

# Performance of air-cathode stacked microbial fuel cells systems for wastewater treatment and electricity production

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## Abstract

Two different air-cathode stacked microbial fuel cells (MFC) configurations under continuous flow were evaluated during municipal wastewater treatment and electricity production at a hydraulic retention time (HRT) of 3, 1, and 0.5 d. Stacked MFC 1 was formed by 20 individual air-cathode MFC units (un-shared reactor). The second stacked MFC (stacked MFC 2) consisted of 40 air-cathode MFC units placed in a shared reactor. Continuous voltage between 50 mV and 680 mV were obtained at an open circuit voltages (OCV) in each individual MFC units. The voltages in series connection, during open circuit mode from stacked MFC 1 was ranging of 420 mV to 700 mV, and for stacked MFC 2, the voltages in series connection are kept around of 410 mV. The maximum voltages produced at closed circuit (1,000  $\Omega$ ) were of 170 mV for stacked MFC 1 and 94 mV for stacked MFC 2. Different power densities in each individual MFC units were obtained due to potential drop phenomenon and the changed of COD concentrations into the un-share and share reactors. The maximum power densities in stacked MFC 1 and MFC 2 in series connection were  $79 \pm 0.65$  mW/m<sup>2</sup> and  $4.2 \pm 0.6$  mW/m<sup>2</sup>, respectively. The voltages by connecting MFCs in series not were increased due to voltage reversal observed in two stacked MFC configurations. The maximum power density of the individual MFC units was up to 1,107 mW/m<sup>2</sup> for stacked MFC 1, and up to 472 mW/m<sup>2</sup> from stacked MFC 2, which were higher than of power densities of the series stacked MFC systems. Power and chemical oxygen demand (COD) removal efficiencies from two stacked MFC configurations were reduced when the HRT was decreased. High removal efficiencies of 84% of COD, 47% of TN, and 30% of TP were obtained in stacked MFC 2 during municipal wastewater at a HRT of 3 d.

## Keywords

Stacked microbial fuel cells; series connection; electricity; wastewater treatment; hydraulic retention time

## INTRODUCTION

Nowadays, the wastewaters represent a source of recyclable water despite the fact that contains many contaminants that may to affect the public health and the environment. The wastewaters contain large amounts of renewable energy in the form of chemical bonds. Domestic wastewater could potentially generate energy up to 2.2 kW/h.m<sup>3</sup> considering that the energy of glucose molecule containing 4.4 kW/h.kg COD (chemical oxygen demand (COD) of 500 mg/L). If this energy is recovered correctly, it could extract electricity from wastewaters (Virdis et al. 2011). Currently, it has been that the microbial fuel cells (MFC) can be utilized as a decentralized wastewater treatment with the advantage to produce bio-energy (electricity, methane and hydrogen) from wastewaters (Logan, 2012). Several

