



Performance of anaerobic fluidized membrane bioreactors using effluents of microbial fuel cells treating domestic wastewater



Kyoung-Yeol Kim, Wulin Yang, Yaoli Ye, Nicole LaBarge, Bruce E. Logan *

Department of Civil and Environmental Engineering, The Pennsylvania State University, 231Q Sackett Building, University Park, PA 16802, USA

HIGHLIGHTS

- MFC–AFMBR combined process was operated over 112 days treating domestic wastewater.
- Impact of HRTs and organic loadings on performance of the AFMBR was evaluated.
- COD removal efficiency of the AFMBR was not affected by variation of HRTs.
- Higher effluent CODs from the AFMBR resulted from a higher organic loading rate.
- TMP could be maintained under 0.18 bar without membrane cleaning over 112 days.

ARTICLE INFO

Article history:

Received 17 January 2016

Received in revised form 15 February 2016

Accepted 17 February 2016

Available online 22 February 2016

Keywords:

Microbial fuel cell

Anaerobic fluidized membrane bioreactor

Organic loading rate

Hydraulic retention time

Transmembrane pressure

ABSTRACT

Anaerobic fluidized membrane bioreactors (AFMBRs) have been mainly developed as a post-treatment process to produce high quality effluent with very low energy consumption. The performance of an AFMBR was examined using the effluent from a microbial fuel cell (MFC) treating domestic wastewater, as a function of AFMBR hydraulic retention times (HRTs) and organic matter loading rates. The MFC–AFMBR achieved $89 \pm 3\%$ removal of the chemical oxygen demand (COD), with an effluent of 36 ± 6 mg-COD/L over 112 days operation. The AFMBR had very stable operation, with no significant changes in COD removal efficiencies, for HRTs ranging from 1.2 to 3.8 h, although the effluent COD concentration increased with organic loading. Transmembrane pressure (TMP) was low, and could be maintained below 0.12 bar through solids removal. This study proved that the AFMBR could be operated with a short HRT but a low COD loading rate was required to achieve low effluent COD.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Anaerobic membrane bioreactors are being increasingly investigated as a way to treat domestic wastewaters as they provide an alternative strategy for a reducing energy demands by avoiding the need for aeration, as well as producing a higher quality effluent without the need for secondary clarifiers (Smith et al., 2013). However, avoiding membrane fouling is a serious challenge for long term operation, as the energy demands and costs can be very high for some membrane processes to control fouling (Liao et al., 2006; Martin et al., 2011). To minimize the membrane fouling and reduce energy use, a two stage anaerobic process was recently proposed that consisted of an anaerobic fluidized bioreactor (AFBR), followed by a secondary membrane process, the anaerobic fluidized bed membrane bioreactor (AFMBR). The membrane reactor contained granular activated carbon (GAC) suspended by recirculation, to

provide a growth support for bacteria (Kim et al., 2011), as well as providing a method for minimizing membrane fouling through the scouring of the membrane by the GAC particles. A low organic loading to the AFMBR and minimal membrane fouling allowed for a relatively short hydraulic retention time (HRT) of only 2.2 h. The AFMBR was further tested as a second stage of an AFBR in a pilot scale test, which showed that this two stage process could reduce the chemical oxygen demand (COD) to <23 mg-COD/L. The use of the fluidized GAC allowed for operation over 485 days without the need for chemical cleaning of the membrane, with a transmembrane pressure range of 0.2–0.5 bar (Shin et al., 2014). One disadvantage of the AFBR, however, is the high concentration of methane in the reactor effluent.

Microbial fuel cells (MFCs) are being investigated as a method for both wastewater treatment and electricity production (Logan and Rabaey, 2012; Rozendal et al., 2008; Wang et al., 2015). In order to be practical for wastewater treatment and energy recovery, MFCs must produce useful power and have HRTs similar to other treatment processes such as activated sludge. In one recent

* Corresponding author. Tel.: +1 814 863 7908; fax: +1 814 863 7304.

