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In situ biofilm removal from air cathodes in microbial fuel cells treating domestic wastewater

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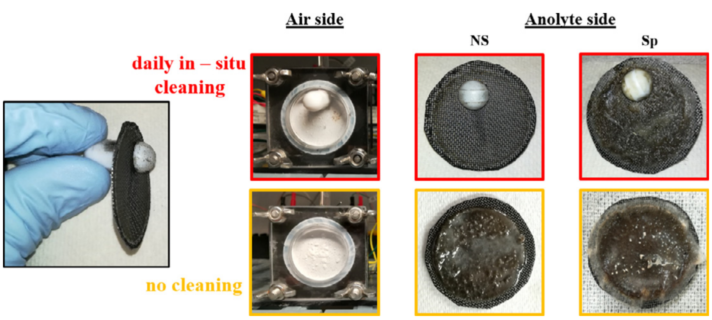
GRAPHICAL ABSTRACT



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ABSTRACT

One challenge in using microbial fuel cells (MFCs) for wastewater treatment is the reduction in performance over time due to cathode fouling. An in-situ technique was developed to clean air cathodes using magnets on either side of the electrode, with the air-side magnet moved to clean the water-side magnet by scraping off the biofilm. The power output of the magnet-cleaned cathodes after one month of operation was 132 ± 7 mW m^{-2}, which was 42% higher than the controls with no magnet (93 ± 4 mW m^{-2}) (no separator, NS), and 110% higher (116 ± 4 mW m^{-2}) than controls with separators (Sp, 55 ± 7 mW m^{-2}). Cleaning cathodes using magnets reduced the biofilm by 75% (NS) and 28% (Sp). The in-situ cleaning technique thus improved the performance of the MFC over time by reducing biofouling due to biofilm formation on the air cathodes.

1. Introduction

The application of microbial fuel cells (MFCs) for wastewater treatment has two major challenges: scaling up the reactor system while maintaining efficient power generation and treatment efficiency (Li et al., 2014b; Cheng et al., 2014); and ensuring the stability of performance over time (Santoro et al., 2017; Logan, 2010). When scaling up an MFC, it is important to use a high packing density of electrodes (area per volume of reactor) to maintain the volumetric power production and thus minimize hydraulic retention times (Logan et al., 2015; He et al., 2016). The stability of power generation is mostly dependent on the performance of the cathodes over time (Liu et al., 2018; Wang et al., 2017; Rismani-Yazdi et al., 2008; Santoro et al., 2017; Kim et al., 2016), as the brush anode behaviour has been shown to be stable for more than a year (Zhang et al., 2014a). Previous studies have shown that the cathode performance is reduced over time for several reasons that include: external fouling due to the biofilm formation (Zhang et al., 2014a; Zhang et al., 2017); and internal fouling, due to the precipitation of salts (An et al., 2017), the adsorption of humic acids (Yang et al., 2016), and the inclusion of metabolic by-products such as extracellular

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polymers and molecules (Liu et al., 2018; Ma et al., 2014). Effective cathode cleaning usually requires a combination of removing the biofilm from the cathode surface to remove external fouling, followed by acid cleaning by immersion of the cathode for several hours in a weak acidic solution (Zhang et al., 2014a; Rossi et al., 2017).

A biofilm readily forms on the solution side of the cathode due to the growth of aerobic microorganisms sustained by oxygen crossover through the cathode and the organic matter in the wastewater (Chung et al., 2011). Once formed, the biofilm can impact the performance in different ways. A biofilm can hinder the ion transport to and from the cathode, reducing the hydroxide ions diffusion from the cathode to the anode or the proton transport from the anode to the cathode, resulting in an increase of the internal resistance of the MFC (Liu et al., 2018; Pasternak et al., 2016; Xu et al., 2012). It has been recently shown that a microbial biofilm on the solution side of the cathode hinder the mobility of the OH^- ions released by the oxygen reduction reaction (ORR), inducing a strong alkalinisation of the environment in the immediate vicinity of the abiotic electrode leading to a large decrease in the cathode open circuit voltage and performance (Erable et al., 2018). It is also possible that the biofilm could act as a separator, reducing the transport of organic molecules into the cathode (Zhang et al., 2009; Yang et al., 2018). However, there does not appear to be any beneficial impact as it has been observed that cathode performance decreases over time, which indicates that biofilm formation has an overall adverse impact on MFC performance; it has also been shown that biofilm removal improves the performance of MFCs (Liu et al., 2018; Zhang et al., 2011b). Several approaches have therefore been developed to reduce biofouling of the cathodes, including adding antimicrobials such as vanillin (Chatterjee and Ghangrekar, 2014), quaternary ammonium salts (Li et al., 2014c), or silver nanoparticles (An et al., 2011) directly in the catalyst, or by using antimicrobial polymers coating (Watson et al., 2011) or antibiotics incorporated into the catalyst layer (Liu et al., 2015). However, all these techniques have only led to small reductions in biofilm formation, and the antimicrobial activity was not constant over time. While it is difficult to remove internal foulants, biofilm removal is easily accomplished in the laboratory by removing the cathode and brushing or scraping off the biofilm. However, cathode removal can be more difficult for larger-scale reactors as it may not be possible to easily detach the cathode for cleaning (He et al., 2016).