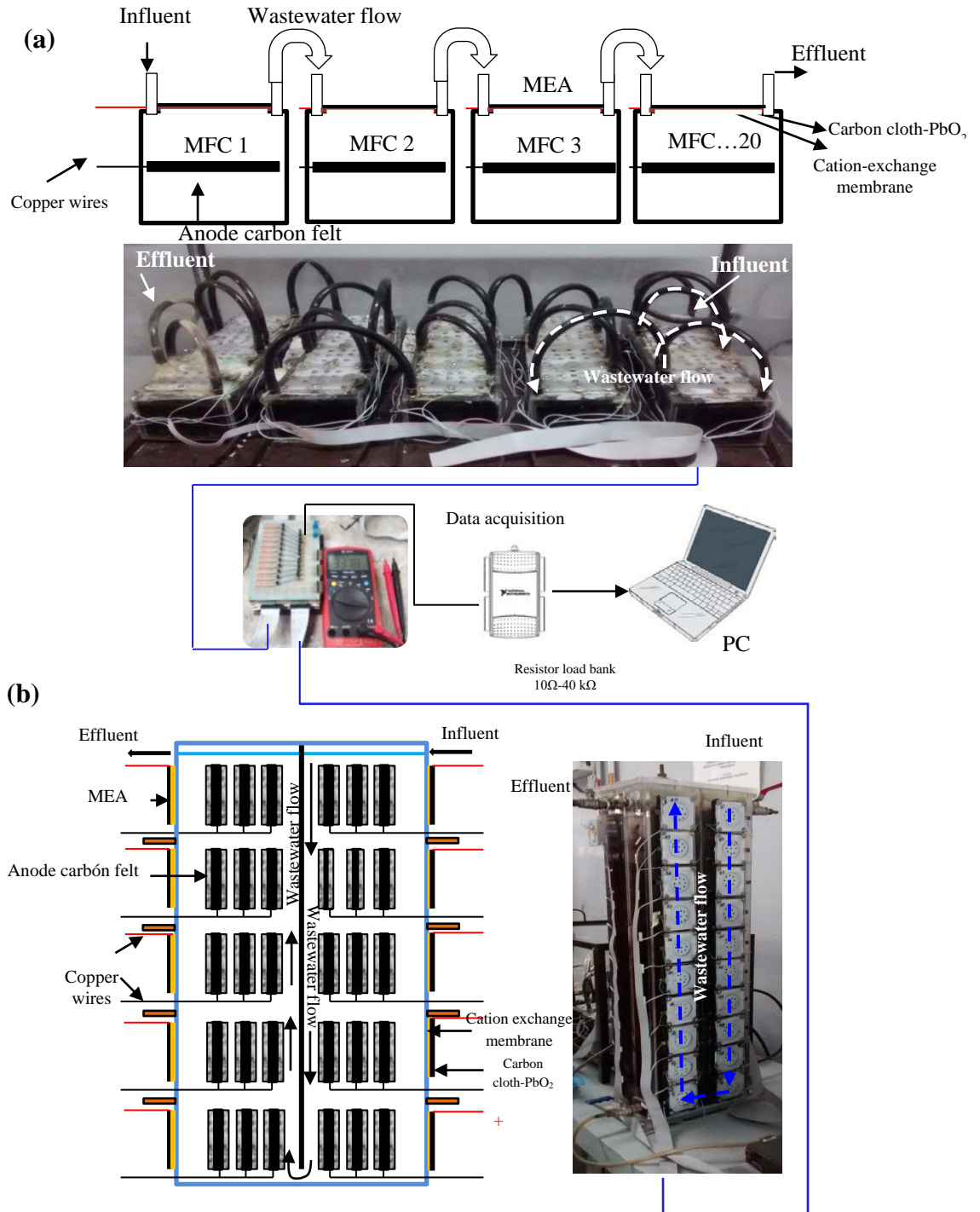
studies have been carried out in single MFC units (smaller-scale) for produced electricity, however, are few examples of MFCs with multiple anodes and cathodes. The scaling-up of MFCs requires process intensification for to reduce the size of the reactor, to generate high power, COD removal, and to reduce the ohmic losses to minimize the voltage reversal into the reactors (Kim et al., 2012; Oh and Logan, 2007). Multi-electrodes MFCs are the best option so far to troubleshoot some these problems (Ahn and Logan, 2012, Jiang et al., 2010). The theoretical voltage at an open circuit mode (OCV) from an individual MFC is ~1.25 V using glucose as electron donor and oxygen as the electron acceptor according to Nerst equation. However, in the practice, the voltages produced in single-chamber MFC are ~0.2 to 0.5 V (Kim et al., 2012; Oh and Logan, 2007). For increasing the voltages from bio-electrochemical fuel cells systems, MFCs could be connected in series units. The study of new architecture for modulation of chamber stacked MFCs are necessary. Xinmin et al. (2016), An et al. (2015, 2015), Yazdi et al. (2015), An et al. (2014), Ieropoulos et al. (2013), Kim et al. (2013), Rahimnejad et al. (2012), Zhuang et al. (2012), Kim et al. (2012), Jiang et al. (2010), Gálvez et al. (2009), Zhuang and Zhou (2009), Ieropoulos et al. (2008), Shimoyama et al. (2008), Aelterman et al. (2006), and Shin et al. (2006) showed a successful use of series stacked MFCs with voltages ranging from 2 V to 23 V at an OCV. However, these studies are limited to 2-6 individual MFC unit connected in series, and few stacked MFCs were operated in continuous flow. The effect of hydraulic retention time (HRT) in a stacked MFC over voltage profiles and organic matter and nutrients removal efficiencies are not studied in stacked MFCs. Therefore, the optimal selection of HRT in a stacked MFC is an important factor to design a larger-scale stacked MFC to removed contaminants and generating electricity production simultaneously. The present study has been carried out to evaluate the performance of two air-cathode stacked MFCs operating in continuous flows during wastewater treatment and electricity production applying three HRT.

## **MATERIALS AND METHODS**

### **Construction, star-up, and operation of air-cathode stacked MFCs systems**

We designed two air-cathode stacked MFC systems of volume ~16 L. The stacked MFC 1 module was prepared by 20 individual MFC units (un-shared reactor) (Figure 1a). Each individual MFC unit had rectangular poly-acrylic plastic shape of volume ~800 mL (11.5 length cm x 11.5 cm width x 6 cm height). An anode electrode (carbon felt (Brunssen Inc. Mexico)) of 6 cm length and 6 cm width (superficial area of 0.0036 m<sup>2</sup>) was inserted into the rectangular anode compartment. Membrane electrode assemblies (MEA) of 6 cm length and 6 cm height were placed between anode and cathode and were exposed to air. MEA consisting of cation-exchange membrane (CMI-7000, Membrane International Inc. USA) and carbon cloth (Brunssen Inc. Mexico) that contained a PbO<sub>2</sub> catalyst layer (0.5 mg PbO<sub>2</sub>/cm<sup>2</sup>). For electron collector, copper wires were utilized. The distance between anode and cathode was of 6 cm. Stacked MFC 1 was fed in continuous cascade mode where the wastewater flow was transported through of the each MFC compartment. Stacked MFC 2 contained 40 units of air-cathode MFC in a shared reactor. Stacked MFC 2 module was formed by four chambers; each chamber of 70 cm height, cm 10 cm length and 5.7 cm width (volume of ~4 L) contained 10 MFC units without separator (Figure 1b). An individual MFC unit was composed by three carbon felt (5 cm length, 5 cm height and 0.65 thickness) and was used as anode (superficial area of 0.0075 m<sup>2</sup>). The three anodes were

connected externally by a single copper wire. A MEA (5 cm length, 5 cm width) was exposed to air and was utilized to separate the anodic chamber from the cathode. The distance between anode and cathode was of 2 cm, and the distance between each MFC unit was 3 cm.