E-mail address: blogan@psu.edu (B.E. Logan).

test, a 90 L stackable MFC produced a relatively high power density for brewery wastewater of $171 \pm 8 \text{ mW/m}^2$ on the basis of cathode projected area (Dong et al., 2015), but it only produced 1.1 W/m^3 on the basis of total reactor volume. In order to produce both a high power density based on both area and volume, it is essential to provide sufficient cathode surface area per volume of reactor (specific surface area; m^2/m^3) as the cathode typically limits power production (Logan et al., 2015). In a recent multi-electrode MFC test, a maximum of $400 \pm 8 \text{ mW/m}^2$ (12 W/m^3) was produced using domestic wastewater by using a reactor with $29 \text{ m}^2/\text{m}^3$ of cathode area (He et al., 2016). One of the main challenges for all MFCs used for wastewater treatment is that at low COD concentrations (~ 100 – 200 mg/L), power densities become very low (Zhang et al., 2015). It is therefore not possible to produce higher power densities at COD concentrations needed for wastewater discharge to the environment. Thus, a post-treatment process is required to further reduce the COD for MFCs.

Several different approaches have been used to combine MFCs and membrane bioreactors to accomplish both low COD concentrations and power production. These include: using an ultrafiltration (UF) or forward osmosis (FO) membrane in the MFC system (Kim et al., 2014; Zhang et al., 2011); adding a membrane module into the MFC reactor (Ge et al., 2013b; Malaeb et al., 2013); and using a two-stage MFC and AFMBR. The UF and FO processes have so far shown problems with sustained treatment due to membrane fouling, and a long HRT is required to meet the levels needed for wastewater discharge. However, the two-stage process of a MFC and an AFMBR was shown to both produce electrical power in the MFC process, and achieve low COD levels needed for discharge with a short HRT by using the AFMBR reactor (Ren et al., 2014). The combined MFCs produced 0.0197 kWh/m^3 , with 92.5% COD removal overall for both processes, and no membrane cleaning was needed during the 50-d study. While this AFMBR study established the feasibility of the combined MFC–AFMBR process, the performance of the AFMBR was not investigated relative to operational parameters such as organic loading, as the reactor was operated at a fixed HRT of 1 h. While there have been previous studies on the AFMBR reactor treating AFBR effluent, the results based on the AFBR primary reactor do not necessarily predict performance using an MFC primary treatment process. For example, the AFMBR operated with the AFBR (1.0–1.9 h HRT) operated at a flux of 6 – $10 \text{ L/m}^2 \text{ h}$ (LMH) with an initial transmembrane pressure of 0.03 – 0.06 bar that increased over time to 0.1 bar . In contrast, the AFMBR (1 h HRT) operated following an MFC produced a flux of 16 LMH , with 0.02 – 0.04 bar needed for treatment, with a 100% increase in pressure over time.

In order to better understand the performance of the AFMBR, we examined the impact of COD loading rate by varying the HRT of the AFMBR. Domestic wastewater (primary clarifier effluent) was first treated in an MFC at a fixed HRT of 8.8 h, and then subsequently treated using an AFMBR at HRTs ranging from 1.4 to 3.8 h to vary the organic loading rate, and then operated under steady conditions at a HRT of 1.2 h. Overall, the AFMBR was tested for performance for 112 d in order to better understand its performance under these different operational conditions.

