Development of a module of stacks of air-breathing microbial fuel cells to light-up a strip of LEDs

S. Mateo¹, A. Cantone², P. Cañizares¹, F.J. Fernández-Morales¹, O. Scialdone², M.A. Rodrigo^{1,*}

¹ University of Castilla-La Mancha, Faculty of Chemical Sciences & Technologies, Chemical Engineering Department, Avenida Camilo José Cela, 12. 13071 Ciudad Real, Spain.

² Università degli Studi di Palermo, Dipartimento di Ingegneria Chimica, Gestionale, Informatica, Meccanica, Viale delle Scienze, Palermo, Italy.

Abstract

This work focuses on scaling-up of the microbial fuel cells technology according to the principle of miniaturization and multiplication. Seven stacks of 16 mini-MFCs (electrode area of 0.86 cm²) were built up leading to a big module of 112 MFCs. The electrical connection among the MFCs in the stacks and among the stacks into the modules was optimized in order to implement this technology. Results show that 1 MFC generates 1.22 mW while the optimization of the electric connection in order to achieve the maximum power results in 6.62 mW compared to the theoretical 182 mW, indicating the existence of large energy losses in the system. However, to light a LED there is not a power threshold but there is a requirement of input voltage (2.6 V) and input current (0.020 mA). For this reason, another optimization of the electrical configuration was carried out to satisfy the threshold values of voltage and current and a strip of 220 LEDs was illuminated for several days. Furthermore, robustness of the MFC technology was confirmed after operating simultaneously 112 MFCs with reproducible performance for 30 days.

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26 **Highlights**

- 27 - Scale-up through stacking miniaturized MFCs is a successful strategy.
- 28 - The parallel-series connection of the MFCs stacked affects the performance.
- 29 - The stacks-modules connection is the key to light up 220 LEDs.
- 30 - Reproducible performance of the 112 MFCs supports its robustness.

31 **Keywords**

32 Microbial fuel cell; miniaturization; multiplication; stack; electrical connection; LED

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34 * Author to whom all correspondence should be addressed: manuel.rodrido@uclm.es

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1. Introduction

The possibility to recover the energy contained in the wastewater through microbial fuel cells (MFCs) has gained prominence in the scientific community during the last fifteen years. Despite of the numerous researches in this field since its conception by Potter in 1910, its real applicability in wastewater treatment and energy sectors is considered very difficult because of the scale-up stage [1]. This stage is limited by the electrode spacing, the surface area to volume, the structure of the MFC, the electrodic materials and the composition of the wastewater, among others [2, 3]. However, from an economic point of view, it has been demonstrated that MFCs can become a real alternative to conventional activated sludge for wastewater treatment, because of their lower operational costs for the COD removal [4].

The implementation of this technology has been investigated from micro-scale MFCs [5-10] up to a size of thousands of liters [11-13]. The first pilot plant of 1 m³ was built in Australia with the purpose of treating brewery wastewater [14] while a MFC of 100 L was created by Ge et al. with the aim of extracting energy for external ultracapacitors [12]. However, both studies were focused on the degradation process of the organic matter instead of on the electricity generation. Other works discarded the increase in the size of the MFC as a strategy because of efficiency loss due to a considerable increase in the internal resistance, which inhibits the electron donation [15]. Recent works point out the replication of miniaturized MFCs and its arrangement in a stack as a viable alternative to carry out the scale-up [5, 16, 17]. Miniaturization of MFCs leads to a reduction of the distance between electrodes, favoring the electron transfer, the control of the nutrients flow and the reduction of the internal resistance [5, 18], limiting the efficiency losses and, therefore, enhancing the MFC's electrical performance. Electrical and hydraulic connections between the MFCs of the stack allow maximizing the performances of the

stack. On one hand, parallel and series connection allows the increase in the current and the potential, respectively [15, 19]. The combination of both electrical connections in a stack can result in an energy dense device. On the other hand, hydraulic connection can enhance the removal of organic matter [20, 21].

The first “real application” of a MFC is probably a train toy named Gastrobot (robot with stomach) [22]. Instead of being supplied by standard chemical batteries or by photovoltaic panels, it was provided with batteries charged with the energy obtained in an abiotic/chemical fuel cells stack. This stack was fed with the products generated when *E. Coli* metabolizes sugar in the artificial stomach of the robot [22]. This creation leads to the first robot powered by MFCs named EcoBot-I fed with glucose. This robot was provided with capacitors that allows the storage of the energy generated by the MFCs and used to run the machine [23]. Many improvements were carried out before developing the first autonomous and self-sustainable robot. This robot was provided with a stack of miniaturized MFCs [24]. MFCs have been also proved to charge mobile phone and it was shown that self-sufficient MFCs stack can supply its own feeding pumps. Other practical implementations of MFCs has been developed in the biosensors field. A change in the current exerted by a MFC can be related to a change in toxicity, the temperature, the concentration of organic matter or of specific compounds such as Cr in a water [25-34]. Synthetic wastewater is frequently employed in MFCs but its operation with municipal sewage as well as industrial wastewaters has also been tested satisfactory [35, 36].