**Figure 1.** Architecture of microbial fuel cells systems. Stacked MFC 1 (a). Stacked MFC 2 (b).

Stacked MFC systems were inoculated with a 50% of raw municipal wastewater from residential housing at the Jiutepec, Mexico, and a 50% of anaerobic granular sludge (previously dissolved) from the UASB reactor at the paper industry wastewater treatment plant. In order to establish a good microbial community in the anode electrodes, the two stacked MFC systems were operated at a HRT of 10 d, and fed with raw wastewater from residential housing. COD concentrations of raw wastewater were  $209 \pm 41$  mg/L, and the total nitrogen (TN), and total phosphorus (TP) concentrations were  $38 \pm 11$  mg/L, and  $15 \pm 3$  mg/L, respectively.

Stacked MFCs were first acclimated under OCV according to Kim et al. (2012) up to ensure a voltage continuous in each MFC unit. After acclimated steady, stacked MFC systems were operated to open circuit mode, and then the open circuit mode was changed to closed circuit mode with external resistor of 1,000  $\Omega$ . For each operational condition, stacked MFCs were worked at a three HRT: 3, 1, and 0.5 d. During all tests, the stacked MFCs systems were operated in shared anolyte mode under continuous flow. For the series connection of 20 and 40 MFC unit, the cathodes of MFCs 1 were connected to anode from MFCs 2 up to final MFC units in each stacked MFC.

### **Analysis and measurements**

We developed an electronic device that can measure the voltages generated from each individual MFC unit and to collect simultaneously the voltages in series connection during the operation of stacked MFC units connected in series. Voltages generated in each individual MFC units were monitored every 5 h, and the voltages in series connection were monitored every 12 h. The measurements of voltages were collected using a data acquisition system based on LabVIEW software connected to a personal computer.

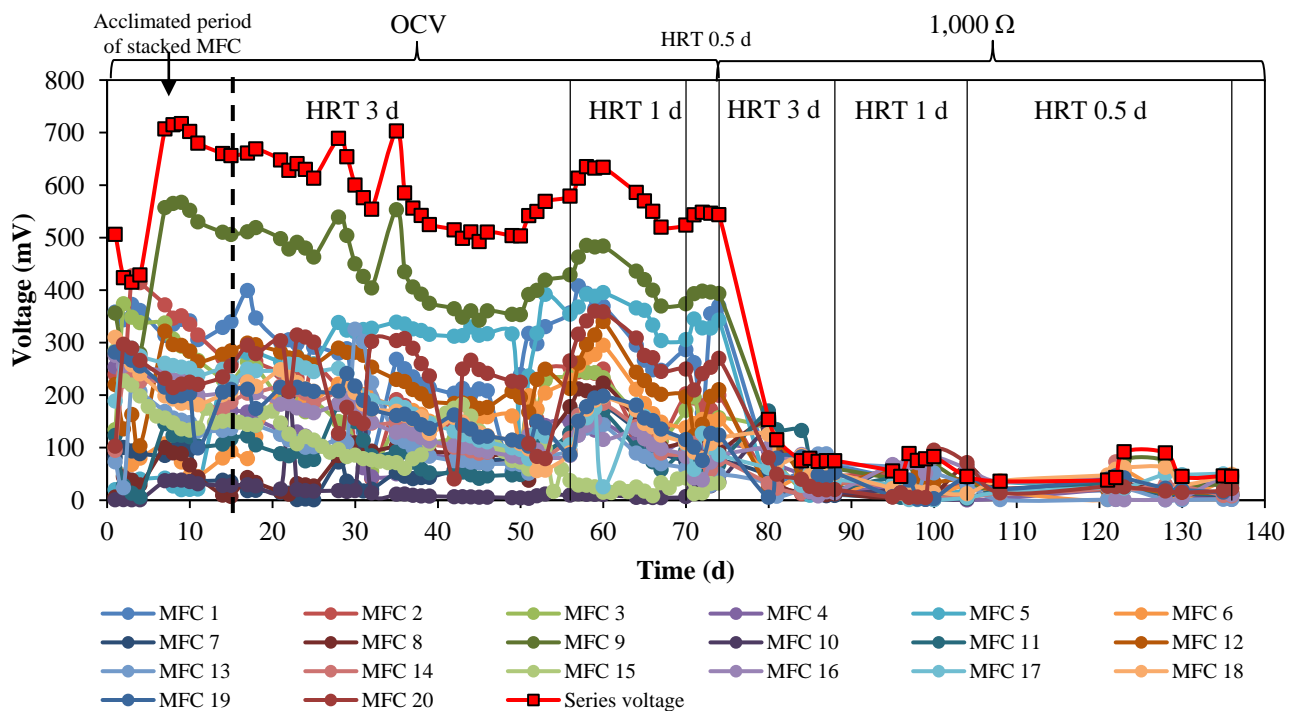
Current,  $I$  (mA), was calculated using  $I=V/R_{ex}$ , where  $V$  (mV) is the voltage and  $R_{ex}$  ( $\Omega$ ) is the external resistor. Power density,  $P$  (mW/m<sup>2</sup>) and current density,  $j$  (mA/m<sup>2</sup>), were calculated according to  $P = IV/AI,000$  and  $j = V/R_{ex}A$ , respectively, where  $A$  (m<sup>2</sup>) is the surface area of the anode electrode. Power density-current density ( $P$ - $j$ ) curves in stacked MFCs were obtained by changing the external resistor from 68  $\Omega$  to 10,000  $\Omega$  (20 min intervals in each resistor) using a resistor portable box/load bank (10 $\Omega$ -40k $\Omega$ ) developed by us. The polarization curves were controlled from LabVIEW software connected to a personal computer. For  $P$ - $j$  curves in series connection, power and current density were normalized based on total superficial area anode electrode (0.0036 m<sup>2</sup> for stacked MFC 1; 0.0075 m<sup>2</sup> for stacked MFC 2, multiplied by the number of MFC unit from each system). COD, TN, and TP were measured using standard methods (APHA, 2005).

