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## ENERGY PRODUCTION FROM WASTEWATER USING HORIZONTAL AND VERTICAL SUBSURFACE FLOW CONSTRUCTED WETLANDS

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### Abstract

The aim of the present work is to study the performance of a constructed wetland for domestic wastewater depuration also working as a microbial fuel cell (MFC) under two different subsurface flow modes in the upper cathodic compartment (horizontal flow, HF, or vertical flow, VF), and to compare the efficiency under both situations. A pilot-scale subsurface constructed wetland treating domestic wastewater was used, and the installation also included the electrodes and devices needed to work as MFC. The system worked continuously during two years: under HF mode (first year) and under VF mode (second year). The Chemical Oxygen Demand (COD) removal results and the cell voltage production were continuously monitored. The wetland successfully worked as MFC generating electric power. The organic pollution was completely oxidized in the lower anaerobic compartment, and aerobic conditions were detected in the upper cathodic compartment, thus producing electrical current. No important differences were observed in COD removal efficiencies working under the two flow modes. The cathodic compartment showed higher redox potential and dissolved oxygen values working under VF mode. The maximum cell voltage measured was approximately 250 mV in both flow modes. Despite the higher oxygenation potential of the VF option, no important differences were observed in voltage generation. The process developed in the present work showed similar cell voltage, power density and current density to other previous works using photosynthetic MFC, benthic MFC and plant-type MFC.

*Key words:* constructed wetland, horizontal flow, microbial fuel cell, vertical flow, wastewater

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### 1. Introduction

Bioelectrochemical wastewater treatment has recently appeared as a promising technology (Logan et al., 2006; Rozendal et al., 2008) which uses organic waste materials to produce electricity according to the fundamentals of Microbial Fuel Cells (MFC), or to obtain valuable products, such as H<sub>2</sub>, according to the fundamentals of Microbial Electrolysis cells (MEC). Both MFC and MEC use electrochemically active microorganisms that are capable of oxidising organic matter to generate electrons, protons, and other metabolic products. The organic matter is oxidized by microorganisms under anaerobic conditions, and they produce extracellular

electron transfer to an electrode (anode) while they are removing the organic pollutants in wastewater. Via an electrical circuit, the electrons are transported to a cathode compartment, where they combine with protons and with oxygen to produce water on a cathode surface. Protons diffuse from the anodic compartment through a membrane. As a result of this electrical connection and the difference in redox potentials, the electrode reactions can occur, and the electrons can flow from the anode to the cathode, allowing electrical current to flow. When the Gibbs free energy change of the overall reaction is negative, electrical energy is produced and the system works as MFC. On the contrary, energy supply is needed and the system works as MEC.

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There are several natural processes which have been used as the base of the design of MFC, in order to extract the energy contained in natural resources, such as the chemical energy of organic matter or wastes. For instance, there are different approaches that integrate photosynthesis with MFC (Rosenbaum et al., 2010), or configurations such as the plant-MFC (De Schampelaire et al., 2008a; Strik et al., 2008). Another approach to harvesting energy from naturally occurring electropotential differences is to locate an anode into sediments in rivers or lakes, with immersion of a cathode in the overlying body of water. Such a set-up is called a sediment microbial fuel cell or benthic MFC (Reimers et al., 2006).

Bioelectrochemical wastewater treatment technologies for nutrient and carbon removal have been recently developed, based on the fundamentals of MFC. All the technologies work through combined anaerobic/aerobic reactors between which there is a difference of redox potentials. There are recent reviews that offer a global state of the art about these technologies (Logan, 2012; Logan and Rabaey, 2012; Mook et al., 2013) and specifically some studies on carbon and nitrogen removal (Virdis et al., 2008; Virdis et al., 2010) or the application of MFC to treat industrial wastewater (Velasquez-Orta et al., 2011). Constructed wetlands (CW) are low-cost systems traditionally used for wastewater treatment.