2. Methods

2.1. AFMBR and MFC construction

The AFMBR reactor (65 mL) was constructed from a transparent polyvinyl chloride (PVC) tube (300 mm long by 16 mm diameter, U.S. Plastic Corp.) as previously described (Ren et al., 2014). Granular activated carbon (GAC) (10 g wet weight; DARCO MRX, 10×30 mesh; Norit) was used as the fluidized particles for scour-

ing the membrane and as a support for bacterial growth. The GAC was rinsed using deionized (DI) water prior to use. The PVC tube was fitted with a membrane module containing eight polyvinylidene fluoride (PVDF) hollow fiber membrane filaments (200 mm long, 2.0 mm outside diameter, 0.8 mm inside diameter, $0.1 \mu\text{m}$ pore size; Kolon Inc., South Korea) that were added to the reactor after the GAC was acclimated as a fluidized bed reactor (no membranes; see details in the Supporting Information). A Hungate tube (10 mL, Bellco Glass Inc., Vineland, NJ) with the bottom cut off was glued onto the top of the PVC reactor body, and the top of the tube was sealed with a thick butyl rubber stopper (20 mm diameter; Chemglass Inc., Vineland, NJ). A gas sampling bag (Calibrated Instruments Inc., NY) was connected using a needle through the rubber stopper to collect gas. A vacuum pressure gauge (Type1490, Ashcroft, Stratford, CT) was installed in the liquid effluent tube to monitor transmembrane pressure (TMP) of the membrane module.

Single-chamber, air cathode MFCs were constructed as previously described (Kim et al., 2015) and used to provide partially treated wastewater to the AFMBR. Each MFC contained 3 anodes (25 mm diameter, 35 mm long) made from graphite fiber brushes with a titanium core (Mill-Rose, Mentor, OH). Cathodes (40 cm^2 projected surface area) were made from a mixture of activated carbon (AC, TYPE and Manufacturer), carbon black (CB, Vulcan XC-72, Cabot Corporation, USA), and a PVDF binder (8.8 mg/cm^2 , 30:3:10) as previously described (Yang et al., 2014). Two layers of a textile cloth (46% cellulose, 54% polyester; 0.3 mm thick; Amplitude Pro-zorb, Contec Inc.) were placed on the cathodes (separators) to reduce fouling on the cathodes and oxygen intrusion into the MFCs. Both electrodes and the separators were acclimated to domestic wastewater in these reactors as part of a previous 9-month long study of MFC performance (Kim et al., 2015). Two MFCs (each with 140 mL working volume) had two cathodes placed on opposite sides of the anodes placed in the middle of the anolyte chamber, with a 0.8 cm gap between the edge of brushes and cathode electrodes (spaced electrodes, 2 cathodes; S2C). The anodes in the other two MFCs (100 mL each) were placed directly on top of the separators (electrodes next to a single cathode; N1C). The N1C brushes were trimmed along their length to form a half cylinder, the flat side was positioned against the cloth separator (0.5 cm from the cathode). Differences in the performance of these two types of reactors as a function of their HRTs was previously reported (Kim et al., 2015). Here, the main function of the MFCs was to provide a partially treated feed to the AFMBR.

2.2. MFC–AFMBR operation

Domestic wastewater was collected from the primary clarifier of the Pennsylvania State University Wastewater Treatment Plant, and stored in a refrigerator (4°C) prior to use. When used as a feed to the MFC, the wastewater was placed in an ice bucket, and then fed to the MFC through a line that warmed to room temperature before entering the MFCs. Each of the two similar types of MFCs were connected in series (2 S2C in series, 2 N1C in series), and then operated in two separate parallel flow paths to provide a combined feed to the AFMBR. The first MFC was labeled the upstream (U) reactor, and the second one the downstream (D) reactor. Domestic wastewater was pumped into the upstream MFCs using two peristaltic pumps (Model No. 7523-90, Masterflex, Vernon Hills, IL), with the two flow rates set to provide a constant theoretical HRT (based empty bed volume) of 8.8 h (31.8 mL/h , S2Cs; 22.8 mL/h , N1Cs).

