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Microbial fuel cell application in landfill leachate treatment

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ABSTRACT

The feasibility of using microbial fuel cells (MFCs) in landfill leachate treatment and electricity production was assessed under high levels of nitrogen concentration (6033 mg N L^{-1}) and conductivity (73,588 μS cm^{-1}). An air-cathode MFC was used over a period of 155 days to treat urban landfill leachate. Up to 8.5 kg COD m^{-3} d^{-1} of biodegradable organic matter was removed at the same time as electricity (344 mW m^{-3}) was produced. Nitrogen compounds suffered transformations in the MFC. Ammonium was oxidized to nitrite using oxygen diffused from the membrane. However, at high free ammonia concentrations (around 900 mg N-NH_3 L^{-1}), the activity of nitrifier microorganisms was inhibited. Ammonium reduction was also resulted from ammonium transfer through the membrane or from ammonia loss. High salinity content benefited the MFC performance increasing power production and decreasing the internal resistance.

1. Introduction

Landfill leachate is generated from the disposal of municipal solid waste. It is highly contaminated with a wide range of organic and inorganic compounds, nutrients, chemical contaminants and heavy metals. Its characteristics depend on many factors including age, precipitation, seasonal weather variation, waste type and composition [1,2].

Biological treatment is commonly used as a simple, reliable and highly cost-effective method by removing organic matter and nitrogen from leachate [3,4]. However, landfill leachate usually exhibits a low carbon–nitrogen ratio and a low level of alkalinity, which make the conventional processes of nitrification and denitrification more difficult. To cope with this factor, it would be necessary to increase the level of alkalinity and/or the organic matter content, thus considerably increasing the cost of treatment [5]. Treatments based on anaerobic ammonium oxidation (anammox) metabolism may provide a more sustainable alternative to the treatment of such wastewater, due to the reduced level of aeration and the lower dosage of external organic carbon that they require [6]. Nitrogen present in leachate mainly in the form of ammonium must be partially aerobically oxidized to nitrite prior to the anammox process. Ganigué et al. [7] demonstrated the feasibility of this process for the long-term treatment of leachate with an extremely high ammonium concentration (up to 5000 mg N-NH_4^+ L^{-1}). The anammox process can thus be applied to treat urban landfill leachate that has been previously subjected to a partial nitritation process with a high level of removal efficiency [8].

In young landfills, leachate contains large amounts of biodegradable organic matter [2]. When a partial nitritation and anammox process is introduced, the organic matter is aerobically oxidized, involving both energy consumption and the production of sludge. Chamchoi et al. [9] and Ruscalleda et al. [10] reported that biodegradable organic matter should also be removed to avoid its negative effects on the anammox process. Therefore, the pre-treatment of leachate to remove an excess of biodegradable organic matter would have a potential niche in the market. In addition, this pre-treatment would become more attractive if electricity was produced through the use of microbial fuel cell (MFC) technology. Various studies [11–13] have reported the production of electricity from the treatment of young leachate. However, some open questions remain regarding the performance of MFCs in relation to the treatment of landfill leachate with high levels of salinity, and the dynamics of nitrogen compounds, among others. The objective of this study is to investigate the feasibility of landfill leachate treatment using MFCs under these conditions.

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Table 1
Leachate characteristics throughout the study.

Compound	Units	Period 1		Period 2	
		Range	Mean ± σ	Range	Mean ± σ
Chemical oxygen demand, COD	mg COD L^{-1}	306–825	507 ± 201	3280–4260	3480 ± 487
Biological oxygen demand, BOD_5	mg BOD L^{-1}	59–137	102 ± 34	68–121	93 ± 22
Total Kjeldahl nitrogen, TKN	mg N L^{-1}	594–1534	936 ± 339	5015–6736	6033 ± 261
Ammonium, NH_4^+	mg N-NH_4^+ L^{-1}	553–1210	802 ± 255	3648–6182	5449 ± 648
Nitrite, NO_2^-	mg N-NO_2^- L^{-1}	0.1–1.2	0.5 ± 0.4	0.0–15.5	2.0 ± 0.7
Nitrate, NO_3^-	mg N-NO_3^- L^{-1}	0.1–4.6	1.7 ± 1.9	0.1–8.9	0.6 ± 0.5
Conductivity	μS cm^{-1}	7220–9176	8583 ± 929	71,700–79,800	73,588 ± 2044
pH	–	8.15–8.70	8.47 ± 0.26	8.39–8.61	8.49 ± 0.07

2. Materials and methods

2.1. Landfill leachate

The landfill leachate concentration range and the mean values of the principal chemical compounds, together with the corresponding levels of electrical conductivity and pH, are summarized in Table 1. The raw leachate presented a highly variable composition, a low BOD/COD ratio (0.02), and a high nitrogen concentration (6033 mg N L^{-1}) and level of conductivity (73,588 μS cm^{-1}). The raw landfill leachate contained 3480 mg COD L^{-1} (with 3% of the influent in the form of BOD_5). Most of the organic matter was thus either slowly biodegradable or totally non-biodegradable.