Wastewater treatment wetlands have been used for decades, and there has been a considerable amount of research activity and literature published on this topic. These systems consist of artificial wetlands that are isolated from the environment around them and receive wastewater. Water purification is achieved by a complex combination of natural physical, chemical, and biological phenomena (Zhi and Ji, 2012). The two major types of CW are surface flow and subsurface flow wetlands (Kadlec and Knight, 1996). Depending on the type of CW, they are formed by different elements including macrophyte plants, a porous solid bed, and a mixed population of microorganisms in the form of biofilms. In a subsurface system the wastewater flows under and through a permeable medium (a gravel bed) which acts as the macrophyte plants rooting media (García et al., 2010). Depending on the flow direction through the permeable medium, the subsurface wetlands are of two types: horizontal flow (HF) and vertical flow (VF). Vertical flow wetlands can work under unsaturated conditions in the gravel bed when the bed is drained.

Different microbiological mechanisms can take place in subsurface wetlands, depending on the redox potential in the different zones of the porous gravel bed (Ojeda et al., 2008). The upper parts of the wetland near the surface can receive oxygen from the atmosphere, as do the inner zones closed to the rhizosphere because of the possible aeration potential of the plants, which are able to transfer oxygen to lower levels in the gravel bed (García et al., 2010).

In addition, VF wetlands can receive more oxygen than HF wetlands because of the unsaturated conditions. In contrast, deep zones in the wetland far from the influence of roots are mainly anaerobic zones. This situation illustrates conditions in which clear differences in redox potentials appear, and wastewater may be treated under different aerobic and anaerobic biological mechanisms.

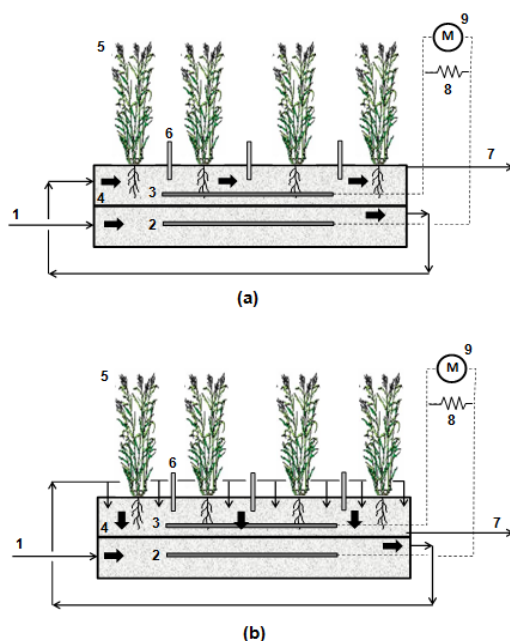
According to the above description, it has been recently proved that CW could also act as MFC, treating wastewater and producing electricity from the oxidation of organic pollutants. There are recent papers in the literature regarding this topic. Some works used vertical flow subsurface systems (Fang et al., 2013; Yadav et al., 2012; Zhao et al., 2013) or surface flow systems with floating macrophytes (Chiranjeevi et al., 2013; Mohan et al., 2011). Regarding the subsurface systems, there are no research works comparing the performance of horizontal and vertical flow systems.

In this context, this work studies the performance of a subsurface flow constructed wetland for domestic wastewater treatment which also works as MFC in order to produce electric power. The objective of the work is to study the effect of the water flow direction in the cathodic compartment. The wetland worked under HF and VF modes and the differences have been discussed.

## 2. Materials and methods

The experimental device consisted of a pilot scale constructed wetland situated in a greenhouse in the Institute for Chemical and Environmental Technology of University of Castilla La Mancha, Ciudad Real (Spain). The wetland was modified to function as an MFC, and it could operate under HF mode (Fig. 1a) or VF mode (Fig. 1b).