The effluents from the MFCs were collected in a glass bottle, and the combined effluent was fed to the AFMBR using a peristaltic pump (inflow). The top of the membrane module was connected to another peristaltic pump (outflow) to extract AFMBR effluent by membrane filtration. These AFMBR pumps were operated at

the same flow rate, with a 10 min on and 1 min off cycle time for periodic relaxation of the membrane. The AFMBR operation was divided into three initial phases based on changing the HRTs: Phase I, HRT = 1.4 h; Phase II, HRT = 2.0 h, Phase III, HRT = 3.8 h (Table 1). Two additional phases (Phase IV and V) were included to accommodate changes in COD concentration of the influent (COD loading) and MFC performance. The HRTs of the two phases were same (HRT = 1.2 h), but the COD loading rates varied due to changes in the average COD of 2.7 ± 0.5 g-COD/L-d (Phase IV), and 2.2 ± 0.3 g-COD/L-d (Phase V) (Table 1). Before Phase V, excess solids in the AFMBR reactor were removed (80% of solution in the reactor was removed and refilled with new MFC effluent), and the separator and cathode surface of the MFCs were cleaned using DI water. The GAC was kept fluidized by recirculation using a peristaltic pump at a flowrate of 235 mL/min, resulting in a bed height of 23–25 cm (80–85% of membrane module was covered by the fluidization bed).

2.3. Analysis and calculations

The voltage across external resistor (200 Ω) for each MFC circuit was monitored every 10 min using a multimeter (Model 2700, Keithley Instruments, USA). Current was calculated using Ohm's law ($I = U/R$) and normalized by cathode surface area of MFCs (S2C: 80 cm², N1C: 40 cm²) to obtain the current density, where U is the measured voltage (V) and R the external resistance (Ω). Energy production (kWh/m³) was calculated as the sum of the power generated by the MFCs divided by the flowrate of the wastewater (Ge et al., 2013a). All COD samples were analyzed following standard methods using HACH COD kits (method 5220, HACH, Loveland, CO). Soluble COD samples were filtered through syringe filters (0.45 μ m pore size, PVDF, 20 mm diameter, Restek Corporation, USA).

3. Results and discussion

3.1. Variability of influent wastewater COD

The COD concentrations of the domestic wastewaters fed to the MFCs were not constant over the length of the study, resulting in variable organic loading rates that reflected those typically experienced by wastewater treatment plants. The influent COD was relatively high and constant for the first 33 days (469 ± 78 mg/L, Phase I), and then within a similar range but overall more variable for the next 10 days (460 ± 136 mg/L, Phase II). The average influent COD was much lower for the following 68 d in the last three phases (316 ± 33 , 329 ± 115 , and 315 ± 83 mg/L) (Table 1). As a result, the average COD loading rate was 9.5 g-COD/L-d for the first two phases, and 6.5 g-COD/L-d for the last three phases (Table 1).

The highest effluent COD (143 ± 17 mg/L) from the MFCs was observed for Phase I, with lower effluent CODs in Phases III (96 ± 9 mg/L) and Phase V (106 ± 14 mg/L each) (Fig. 1). Although the effluent COD of the MFCs was relatively high for Phase IV

(135 ± 24 mg/L), the effluent COD was reduced to 106 ± 14 mg/L in Phase V following cleaning of the MFC cathodes.

3.2. Effect of COD loading and HRT on COD removal by the AFMBR

An average effluent COD of the combined MFC and AFMBR was 36 ± 6 mg/L over the 112 days operation, with an average influent of 358 ± 98 mg-COD/L (Fig. 1). Assuming a ratio of 0.5 for the 5-day biochemical oxygen demand (BOD₅) to COD, based on previous direct comparisons of BOD₅ and COD for this site (Hays et al., 2011), the effluent COD from the AFMBR would meet secondary treatment discharge standards in the United States for BOD₅ <30 mg/L (EPA, 2010). Gas production from the AFMBR was minimal (little observed inflation of the gas bag), consistent with previous AFMBR tests where we found little gas production (Ren et al., 2014), and therefore gas production was not quantified.

