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From single MFC to cascade configuration: The relationship between size, hydraulic retention time and power density



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ABSTRACT

Achieving useful electrical power production with the MFC technology requires a plurality of units. Therefore, the main objective of much of the MFC research is to increase the power density of each unit. Collectives of MFCs will inherently include units grouped in cascades, whereby the outflow of one is the inflow to the next unit; such an approach allows for better fuel utilisation. However, such a configuration is subject to some important considerations, including: the size of the MFCs; the number of units i.e. the length of the cascade; hydraulic retention time; fuel quality; and optimisation of anode surface and microbial colonisation. In the present study, optimisation of the aforementioned aspects has been investigated in order to establish the most appropriate cascade design. Results demonstrate that an increased flow rate of treated urine achieved equal power density with the same setup when fed with fresh urine at a lower flow rate. The independent investigations of these parameters have led to the design of a cascade that maintains uniformity with regard to the aforementioned parameters, by incorporating units of decreasing size, thus allowing locally shorter hydraulic retention times and therefore leading to increased power density levels.

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Introduction

Microbial fuel cells are energy transducers consisting of an anode, a cathode and typically a cation exchange membrane. Microorganisms consume organic fuel as their carbon-energy and electron source, with the anode electrode serving as their endterminal electron acceptor. Microbial anaerobic respiration in the anodic compartment, also releases cations that flow through the membrane into the cathodic compartment. The electrons and the cations such as protons react together and reduce the oxidant present in the cathode (e.g. oxygen) [1,2]. The first report of a functioning MFC was published in 1911 [3], however, research mainly gained significant interest during the two last decades [4–6]. In the context of reducing energy consumption, MFC technology is of interest since it can treat organic waste from various sources (e.g. agricultural, industrial, anthropogenic) without having to spend energy, as is the case for existing wastewater treatment

processes [7]. Treating organic waste at low cost is an important aspect in countries of the Developing World, since water pollution by humans and animals is one of the major causes for gastrointestinal disease outbreaks. Recent studies have demonstrated the potential of using urine directly as a fuel for electricity generation, [8,9]. Ongoing work in scaling up the microbial fuel cell technology towards out-of-the-lab scale is regularly reported, with a number of approaches being pursued.

One of these approaches is the stacking of small-scale MFC units for useful electricity generation [10]. The approach employed in this study is similar to the one described previously [11,12], where collectives of MFCs are assembled in a cascade manner. Such setup allows for a better utilisation of the organic matter into electricity because of shorter diffusion distances [10–12]. By using a plurality of cascade modules connected together, practical levels of power – at operational voltages – can be achieved. Such configuration is subject to some important considerations, including (i) size of MFCs; (ii) number of units/length of a cascade; (iii) hydraulic retention time; (iv) fuel quality; and (v) optimisation of anode surface and microbial colonisation. All of these aspects directly influence the power density of each MFC, thus the efficiency of the system as a whole.

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In addition to the structural material of a MFC, a limitation also appears regarding the size of MFCs, whereby large-volume units tend to have lower power densities and hence decreased efficiency [10]. Small scale units on the other hand produce higher energy densities, and this can be explained by the increase of surface area-to-volume ratio (SA:V; of the electrode macro-surface area to chamber volume), as well as the distance between the anode and cathode electrodes. This includes the proton diffusion distances between an anode and a cathode, the rate of supply and diffusion of fuel molecules into the anodic biofilm electrode and the electron paths from the electro-active organisms both in the biofilm and planktonic phases. As for the cascade length (number of MFCs within a cascade, or length of elongated units), this is also important, due to the inevitable gradients emerging from the downstream treatment process, where MFCs at the latter stages, receive depleted fuel. For a given fuel quality or concentration. the power densities of the last units of a cascade are likely to decrease, thus requiring an optimisation strategy. For example, increased flow rates would ensure a more uniform distribution of growth-limiting nutrients along the cascade, or a dynamic reconfiguration system could bypass (disconnect) underperforming units in the cascade, in response to changes in fuel quality. In the present study, optimisations of the aforementioned aspects have been investigated in order to establish the most appropriate design of cascade.