The wetland consisted of a 1.15 m × 0.47 m plastic channel with a bed depth of 0.50 m and a longitudinal slope of 1%. It was filled with gravel with an average particulate diameter of 9 mm, apart from the left and right five-centimetre vertical layers, for which the average particulate diameter was 15 mm to improve the distribution of wastewater in the wetland. The bed porosity was 0.4. Sampling points were placed along the wetland at  $\frac{1}{4}$ ,  $\frac{1}{2}$ , and  $\frac{3}{4}$  of the total length. They consisted of vertical plastic tubes (3 cm diameter, and 10 cm length) perforated with several small holes (0.5 cm diameter) at different positions. The sampling point allowed introducing dissolved oxygen or redox probes. *Phragmites australis* (Reed) bought in a commercial greenhouse was planted in the wetland in autumn 2011 (20 plants m<sup>-2</sup>). The necessary elements of a MFC were also included during the wetland construction. A horizontal, rectangular (0.70 m × 0.15 m and 0.03 m thickness) graphite anode was located in the gravel bed, 12 cm above the bottom of the wetland, and an identical graphite cathode was also located 12 cm below the gravel surface.



**Fig. 1.** Experimental installation working under (a) Horizontal Flow mode and (b) Vertical Flow mode. (1) Wastewater feeding; (2) Anode; (3) Cathode; (4) Bentonite layer; (5) Reed plants; (6) Sampling points; (7) Treated effluent; (8) Resistance; (9) Multimeter

The distance between electrodes was 25 cm. Insulated copper wires were used to connect both the electrodes, including a 120  $\Omega$  resistor between them.

A 0.02 m thickness layer of calcium bentonite (*Bentonil A*, from *Süid-Chemie*) was placed in the middle depth of the gravel bed to separate the anode and cathode compartments and limit the growth of roots only to the upper area in which the cathode was located. The wastewater flow entered the system at the bottom anode compartment, passed horizontally through the completely flooded compartment, and left through the opposite end. Subsequently, the wastewater was pumped to the cathodic compartment, and passed through in two ways: via horizontal subsurface flow (Fig. 1a) or via vertical flow (Fig. 1b), and finally left the wetland.

The wetland was continuously fed with domestic wastewater through a peristaltic pump. Synthetic wastewater was always used in order to have control of the wastewater characteristics. The average inlet wastewater composition was: glucose (160 mg l<sup>-1</sup>), CH<sub>3</sub>COONa·3H<sub>2</sub>O (160 mg l<sup>-1</sup>), NaHCO<sub>3</sub> (111 mg l<sup>-1</sup>), KH<sub>2</sub>PO<sub>4</sub> (44.5 mg l<sup>-1</sup>), MgCl<sub>2</sub>·6H<sub>2</sub>O (37.1 mg l<sup>-1</sup>), CaCl<sub>2</sub>·2H<sub>2</sub>O (30.1), (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (112 mg l<sup>-1</sup>), (NH<sub>4</sub>)<sub>2</sub>Fe(SO<sub>4</sub>)<sub>2</sub>·6H<sub>2</sub>O (84.2 mg l<sup>-1</sup>). The main inlet wastewater parameters were as follows: Total Suspended Solids-TSS (35 mg l<sup>-1</sup>), Chemical Oxygen Demand-COD (230 mg l<sup>-1</sup>).

The experimental pilot plant continuous process started in January 2012 by feeding wastewater and seeding microorganisms by adding a small amount of biological sludge from the anodic compartment of a laboratory-scale MFC treating similar wastewater (González del Campo et al., 2013). The seed was added to the lower compartment

of the wetland. The wetland worked under a continuous operation mode, and the water flow direction in the cathodic compartment was the only variable under study: the upper part of the wetland worked as an HF system during one year (2012) and then it worked as a VF system during a second year (2013).

A constant wastewater flow of 30 l d<sup>-1</sup> was used to maintain a hydraulic residence time of 3.2 days. The wastewater pH was always maintained at approximately 7.4 by means of the buffer capacity of the synthetic medium. Despite the air-conditioning system available, temperature fluctuations of the ambient air in the greenhouse ranged between 18 and 31°C, depending on the external weather. It caused wastewater temperature fluctuations between 20 and 26°C.