There were no significant difference in COD removal efficiencies in the first three phases of study, despite the use of three different HRTs (1.4 h, 2.0 h, and 3.8 h) and resulting different COD loading rates (2.8 ± 0.3 g-COD/L-d at a 1.4 h HRT; 1.3 ± 0.3 g-COD/L-d at a 2.0 h HRT; and 0.8 ± 0.3 g-COD/L-d at a 3.8 h HRT) (Fig. 2). For example, $68 \pm 4\%$ COD removal was obtained in Phase III (3.8 h HRT), which was comparable to COD removal in Phase I ($70 \pm 4\%$) even with a shorter HRT (1.4 h HRT) and lower COD loading rate. These results indicate that increasing HRTs for AFMBR operation would not be effective to improve the performance of the AFMBR in terms of COD removal efficiency.

In contrast to the similar COD removal efficiencies, different concentrations of COD in the effluents were obtained from the AFMBR due to the different COD loading rates in each Phase (Fig. 2). When the COD loading rate was 2.8 ± 0.3 g-COD/L-d in Phase I, a higher effluent COD was of 43 ± 6 mg/L, than the

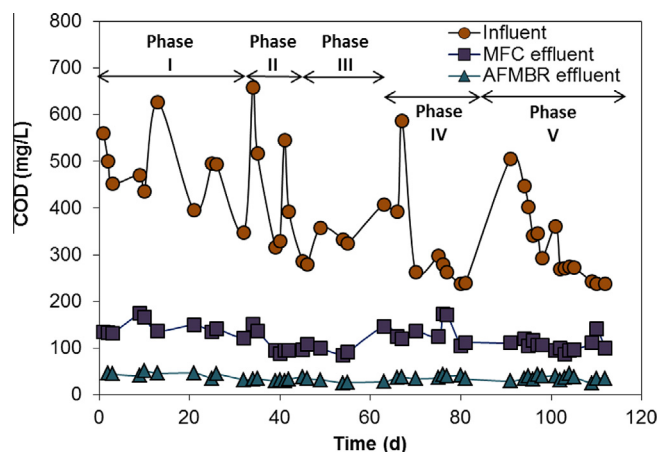


Fig. 1. Variation of influent wastewater (circle) COD and effluent COD of the MFCs (square) and the AFMBR (triangle) according to the influent COD levels during MFC–AFMBR operation for 112 days.

Table 1
Operational conditions of the MFCs and AFMBR connected in series over 112 days. The whole period was divided into five phases based on different operational conditions for the AFMBR operation.

Reactor	Conditions	Phase I (0–33 days)	Phase II (34–44 days)	Phase III (45–62 days)	Phase IV (63–84 days)	Phase V (85–112 days)
MFC	HRT (h)	8.8	8.8	8.8	8.8	8.8
	Influent COD (mg/L)	469 ± 78	460 ± 136	316 ± 33	329 ± 115	315 ± 83
	COD loading rate (g-COD/L-d)	9.6 ± 1.5	9.4 ± 2.5	6.3 ± 1.9	6.7 ± 2.3	6.4 ± 1.7
AFMBR	HRT (h)	1.4	2.0	3.8	1.2	1.2
	Influent COD (mg/L)	143 ± 17	110 ± 27	96 ± 9	135 ± 24	106 ± 14
	COD loading rate (g-COD/L-d)	2.8 ± 0.3	1.3 ± 0.3	0.8 ± 0.3	2.7 ± 0.5	2.2 ± 0.3
	Membrane flux (L/m ² h)	13.2	8.0	5.6	13.6	13.6

