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Impact of disinfectant on the electrical outputs of urine-fed ceramic and membrane-less microbial fuel cell cascades

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ABSTRACT

The application of microbial fuel cells in sanitation has demonstrated feasibility in supplying electricity and providing safety in underserved communities, especially at toilet blocks. Two different designs of urine fed MFC cascades, ceramic MFCs (***c***-MFC) and self-stratifying MFCs (***s***-MFC), have been employed in large-scale feasibility studies. As part of a pre-commercialisation approach, this study verified the resilience of each design when a commercial disinfectant was introduced into the system. Five different conditions, varying in concentrations (24.2 mM–604.5 mM) and the total volume (50–500 mL) of sodium hypochlorite disinfectant introduced, were tested. Upon adding the disinfectant, both types of MFC-cascades exhibited rapid power drops with response times lower than 5 min in all tested conditions, followed by relatively swift recovery times of up to 250 min. The volume of disinfectant introduced had a greater impact on power output than its concentration or dose. Comparing the two designs, the ***c***-MFC demonstrated a much larger voltage drop, up to 0 mV, and shorter recovery time compared to the ***s***-MFC under most test conditions, mainly attributed to the presence (***c***-MFC) or absence (***s***-MFC) of a membrane. Overall, both types of MFCs exhibited strong resilience to sodium hypochlorite additions, thereby highlighting the commercial potential of the technology towards safe off-grid sanitation.

1. Introduction

Urban off-grid sanitation based on ecological circularity, is emerging as a complementary alternative to the conventional centralised water-flush toilets [1]. The progress addressing Sustainable Development Goals for global access to safe sanitation and advance in the protection of public health to avoid pathogen exposure at the source of production is one of the key elements to explore. On-site sanitation systems need to be simple to operate and maintain, reliable, and resilient to unreliable electrical service [2].

The capacity of the microbial fuel cell (MFC) to generate energy whilst treating wastewater has led to a lot of progress in the development and implementation of the technology during the past decade [3]. Using the criteria from Bird et al. [3], 30 out of the 11,414 studies on the matter have reached pilot-scale stage. Alongside the treatment of municipal [4–7] or farming [8] wastewaters, the technology has been implemented as a sanitation solution for decentralised areas. The latter has achieved the deployment of pilot-scale systems converting the organic content of urine into electricity to power lights and bring safety in dark areas that are not connected to the grid [9–12].

Amongst the tested urine fed MFC designs, only two reached the pilot stage. The first design is known as ceramic MFCs (***c***-MFC) and exploits ceramic as both the membrane and a structural feature [13]. This design, in addition to a low production cost, benefits from allowing the disposition of a plurality of internal cathodic compartments surrounded by multiple anodes submerged in the same module. These are the two key aspects in the scaling up of the technology [9]. The second design is known as self-stratifying MFCs (***s***-MFC). The s-MFC type eliminates the use of physical membrane components to enhance cost-effectiveness and design simplicity, instead, it exploits the capacity of microorganisms to influence/structure biogeochemical gradients in water column, serving as a bio-exchange membrane between the anodes and cathodes [14]. This design allows for a high surface area of electrode pairs (i.e., cathodes and anodes) per volume of reactor. Similar to ***c***-MFCs, ***s***-MFC also permit the scaling up of the technology at low cost precisely because it is a membrane-less design. The scalability of both designs allowed for field

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trials to be carried out with the MFC being the core of autonomous sanitation systems. These systems comprised stacks of either ***c***-MFC or ***s***-MFCs employed to treat urine whilst generating electricity to power purposed built LED lighting [9–12].

Although such a system drastically reduces water consumption, hygiene requires running water for cleanliness. Information gathered during the trials indicates that cleaning personnel often use the same solution for cleaning both floors and troughs, thus, introducing disinfectant into the systems. The aim of the present study is to evaluate the impact of disinfectant on the power output of cascading microbial fuel cells, under controlled conditions. The MFC designs used here (i.e. ***c***-MFC and ***s***-MFC) are similar as to the ones previously deployed in the field. Our objective was to provide evidence of whether disinfectants could still be used, maintaining hygiene and study its impact on the stack performance in the field. To our knowledge, this study marks the initial exploration of the impact of disinfectant on urine-fed MFC designs implemented at a pilot-scale. The findings from this study will provide valuable insights for the next stage of technology development and commercialisation. Increasing concentrations of the most common disinfectant used, sodium hypochlorite (a.k.a. household "bleach"), were added directly in the top MFC-module of cascades comprising three modules. The power output of the cascades was used as the parameter to evaluate the impact disinfectant would have on a MFC-based sanitation system.