In this study, an easy and scalable in-situ method was developed to remove the external biofilm fouling, i.e. without the need to remove the cathode from the reactor. Two magnets were placed on either side of the cathode, so that the magnet on the anolyte side was held against the cathode by the magnet on the air side. By moving the air side magnet across the cathode, the magnet on the solution side is also moved across the cathode which physically scrapes the biofilm off the cathode surface. This approach of using a magnet to clean a surface is routinely used in an aquarium to clean the glass of the tank. A magnetic cleaning procedure can be repeated as often as needed without removing the cathode or interrupting operation of the MFCs. To test this cleaning process, MFCs were operated using domestic wastewater (Zhang et al., 2014b). The cathodes in the MFCs were operated without a separator (no separator, NS), or with a cloth separator on the cathode (separator, Sp) as the use of a separator has been shown to reduce fouling and result in improved performance over time (Yang et al., 2018; Li et al., 2014a). Performance was compared with control reactors (no cathode cleaning) over time, with the MFCs operated in fed-batch mode, based on polarization and power density tests. Following operation of the MFCs for over one month, the biofilm growth on the cathode or separator surface was quantified based on extractable protein.

2. Materials and methods

2.1. MFC construction and operation

MFCs were single-chamber, cubic-shaped reactors constructed from a polycarbonate block 4 cm in length, with an inside cylindrical chamber having a diameter of 3 cm (Rossi et al., 2017). The anodes were graphite fiber brushes (2.5 cm in both diameter and length) wound using two titanium wires that were heat treated at 450 °C in air for 30 min prior to use, and placed horizontally in the middle of MFC chambers (Logan et al., 2007; Vargas et al., 2013; Shi et al., 2012). Anodes were acclimated in MFCs for over two years at a fixed external resistance of 1000 Ω, at a constant temperature (30 °C) prior to use here with new cathodes. Commercially available VITO CORE® cathodes were manufactured by VITO (Mol, Belgium) and contained an activated carbon catalyst, a 70% porous diffusion layer (70% DL) made of a thin layer of polytetrafluoroethylene (PTFE), and a stainless steel current collector (Pant et al., 2010, Zhang et al., 2011b). Domestic wastewater was collected once a week from the primary clarifier of the Pennsylvania State University Waste Water Treatment Plant, and stored at 4 °C prior to use. All reactors were operated in fed batch mode at 30 °C in a temperature controlled room.

The magnets used for cleaning the cathodes were PTFE covered, rounded cylindrical stirring bars (VWR) 2 cm long with a diameter of 1 cm. The bars were cut into shorter (0.5 cm) and longer (1.5 cm) pieces, with the shorter magnet inserted on the solution side of the cathode and the longer magnet on the air side (see photographs in the Supporting Data). The electrode was cleaned daily by moving the magnet on the air side across the whole area of the electrode, which moved the magnet on the water side to scrape off the biofilm. Four control MFCs were operated in the same configuration; one pair of MFCs had a magnet on the cathode that was never moved to provide a comparison of the impact of the lower active area of the cathode; and a second pair of MFCs were operated without any magnet on the cathode. One set of experiments was conducted with no separator on the cathode (NS); a second set of experiments was conducted using the same MFC configuration with a cloth separator on the cathode, so that the magnet scraped the separator and not the cathode (Sp). A pre-cut wipe cloth (PZ-1212, Contec, USA) with the same area of the cathode (11.3 cm^2) was used as a separator in the Sp configuration (Zhang et al., 2014b).