The first step was to investigate the relationship between the anode density (amount of anode per volume of anolyte) and power-density (amount of power per volume of anodic chamber). The second step was to study the relationship between the power density and the SA:V ratio. It was hypothesised that if SA:V ratios remain constant, the power density should remain the same per unit length or volume. The relationship between the number of passages of a fixed volume of urine through an MFC, the hydraulic retention time (HRT) and the power density, were then investigated.

In geometry, it is well known that for shapes like cylinders, the ratio of internal surface area to volume increases, with decreasing size. For rectangular shapes, this is also the case when membranes or electrodes occupy the two largest surfaces of the container. However in this case, the SA:V ratio also remains constant if the height is constant, when both length and width are varied equally. It is because of these principles that the study started with cylindrical MFCs and then moved to rectangular shapes.

Materials and methods

Tubular MFCs: anode sizes

Tubular MFCs were made of ceramic tubes (EM80P; Anderman Industrial Ceramics, Kidderminster, UK) serving as both structural and membrane materials separating the inner anode from the outer cathode. Each tube was 100 mm long, 26 mm wide and 18 mm in internal diameter (25 mL). The cathode was applied as a conductive coating using a mixture (conductive plasti-dip, CPD) consisting of 3 g of graphite powder, 0.6 g dry weight of synthetic rubber coating (Plasti Dip International, Minnesota, USA) and petroleum ether (80°-110°) as solvent. All MFCs were coated with $3.75 \text{ g} \pm 0.25 \text{ g}$ (final dry weight) of the conductive mixture. The anode sizes that were investigated ranged from 200 cm² to 1400 cm² of 20 g m⁻² carbon fibre veil electrode (PRF Composite Materials Poole, Dorset, UK). Anodes had a tubular shape by wrapping the material around an inner stainless steel current collector, which was then placed within the ceramic tubes. Since the internal diameter of the ceramic tube was fixed, the volume occupied by the anodes increased with increasing electrode surface area (200 cm², 400 cm², 800 cm², and 1400 cm²): the smallest anode (200 cm²) was forming a thin film of 2 mm, whereas the largest anodes were completely utilising the available space. Each condition was tested in triplicate resulting in 12 independent MFCs.

The MFCs with $1400\,\mathrm{cm}^2$ anodes were also employed in the investigation of urine recirculation into the same MFCs. For the first passage, fresh urine was pumped at a flow rate of $1.9\,\mathrm{mL}\,h^{-1}$ (HRT = 13h10), which was the same as for the anode size experiment above. For the second passage of urine, the flow rate was initially left at $1.9\,\mathrm{mL}\,h^{-1}$ and was then increased to $8.1\,\mathrm{mL}\,h^{-1}$ (HRT = 3h00) until fuel exhaustion. For the third passage, the flow rate was increased to $16.5\,\mathrm{mL}\,h^{-1}$ (HRT = 1h30), which was retained for the subsequent fourth, fifth and sixth passages.

Tubular MFCs: membrane surface area to chamber volume ratio (SA:V)

Diameter investigation

Tubes (same as above) of 100 mm lengths but of different diameters were employed to investigate whether the power density increases with the reduction of size, as previously suggested [10]. The larger MFCs (n=2) had an internal diameter of 26 mm, whilst the smaller units had an internal diameter of 18 mm (n=3). Both MFCs had tubular carbon-veil anodes of 800 cm², as described above. The surface area to chamber volume (SA:V) ratio was $1.54 \, \mathrm{cm}^{-1}$ and $2.26 \, \mathrm{cm}^{-1}$ for the larger and smaller MFCs, respectively. The outer surfaces of the MFCs were coated with the same loading CPD. The flow rate of urine was set at 1.9 mL h $^{-1}$.