In this context, the main of this work is to create a module of 7 stacks, with 16 mini-MFCs for each stack, capable to light up a strip of LEDs. Different electrical combinations were tested in the stacks and in the module with three purposes: maximizing the energy output, lighting-up a chain of 220 LEDs and demonstrating the real applicability of this technology in order to power low energy demanding devices.

2. Materials and Methods

2.1. Stack construction. The experimental set-up consisted of a module of 112 MFCs.

The 112 MFCs were arranged in 7 stacks, with 16 mini-MFCs for each stack. The arrangement of the MFCs in a stack and the dimensions expressed in centimetres are shown in Figure 1. Each stack was built up assembling four polymeric material boards of 4 mm thick: one board for the inlet of the wastewater to the anodic chamber according to the Fig 1.A, two boards to form the anodic chamber and one board to create the cathodic chamber following the structure of Fig 1.B.

Figure 1. a) Design of a stack of 16 MFCs: Inlet board for the anodic chamber (the larger holes represent the bolts, while the smaller holes represent the inlets and outlets of each

MFC). b) Design of a stack of 16 MFCs: Anodic and cathodic inner boards. c) Real view of a stack of 16 MFCs. d) Scheme of a single MFC.

For an easier comprehension, the construction of one MFC is going to be studied in detail. Each single MFC was based on air-breathing carbon paper (Freudenberg C2) cathode presenting 0.866 cm^2 of active area in direct contact with the air of the atmosphere. The cathode contains 10% of Teflon and its surface was modified with the addition of a load of $0.5 \text{ mg Pt cm}^{-2}$ [37]. The cathode was separated from the anode with a Nafion N117 proton exchange membrane (DuPontTM Nafion PFSA Membrane), due to its film thickness of $183 \text{ }\mu\text{m}$ and good performance. Anode, membrane and cathode were placed as close as possible to reduce the internal resistance. The anode consisted of carbon felt [38] with an active area of 0.866 cm^2 , 3 mm of thickness, $35.3 \text{ m}^2 \text{ g}^{-1}$ of specific area, 500 g m^{-2} of area weight, 95% of porosity and $30 \text{ }\mu\text{m}$ of roughness. The anode was located in the anodic chamber facing to the wastewater and bacteria by the side not in contact with the membrane. The anodic chamber had a volume of 0.695 cm^3 . Anode and cathode were externally connected by a $120 \text{ }\Omega$ resistance.

2.2. Stack operation. Cascades of four MFCs in horizontal position formed the hydraulic connection. The vertical arrangement was discarded in order to avoid large pressure drops, which caused hydraulic problems. Each cascade was connected to an auxiliary anodic tank of 115 mL. This tank was filled with electrogenic bacteria from an on-going MFC, which was used as the inoculum and was recirculated to the MFC during the first day. The second and third days, the 50% of the volume was removed and replaced by fresh sludge. From the fourth day, a sludge age of 2.5 days was established: 46 mL of the volume was purged and replaced by fresh wastewater in a daily basis [39]. It is important to remark that the initial seed was collected from the aerobic reactor of the Wastewater

Treatment Plant of Ciudad Real described elsewhere [40] and treated according the same criteria to inoculate the on-going MFCs [41]. During the experiments, the temperature was kept at 23°C. Wastewater was prepared in the laboratory to ensure the same conditions and composition. It contained 16.10 g L⁻¹ of sodium acetate, 2.77 g L⁻¹ of sodium carbonate, 1.11 g L⁻¹ of potassium dihydrogen phosphate, 1.25 g L⁻¹ of calcium chloride, 0.92 g L⁻¹ of hexahydrate magnesium chloride, 1.85 g L⁻¹ of ammonium sulphate and 0.07 g L⁻¹ of ferric ammonium sulphate. Before being fed to the MFCs, the synthetic wastewater was autoclaved at 120° C for 15 minutes [42].