Single cycle polarization tests were conducted by varying the external resistance (1000, 500, 200, 100, to 75 Ω at 20 min intervals) after filling the reactor with fresh solution and holding it in open circuit conditions for 2 h, for a total test duration of 3.7 h. The voltage drop (U) across an external resistor was recorded by a computer-based data acquisition system (2700, Keithley Instrument, OH). Current densities (i) and power densities (P) were normalized to the total exposed cathode projected area (A = 7 cm^2), and calculated as $i = U/RA$ and $P = iU/A$, where R is the external resistance. The presence of the magnet on the cathode reduced the exposed area of the electrode. Therefore the power density was also reported in terms of the active area (AA = 6.2 cm^2) of the cathode in the presence of the magnet ($P_{AA} = iU/A$). During each polarization test, anode and cathode potentials were also recorded using reference electrodes to track the change of single electrode performance in MFCs. The reference electrode was placed close to the anode brush and used to measure the anode potential (E_{An}). The cathode potential (E_{Cat}) was estimated using the cell voltage as $E_{Cat} = U + E_{An}$, and thus are slightly underestimated (more negative) due to inclusion of ohmic losses. All results are presented as the average and standard error of tests using duplicate reactors.

2.2. Cathodes and separators surface biomass analyses

Biomass on the bare cathodes and separators was measured based on total protein concentrations using a bicinchoninic acid protein assay kit (Sigma Aldrich) (Ishii et al., 2008). Biomass was extracted from the separator or cathode using 0.2 N NaOH. Each sample was placed in a petri dish with 5 mL of 0.2 N NaOH. The NaOH solution was withdrawn into a 10-ml syringe and flushed over the surface of the electrode six to eight times over a 1-h extraction period. This liquid was removed and weighed, and the electrode was further rinsed with an equivalent

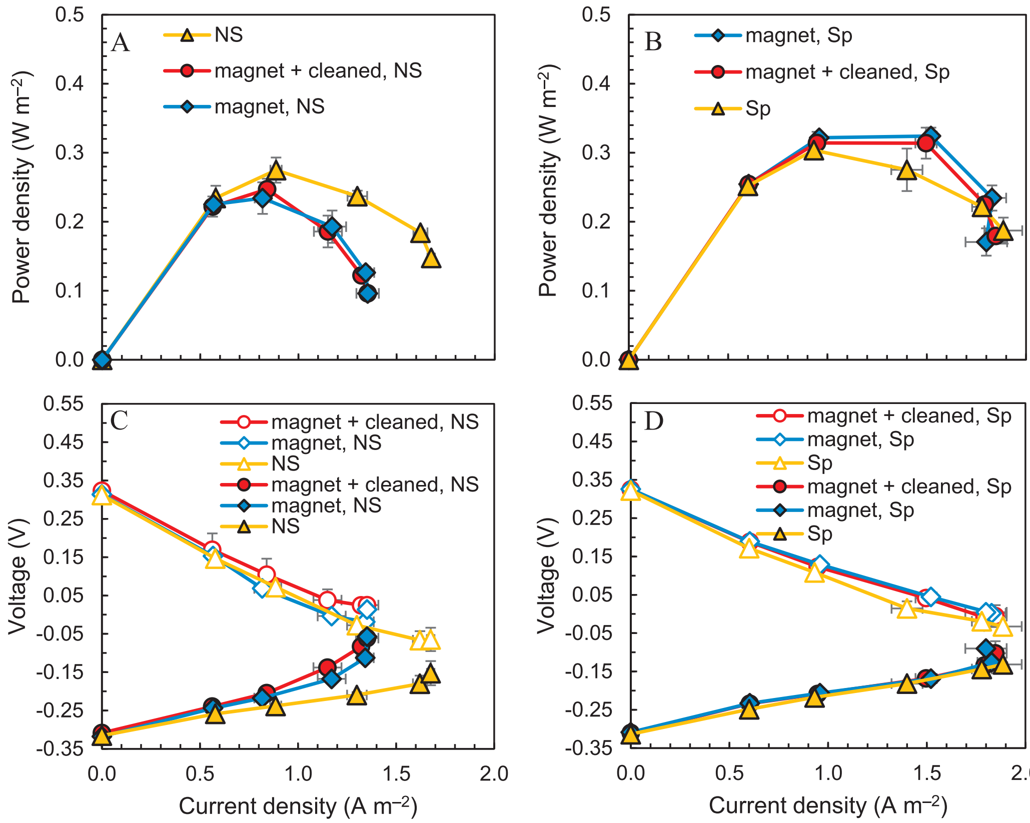

Fig. 1. Power density curves (A) without (NS) and (B) with the separator (Sp), and electrode potentials (C) without (NS) and (D) with the separator (Sp) for the three MFC configurations using well acclimated anodes and new cathodes (day 1). Cathode potentials (open symbols) and anode potentials (closed symbols).

amount of deionized water. The liquids were pooled, yielding a sample in 0.1 N NaOH, which was frozen (–20 °C) and then thawed at 100 °C for 10 min. This freeze–thaw cycle was repeated three times, and 0.1 mL of the extracted solution was used for the protein analysis by the bicinchoninic acid method against a bovine serum albumin standard in 0.1 N NaOH (Bond and Lovley, 2003).