The whole system was weekly monitored during the period between march-september in both cases. The influent flow was measured. Samples of the influent and effluent water flow were taken, and soluble COD was analysed in the laboratory according to the standard methods (APHA-AWWA-WPCF, 1998). The dissolved oxygen level (DO), and redox potential (ORP) in the cathodic compartment were monitored *in situ* using the sampling points. The ORP was measured by a Redox-pH-meter (PCE-228). DO was measured using a YSI 5000 dissolved oxygen probe.

During normal operation, the anode and the cathode were connected by means of wires and the resistance. The potentials between the edges of this resistance were continuously monitored. A digital multimeter was connected to the system to continuously monitor the value of the cell potential.

### 3. Results and discussion

#### 3.1. Organic pollution removal

Fig. 2 shows the wetland influent and effluent COD concentrations. Although some fluctuations were observed, an average COD value of  $230 \text{ mg l}^{-1}$  was continuously fed to the wetland during the two operation periods. The first experimental period used horizontal flow (HF). After seeding the system with microorganisms from an MFC, the start-up stage lasted for approximately three months.

The COD measurements started at day 90 showing a removal efficiency of approximately 65%, and COD was nearly completely removed since day 125 to the end of the HF period. The second period used vertical flow (VF), and the COD removal efficiency values were low at the beginning, but they quickly increased and the effluent COD concentration reached similar values to the HF period in approximately two months. Thus, the wetland worked with important organic pollution removal efficiencies in both cases and no significant differences were observed.

#### 3.2. Operating conditions inside the wetland

Fig. 3 shows the operating conditions into the cathodic compartment during the two periods (Fig. 3a shows the redox potential values, and Fig. 3b shows the dissolved oxygen levels). The data plotted in Fig. 3 are mean values of the ones obtained at the same time in the different sampling points (Fig. 1). It can be noted important differences in both ORP and DO values depending on the water flow direction in the cathodic compartment. Approximately constant levels of dissolved oxygen ( $2 \text{ mg l}^{-1}$ ) and redox potential ( $-30 \text{ mV}$ ) could be observed during the HF operation period, but clearly higher ORP (Fig. 3a) and DO (Fig. 3b) values were observed during the VF period.

Differences between the pollutants removal capabilities of HF and VF subsurface wetlands have been extensively studied. Some recent works have been published by Yalcuk and Ugurlu (2009), Dan et al. (2001) or Abou-Elela et al. (2013). They found that the VF usually offers better nitrification results, and sometimes, better organic matter removal efficiencies. Pandey et al. (2013) reported also higher ORP values inside the VF compared to HF subsurface wetlands because of the penetration of more oxygen in the unsaturated gravel bed. The results in the present work show greater variability and fluctuations in the DO and ORP measurements during the VF operation mode than the HF mode.

This behaviour could be also related to the heterogeneity of the unsaturated gas-liquid media in the cathode compartment.

#### 3.3. Energy production

Fig. 4 shows the cell voltage values measured during the two operation periods. As indicated in

section 3.1, the start-up stage lasted for approximately three months in the HF period. The voltage measurements started at day 90, but it was not detected until day 110 approximately and then increased while the effluent COD was simultaneously decreasing (Fig. 2). It reached the maximum value at approximately  $250 \text{ mV}$  after 160 days. The dissolved oxygen level in this type of wetlands is usually very low (García et al., 2010); and a  $2 \text{ mg l}^{-1}$  concentration in the present work can be considered a relatively high level. Thus, it could be assumed that the wastewater organic matter was completely oxidised in the lower wetland compartment (working under anaerobic conditions) and that the upper wetland zone worked under slightly aerobic conditions. The upper part of the wetland was in contact with the atmosphere and was further supplied by the rhizosphere, which should transfer oxygen from the air. Specifically, the aeration potential of *Phragmites australis* in such wetlands has been measured in previous investigations, and different values were obtained. Armstrong et al. (1990) reported oxygen release values of  $5\text{-}12 \text{ g O}_2 \text{ m}^{-2} \text{ d}^{-1}$ , while Gries and Garbe (1989) reported values of  $1.5\text{-}3 \text{ g O}_2 \text{ m}^{-2} \text{ d}^{-1}$ . Therefore, a potential difference should be generated between both zones causing electric current.