Length investigation

The MFCs had identical diameters but variable lengths (5 cm, 10 cm and 40 cm), resulting in a constant SA:V ratio of 2.26 cm⁻¹. Anodes were wrapped around an inner current collector (stainless steel grid) as described above. The total surface area of the anodes was 400 cm² for the 5 cm long MFCs, 800 cm² for the 10 cm long MFCs, and 2800 cm² for the 40 cm long units (rather than 3200 cm², which was not feasible due to physical limitations). For the effect of length experiment, the three 5 cm long MFCs were paired electrically in series or parallel, and all combinations were investigated, i.e. MFC1-MFC2: MFC1-MFC3 and MFC2-MFC3. The error bars were therefore generated from these three combinations, and for both configurations, under polarisation experiment conditions (see below). MFC cathodes were coated with the same loading CPD but obviously at loadings proportional to length. These MFCs were inoculated, matured and run in batch-fed mode to allow homogeneous mixing of the nutrients all along the anode of each MFC.

Flat MFCs: comparison of two cascades; control cascade (CC: 4 MFCs of same size) versus test cascade (TC: 4 MFC of various sizes)

For the control cascade (CC), MFCs were made of acrylic frames with an internal anodic compartment of 10 mm height, 100 mm length and 60 mm width (Fig. 1). The two baffles at the input and output of each MFC (Fig. 1a) were incorporated into the design to enable laminar flow conditions. The aim of having laminar flow was to ensure a homogenous distribution of nutrients along the entire width of all MFCs, in order to make comparisons valid between the MFCs of various widths. The anodes (40 cm² cm⁻³) were made of carbon veil (20 g m⁻²; PRF Composite Materials Poole, Dorset, UK) folded down to fit inside the chambers and a stainless steel wire served as the current collector. Flat ceramic membranes (2.5 mm thick) were fixed to the largest faces of the acrylic frame, thus sealing the internal anodic compartment. The cathode consisted of conductive plasti-dip (CPD) coating, with the same carbon-to-PlastiDip ratio as for the tubular MFCs, which was painted $(6.0 \text{ g} \pm 0.150 \text{ g})$ over a single sheet of carbon veil. This sheet of CPD was then put on both sides of each MFC.

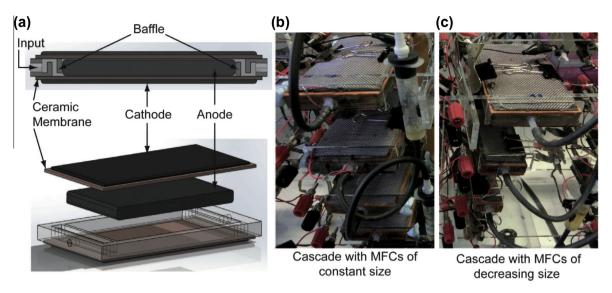


Fig. 1. Setup of the cascades. (a) A flat MFC employed in the building of a cascade (b) A cascade with constant size MFCs. (c) A cascade with decreasing sizes MFCs.

For the test cascade (TC) 4 MFCs were employed with different widths, 60 mm, 30 mm, 15 mm and 7.5 mm for MFC 1–4 respectively; these MFCs were also coated with the same CPD loading. The two types of cascade (CC and TC) each of four MFCs were then assembled in triplicate (Fig. 1b and c). Air/gas-gap fluid drippers were placed between each MFC to avoid fluidic connection through the anolyte, thus allowing the monitoring of each individual MFC. Each cascade was fed with fresh urine individually through a peristaltic pump (205CA cassette, Watson-Marlow Inc, UK). The flow rate was set at 8.85 mL h $^{-1}$ per cascade, giving an HRT of 6h40 for the fixed size MFCs and 3h20, 1h40 and 50 min for the decreasing size MFCs.

MFCs operation and monitoring

Feedstock and MFC enrichment

The MFCs were inoculated with the anolyte effluent from other existing MFCs running under long-term continuous flow conditions with urine. Urine (pH between 6.4 and 6.7; conductivity between 18 and 25 mS) was collected from an anonymised pool of healthy individuals, with no known previous medical conditions, on a daily basis. Following the inoculation, the subject MFCs were fed with this fresh neat urine on the day of collection. The MFCs were allowed to reach steady state conditions after 10 days, and the polarisation experiments were performed 3 days later. All MFCs had matured under a load (y) proportional to their total anode surface area (x) based on the relationship between the load for maximum power transfer and the corresponding anode surface area. The applied equation ($y = 561379x^{-1.125}$) was derived from the power curves obtained from different designs of ceramic MFCs.