## **RESULTS AND DISCUSSION**

### **Star-up and performance of stacked MFC systems at three HRT**

Figure 2 and Figure 3 shows the voltages obtained during the stacked MFC systems operation at a three HRT under open and closed circuit mode. During the two stacked MFC systems star-up (acclimated period), the individual MFC units produced voltage at OCV in

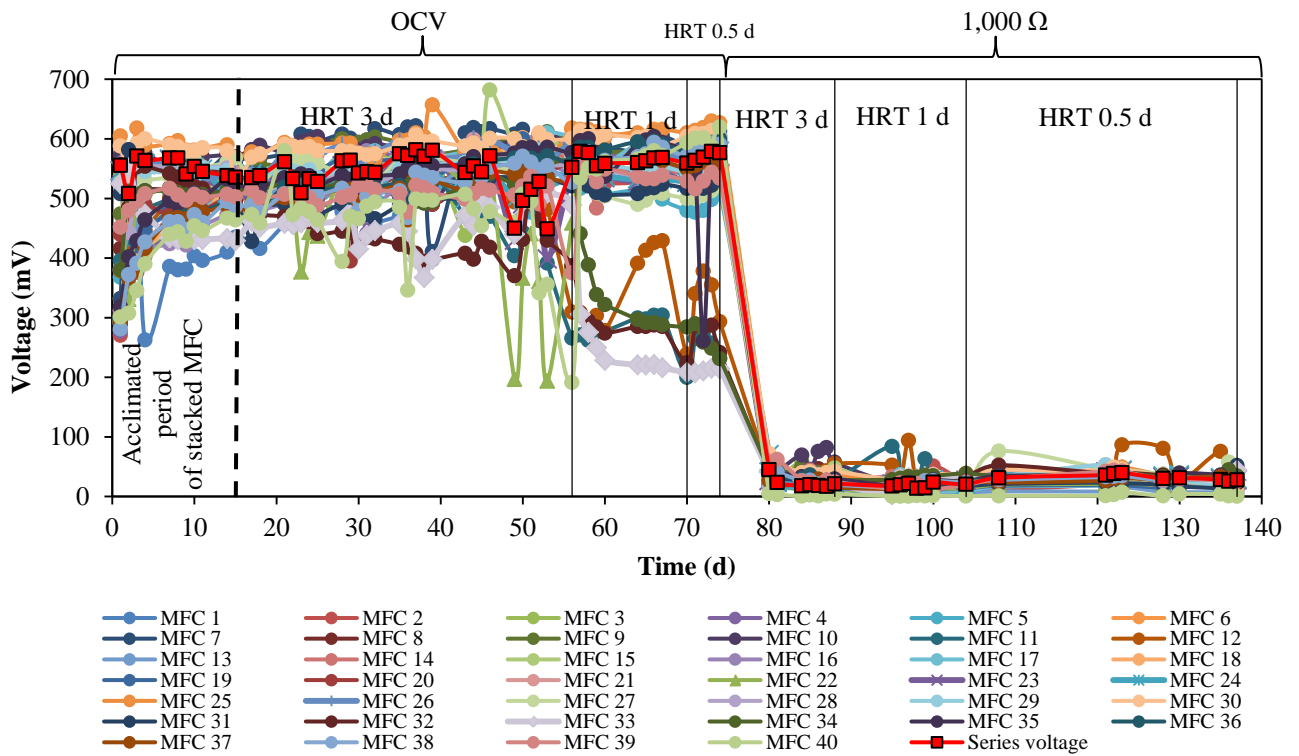
ranging from 0.7 to 567 mV for stacked MFC 1 and from 263 hasta 600 mV by stacked MFC 2. All individual cell units were acclimated under OCV noting a lag phase during the first five days. Similar observed have been reported by Ahn and Logan (2012), Hong et al. (2011), and Zhang et al. (2011a), however, in these previous studies, the MFCs were acclimated under closed circuit mode. From 6 to 14 days, the voltages were increased up to reach voltages steady. High voltages production in individual cell units were obtained in stacked MFC 2 compared to stacked MFC 1. Different voltages were obtained in each individual MFC units indicating that into to the two systems were generated different oxidation-reduction reaction rates and potential difference on the anode and cathode due to the decreased the COD concentrations in each MFC and competition between neighboring cells for the harvest of the electrons and protons. When the individual MFC units were switched to a series connection, the maximum voltage during acclimated period was 707 mV for stacked MFC 1 and 568 mV for the stacked MFC 2. When the stacked MFC 1 was switched to different HRT, the voltages in all individual MFC units decreased. For stacked MFC 2, four individual MFC units were affected by the change of HRT. The electricity productions were generated steadily in all individual MFC units and were remained positive. Electricity production under OCV from stacked MFC 1 and stacked MFC 2 were  $165 \pm 51$  mV and  $535 \pm 30$  mV, respectively.