2. Experimental

2.1. Microbial fuel cells

Two types of MFC designs were investigated, ceramic membrane dual compartment MFC (***c***-MFC) and self-stratifying membrane-less MFC (***s***-MFC). The ***c***-MFC and ***s***-MFC employed here were of the same as the ones used for a previous study on the impact of feedstock dilution on the power and treatment performance [15]. Three modules of each design were assembled in a cascade configuration, whereby the effluent of one module directly feed into the following one (Fig. 1).

The reader is referred to Ref. [15] for more detail on the specificity of each design. Overall, each ***c***-MFC module comprised of 8 individual MFC with 47 mm tall ceramic cylinders (Laufen, Austria) acting as membrane (Fig. 1). Each ***s***-MFC module comprised of a set of 28 cathodes positioned 5 mm above a set of 28 anodes enclosed within the same cylindrical vessel. Both sets were built on the same design principle: 28 anodes and 28 cathodes (height: 2 cm) were disposed on a 155 cm long 316 stainless-steel mesh acting as the current collector. The mesh was folded into a concertina shape to fit into the vessel (Fig. 1). Both designs had the same total anode surface area of 10,080 cm^2 (10 g m^{-2} carbon veil), which were loaded with 1.25 ± 0.1 mg cm^{-2} (***c***-MFC) and 1.19 ± 0.2 mg cm^{-2} (***s***-MFC) of activated carbon (AC): polytetrafluoroethylene (PTFE) mixture (95:5; w/w). The ***c***-MFC cathodes had a surface area of 248 cm^2 and the ***s***-MFCs had a surface area of 263 cm^2. The ***c***-MFC cascade had a total displacement volume of 1.305 L and the ***s***-MFC cascade had a total displacement volume of 1.575 L. All the modules within each cascade were electrically connected in parallel. Both cascades had been in operation for more than a year at the time of the experiment [15].

2.2. Operating conditions

Both cascades were fed continuously with neat human urine using a peristaltic pump (39.8 ml h^{-1}). The fed urine was collected daily from a tank pooling together the urine donated by anonymous individuals. Urine had undergone partial hydrolysis within the collecting tank, with the resulting pH of the hydrolysed urine feeding the cascade ranging between 8.2 and 8.9. Following results from the previous study [15], the hydraulic retention time (HRT) was set to 12h per module. Due to the slight difference in displacement volume, the ***c***-MFC cascade had a HRT of 32.7h and the ***s***-MFC cascade had a HRT of 39.6h. Five conditions were tested: (**a**) the addition of 50 ml of disinfectant (sodium hypochlorite, commercial "bleach") following the commercial disinfectant manufacturer's recommended final concentration for a "disinfection solution" aimed at floor cleaning (1.8 g l^{-1} NaClO; 24.18 mM); (**b**) the addition of 200 ml of 24.18 mM NaClO; (**c**) the addition of 500 ml of 24.18 mM NaClO; (**d**) the addition of 500 ml of 48.36 mM NaClO; and (**e**) the addition of 50 ml of undiluted disinfectant (45 g l^{-1} NaClO; 604.51 mM). These tested concentrations were chosen to simulate a common scenario where someone might use bleach to clean a toilet, introducing NaClO to the MFC system receiving the toilet discharge. Due to the difference in volume between the two designs, this corresponded to a final concentration within the first module of (**a**) 2.78 mM (0.21 g l^{-1}) and 2.30 mM (0.17 g l^{-1}), (**b**) 11.12 mM (0.83 g l^{-1}) and 9.21 mM (0.69 g l^{-1}), (**c**) 24.18 mM (1.8 g l^{-1}) and 23.03 mM (1.71 g l^{-1}), (**d**) 48.36 mM (3.6 g l^{-1}) and 46.06 mM (3.43 g l^{-1}), and (**e**) 69.48 mM (5.17 g l^{-1}) and 57.57 mM (4.29 g l^{-1}) for the ***c***-MFC and the ***s***-MFC, respectively (Table 1). The above concentrations were calculated using the disinfectant's stated concentration of 4.5 g per 100 mL and the molar mass of 74.44 g mol^{-1}. It was decided to have increasing concentrations to avoid having to re-inoculate the system if a tested concentration was to permanently damage a significant number of electroactive microorganisms.