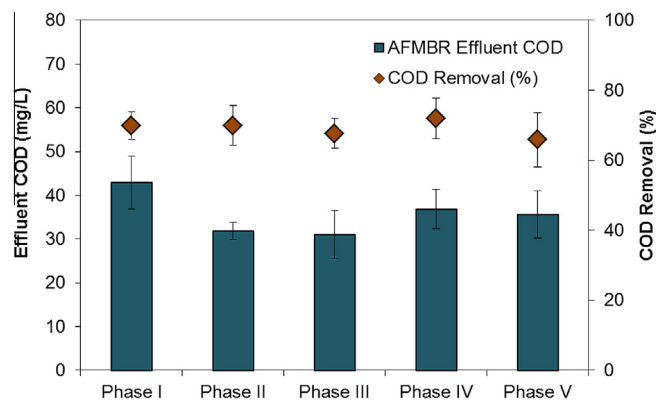


Fig. 2. Effluent COD level (mg/L) and COD removal efficiencies (%) in the AFMBR at each Phase (with different HRTs and COD loading rates).

31 ± 5 mg/L obtained in Phase III with 0.8 ± 0.3 g-COD/L-d. The level of COD in the AFMBR effluent varied from 27 mg/L to 51 mg/L, following the trends in COD loading rates over time. These results showed that the lower level of influent COD will be required to achieve a lower COD effluent from the AFMBR.

These results showing little change in COD removal efficiencies at different HRTs are in accordance with those previously reported for AFMBRs treating AFBR effluent. Yoo et al. (2012) reported similar COD removal efficiencies for the AFMBR at different HRTs (65 ± 10% at 3.4 h; 64 ± 9% at 2.3 h), at organic loading rates (1.0–1.2 kg COD/m³-d) comparable to those used here. Bae et al. (2014) obtained 13 ± 5 mg/L and 9 ± 4 mg/L effluent COD from an AFMBR, but the influent COD concentrations were only 54 ± 10 mg/L (1.5 h HRT) and 42 ± 16 mg/L (1.3 h HRT). These results and our findings here indicate that, unlike other processes, a longer HRT for the AFMBR will not be effective for reducing effluent COD concentrations. Fortunately, the AFMBR treatment is accomplished at very low HRTs, such as the 1.2–1.4 h used here. In order to achieve a lower level of COD effluent from the AFMBR, a lower influent COD (from an MFC or AFBR) will be required.

Overall soluble COD removal by the MFC-AFMBR was on average 76 ± 4% (effluent COD: 35 ± 6 mg/L), with 51 ± 7% of soluble COD in the wastewater (influent) removed by the MFCs, and 50 ± 8% removed by the AFMBR (Table S1). A higher COD effluent (35 ± 6 mg/L) was observed in this study compared to 16 ± 3 mg/L in a previous MFC-AFMBR study (Ren et al., 2014). This difference might be due to the different amount of soluble COD in the AFMBR influent (MFC effluent), as the soluble COD of the MFC effluent was 57 ± 14 mg/L in the previous study, which was about 19% lower than here (70 ± 6 mg/L).

Nutrient removal in completely anaerobic systems is quite challenging, and although it is of great interest there have been few studies that have included nutrient analyses for MFCs or AFMBRs. Several emerging biological technologies were recently reviewed for treatment of anaerobic reactor effluent for nutrient removal, but all are still under development (Delgado Vela et al., 2015). There are also abiotic alternatives such as electrochemical, precipitation and coagulation processes. For example, nutrient removal using air cathode electrocoagulation (ACEC) with a sacrificial aluminum electrode showed 99% removal of both ammonia and phosphorus in 4 h, which required 1.8 kWh/m³, but this was lower than many previous approaches (Tian et al., 2016).

3.3. TMP variation in the AFMBR for 112 days operation

The TMP measured for the AFMBR did not appreciably change during the first three phases, ranging from 0.04–0.07 bar (Fig. 3). However, in Phase IV a steady increase in TMP was observed to

0.18 bar. Up until Phase IV, solids had not been removed from the reactor. When the excess solids (not associated with GAC) were removed, the TMP decreased to 0.09 bar without membrane cleaning. The TMP was then fairly constant at 0.09–0.12 bar in Phase V (Fig. 3).