2.3. Electrical connection. MFCs were seeded and started under close circuit conditions by an external resistance of 120 Ω. Each MFC was connected to an external load, so the electrical connection was individual. Once the steady state was reached, parallel, series and combination of both electrical connections were tested. The parallel connection was carried out by connecting all the anodes leading to a unique anodic output while in series, the cathode of the first MFC is connected to the anode of the second MFC and so on until the last MFC of the stack. In this case, the anode of the first anode and the cathode of the last MFC remain unconnected so the electrical circuit is closed by connecting them.

2.4. Electrochemical measurements. The electricity production was measured continuously every 5 minutes in terms of potential difference under close circuit conditions with a Keithley 2000 Multimeter. Polarization curves were obtained by carrying out a potential sweep from the OCV to 0.001 V with a potentiostat/galvanostat AUTOLAB PGSTAT30 at a scan rate of 0.001 V s⁻¹.

3. Results and Discussion

3.1. Performances of individual MFCs. First, the stability and reproducibility of all the MFCs connected in each of the seven stacks was evaluated.

Figure 2 shows the relationship between the stationary current density of single MFCs under an external load of $120\ \Omega$ each one and the time required to achieve it by each MFC. The results obtained at the steady state are shown as the average value reached by the single 16 MFCs of each stack.

Figure 2. Stationary current density under an external load of $120\ \Omega$ versus the time required to achieve it. Data represents the average and the standard deviation of the single 16 MFCs arranged in a stack for each point.

It can be observed that the biofilm formation took between 18 and 22 days in all the single MFCs of the stacks evaluated while the current densities at the stationary phase ranged from 6.0 up to $10.5\ \text{A m}^{-2}$. Both facts are very important from the point of view of the operation reproducibility, because there were no great divergences between the

MFCs contained in the seven stacks studied. In addition, considering the average of the single MFCs per stack, the higher current densities were usually observed for the stacks that needed lower time for the start-up of the bioelectrogenic activity. This relationship may be explained in terms of a more successful colonization of the anodic surfaces by the inoculum in stacks 7 and 4 as compared to stack 3 and 2. However, results were very close and their dispersion was low enough to be considered.

Figure 3 shows the results obtained during the electrochemical characterization for the individual MFCs of each stack by polarization curves. Each point is represented by the average of the single MFCs that make a stack up, electrically operated individually. It can be observed that regardless the electrochemical parameter, there were not relevant differences between the seven stacks evaluated as the average of their single MFCs, indicating again the robustness of the stacks system.

Figure 3. Performances of the individual MFCs per stack. a) OCV (●) and maximum current density (□). b) Maximum power density (●) and current density at the moment at which the maximum power is achieved (□). Each data point represents the average and the standard deviation of the single 16 MFCs arranged in a stack for each point.

OCVs show values within the range of 0.41-0.47 V while the maximum current density changes between 11.2-18.1 A m⁻². These values are very remarkable for this type of MFC, in particular when comparing with other studies, which obtained maximum current densities of 0.48 A m⁻² [43], 0.8 A m⁻² [44], 9.0 A m⁻² [45], 0.0073 [5], 2.2 A m⁻² [46], clearly under the values reached by the MFCs tested in this work. These results reflect how powerful and robust the MFCs of this study are. Maximum power values range between 1.36-2.29 W m⁻².

3.2. Assessment of the electrical configuration of individual stacks. Basic conventional circuit theory points out that the output of the parallel connection of power supply devices is the sum of the individual currents produced by each of them, while the sum of the potentials is obtained with their connection in series. Shifting this statement to the particular case of the microbial fuel cells (considered as power supply devices), the combination of the performances of individual MFCs should may help this technology to be applicable to real-life with the production of more important amounts of energy. Thus, it is important to find the best way to achieve the best output: parallel, series or a combination of both. Theoretically, regardless the configuration, the output power must remain constant. In practice, the parallel arrangement is expected to get an overall resistance equals to the lowest individual resistance, while the series configuration causes reversal voltage problems [15]. The combination of both may help to solve the reversal problems, despite a slight increase in the internal resistance could happen. For this reason, this section is focused on the electrical connections of the 16 MFCs of each stack, in order to find the best configuration. All stacks have 16 MFCs connected in different ways. In order to sum up the type of connections into a single number, the parameters P-S = (parallel connections related to MFCs/series connections related to groups of

MFCs)/number of cells is used. A P-S=0 was used to identified clearly all the MFCs in series while a P-S=1 indicates that all the MFCs are connected in parallel.

Table 1. Electrical configurations of a stack of 16 MFCs.