Data capture and polarisation experiments

Voltage output was monitored against time using an Agilent LXI 34972A data acquisition/switch unit (Farnell, UK). Measurements were recorded every two minutes. Recorded raw data were processed and analysed using Sigma Plot v11 software (Systat Software Inc., London, UK). Polarisation performance levels were recorded for each individual fuel cell and then averaged (n = 3). The polarisation experiments were performed, using an automated resistorstat [13], and the resistive load values ranged from 38,000 Ω to 11 Ω and comprised 33 steps. Each resistor was connected for a period of 5 min.

Results and discussion

Anode size

Increasing the total surface area of anode within a fixed volume chamber of a tubular ceramic MFC, increased the volumetric power density of the unit (Fig. 2a), but this increase is not linear (Fig. 2a). Over a certain size, the available anodic volume gets filled with anode material and MFCs start to be limited by local masstransfer limitations. Normalising the power density by the total anode surface area indicates that the smaller anode surface area has a higher power density (Fig. 2b). However, absolute power was higher for the larger surface area of electrode, when the ceramic membrane area was identical. For this reason, it is believed that the volumetric normalisation of power density is more accurate with regard to the scaling up of MFCs. In the present experiment, 1400 cm² was the largest surface area that could be physically accommodated into the 25 mL chamber without blocking the perfusate flow. Moreover, it represented the maximum amount that should be employed since it could also increase the electrostatic potential of the analyte [14]. Thus, a ratio of anode between 32 cm² cm⁻³ and 56 cm² cm⁻³ appeared to give a good trade-off between size and hydrodynamic limitations. Based on these results, an anode density of 40 cm² cm⁻³ was employed in the other MFCs.

Power-density and surface area to volume ratio

When building a stack of MFCs, the size of each unit has to be defined. The comparison between the two controls, i.e. the large diameter MFCs (2.6 cm ID; n=2) and the small diameter MFCs (1.8 cm ID; n=3), showed that the absolute power of the larger MFC is greater than the smaller one. However, in terms of volumetric power density, the smaller MFC outperformed the larger. This confirmed that a higher SA:V ratio increases the power density for a given anode surface area (800 cm²) and a given length (10 cm) (Fig. 3a). When the anode surface-area of the small diameter MFC was divided by 2 (400 cm²), for an identical length of 10 cm, the power density was again higher at 5.8 W m $^{-3}$ (n=3) compared to 4.4 W m $^{-3}$.

The finding that power density was higher when decreasing the diameter (and increasing the SA:V ratio), suggested that elongation of a fixed diameter tube (which would maintain the SA:V ratio

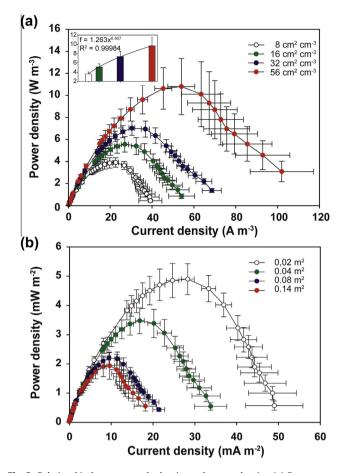


Fig. 2. Relationship between anode density and power density. (a) Power curves normalised to the volume of the anodic chamber (25 mL; tubular MFCs). The labels specify the surface of anode per mL of anodic volume. Error bars represent standard deviation between independent MFCs (n = 3). The inset shows the relationship between the maximum power density produced by identical MFCs fitted with increasing size anodes. (b) Power curves normalised to the anode surface area. Error bars represent standard deviation between independent MFCs (n = 3).