3. Results and discussion

3.1. Evaluating the impact of the blocked cathode area and separator.

The initial (day 1) maximum power density based on polarization tests of the MFCs without separators (NS) that contained a magnet and would be cleaned daily was 247 ± 15 mW m^{-2}. The controls with magnets that were not going to be moved produced 234 ± 23 mW m^{-2}, while the controls that did not contain magnets produced 275 ± 18 mW m^{-2} (Fig. 1). These differences in maximum power densities are within a range expected for MFCs (~15%) (Yang et al., 2017). However, the presence of the magnet on the cathode, which reduced the active area by 11%, could have lowered the power output of the MFCs. If these power densities are normalized by the active area of the electrode (P_{AA}), due to the space occupied by the magnet, the power densities produced by MFCs with magnets were 280 ± 17 mW m^{-2} for the MFCs that would be cleaned daily, and 260 ± 26 mW m^{-2} for the MFCs that would not be cleaned. These power densities corrected for cathode surface area thus produce power densities more similar to the MFCs lacking the magnet. Cathode potentials for the MFCs without the separators were all similar to each other, but the anode potentials in the presence of the magnet were slightly more positive than the controls without magnets at current densities higher than 0.8 A m^{-2} (Fig. 1).

The initial (day 1) maximum power densities produced in MFCs with the separator placed on top of the cathode were higher than in the absence of the separator. The initial power density of the MFCs with separators and magnet that would be moved was P = 314 ± 16 mW m^{-2} (Sp) (P_{AA} = 354 ± 18 mW m^{-2}). This power density was similar to the initial maximum power density of 324 ± 12 mW m^{-2} (P_{AA} = 365 ± 13 mW m^{-2}) from control reactors with magnets that would not be moved for cleaning. The power density produced by controls without magnet was 304 ± 9 mW m^{-2}, which was lower than that of the MFCs without magnets. The higher power density for the MFCs with the magnets was likely due to the magnets pressing the separator tightly against the cathodes. It was shown that a gap between the separator and cathode, which can contain immobile water, can reduce power (Wei et al., 2013). Thus, it is likely that the magnet helped to press the separator against the cathode and avoid the deleterious impact of this water gap on performance (Zhang et al., 2011c). It is also possible that there were variations between the MFCs due to small changes in the composition of the wastewater used for the tests since the polarization tests for the two configurations (Sp and NS) were conducted in different days (see additional data in the Supporting data).

The greater initial power densities for the MFCs with the separators compared to the MFCs without separators was due to cathode performance, as the cathode potentials were more positive in the presence of the separator than in the absence of the separator. For example, in the MFCs with magnets that would be cleaned daily, the cathode potentials were 123 ± 5 mV (Sp, 0.95 ± 0.02 A m^{-2}), which was 18 mV higher than that of the MFCs without separators (NS, 105 ± 42 mV, NS, 0.84 ± 0.03 A m^{-2}). Similarly, the cathode potentials for the MFC cathodes with magnets that would not be cleaned daily were 60 mV higher (Sp, 129 ± 2 mV at 0.96 ± 0.01 A m^{-2}; NS, 69 ± 11 mV at 0.82 ± 0.04 A m^{-2}), and 38 mV greater for the cathodes with no magnets (Sp, 110 ± 12 mV at 0.93 ± 0.01 A m^{-2}; NS, 72 ± 14 mV at