After the change to VF conditions, the cell voltage was initially low but slowly increased to finally reach similar values to the ones obtained under HF conditions. According to the above discussion (section 3.2), more favorable conditions (higher aeration in the cathode compartment, and a higher ORP difference between anode and cathode compartments) would be expected under VF mode, supposing that the lower compartment remained under anaerobic conditions, and thus higher cell voltage would be obtained.

However, it was not the case and the results were closed to the HF mode. Some discrete measurements of DO and ORP were done in the outlet flow of the lower compartment. Values between  $1$  and  $2 \text{ mg O}_2/\text{L}$  and between  $-10 \text{ mV}$  and  $0 \text{ mV}$  approximately were respectively detected, indicating that the VF mode caused also slight oxygen penetration in the anode compartment. Thus, the VF wetland did not improve the performance of the cell. The VF operation mode showed again greater variability and fluctuations in the voltage measurements, as occurred with ORP and DO, due to the gas-liquid medium heterogeneity.

Different performance parameters (coulombic efficiencies, power density and current density) were calculated from the experimental results. Calculations were performed according to Logan et al. (2006). These parameters provide information about the electric power generation capability of the wetland. Very low coulombic efficiencies were obtained (0.1%) compared with several previously reported values obtained in conventional biological wastewater treatment systems with an MFC assembly.

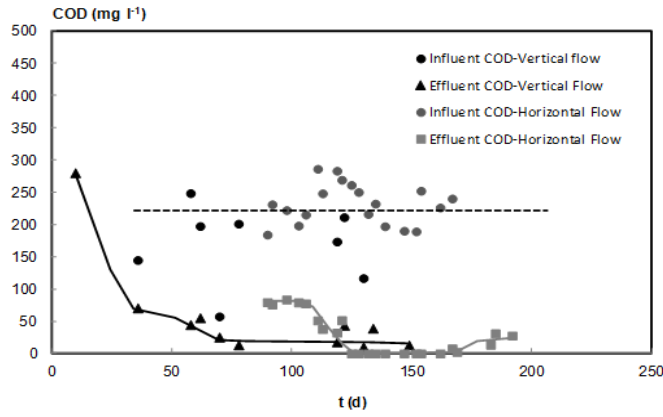


Fig. 2. Influent and effluent COD concentrations during the two experimental periods