independent of length) would result in constant power density per unit of length, and therefore twice the length would give twice the power. Comparing units of various lengths and joining smaller units together tested this hypothesis. Polarisation experiments were conducted on the 5 cm MFCs when connected in parallel and in series. Such analysis allowed for a direct comparison between different setups with an identical treating volume (25 mL): one MFC of 10 cm or two MFCs of 5 cm connected in parallel or series. As can be seen in Fig. 3b, the peak power between the 10 cm and the two 5 cm configurations was identical (5 cm series 174 μ W \pm 2 μ W; 5 cm parallel 194 μ W \pm 12 μ W; 10 cm $187 \mu W \pm 22 \mu W$), and thus the polarisation confirmed that both setups were giving equivalent performance, since they totalled the same volume (Fig. 3b). If doubling the MFC length from 5 cm to 10 cm did not influence power density, then a further increase in length would also be expected to give the same power density but increase absolute power. Due to the physical limitations of the electrode conformation and the hydrodynamic pressure on the vertical ceramic tube, a 10-fold increase in length could not be achieved, therefore the length was only increased by 8 times (40 cm). Results showed that the power density of all MFCs was equivalent, independent of length under these conditions (Fig. 3b), since they all had identical SA:V ratios. The series electrical connection showed as expected, higher working voltage, but lower current. Thus, depending on the target voltage, a number

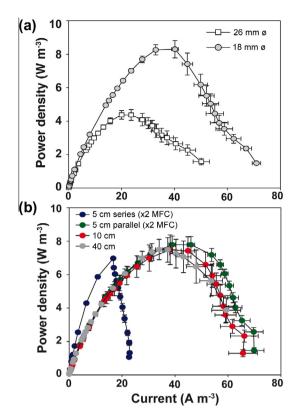


Fig. 3. Comparison of the power density of MFCs having different diameter or length. (a) Polarisation curves, normalised by the internal volume, of the large (2.6 cm \emptyset) and small (1.8 cm \emptyset) tubular MFCs of equal length (10 cm). (b) Power density comparison from the different lengths and configurations tested, at equal diameter (1.8 cm \emptyset). Error bars represent standard deviation between independent MFCs (n = 3).

of elongated MFCs can be employed in parallel, for increasing current, and then sets of these elongated MFCs (or cascades of MFC) can be joined in series, in order to increase the voltage, with little negative effects on power density.

Relationship between hydraulic retention time, number of urine recirculations and power

The above-mentioned results showed that any length of MFC, within the tested range, would end-up having an equivalent power density. However, this hypothesis only holds true if the fuel quality is uniform for all stages in the cascade. In a configuration whereby the system is under a continuous flow, the output part of a cascade will receive fuel that is partly depleted of carbon-energy (C-E) source. An experiment was designed to investigate a means of maintaining a level of power output efficiency despite decreases in the C-E fuel quality along a cascade of MFCs.

The experiment was to increase the flow rate (by media pump rpm) as a means of decreasing the hydraulic retention time (HRT), in order to maintain a constant power density along the cascade. As shown in Fig. 4a and b, urine that has already been treated once produces less power than fresh urine at equivalent flow rates. However, the increased flow rate (decreased HRT) favourably enhanced power production. Fig. 4c shows that increasing the flow rate of the urine increased the power density of the MFC to the same level as with fresh urine. The third treatment of urine at a further doubling of the flow rate also resulted in equivalent power density compared with fresh urine (Fig. 4d). In contrast, when the flow rate was kept identical for ageing and depleted urine (Fig. 4e–g) there was much less response by the 6th re-circulation run.

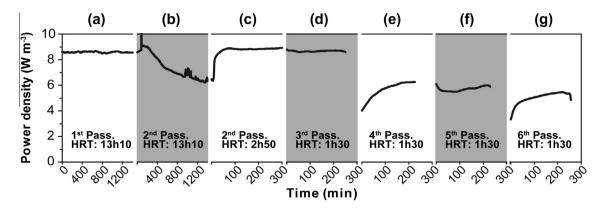


Fig. 4. Power density of MFCs fed with treated fuel (urine) at different flow rates. Number of re-circulation passages indicates how often the fuel has been treated by an MFC. The hydraulic retention time (HRT) reflects the applied increasing flow rate. The indicated time is relative to each new condition (a–g). The presented data are the average of independent MFCs. Standard deviation has been applied but the error bars are smaller than the line thickness (n = 3).