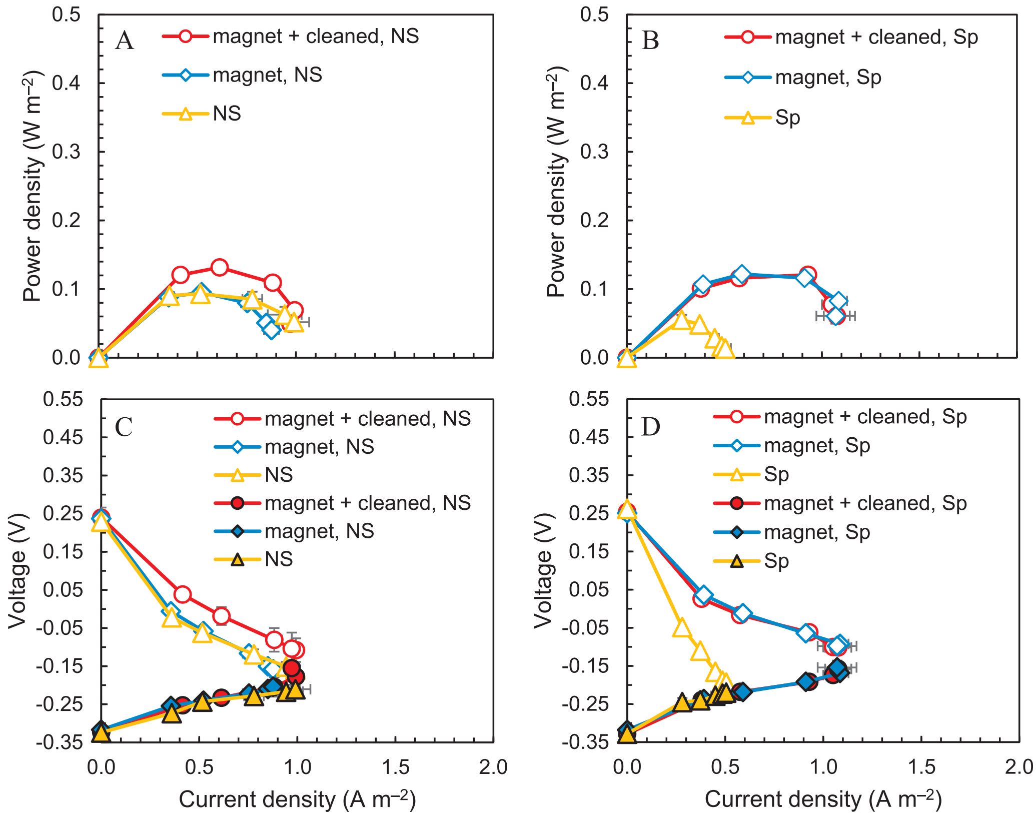

Fig. 2. Power density curves (A) without (NS) and (B) with the separator (Sp) and electrode potentials (C) without (NS) and (D) with the separator (Sp) for the three MFC configuration using well acclimatized anodes and used cathodes (day 30). Symbols of cathode potentials (open symbols) and anode potentials (closed symbols) corresponds to the electrode potentials in the same configuration at day 1.

$0.89 \pm 0.03\,A\,m^{-2}$). The difference in the anode potential between the two configuration (NS and Sp) was negligible at low current densities, but a substantial difference was observed between the MFCs with the magnets and the controls without it at current densities above $1\,A\,m^{-2}$ in the absence of separators. However, the maximum power density point was reached at current densities between $0.82 \pm 0.04\,A\,m^{-2}$ and $0.89 \pm 0.03\,A\,m^{-2}$, thus the impact of the lower performance of the anodes on the maximum power density was negligible. Small differences in the composition of the wastewater affected with the same magnitude the cathode performance in both configurations but the presence of the magnet adversely affected the anode performance only in the absence of a separator. (See additional supporting data in the Supporting data.)

3.2. MFC performance in wastewater after one month

After one month of operation, power output had decreased for all MFCs due to cathode fouling (Fig. 2). For the MFCs without separators, the maximum power density from polarization tests with the cleaned cathodes was $132 \pm 7\,mW\,m^{-2}$, which was 39% higher than the MFCs with cathodes with a magnet and no cleaning ($95 \pm 6\,mW\,m^{-2}$) or 42% higher than the control with no magnet ($93 \pm 4\,mW\,m^{-2}$). Adjusting the power from the cleaned-cathode MFCs for the portion of the cathode covered with the magnet produces a maximum power density of $P_{AA} = 148 \pm 8\,mW\,m^{-2}$. While this is 53% of the power initially produced with the new cathodes on day 1 for the cleaned cathode MFCs, the maximum power production of the control with no magnet was reduced by 66% compared to that on day 1.