2.2. Experimental procedure

The experimental study was divided into two periods. The first 46 days (Period 1), the MFC was fed with diluted urban landfill leachate (20%, v/v raw leachate). The dilution was done to minimize the risk of any inhibitory effect. The MFCs treated 0.76 ± 0.06 L d^{-1} at a hydraulic retention time (HRT) of 8.3 ± 0.7 h.

In order to increase the biodegradable organic matter available for exoelectrogenic bacteria, the organic loading was increased in Period 2 (Table 1) by feeding with a 100% raw leachate. Once power production was observed, the organic load was progressively increased by decreasing the HRT from 7.4 to 3.2 h. Accordingly, the daily flow increased to 1.51 ± 0.38 L d^{-1}. This period lasted 110 days.

2.3. MFC configuration

Fig. 1 shows a diagram of the air-cathode MFC used to treat the landfill leachate. The air-cathode MFC consisted of anode and cathode frames placed on opposite sides in a methacrylate rectangular chamber 32 × 27 × 400 mm (with an empty bed volume of 343 mL). The anode frame was filled with 6 mm granules of graphite (Alfa Aesar, Germany), reducing the compartment volume to 167 mL (net anodic compartment; NAC). The electrodes were washed in 1 N HCl and 1 N NaOH to remove possible metal and biomass contamination [14]. A thin graphite electrode (30 × 35 mm, Sofacel, Spain) was used to connect the MFC to an external electrical circuit. The cathode electrode selected was C-cloth 0.35 mg cm^{-2} of Pt catalyst with 30% wet-proofing (Clean Fuel Cell Energy LLC, USA). A 90 cm^2 cation exchange membrane (CEM, Nafion® 117, Dupont) was treated according to Liu and Logan [15] and used between the anode and cathode frames. The anode and the cathode were connected through an external resistor (4610 Ω) to close the circuit. Landfill leachate wastewater was continuously fed to the recirculation loop to (i) obtain the desired volumetric loading rate, (ii) maintain well-mixed conditions, (iii) avoid concentration gradients and (iv) avoid clogging of the granular matrix. The system was thermostated at 23 ± 2 °C, and pH was monitored during the study. The MFC was inoculated with 50 mL effluent from a parent MFC treating synthetic wastewater (mainly composed of sodium acetate and a buffer solution). The MFC operated for 120 days at a mean organic loading rate of 1.02 kg COD m^{-3} d^{-1} (COD removal efficiency 90%) producing 8.5 W m^{-3}.

2.4. Analyses and calculations

Standard wastewater measurements for organic matter (chemical oxygen demand (COD) and biological oxygen demand (BOD_5)) and for nitrogen (total Kjeldahl nitrogen (TKN), ammonium (NH_4^+), nitrites (NO_2^-) and nitrates (NO_3^-)) were taken twice a week and analysed according to the APHA Standard Methods for the Examination of Water and Wastewater [16]. The organic and nitrogen loading rates (OLR and NLR, kg COD m^{-3} d^{-1} and g N m^{-3} d^{-1}) were calculated as the daily influent/effluent organic matter or nitrogen concentrations (mg COD L^{-1} and mg N L^{-1}) per hydraulic retention time (HRT; days). Finally, the organic and nitrogen removal rates (ORR and NRR, kg COD m^{-3} d^{-1} and g N m^{-3} d^{-1}) were calculated as the difference between the influent and effluent loading rates.