**Figure 2.** Evolution of voltages to different HRT from the stacked MFC 1.

In series connections, the voltage obtained from stacked MFC 1 was  $580 \pm 65$  mV using 20 individual cells. For stacked MFC 2 in series connection, the voltage was of  $540 \pm 35$  mV, which was similar to the generated in each individual MFC unit. According to the law of

conservation of energy, when the cells units are connected in series, it is expected that the voltages generated in each cells is the sum of all voltages generated from all cell units ( $V_{total} = V_{MFC 1} + V_{MFC 2} + V_{MFC n+1}$ ). This principle was not observed in the two stacked MFC systems proposed either open or closed circuit mode due to a voltage dropped generated into the systems. This behavior has already been reported by others studies from stacked systems during series connections (Kim et al., 2012; Zuang et al., 2009; Wang and Han, 2009; Oh and Logan, 2007). When the individual MFC units are connecting in series a voltage drop phenomenon or voltage reversal are presented due to different factor as substrate concentration gradient between cells and cells, insufficient oxygen at the cathode, insufficient fuel, impedance differences, and a lack of catalyst and higher internal resistance (ohmic loss) (Oh and Logan, 2007). According to Kim et al. (2012), when the each individual MFC unit is not connect, the electrons formed in anode by bacteria activity are statics, then when anode and cathode are connected exist a electrons transfer from anode to cathode. The protons generated in anode