The higher power output of the MFCs with cleaned cathodes was clearly due to better performance of the cathodes (Fig. 2). For example, at a current density of $0.88 \pm 0.04\,A\,m^{-2}$ the cathode potential was -81 ± 31 mV, which was 46% more positive than the uncleaned cathode potentials of -151 ± 3 mV at a slightly lower current density of $0.85 \pm 0.01\,A\,m^{-2}$. There was no change in the anode potentials after one month of operation, although the maximum current densities for the anodes was lower due to the reduced performance of the cathodes. Differences in anode potentials noted for the MFCs with the magnets (NS) on day 1 were absent after one month of operation as all the anodes had similar performance.

Cleaning the separators on the cathodes using the magnet did not improve performance of the MFCs. The maximum power density of the MFCs that were daily cleaned after one month was $P = 121 \pm 4\,mW\,m^{-2}$ (P_{AA} $136 \pm 4\,mW\,m^{-2}$), which was not different than that obtained when the magnet was placed on the cathode but not moved ($P = 122 \pm 1\,mW\,m^{-2}$, P_{AA} $137 \pm 1\,mW\,m^{-2}$). These maximum power densities using a separator were both lower than that obtained with cleaning of the cathodes for MFCs without separators ($132 \pm 7\,mW\,m^{-2}$). The control reactors that had separators, but no magnet, produced only $55 \pm 7\,mW\,m^{-2}$, which was 55% lower than that obtained with the magnet and cleaning, and an overall reduction of power of 82% on day 1. Thus, directly cleaning the cathode using the magnet improved performance relative to MFCs with or without separators.

The decrease in the power densities of the MFCs with separators was clearly due to decrease in cathodes potentials. For example, the cathode potential with the separators was -17 ± 2 mV for the cleaned cathodes and -12 ± 8 mV for the cathodes that were not cleaned at $0.58 \pm 0.01\,A\,m^{-2}$ for MFCs with magnets on the cathodes. In the absence of the magnet, the cathode potential dropped to -49 ± 1 mV at $0.28 \pm 0.02\,A\,m^{-2}$ (Fig. 2). The anode potentials did not change after one month of operation, indicating that changes in the cathode potentials reduced MFC performance.

3.3. Cathodes and separators surface biomass analysis

Based on the protein analyses after one month of operation, there was less biofilm on the cathodes that were cleaned daily, as well as the separators that were cleaned daily, compared to the controls. The biomass content of the cleaned cathodes from MFCs without separators was 110 ± 29 μg cm^{-2}, which was 75% less than that of the controls (441 ± 26 μg cm^{-2}) that had a magnet but were not cleaned daily (photographs in the Supporting data). The protein content on the cleaned separators was 327 ± 17 μg cm^{-2}, which was lower than that of the separators that were not cleaned but had a magnet (453 ± 61 μg cm^{-2}), and 45% less than that of the control without a magnet (596 ± 27 μg cm^{-2}) (see also photographs in the Supporting data). The biomass on the cleaned separator was the lowest but significantly higher than that observed on the electrode in the absence of a separator, thus different methods for the removal of the microbes within cloth separators need to be developed.

3.4. MFC performance in wastewater after biomass removal without the separator

To investigate whether cleaning the fouled cathodes after one month could restore power to the same level as that obtained by cleaning the cathode daily, the biofilm was removed from the cathodes with the magnet after one month of operation. Removal of this biofilm after one month resulted in a maximum power density of 133 ± 7 mW m^{-2}, which was the same as that of 129 ± 23 mW m^{-2} for the cathodes cleaned daily (Fig. 3). The change in performance was due to an increase in the cathode potentials as the anode potentials were not affected by the biofilm removal on the cathode. Considering the lowest power density of 93 ± 4 mW m^{-2} for the cathode without the magnet to be due to both internal and external fouling, for a total reduction of a 66% in performance, 48% of the reduced power production was due to internal cathode fouling (from 275 ± 18 mW m^{-2} to 133 ± 7 mW m^{-2}), and 18% was due to external (biofilm) fouling (from 133 ± 7 mW m^{-2} to 93 ± 4 mW m^{-2}). Previous studies reported an improvement after the biofilm removal in the range 3%–5% after 5.5 months (Zhang et al., 2014a) and 7% after 1 year (Zhang et al., 2017), indicating a larger impact of the internal fouling after long period of time.