2.3. Data capture

A purpose-built circuitry was maintaining constantly the cascade under a potentiostatic condition of 400 mV. This electronic board (details in Ref. [16]) converted the measured current into a voltage that was recorded by an Agilent Data Acquisition System (Agilent LXI 34972A; Farnell, UK). Measurements were recorded every 5 min. The current I in Amperes (A) was calculated using conversion formula [16], I = (Vm)/19.8, where Vm is the measured voltage in Volts, and 19.8 the conversion factor measured for the electronic board. This conversion

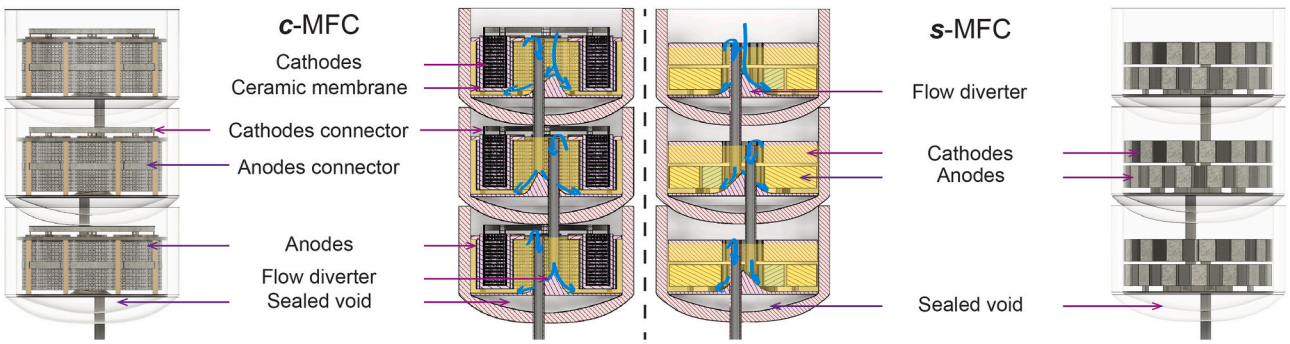

Fig. 1. Cross section view of the ceramic-based (***c***-MFC; left) and the membrane-less MFC (***s***-MFC; right) cascades used in this study.

Table 1
Concentrations of the added disinfectant (NaClO) for each condition and cascade.

Condition	Added NaClO solution			Dose (g)	Final conc. *c*-MFC		Final conc. *s*-MFC	
	Vol. (ml)	Conc. (g/l)	Conc. (mM)		g/l	mM	g/l	mM
a	50	1.8	24.2	0.09	0.21	2.78	0.17	2.30
b	200	1.8	24.2	0.36	0.83	11.12	0.69	9.21
c	500	1.8	24.2	0.90	1.80	24.18	1.71	23.03
d	500	3.6	48.4	1.80	3.60	48.36	3.43	46.06
e	50	45	604.5	2.25	5.17	69.48	4.29	57.57

factor is a unique number for the bespoke electronic board made in house. The power output P in Watts (W) was calculated as $P = I \times V$, where V is the constant voltage (400 mV) in Volts (V) and I the calculated current in Amperes (A).

3. Results and discussion

Both the ***c***-MFC and the ***s***-MFC were fed urine for a week prior testing the first experimental condition. The disinfectant was prepared with tap water as the eluent and then fed at once to the cascade: the first conditions consisted of a 50 ml addition into the 1st module of each cascade whilst urine was pumped at 39.8 ml h^{-1}; and the same was done for each condition (Table 1). A sequential dilution was occurring as the content of the first module was traveling to the two downstream ones within each cascade. Hence, the first module in each cascade was facing the highest concentration of disinfectant. Overall, each added volume of disinfectant-urine mix exited the cascades after 32.7h and 39.6h for the ***c***-MFC the ***s***-MFC designs, respectively.