travel through of wastewater (anolyte) and cation-exchange membrane to the cathode for electroneutrality. This oxidation-reduction reaction is faster in the anode that cathode. For this reason, when the individual MFC unit are connected in series and share the same anolyte or when the individual cell are in shared reactor the voltage drop is caused by ionic cross-conduction between units. This phenomenon was observed in the two stacked MFCs. Another factor that caused the voltage drop between units was the architecture of microbial fuel cells mainly of the stacked MFC 2. The individual cells that formed the stacked MFC 2 were placed in a shared reactor and these cells were not divided by separator.



**Figure 3.** Evolution of voltages to different HRT from the stacked MFC 2.

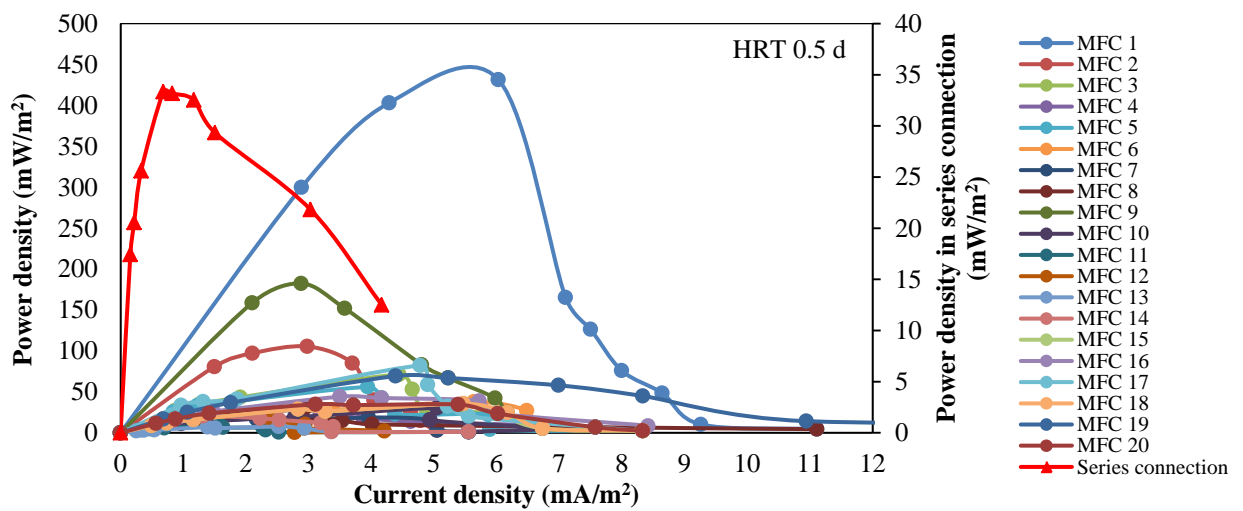
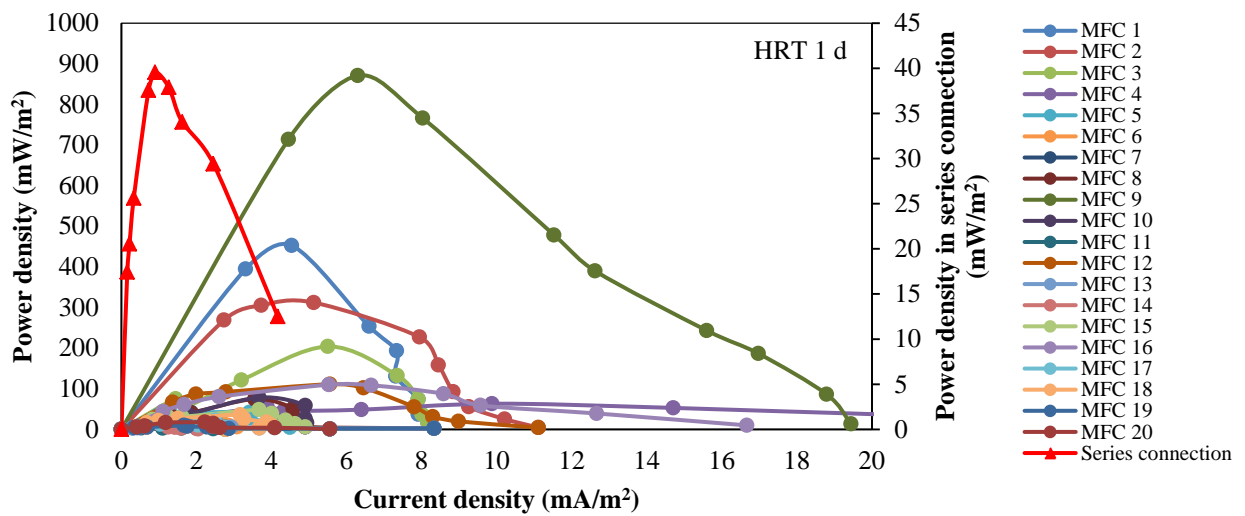
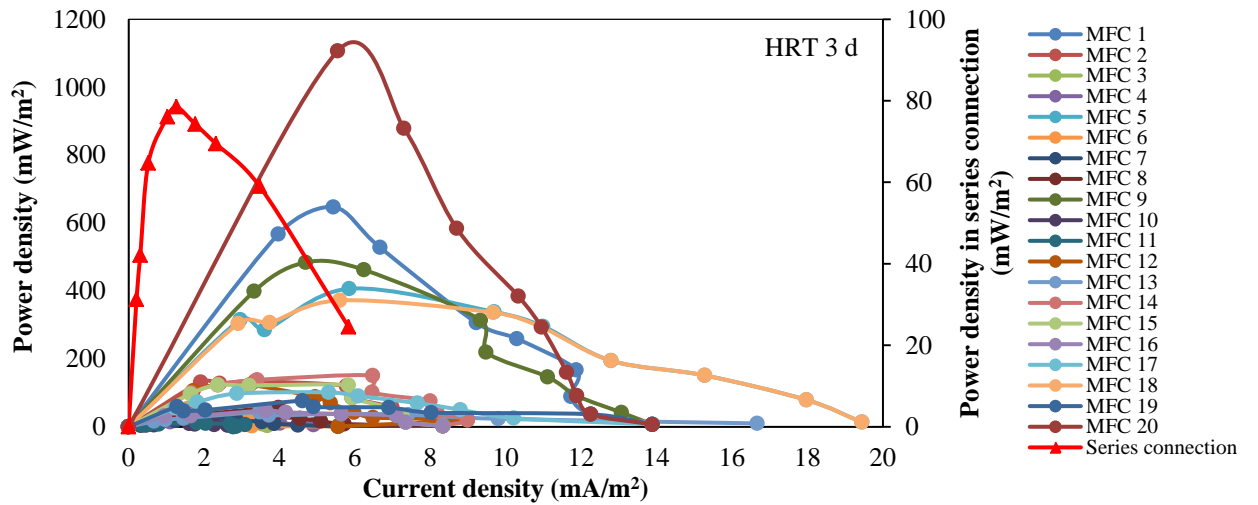
When the MFC units are sharing the same reactor and the distance between each unit is short, the force with which travel the protons through anolyte from anode to the cathode is low compared to the potential difference between the anode and cathode from their respective MFC ( $\text{cell}_n$ ). To increase the force to transport a proton from anode of the  $\text{cell}_n$  to cathode of the  $\text{cell}_{n+1}$  would have to be away the distance between the  $\text{cell}_n$  and the  $\text{cell}_{n+1}$ . However, it would not be a practical approach since the reactor design would be very large impacting directly on the operating costs of the system.