3.5. MFC performance after replacement of the separators

The presence of the separator adversely impacted the performance of the control MFCs without the magnet after one month of operation. The maximum power density of the MFCs with separators after one month was 55 ± 7 mW m^{-2}, which was 41% lower than that obtained without separators (93 ± 4 mW m^{-2}). Therefore, the separators on all the MFCs were replaced with new separators after one month to determine their impact on power production (Fig. 3). All MFCs produced higher power densities with the new separators. The reactors that had been cleaned with the magnet had an increase of 12% in the maximum power density to 136 ± 1 mW m^{-2}, the MFCs with the magnet that were not cleaned increased to 143 ± 3 mW m^{-2}, while the controls without the magnets produced 131 ± 19 mW m^{-2}. These maximum power densities were also similar to those obtained with the MFCs without separators when the external biofilm was removed. Thus, removing the biofilm on the cathode or replacing the separator removed the external fouling of the MFCs, resulting in similar maximum power densities that were limited by internal fouling.

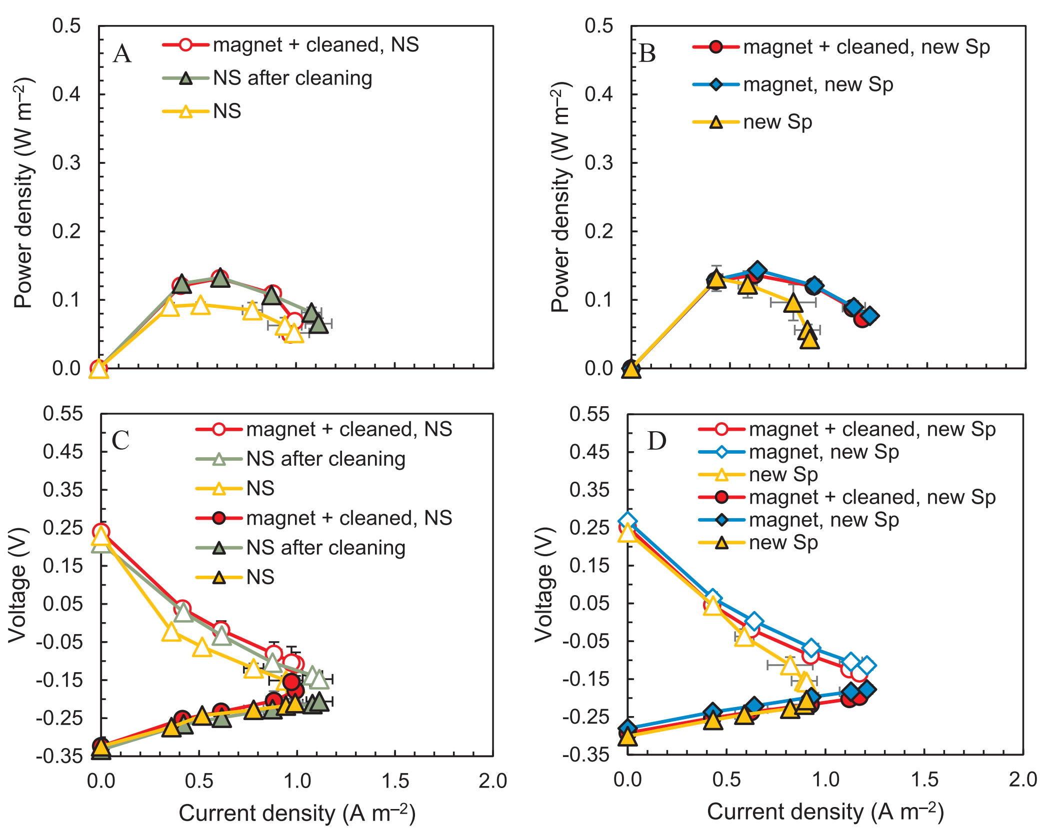

Fig. 3. Power density curves (A) without (NS) and (B) with the separator (Sp) and correspondent electrode potentials (C) without (NS) and (D) with the separator (Sp). (A, C) Before (yellow) and after the removal of the biofilm (red) daily or (green) after one month. (B, D) After the replacement of the 30 days old separators in the three MFC configuration. Symbols of cathode potentials (open symbols) and anode potentials (closed symbols) corresponds to the electrode potentials in the same configuration at day 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

