carbon load (COD content less than 1 g/L) wastewater treatment or in low temperature waste treatments. Nevertheless, a combination of both AD and MFCs seems feasible [19][36]. The application of MFC to wastes treatment has been mainly focused on leachate and both municipal and industrial sewage treatment but, recently, some researchers proved the reliable utilization of composite solid wastes as well as the organic fraction of municipal solid wastes as suitable MFC fuel [8] [35].

4.1 Challenges and Future Perspectives

The real challenge in MFCs application to waste treatment is linked to the reduction of energetic losses (Columbic losses, for example), with an improvement of the MFCs overall performance. According to Mc Carty *et al.* (2011), about 44% of soluble organics in wastewater could be effectively converted into electricity by a MFC, which is more than achieved by an AD, whose conversion efficiency is estimated to be about 37%. According to the same authors, a MFC system had been estimated to cost 800 times more than an anaerobic system, based upon available technologies [38]. Nevertheless, this assertion doesn't take into account the huge increase of scientific research at all levels (chemical, biological and technological), even focused on the utilization of less expensive materials and production of valuable by-products. Considering that MFCs avoid CH_4 production and combustion, the environmental advantages could compensate a higher cost of production.

Although a complete comparison of BES and AD technologies by means of a Life Cycle Assessment approach is still to be done, preliminary LCA results suggest that based on current knowledge and existing materials MFCs compare very well with the existing treatment methods such as AD [39]. Further, given the huge research efforts worldwide, breakthrough results are expected very soon thanks to cheaper materials and better microorganisms [39][40]. The research for better microorganisms is also leading to an increasing interest for synthetic microbiology to manipulate electron flows and to establish a catalytic interface between the electrodes and the bacteria [40].

5. Discussion and Conclusions

According to recent technological progresses, MFCs technology is expected to become of practical use in the near future. MFCs, in fact, could represent a cost-effective low-energy waste treatment and potentially lead to a waste treatment cost reduction and less environmental impacts. Nevertheless, large-scale application of MFCs to wastes treatment is still a challenging option. Different affecting factors constitute a real problem for researchers, as MFCs performances vary according to the fuel and operational conditions.

MFCs should be designed according to the fuel used, while a specialized micro-flora selected from the fuel itself should be used. In many cases, in fact, MFC using indigenous micro-flora showed better results than MFC working with exogenous bacteria. Inocula deriving from previous MFCs give higher power outputs and overall performances [21].

The study of SMFCs implies the estimation of the COD content of the biomass attached to solid substrates of the fuel itself, in order to evaluate the energy conversion efficiency of the cell. But, while it is possible, for example, to determine proteins amount of the anodic biofilm and, indirectly, its COD content [24], a different method should be considered for SMFCs, as solid organic matter in the anodic compartment could contain residual vegetable or animal proteins, which may interfere with the COD measure of attached biomass. Another issue is represented by the criteria and the units of measure adopted to evaluate MFCs performances. Most often it is not possible or is, at least, difficult to compare the performances of MFCs with different layouts in terms of PD or CD for m^2 of cathodic or anodic surface.

A possible solution could be to refer the electrical outputs to the volume of the anodic compartment in the case of double chamber MFCs or to the volume of the cell for single-chamber MFCs. For solid matter, the weight of the fuel could be considered as well. In any case, a standardization of criteria and measures is needed for comparison. Considering the progresses achieved by scientific research and the increasing number of scientific publications about microbial electrochemistry technology, the practical implementation of MFCs (and BES in general) in waste treatments is becoming day-by-day more reliable.

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