Results indicate that the power output of the ***c***-MFC reach pre-addition power level within 50 min, for concentrations roughly between 3 and 11 mM NaClO (Table 1, Fig. 2a and b). With a 24 mM final concentration in the first module of the ***c***-MFC, the power dropped to 0 mW in less than 5 min (sampling rate; Fig. 2c). As the added disinfectant was prepared using tap water, the measured sudden power drop could be due to either the whole volume being replaced with a solution lacking organic matter (feedstock), or to the NaClO concentration reaching a threshold affecting the electroactive microbial communities. Although the ***c***-MFC cascade reached steady state 160 min after additions, this new steady state was roughly 20 % lower than the initial one. Moreover, this new steady state being stable over a period longer than the HRT of the cascade (32.7h) would suggest that the NaClO concentration has more impact on the system than the volume added to the first module. A similar behaviour was observed when adding 500 ml of a solution having twice the concentration (final concentration of 48.36 mM; Fig. 2d). The power output dropped to 0 mW within less than 5 min and steady state was recovered after roughly 250 min. These results suggest that the drop to 0 mW is due to the volume added and the recovery time is influenced by the concentration of NaClO. Following these findings, the addition of 50 ml of concentrated solution (69.48 mM final concentration) was tested. The hypothesis was that the recovery time would be longer than the previous test condition due to a higher concentration, whilst the power drop would be less pronounced. Results show that this was not the case. Although the power drop occurred in approximatively 15 min (40 %), the recovery of power output occurred in 105 min (Table 2). These results show that the volume of disinfectant introduced in a cascade has more impact on the power output than its concentration, under the tested conditions. Moreover, the results show that the cascades were able to rapidly recover from the addition of NaClO concentration that was three time higher than the concentration recommended by the manufacturer to use.

Table 2
Latency and recovery time for both cascades.

Condition	*c*-MFC Time (min.)			*s*-MFC Time (min.)		
	Latency	Recovery	Total	Latency	Recovery	Total
a	10	10	20	10	5/30[a]	15/40[a]
b	5	45	50	40	60/225[a]	100/265[a]
c	10	235	245	25	155	180
d	25	205	230	35	35/235[a]	70/270[a]
e	10	95	105	40	65	105

[a] Time to reach 100 %/time to reach steady state.