After 53 days of stacked MFC systems operation under OCV, the HRTs were reduced to 1 and 0.5 d. In both stacked MFC systems, the reduction of HRT were not affected slightly the electricity production in each individual MFC units and in series connection. When the systems were operated at closed circuit mode (resistor of 1,000  $\Omega$ ), the maximum voltage achieved in stacked MFC 1 was  $46 \pm 28$  mV at a HRT of 3 d. At a HRT of 3 d, the average voltage was  $30 \pm 8$  mV lower than the staked MFC 1. The voltage data in series connection from two stacked cells showed that the voltages drop phenomenon was also observed in the closed circuit mode. Under closed circuit mode, the voltages decreased slightly due to the high internal resistance. When the HRT decreased during the stacked systems under a resistance, the voltages were affected with values below 30 mV. The polarization curves of the individual MFC units (Figures 5 and Figure 5) show clearly this effect. These results indicate that the selection of HRT in stacked MFCs is an important factor for scalable design of stacked systems (Ahn and Logan, 2012) and will need considered the external resistance value as a variable of design. Overall performance under closed circuit mode of the two stacked MFC systems were slightly that when were operated in open circuit mode.

### **Polarization curves in non-stacked and series connection from MFCs at three HRT**

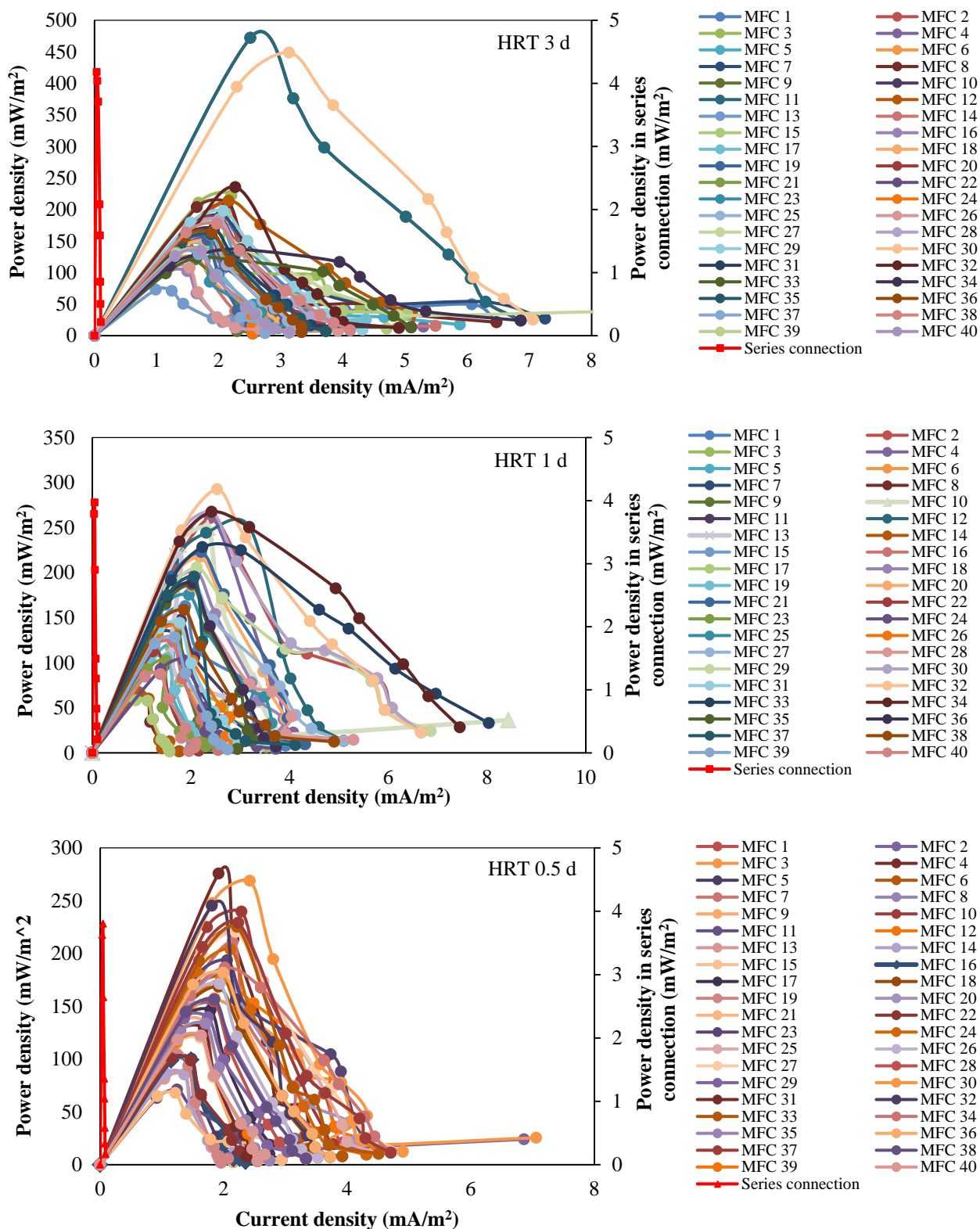
Figure 4 and Figure 5 shows  $P$ - $j$  curves obtained from non-stacked units and the stacked MFC in series connections. The polarization curves were performed after running at least three HRT during continuous flow operation stacked systems. Likewise, the  $P$ - $j$  curves were realized in continuous flow mode. The power and current densities generated from each individual cell under different HRT were not the same, but showed nearly the same polarization curves. For de two stacked MFC systems, the maximum power densities were determined at HRT of the 3 days. For individual MFC unit from stacked MFC 1, the maximum power density was  $1,106 \pm 1.2$  mW/m<sup>2</sup> that corresponding to the last individual MFC unit (MFC unit 20). The individual cell that showed a low power density (peak of power density) was MFC unit 10 ( $10 \pm 1$  mW/m<sup>2</sup>). In series connection from stacked MFC 1, the maximum power density observed was  $79 \pm 0.65$  mW/m<sup>2</sup> for  $14 \pm 2$  mV working voltage at external resistor of 680 ohms. The current density was  $1.3 \pm 0.4$  mA/m<sup>2</sup>. The polarization curves of the individual cells from stacked MFC 2, showed a maximum power density of  $473 \pm 2.5$  mW/m<sup>2</sup> for individual cell 10. Individual MFC unit 13 was the cell that presented a low power density (maximum power  $73 \pm 0.5$  mW/m<sup>2</sup>). Lower current densities (below 1 mA/m<sup>2</sup>) were obtained when the individual MFC units were connected in series. The maximum power density obtained from the stacked MFC 2 in series connections was  $4.2 \pm 0.6$  mA/m<sup>2</sup>. The magnitude of the voltage drop observed in the two stacked were greater when the individual MFC units were connected in series. The  $P$ - $j$  curves suggests that the two stacked MFC systems in series connections, the ohmic and activation losses

were the main action mechanism during the voltage dropped in the two systems (An et al., 2015, Gurung and Oh 2012; Ieropoulos et al., 2008; Oh and Logan, 2007).