Cell potential (V) in the MFC circuit was monitored at five-min intervals using an on-line multimeter (Alpha-P, Ditel) with a data acquisition system (Memograph® M RSG40, Endress+Hauser). Current (I) and power ($P = IV$) were calculated according to Ohm's law. Power density was calculated by dividing power by the net anodic volume (mW m^{-3}). Polarization curves were obtained by varying

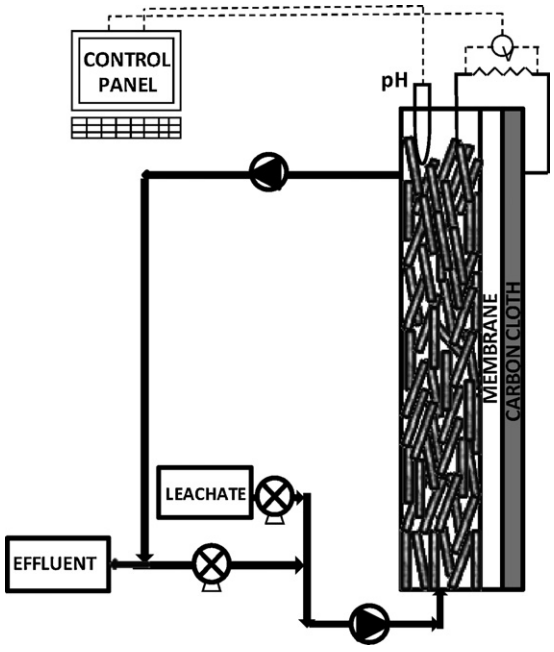

Fig. 1. Diagram of the air-cathode MFC used to treat leachate.

the external resistance in the circuit and measuring the voltage according to Logan et al. [17].

2.5. Microbiology analysis

Fluorescent in situ hybridization (FISH) analysis was performed as specified by Amann [18] using the Cy5-labelled EUBMIX probe [19] to target the entire bacterial community and the Cy3-labelled SRB385 probe for the exoelectrogenic bacteria *Geobacter sulfurreducens* [20]. For the identification of nitrifying bacteria, FITC-labelled NSO1225 and NSO190 probes were used to identify nitrifying bacteria – ammonium oxidizing bacteria (AOB) [21] – and Cy3-labelled probes consisting of NIT3, compNIT3 [22], Ntspa-0662 and compNtspa-0662 [23] were used to identify nitrite oxidizing bacteria (NOB). The probed sludge was examined using a Leica® confocal laser microscope. The area containing specific labelled probe cells was quantified as a percentage of the area of the entire bacterial population (EUBMIX).

For scanning electron microscopy (SEM) analysis, small pieces of anode electrodes with biofilms were taken from the air-cathode MFC and immersed in 2.5% (wt/v) glutaraldehyde in a 0.1 M cacodylate buffer at pH 7.4. The samples were then washed and dehydrated successively in ethanol series. The fixed samples were dried with a critical-point drier and sputtered with a 40 nm gold layer. The coated samples were examined with a SEM (model DSM-960; Zeiss, Germany) at 20 kV and the images were captured digitally.

3. Results and discussion

3.1. MFC performance: organic matter removal and electricity generation

Fig. 2 presents the organic loading removal rates (OLR and ORR, kg COD m^{-3} d^{-1}) and the power density evolutions in the air-cathode MFC treating landfill leachate during the experimental period.

During the first 46 days of the operation (Period 1) the influent organic matter content was 507 mg COD L^{-1} (102 mg BOD L^{-1}; Table 1) at an HRT of 8.3 ± 0.7 h. During this period, 0.47 ± 0.15 kg COD m^{-3} d^{-1} was removed (32% of removal efficiency), producing 6.1 ± 4.2 mW m^{-3} of electricity. When raw landfill leachate was fed in (Period 2), the OLR jumped from 1.48 ± 0.53 to 21.9 ± 6.2 kg COD m^{-3} d^{-1} by decreasing the HRT

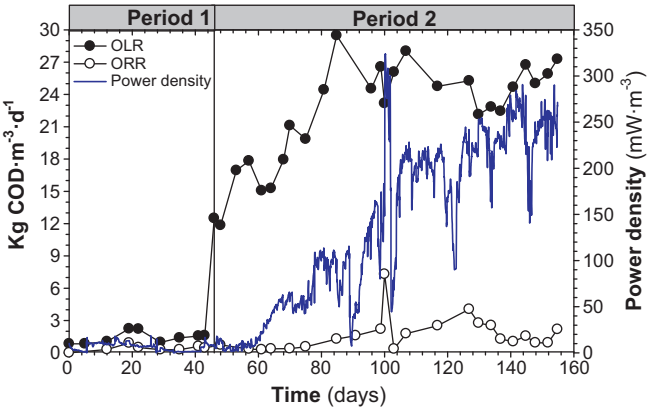

Fig. 2. Organic loading and removal rates (OLR and ORR, kg COD m^{-3} d^{-1}) and power density (mW m^{-3}) evolutions in the air-cathode MFC treating landfill leachate.