Because the available quantity of urine was limited, the flow rate was not increased after the 3rd passage, since the MFCs would not have the time to reach steady-state before running-out of media. These results demonstrate that, within a cascade that treats the same fuel, the power density may be maintained by increasing the flow rate of a depleted fuel at each stage of the cascade. Therefore, to maximise the power output, whilst avoiding the recirculation of urine multiple times at an increasing flow rate, the size of different treating stages (MFCs) in a cascade could be decreased in order to reduce locally the HRT. In other words, having a cascade where each stage matches the quality of the fuel.

Implementation of findings: assembling a cascade of MFCs to treat urine

Results on the relationship between the anode size and the power density indicate that the trade-off between power density and hydrodynamic limits is obtained with an anode density ranging between 32 and 56 cm² cm⁻³ of anolyte (Fig. 2). On the one hand, constant or increasing SA:V ratio results in constant or increasing power density (Fig. 3), and increasing the flow rate allows each unit to maintain constant maximum power density, despite the C-E source depletion (Fig. 4). One method to achieve an increase of flow rate supply in a cascade with constant flow is to decrease the size of successive downstream MFCs. The smaller volume, gives rise to a faster flow rate/dilution rate. However, when employing cylindrical MFCs, any change in diameter also directly affects the SA:V ratio (Fig. 3a), conflating the analysis of the effects of both flow rate and SA:V ratio. For this reason, the experiment employed flat plate MFCs whereby a constant SA:V ratio was maintained for constant length/constant depth by only changing the width of MFC units, and investigated whether doing so would improve the power density of the cascade stack. The first control cascade, (CC) consisted of four units of constant size, all using an anode packing density of 40 cm² cm⁻³ (Fig. 1b). The second test cascade (TC) consisted of four units of decreasing width, resulting in successively halving the volume, but still retaining the same SA:V, giving a two-fold increase in the cross section velocity (Fig. 1c). The first MFCs of each cascade were identical in terms of size, volume and supply rate of nutrients (fresh urine). and the power output was observed to be similar (Fig. 5a). Both cascades were under the same flow rate.

In terms of absolute power, both cascade-types were producing similar levels (Fig. 5a) but the TC had half the volume of the CC, and hence the power density as a stack was higher (Fig. 5b). As shown, this increase was linked to the decreasing HRT (Fig. 5c) due to the decreasing size of units in cascade. However, as the TC occupied a

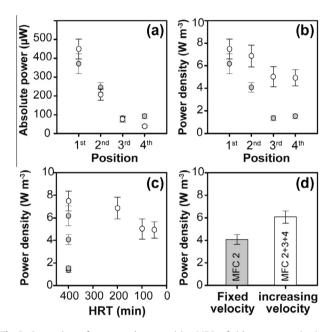


Fig. 5. Comparison of two cascades comprising MFCs of either constant size (grey circles) or decreasing size (white circles). Absolute power (a) and power density (b) of each MFC depending on its position within the cascade. (c) Power density of each MFC depending on the hydraulic retention time. (d) Power density produced after the 1st MFC for an equal volume of anolyte: either 1 MFC of 60 mL (2nd position), or 3 MFCs of decreasing HRT (51.5 mL; 2nd, 3rd and 4th). Error bars represent standard deviation (3068 time points in triplicate).

smaller volume, the comparison between the two types of cascades should be performed at equal treated volume: in CC the urine received by the third MFC of the cascade has already been treated by 120 cm³ of electroactive biofilm that corresponded to the totality of the treatment surface of the TC (112.5 cm³). Since the first MFCs were the same in both types of cascade, the comparison was focussed on the 2nd MFC of the CC and the combined 2nd, 3rd, and 4th MFCs of the TC, corresponding to 60 mL and 52.5 mL, respectively. The absolute power produced by all the MFCs of the TC was higher than the absolute power produced by the second MFCs of the CC (319 \pm 29 μ W and 244 \pm 26 μ W respectively). These results confirmed that a cascade comprising MFCs of decreasing size optimises the harvested power for a given volume of fuel (here urine; Fig. 5d), keeping in mind that the volumetric flow rate for both cascades, as a whole, was the same.