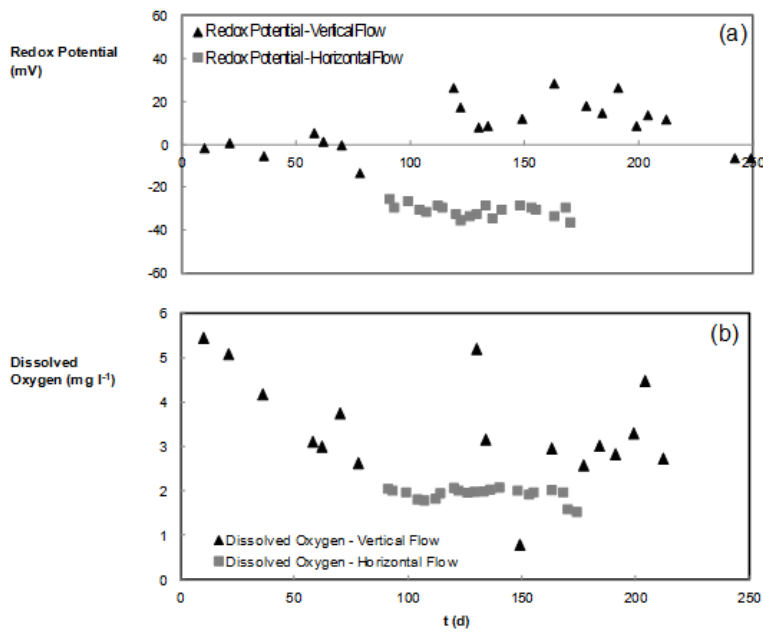


Fig. 3. Redox Potential (a) and Dissolved Oxygen (b) levels in the upper wetland compartment

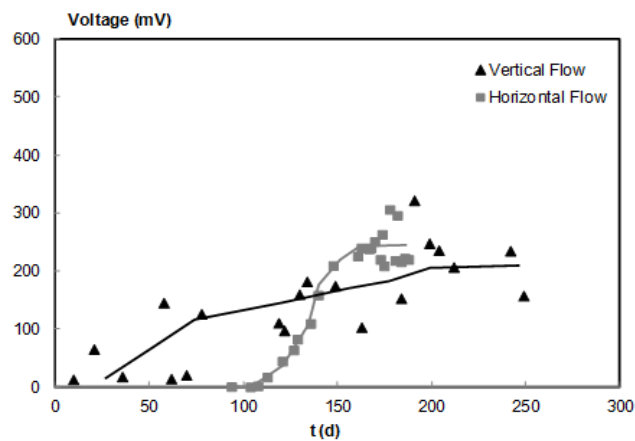


Fig. 4. Cell voltage values obtained during the two experimental periods

He et al. (2005) reported coulombic efficiencies between approximately 1% and 7%, and Virdis et al. (2008) reported values between 15% and 45%. In contrast, the cell voltage, power density, and current density values obtained in this work (250

mV, 2.0 mW m<sup>-2</sup> and 8.0 mA m<sup>-2</sup> respectively) are similar to the ones obtained in previous works using photosynthetic MFC, sediment-type MFC, and plant-type MFC. Zou et al. (2009) reported power density values of approximately 1 mW m<sup>-2</sup>, and current

densities up to 30 mA m<sup>-2</sup> using a photosynthetic MFC. Strik et al. (2008) reported 67 mW m<sup>-2</sup> as maximum value, whereas De Schampelaire et al. (2008a) reported values between 10 and 20 mW m<sup>-2</sup>, with both works using plant-type MFC.

Regarding the sediment-type MFC, Logan et al. (2006) indicated that this type of MFC can produce a power density of up to 28 mW m<sup>-2</sup>, and De Schampelaire et al. (2008b) indicated that the average power values are on the order of 10 to 20 mW m<sup>-2</sup>. The few known systems based on constructed wetlands reported 35 mW m<sup>-2</sup> as the maximum power density value (Yadav, 2010), 16 mW m<sup>-2</sup> as the maximum power density value and current density values up to 70 mA m<sup>-2</sup> (Yadav et al., 2012), and power density values between 17 and 80 mW m<sup>-2</sup> (Mohan et al. 2011).

Lower values than the obtained in the present work were reported by Zhao et al. (2013). Based on these results, HSSF-CW coupled with MFC would produce similar electric power to the one produced by conventional sediment-type MFC and plant-type MFC.

#### 4. Conclusions

The subsurface flow CW worked under two flow modes (HF and VF) in the cathodic compartment. Similar and high COD removal efficiencies were obtained under both modes. Higher ORP and DO values were observed in the cathodic compartment working under VF direction. The maximum cell voltage obtained was approximately 250 mV under the two flow modes.

Despite the better oxygenation capability of the VF wetland, no significant differences were observed in the power generation. Similar cell voltage, power density, and current density values were obtained compared to the ones obtained in previous works using photosynthetic, sediment-type or plant-type MFC.

#### Author Disclosure Statement

The authors declare no competing financial interest.

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