to 5.1 h. By feeding in raw leachate and as a result of bacterial growth, power density increased to 60.7 mW m^{-3} on day 67. At that moment, HRT was decreased again to 3.7 h with the corresponding OLR rise to 24.42 kg COD m^{-3} d^{-1}. 1.50 kg COD m^{-3} d^{-1} was removed producing 106.5 mW m^{-3} on day 81. The maximum level of ORR was achieved on day 100 at 8.51 kg COD m^{-3} d^{-1} (37% removal efficiency) with a power density of 344 mW m^{-3}. On days 90, 102 and 122 the MFC suffered several mechanical problems that influenced its performance, but it recovered fast. Towards the end of the experimental period (i.e. the last 30 days), the power density remained stable at values higher than 258 mW m^{-3}, treating 2.2 kg COD m^{-3} d^{-1}(10% of removal efficiency) The air-cathode MFC thus converted chemical energy (the biodegradable organic matter) directly into electricity. However, the coulomb efficiency (CE) was below 2% indicating the loss of substrate in a non-electricity generation process. In spite of the low CE, the power density produced was higher than reported in the literature [11,13], although these figures corresponded to the treatment of domestic landfill leachate with higher BOD_5 concentrations (BOD_5 between 4116 and 6300 mg BOD L^{-1}).

3.2. Power generation from landfill leachate

Electricity from landfill leachate was produced during the experimental period (Fig. 1). In order to monitor the electricity production, polarization curves were obtained on different operational

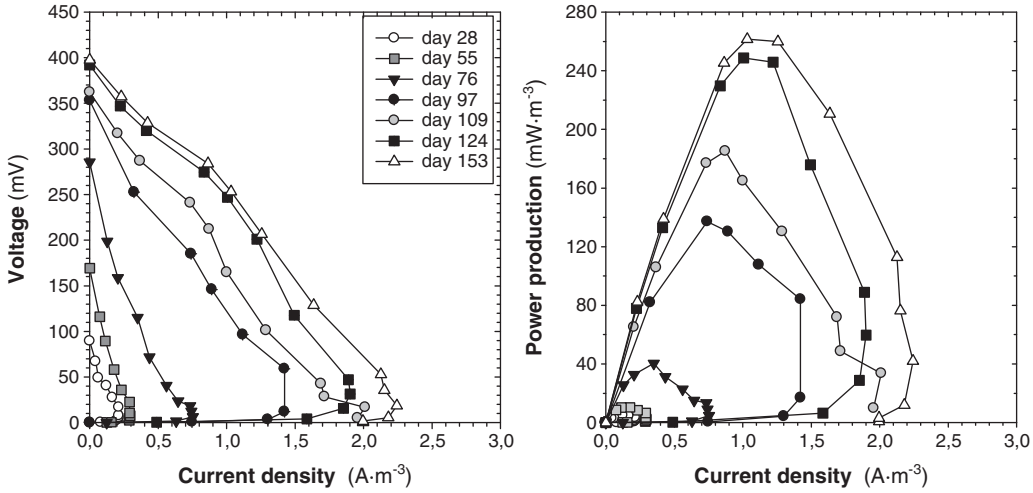

Fig. 3. Polarization curves of the MFC treating landfill leachate on different days of operation. Cell voltage (*left*) and power production (*right*) are in function of the current density.

days (Fig. 3). The maximum power density and open cell voltage (OCV) increased with time, reaching their maximum values on day 153 (278.2 mW m^{-3} and 398 mV, compared to day 28: 5.3 mW m^{-3} and 89 mV). From days 76 to 97, the MFC evolved significantly, and power density increased by 340%. This increment was also in accordance with the increase in the ORR, as shown in Fig. 2. Therefore, once raw leachate was fed in (on day 47), the system evolved faster. Not only organic matter availability of raw leachate favoured the MFC performance, the electrical conductivity increased from 8583 to 73,588 μS cm^{-1} (Table 1). As Liu et al. [24] demonstrated that the power density increases due to ionic strength and electrode spacing resulted from a decrease in the internal resistance (ohmic resistance). In this way, the external resistance of the air-cathode MFC was lowered from 4610 Ω to 1970 Ω (on day 70) and finally to 1462 Ω (on day 97) which resulted in a significant increase of the continuous current generation.