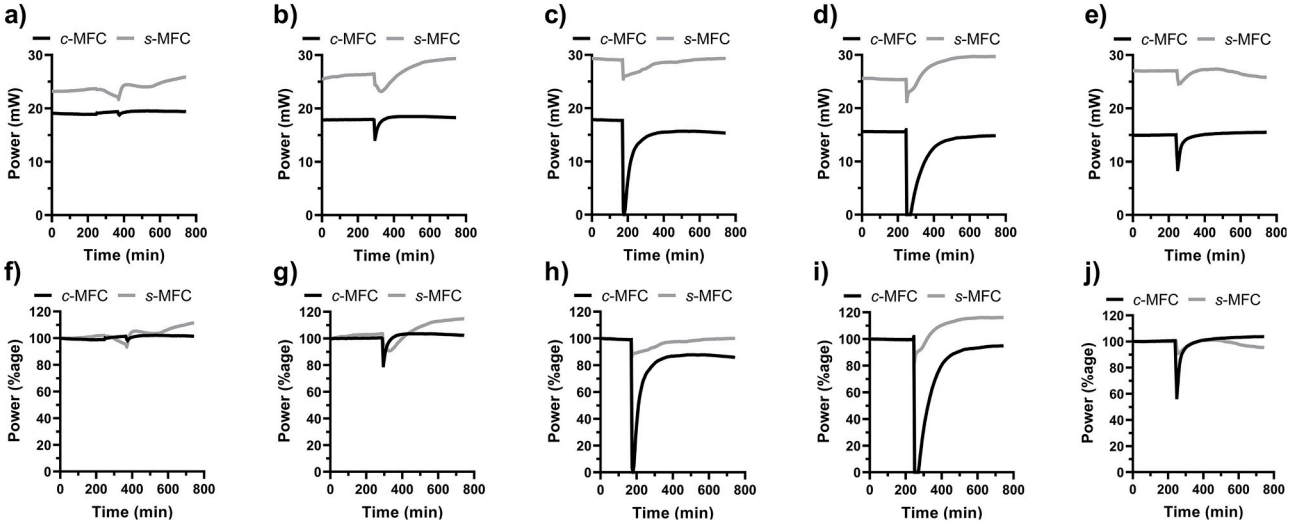

Fig. 2. Absolute power outputs under the tested conditions (**a,f**), and percentage of the output based on the average power produced during the 60 min prior additions. Response of the cascades to the additions of: (**a,f**) 50 ml of 24 mM NaClO, (**b,g**) 200 ml of 24 mM NaClO, (**c,h**) 500 ml of 24 mM NaClO, (**d,i**) 500 ml of 48 mM NaClO, and (**e,j**) 50 ml of 605 mM NaClO.

Normalising the power outputs into percentage of the initial condition enables a rough comparison between the two cascades' responses to NaClO additions. Interestingly, if both cascades have a similar power response to NaClO additions, a sharp drop, the recovery behaviour differs between the ***c***-MFC and ***s***-MFC cascades. The recovery curve of the ***s***-MFC cascade have a more pronounced latency following the power drop and then progressive increase toward a new steady state. The latency time of the of the ***s***-MFC was nearly three time higher compared to the ***c***-MFC, with an average of 35 ± 5 and 12 ± 5 min, respectively. Under the test condition **b** and **d** (Tables 1 and 2) two recovery times could be considered for the ***s***-MFC cascade: first the time to reach initial power output, and secondly the time to reach steady state, which in these test conditions were above the initial value (Fig. 2g–i). Indeed, the ***s***-MFC cascade displayed a tendency to establish a steady state higher than its initial one (Fig. 2f,g,i), whereas the ***c***-MFC cascade showed a tendency to have either the same steady state (Fig. 2g–j) or a lower steady state after additions (Fig. 2h and i). The ***c***-MFC's overall recovery time, from steady state to steady state, was shorter than the ***s***-MFC cascade for nearly all test conditions (except condition **c** and **e**). However, when considering the time needed to reach initial value, the ***s***-MFC cascade was displaying speed either similar (condition **a**, and **e**) or slightly faster (condition **c** and **d**; Table 2). The main difference resides in the power drop and the slope of the power recovery: the ***c***-MFC had much higher voltage drop and steeper slopes, during recovery, than ***s***-MFC's (Fig. 2).

As shown during a previous study [15], the electrical outputs of ***c***-MFC cascades are more stable over time than current output of the ***s***-MFC cascade, which fluctuate more, without considering the impact of the test conditions (Fig. 2). Regarding the steady state power output of the cascade, results indicates that ***c***-MFC cascade's output decreased by roughly 21 % between the beginning and end of the experimental runs. At the beginning, under the two first incubating conditions (Fig. 2f and g), the output was of 19 ± 0.5 mW. However, after the addition of 500 ml NaClO (24 mM), the output felt at 15 ± 1.2 mW (Fig. 2c) which became the new steady state level for the ***c***-MFC cascade (Fig. 2d and e). Conversely, the steady state power output from the ***s***-MFC cascade increase from 23 ± 1.0 mW (Fig. 2a) to 29 ± 1.0 mW (Fig. 2d) and then decrease to 26 ± 1.0 mW (Fig. 2e). Since the ***s***-MFC design is membrane-less and the electrolyte is successively in contact with the anodes and the cathodes, the addition of NaClO should affects the electroactive communities of both the anodes and the cathodes, in addition to the commensal microbial communities. The 26 % power increase of the ***s***-MFC cascade implies that the disinfectant had more impact on microbial communities that were otherwise limiting the power generation. Hence, results suggest that the power generation in ***s***-MFC is limited by the activity of commensal microbial communities. Conversely, as the ***c***-MFCs have membranes, the addition of NaClO only affect the anodic communities. Hence, the power output decrease of the ***c***-MFC cascade indicate that the either the anodic commensal microbial communities in ***c***-MFC participate to power generation, or part of the anodic electroactive communities is sensitive to NaClO. As in ***c***-MFC the anodic electroactive communities form thick biofilm on the anodes, results could also indicate that only the communities on the external part of the biofilm were affected by the NaClO additions. Our study is unique as previously, the hypochlorite was used as catholyte at 3 g/L of available chlorine dose where the anodic effluent was treated and disinfected [17] which emphasises the need for further exploration. As the pathogen inactivation is inherent part of safe sanitation systems there is a growing number of studies exploring this area. An ultraviolet C light-emitting diode (UV-C LED) water disinfection system activated by microbial fuel cells (MFCs) was recently tested which focused on pathogen inactivation [18]. Such complementary approaches explore the MFC systems use in different ways to improve off grid self-sustainable sanitation facilities. Providing hygienic conditions of the sanitary units as presented in our study is focusing on real-world application for protecting human health and opens new dimensions towards its field applications. As the main concerns related to off-grid sanitation were identified as safety, dignity, functionality, smell, accessibility, size of the system, environmental impacts, and reliance on water and electricity [1], this opens up an important route towards safe off grid sanitation systems.