Configuration in Series (number of packs)*	Configuration in Parallel (number MFCs)**	Scheme	Ratio Parallel-Series referred to the MFCs
16	1		0
8	2		0.016
5	3		0.037
4	4		0.0625
2	8		0.25
1	16		1

*Number of packs connected in series; **Number of MFCs connected in parallel that forms a pack. (Design: parallel → dashed line; series → continuous line)

In our study, the connections studied leads to six combinations as shown in Table 1. For example, a P-S=0.25 indicates that there are 2 groups of 8 MFCs. The 8 MFCs are connected in parallel (dashed line in the scheme) leading to two groups which are

connected in series (continuous line in the scheme). Figure 4 reports the OCV, the maximum current and power and the internal resistances obtained from the polarization curves carried out for the seven stacks when they were operated at the steady state.

Figure 4. a) Evolution of the OCV (●) and the maximum current (○) with the P-S ratio.
b) Evolution of the maximum power (●) and internal resistance (○) with the P-S ratio.

It can be observed that the OCV decreases when the P-S ratio tends to high values (parallel connections). A maximum voltage of 6.44 V is achieved by arranging all the MFCs in series. This value is much higher than the 0.39 V reached in parallel. Otherwise, as expected, the enhancement of the P-S ratio (i.e., the increase in the number of the cells connected in parallel) results in an improvement of the maximum current from 0.48 (all MFCs connected in series) up to 6.80 mA obtained when all the cells were connected in parallel. According to Figure 4b, the power produced is favoured with the parallel connection: when all the cells were connected in series or in parallel, the power was 0.76 and 1.33 mW, respectively, because of the dramatic decrease of the internal resistance achieved for the parallel connection (fig. 4b). Indeed, the internal resistance [47] registered a value of 8305 Ω for a P-S of 0 (all MFCs connected in series) and a value of 45.9 Ω when the ratio is 1.00 (all cells connected in parallel), respectively. When a mixed arrangement was used, the best results were

achieved for a value of the P-S ratio of 0.25 (two packs connected in series, each pack containing 8 MFCs connected in parallel); with this arrangement, the power experienced a quite high value of 1.3 mW, very similar to the theoretical value of 1.51 mW, and to that achieved with all MFCs connected in parallel. Conversely, the arrangements with a high number of cells connected in series gave the worst results. As an example, the arrangement with a P-S ratio of 0.25 (achieved with 2 packs connected in series, each pack formed by 8 MFCs connected in parallel) results in a maximum power of 0.214 mW drastically lower than that achieved with a P-S ratio of 0.25. This performance can be explained by the cell potential and current achieved. Under maximum power conditions, the voltage reached was 0.942 V instead of the theoretical value of 1.4 V corresponding to the electrical connection. These energy losses probably can be a consequence of the weak MFCs, whose voltage tends to decrease quickly under low external resistances [43].

Furthermore, not only the voltage is quite different from that expected, but also the current, which is 0.266 mA value much lower than the theoretical 1.079 mA that should be reached. In this case, the internal resistance experienced a considerable value reaching 4.16 k Ω .

More in general, on the bases of the results reported in Fig.4, the more appealing configurations seems to be that with a large number of cells connected in parallel.

3.3. Connection of stacks to form modules. Once it has been evaluated the influence of the electrical connections of the MFCs into a single stack, the following point to be covered in this work focuses on the connection of stacks to form modules. For this study, P-S ratios of 0.016, 0.0625 and 0.25 have been used to connect single MFCs inside each single stack, which is aimed to be further connected into modules.

The connection among the stacks was carried out, with a similar approach to that used to connect MFCs in the single stack, connecting stacks in parallel to form packs and connecting the packs in series, trying to reach the best electric output as possible. In the standard configuration, three packs (consisted of 2, 2 and 3 stacks connected in parallel) were used with three independent electrical outputs. Then, the 3 packs were connected in series (see table 2, “Standard configuration”). The inverse configuration was also tested for comparison purposes: two individual pairs of two stacks and three stacks in series leading to three packs of stacks whose outputs were connected in parallel (see table 2, “Inverse configuration”).

Table 2. Electrical connection between the stacks of the module.

The performance of the different combinations has been evaluated in terms of internal resistance, OCV, maximum current and power from the polarization curves and it is reported in Figures 5 and 6.

Figure 5. Evolution of the internal resistance (5a) and of OCV (5b) with the P-S ratio inside the stack. (■) Experimental data for Inverse Configuration; (●) Experimental data for Standard Configuration. (□) Theoretical data for Inverse Configuration; (○) Theoretical data for Standard Configuration.