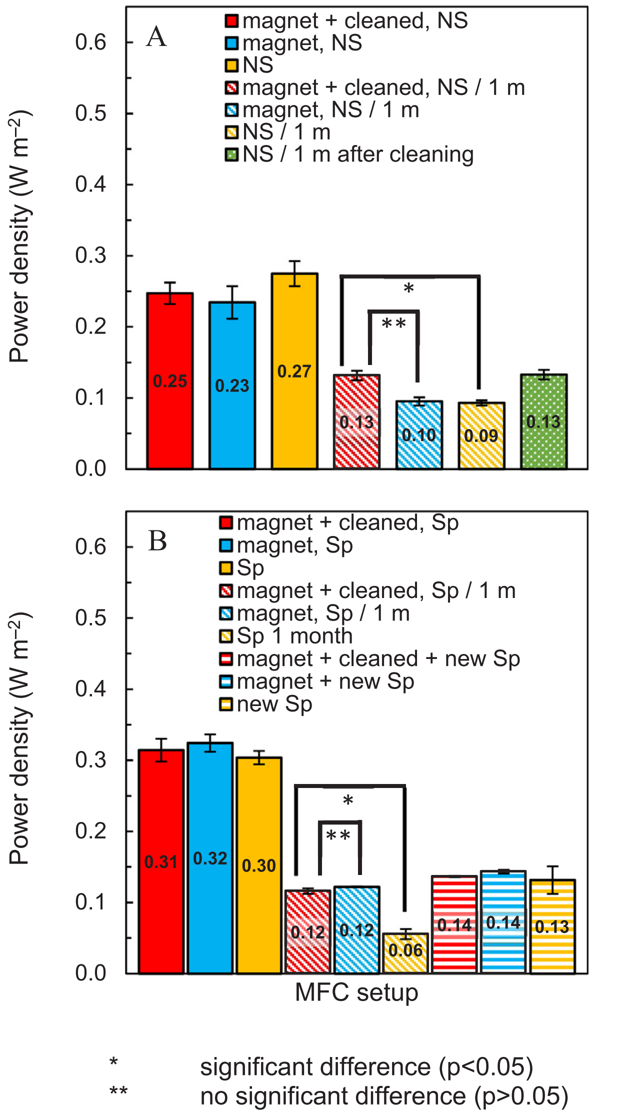

Fig. 4. Maximum power density (A) without a separator (NS) and (B) with separators (Sp) on day 1 (1 d) and after one month of operation (1 m), and after cleaning the separator, the cathode surface, or replacing the separator for the three different MFC configurations. Significant differences were analyzed using two tails *t-test* with an alpha value of 0.05.

3.6. Implications for maintaining MFC performance over time

The low stability of the cathode performance over time can hinder the applications of the MFCs for wastewater treatment (Liu et al., 2018; Santoro et al., 2017), and biofilm growth on the solution side of the cathodes is one of the main reasons for reduced MFC performance (Yang et al., 2018; An et al., 2017; Li et al., 2014c). As shown here, an in-situ cleaning of the biofilm on the solution side cathode reduced the biofilm growth on the cathodes, and improved performance by 39% relative to controls (no cleaning) after one month of operation (Fig. 4). When separators were used on the cathode, there was no beneficial impact of cleaning the separator on the cathode compared to the control with a magnet that was not moved. However performance of the MFCs with the separators and magnets were improved relative to the MFCs with separators and no magnet, due to the magnet keeping the separator pressed against the cathode. This finding is consistent with previous reports showing that better performance is obtained in MFCs when separators are bonded (Yang et al., 2018) or pressed (Wei et al., 2013) onto the cathode to minimize the gap between the separator and cathode.

Even though the daily cleaning of the cathodes improved the performance of the MFCs, the power output with cleaned cathodes after one month of operation was still only 53% (NS) or 38% (Sp) of that obtained at the beginning of the tests with new cathodes (Fig. 4). For example, MFCs with cathodes that were cleaned daily (NS) produced a maximum power density of 247 ± 15 mW m^{-2} on day 1, but only 132 ± 7 mW m^{-2} after one month of operation. Removing the biofilm from the cathodes of the MFCs without separators, or using new separators on the MFCs that had been operated with separators, all resulted in maximum power densities that were ~ 50% lower than that initially obtained with new cathodes. Thus, this decrease in power was due to internal fouling of cathodes. This will require that other in-situ methods be developed to remove the internal fouling, or that the cathodes will need to be removed and cleaned using a chemical method such as soaking in acid (Liu et al., 2018; Zhang et al., 2014a; Rossi et al., 2017).

4. Conclusions

Daily cleaning the cathode maintained better power production over time compared to no biofilm removal. MFCs with cleaned cathodes generated 42% higher power densities after one month of operation by reducing the biofilm on the cathode. Cleaning separators did not improve the MFC performance, but the firm adhesion of the separator to the cathode by using the magnet reduced the separator fouling, increasing the power produced by 110% after one month. Replacing the separators in control reactors after one month of operation resulted in performance similar to that produced by the MFCs cleaned daily with the magnet.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.biortech.2018.06.008.

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