3.3. Nitrogen removal in the air-cathode MFC

The MFC successfully minimized the biodegradable organic matter present in the landfill leachate. However, the dynamics of nitrogen in the air-cathode MFC were unknown before this study. Nitrogen compounds (in the form of TKN, ammonium, nitrite and nitrate) were therefore monitored during the experimental period. Fig. 4 shows the evolution of the nitrogen loading rate (NLR, g N m^{-3} d^{-1}), the free ammonia concentration (FA), the effluent ammonium removed and the nitrite produced loading rates of the MFC treating landfill leachate.

During Period 1, the landfill leachate was diluted, giving a NLR of 2.71 ± 0.82 kg N m^{-3} d^{-1} and a salinity of 8583 μS cm^{-1}. The average ammonium removed in the effluent was 0.44 ± 0.30 kg N-NH_4^+ m^{-3} d^{-1} (16% removal efficiency). Nitrites (0.25 ± 0.20 kg N-NO_2^- m^{-3} d^{-1}; 124 mg N-NO_2^- L^{-1}) measured in the effluent suggested that partial nitritation (57% efficiency) took place. In this biological process carried out by AOB, ammonium is oxidized under aerobic conditions to nitrite. Since oxygen was not supplied in the MFC, it must be introduced by diffusion through the membrane [25]. Kim et al. [25] demonstrated that Nafion (the CEM used in this study) was the most permeable membrane to oxygen.

On the other hand, the ammonium not oxidized to nitrite (0.19 ± 0.30 kg N-NH_4^+ m^{-3} d^{-1}; 43% of the total ammonium removed) could be lost through the membrane [25,26], or through an ammonia liquid–gas transfer. Kim et al. [25] found that the volatilization of ammonia is enhanced by localized pH increase at the cathode in a single chamber MFC. This result was confirmed by checking the pH in the air-cathode MFC treating landfill leachate.

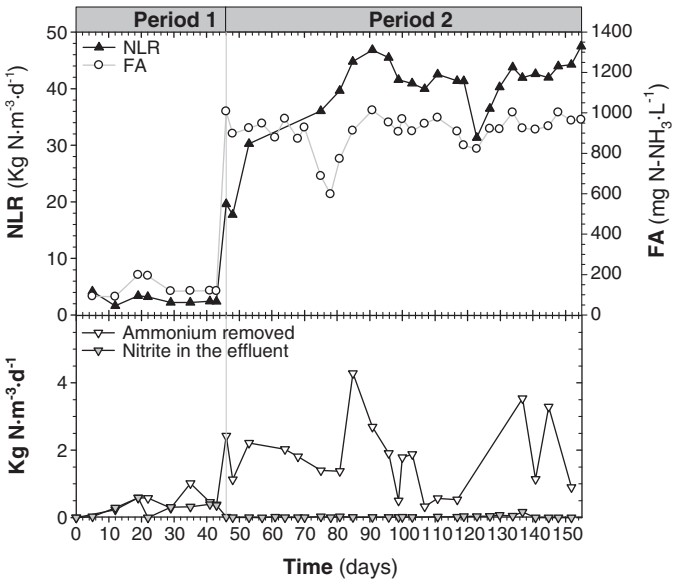

Fig. 4. Nitrogen loading rate (NLR), free ammonia (FA), levels of ammonium removed and effluent nitrate load evolutions in the air-cathode MFC.

The pH was higher than 11. At this pH, all ammonium present in the cathode is quickly converted to ammonia.

Once raw leachate was fed in (Period 2), the NLR increased to 19.6 kg N m^{-3} d^{-1}, reaching a maximum of 46.8 kg N m^{-3} d^{-1} and a salinity of 73,588 μS cm^{-1} on day 90. From that moment onwards, neither nitrites nor nitrates were detected in the effluent. According to Anthonisen et al. [27], the high nitrogen concentration plus the operational conditions (a temperature of 23 °C and a pH of 8.6) resulted in 20% of the nitrogen (892 mg N-NH_3 L^{-1}) being present as free ammonia (FA, NH_3). It has been observed in the relevant literature that this high value inhibits the performance of the nitrogen organism [27]. Therefore, the nitrite absence (<0.01 ± 0.01 kg N-NO_2^- m^{-3} d^{-1}; 2.1 mg N-NO_2^- L^{-1}) must have been caused by FA inhibition. However, ammonium was still removed from the MFC (1.59 ± 1.07 kg N-NH_4^+ m^{-3} d^{-1}; 4% removal efficiency). This may have occurred as a result of physical–chemical processes [24–26]. Moreover, the ammonium lost was increased in the Period 2 when the MFC produced more power. This fact indicates that ammonium is being transported across the membrane to maintain the charge-balance [25].