Figure 6. a) Evolution of the maximum current (6a) and of the power (6b) with the P-S ratio inside the stack. (■) Experimental data for Inverse Configuration; (●) Experimental data for Standard Configuration; (□) Theoretical data for Inverse Configuration; (○) Theoretical data for Standard Configuration.

As expected the module with Standard Configuration outperforms the other module tested. However, values are much lower than the maximum theoretical values expected, indicating the existence of large energy losses in the system. Experimental values approached to the theoretical ones when the stack P-S ratio increased for the OCV and at low stack P-S ratios for the current and power. It is very interesting to highlight the experimental trend observed for the maximum current and power in Figure 6, which is very similar regardless the connection of the cells in the stacks. On the other hand, this connection affects considerably the outputs values at low P-S ratios of the MFCs.

3.4. Connection of stack modules to light up a stripe of LEDs.

Once characterized the electrical performance of the stacks and modules, and in order to check the applicability of the staking of MFC, it was decided to use the modules to light up a stripe of LEDs.

Before the study, one of the LEDs of the stripe was characterized (Fig 7). The characteristic curve shows the operational voltage and current. For a proper performance, the LED must be supplied with the optimum values of voltage and current of 3.4 V and 0.029 mA, respectively, to work in its best conditions and higher values must be avoided, because from this value on, the current increased quickly and the LED can be damaged and burnt.

Figure 7. Characterization curve of the LED used.

The relevant information of this Figure is in the threshold values. The LED required 2.6 V and 0.020 mA to start lighting, as checked experimentally. For this reason, the module was connected with an electrical configuration that ensures the threshold current and voltage. It is important to take into account many considerations: 1) the internal resistance of the stack entails an important energy loss and must be reduced according to Eq. 1; 2) the system should work under the required voltage and the resulting current when closing

the circuit with the LEDs; and, finally, 3) the connection between the LEDs. A stripe of 21 LEDs was the initial goal stated. The LEDs connected in series require a voltage of 54.6 V and a current of 0.020 mA, while a parallel connection implies a current of 0.42 mA and a voltage of 2.6 V.

$$V_{\text{inlet}} = I_{\text{threshold}} \cdot R_{\text{int MFC}} + I_{\text{threshold}} \cdot R_{\text{LED}} + V_{\text{threshold}} \quad (1)$$

where R_{LED} can be considered negligible.

According to theory, series connections will provide the voltage but the current remains constant. At OCV (maximum voltage) conditions, the MFC produced 0.531 μA , value unsuitable for lighting up the LEDs. On the other hand, the total parallel would not produce an increase in the voltage higher than 0.450 V. Hence, proper combinations of series-parallel arrangements are necessary.

These combinations were studied, for single MFCs, in the previous sections of this work. For each combination, the requirement of voltage, taking into account the losses due to the respective internal resistance, was checked as well as the current established at the resulting voltage. In this way, most of the combinations were discarded because one of the requirements was not achieved. Both requirements were reached only operating with a ratio P-S of 0.0625 between the MFCs of each stack and a ratio P-S of 0.111 between the stacks of the whole module. This combination leads to an internal resistance of 464 Ω . To overcome this resistance, an input of 2.81 V must be supplied to light-up the LED strip; the OCV was 3.61 V while the current that circulates through the system at the ignition voltage of the LEDs of 2.6 V was 1.73 mA, value much higher than the requirement of 0.42 mA. Indeed, when the stack was operated under these conditions, the strip allowed the illumination of the 21 LEDs. The lighting time was monitored and it shined continuously for 20 days, moment at which more LEDs were added to the strip

(Figure 8). Every day, 10 more LEDs were added up to 220 LEDs. As shown in the photos, the brightness diminishes upon increasing the LEDs in the chain while the voltage passing through the chain declines very slowly. The illumination of the LEDs was maintained during 5 days before stopping the experiment, showing a very successful result of the test performed.

Figure 8. a) Evolution of the voltage circulating through the LED chain. b) Number of LEDs added to the chain with the time.

Conclusions

From this work, the following conclusions can be drawn:

- The miniaturization and multiplication of MFCs is a successful scale-up strategy. By using this approach with a device consisting of 112 MFC (a module with 7 stacks, each one composed by 16 MFCs), up to 220 LEDs were lighted up simultaneously and continuously for several days.
- Robustness of the MFC technology has been confirmed after operating simultaneously 112 MFCs with reproducible performances for a very long period.
- The energy output of a stack of MFCs is affected by the electrical connection and favoured by the parallel connection. The smart combination of parallel and series connection is necessary for practical applications. Strategy followed in this work (connecting MFC into stacks and stacks into module) was successful.

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