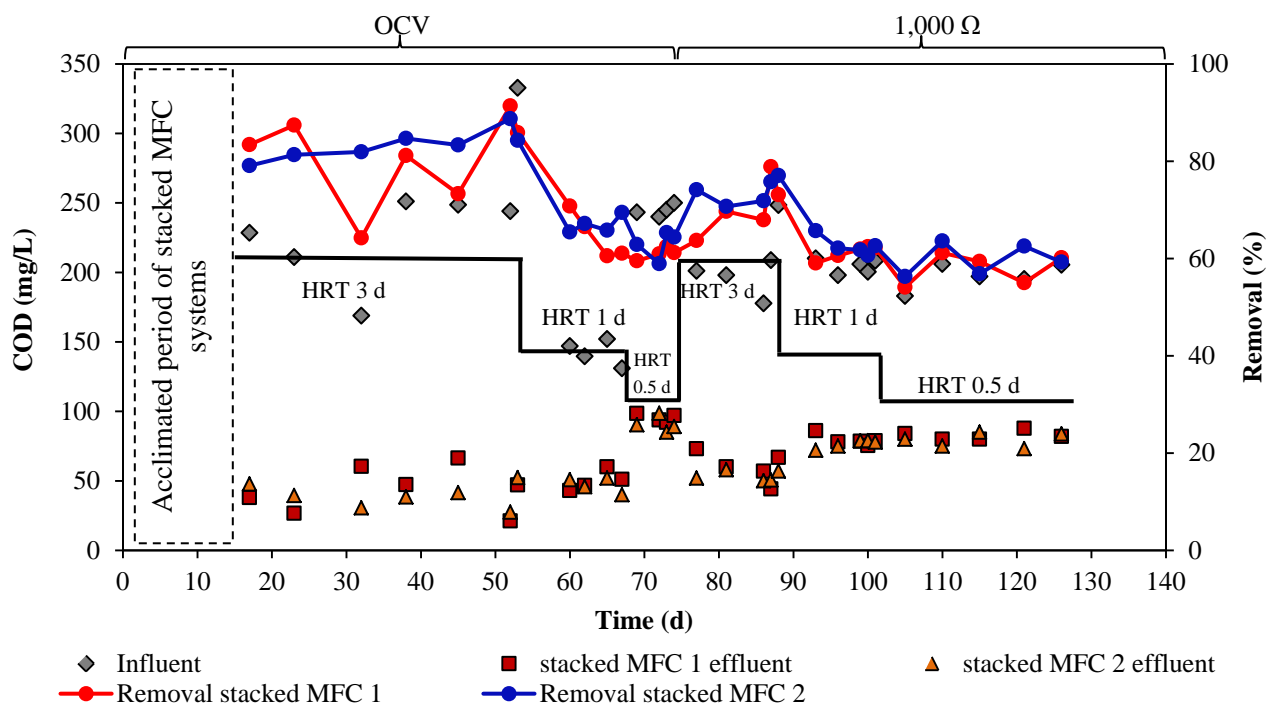
**Figure 4.** *P*-*j* curves obtained from non-stacked units 1-20 and stacked MFC 1 in series connection using 10 Ω, 68 Ω, 180 Ω, 330 Ω, 680 Ω, 1 kΩ, 3 kΩ, 6 kΩ and 10 kΩ at different HRT (3, 1, and 0.5 d).



**Figure 5.** *P-j* curves obtained from non-stacked units 1-40 and stacked MFC 2 in series connection using 10  $\Omega$ , 68  $\Omega$ , 180  $\Omega$ , 330  $\Omega$ , 680  $\Omega$ , 1 k $\Omega$ , 3 k $\Omega$ , 6 k $\Omega$  and 10 k $\Omega$  at different HRT (3, 1, and 0.5 d).

### COD removal at different HRT

The COD removals at different HRT from the stacked MFC systems are shown in Figure 6. The COD in the two stacked system influents had a  $210 \pm 42$  mg/L during the tests. The results showed that the COD removal was increased when the organic load was reduced from  $0.45 \pm 0.06$  g/L-d (HRT 0.5 d) to  $0.075 \pm 0.008$  g/L-d (HRT 3 d). At HRT of 3 days, maximum COD removals were obtained in both systems. When the cell systems were operated under open circuit mode, the COD removal for stacked MFC 1 was  $81 \pm 12\%$ , and for stacked MFC 2 was  $84 \pm 8\%$ . Under closed circuit mode, the COD removal efficiencies in the two systems decreased slightly with the three HRT tested. Previous studies found that, the influence of the external resistance applied on performance of the MFC nor to have a significant influence on COD removal, however, the bacteria diversity and metabolism on MFCs is modified when the MFCs is operated at closed circuit mode suggesting that fermentative metabolism dominated over anaerobic metabolism when the cells are operated under external resistor (Katuri et al., 2011; Rismani-Yazdi et al., 2011). The TN and TP concentrations in the stacked MFC systems were in the ranging from 23 to 37 mg/L for TN and from 13 to 22 mg/L for TP. The nutrients were slightly effectively removed in each cell system (<47%). Effluent concentrations of nitrate were also low in all tests (less than <4 mg/L). At different HRT (3, 1 and 0.5 d) the removal of TN and TP in two stacked MFCs was not significant.



**Figure 6.** COD removal during wastewater treatment from stacked MFC systems at different HRT (3, 1, and 0.5 d).

## CONCLUSIONS

The two stacked MFC systems tested were not effective for power production in series connection under OCV and closed circuit mode. The OCV and closed circuit voltage of the two cells systems during series connection were not equal to the sum of the voltages produced in each individual MFC units. Voltages dropped (voltage reversal) in the two systems were generated due to the architecture of the systems (shared reactor) and the same anolyte in all MFC units. The maximum power density in series connection of the stacked MFC 1 was  $79 \pm 0.65 \text{ mW/m}^2$  (current density of  $1.3 \pm 0.4 \text{ mA/m}^2$ ). For the individual MFC unit (no-series connection), the maximum power density was  $1,106 \pm 1.2 \text{ mW/m}^2$  (current density of  $5.5 \pm 0.6 \text{ mA/m}^2$ ) at HRT of 3 d. Power production in stacked MFC 2 ( $4.2 \pm 0.6 \text{ mW/m}^2$ ) and the current density ( $0.04 \pm 0.006 \text{ mA/m}^2$ ) were lower compared with the power generated from stacked MFC 1. The results showed that the COD removal were increased when the HRT were increased from 0.5 to 3 d.

## ACKNOWLEDGMENT

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