Based on these results, in the future applications of self-sustainable toilets systems, the pathogen control of the sanitary units could be easily maintained by using the commercial disinfectants. This poses a further investigation path towards MFC - synthesised catholyte that previously shown disinfecting properties and its potential use as disinfecting agent for sanitation [19,20]. Further investigation of MFC-produced disinfectant and its direct reuse in a circular and sustainable manner on the MFC systems is an important element in self-sustainable approach towards improving the sanitation facilities in a field-scale, off grid applications of this technology. The systems would be not only applicable for energy recovery from human waste but contributing to reuse and recovery improving current sanitation systems [21].

4. Conclusions

The development of MFC technology for various applications has made great progress. Implementation and commercialisation through scale-up as an engineering solution should not be overlooked since it should be the ultimate goal of all technologies. Recently, large-scale (>100L) MFCs systems that generate electricity whilst treating urine have proven their merit within the sanitation context. Our previous pilot tests [9,11,12] have successfully demonstrated that MFC technology can provide additional safety in addition to off grid energy sources and waste treatment. In the course of technology development, rigorous testing in real-world operating conditions with real-scale systems is essential. In this study, sodium hypochlorite, the main component of most common commercial disinfectants typically used to clean toilets, was intentionally introduced into the MFC cascades. By changing the concentration and the added volume of the disinfectant, we aimed to test the resilience of two different MFC systems that have been used in large-scale pilot trials. The results showed that although there was a difference in how the disinfecting agent affects the two MFC prototypes, both systems responded instantly to the introduction of disinfectant, resulting in an instantaneous decrease in power output. However, the power output of the two systems quickly recovered to a new steady state, which was not far away or slightly lower or higher than the previous level. This is due to the resilience of anodic biofilms formed on the surfaces of MFC-anode electrodes and it is in agreement with literature reporting resistance of biofilms to changes in the external environment compared to planktonic communities [22–24]. This result is important for the commercialisation of MFC technology, because it demonstrates that the system is sufficiently robust for usual domestic implementations, further supporting a previous study showing the use of the MFC technology for household wastewater [25]. These systems still need to undergo further investigation to establish user and operator guidelines. Notably, this study examined the MFC system's response to a single exposure to a commercial disinfectant containing sodium hypochlorite, which represents a relatively short-term exposure (HRT of c-MFC cascade and s-MFC cascade: 32.7h and 39.6h, respectively). Future studies may include long-term exposure to harsh chemicals, system reset and the influence of the external temperature changes.

Credit authorship contribution statement

Jiseon You: Investigation, Analysis and Interpretation, Writing – original draft, Writing – review & editing. Xavier Alexis Walter: Supervision, Conceptualization, Methodology, Data curation, Analysis and Interpretation, Writing – original draft, Writing – review & editing. Iwona Gajda: Methodology, Analysis and Interpretation, Writing – review & editing. John Greenman: Writing – review & editing. Ioannis Ieropoulos: Supervision, Conceptualization, Funding acquisition,

Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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