After the individual investigation of each MFC, the units within the cascades were connected electrically, to produce a single stack:

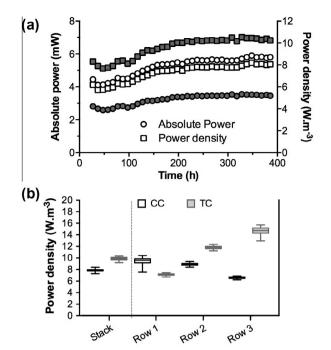


Fig. 6. Power density comparison between the two stacks. (a) Long time power monitoring of the CC stack (units of identical size; white symbols) and the TC (units of decreasing size; grey symbols). Both absolute power and power densities are plotted. (b). Boxplot illustrating the variation and mean of the power densities of each row of the cascade (n = 12,000, the last 200 h of (a)). White boxes are for the CC stack and grey boxes are for TC stack.

one stack of MFCs with identical sizes (CC, total volume of 720 mL), and one stack of MFCs with decreasing sizes (TC, total volume of 337.5 mL). Each stack comprised 3 independent groups of four cascading MFCs. For each stack, all MFCs having the same position in their respective cascade (row of 3 MFCs) were electrically connected in parallel. At first, the four rows, within each stack, were connected in series (TC: 570 Ω ; CC: 335 Ω). Results showed that the series connection was not stable for either type of stack (data not shown). This was particularly true for the TC stack cascading decreasing units: the last cells of the cascade (forth row) reversed after less than an hour. The CC stack, cascading MFCs of constant size, was stable for a longer period, but the last MFCs of the cascade did reverse after 12 h:30. Hence, all MFCs of the last 2 rows in each stack were connected in parallel and then connected in series with the first two rows, whose resistance was set accordingly (TC: 335 Ω ; CC: 151 Ω). As illustrated by the long-term monitoring, both cascades had a stable power output and no cell reversal was observed (Fig. 6a).

The absolute power of the TC stack was lower than the CC stack, $\sim\!3.50\pm0.05$ mW and $\sim\!5.75\pm0.08$ mW, respectively (Fig. 6a). The power density of the TC stack was higher than the CC stack, $\sim\!10.00$ W m $^{-3}$ and $\sim\!8.00$ W m $^{-3}$ respectively (Fig. 6b). This 25% increase of power density is a significant improvement given the number of sub-units implied in the implementation of MFC technology for applications. As illustrated by the power density of each row (position in the cascade), the power density increase of the TC stack is due to the power density increase of each row: from 8 to 15 W m $^{-3}$ (Fig. 6b). On the contrary, the power density of each row in the CC stack, decreased with its position in the cascade (Fig. 6b). It is known that smaller MFCs have a greater power density than larger vessels because of their higher SA:V ratios [10,15]. In this case however, all MFCs of both stacks had the same SA:V ratio. Hence, the presented results demonstrate that having a cas-

cade of decreasing size MFCs enhances the stack power density because of the local decrease in HRT.

Conclusions

This study focused on the different aspects that affect the power density of a cascade fuelled by urine: anode SA (density of packing), SA:V ratio, MFC shape and size, hydraulic retention time, and urine age. The independent investigations of these aspects led to the design of a cascade that maintains equilibrium between these parameters and resulted in an optimum anode density of 40 cm² cm⁻³ for flat ceramic membranes. A constant SA:V ratio was maintained whilst decreasing the size of the subsequent MFCs, which decreased the local HRT. This approach gave rise to the TC stack, which occupied a smaller volume than the control stack and produced higher power density in terms of volumetric footprint. This is particularly important if scaling up using a plurality of cascades is envisaged. To the authors' knowledge, this is the first study that brings together the different aspects influencing power density and empirically implementing these findings to produce and test a new cascade design consisting of MFC units of decreasing size.

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