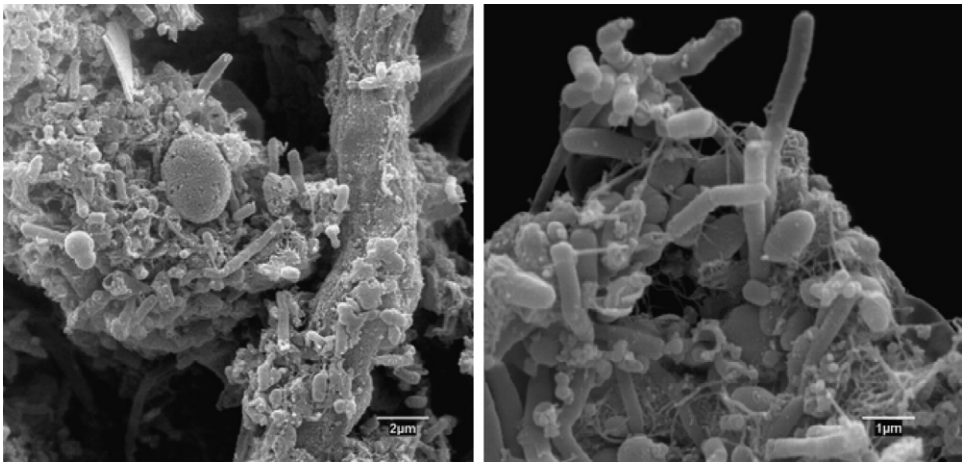

Fig. 5. SEM images showing the anode biofilms taken from the MFC on days 43(left; Period 1) and 154 (right; Period 2).

3.4. Microbiology

Landfill leachate treatment and energy production were also assessed using microbiology techniques, i.e. SEM and FISH analysis. SEM observations from the biofilm (Fig. 5) demonstrated the predominance of microorganisms with bacilliform morphology. The aggregated microorganisms were firmly attached to the surfaces of the electrode with nanowire-like filamentous appendages [28]. Fig. 5 shows the exoelectrogenic bacteria that evolved during the experimental period, transporting the electrons through their nanowires. Apart from the geobacter species, other types of microorganisms were indentified, suggesting that the biofilm was not a pure culture. The high conductivity of the leachate induced salt precipitation, as shown in Fig. 5.

FISH analysis was used to identify *Geobacter sulfurreducens* and the ammonium and nitrite oxidizer bacteria (AOB and NOB). At day 90, the FISH analysis showed the presence of AOB ($4.5 \pm 0.2\%$) and NOB ($2.5 \pm 0.5\%$) in the sludge. AOB are responsible for oxidizing ammonium to nitrite, after which NOB oxidize nitrite to nitrate, as observed in Fig. 4 (Period 1). However, in Period 2, in spite of their presence, they were nonetheless inhibited by high FA concentrations.

Regarding exoelectrogenic bacteria, *Geobacter sulfurreducens* was identified in the sludge. The Geobacter population increased from $5.0 \pm 0.4\%$ to $15.4 \pm 0.7\%$ between days 0 and 90. This increment positively correlates with the power density profile (Fig. 2). It is important to take into account that other exoelectrogenic bacteria could be present in the sludge. Therefore, further microbiological studies should be done to identify the bacteria concerned.

4. Conclusions

The use of MFC led to the treatment of the biodegradable organic matter of landfill leachate and the production of electricity even with high nitrogen content and salinity. A maximum amount of organic matter ($8.5\,kg\,COD\,m^{-3}\,d^{-1}$) was removed with a power density of $344\,mW\,m^{-3}$. The *Geobacter sulfurreducens* population increased during the experimental period.

The nitrogen content in leachate suffered transformations inside the MFC. Ammonium was removed from the leachate resulted from ammonium transfer through the membrane or from ammonia loss. Neither nitrites nor nitrates were detected in the effluent when raw leachate was fed in, due to the high free ammonia content which inhibited nitrifier bacteria. High salinity content benefited the MFC performance increasing power production because decrease the internal resistance.

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