The TMP variation until day 62 in this study was similar to that reported by Ren et al. (2014) for the AFMBR (HRT = 1 h) treating MFC effluent, which ranged from 0.02 to 0.05 bar over 50 days operation, with an average flux of 16 LMH. Increasing the reactor HRT reduced the flux but did not appreciably impact the TMP, with a flux of 13.2 LMH in Phase I at an HRT of 1.4 h, decreasing to 8.0 LMH in Phase II, and 5.6 LMH in Phase III due to the longer HRTs. While a decrease in the flux might be expected to reduce the rate of membrane fouling, the rapid increase in TMP when the flux was increased to 13.6 LMH showed the reactor conditions had changed. Since accumulated solids had not been removed from the AFMBR reactor until Phase IV, this rapid increase was thought that the result of excess solids in the reactor for over 80 days operation. In a previous study using AFBR effluent (Kim et al., 2011), the TMP rapidly increased over 0.35 bar, and could be decreased to only 0.25 bar with membrane cleaning using chemicals. However, the TMP was successfully reduced and maintained to <0.1 bar by the same membrane cleaning followed by a daily removal of excess solids. Yoo et al. (2012) and Ren et al. (2014) periodically removed excess suspended solids by withdrawing reactor fluid from the recirculation line or reactor, and Shin et al. (2014) discarded solids after 101 days of operation by withdrawing bulk liquid from AFMBR reactor using peristaltic pump. Since reducing the solids in the reactor greatly decreased the TMP, and no membrane cleaning was needed, it was concluded that proper solids retention in the AFMBR will be critical for maintaining a low TMP and ensuring good performance.

3.4. MFC performance

The average COD of the MFC was 118 ± 25 mg/L over 112 days, for an overall removal efficiency of 65 ± 11%. The highest COD removal efficiency was observed in Phase II (75 ± 4%), when the influent COD was 460 ± 136 mg/L while the lowest and unstable COD removal efficiency was obtained in Phase IV (56 ± 14%) with 329 ± 115 mg-COD/L influent (Fig. 4). There was no significant change in COD removal efficiency (65 ± 10%) by the MFCs after cathode and separator cleaning (rinsing using DI water).

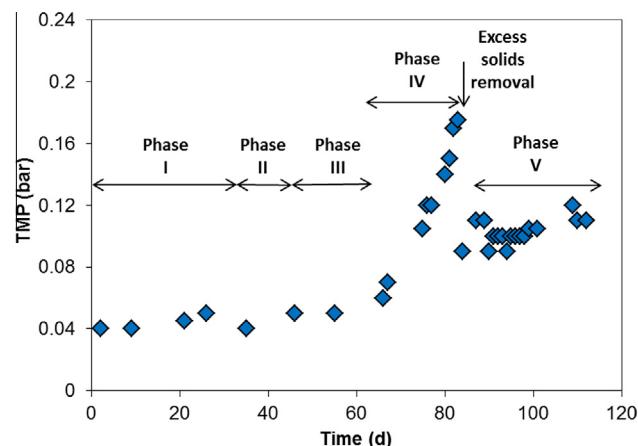


Fig. 3. The TMP variation in the AFMBR during 112 days operation. The excess solids were removed from the AFMBR reactor at day 83, and no additional membrane cleaning process was conducted over the operation except the GAC fluidization.

The COD removal efficiencies obtained here are a little lower than those reported by Kim et al. (2015) (64–69%) using the same MFC designs and an HRT of 8.8 h. Although the cathodes in this study have been used in the MFCs fed with domestic wastewater over 1 year, the MFCs were still showing a comparable COD removal efficiency ($65 \pm 11\%$). This result supported the fact that cathode contamination, which severely affects power generation of MFCs, does not seriously impact COD removal efficiencies of MFCs. The COD removal efficiencies are higher than those reported in previous MFC studies using slightly different reactor designs. For example, Ahn and Logan (2013) achieved 57% of COD removal for domestic wastewater using a single MFC with an 8 h HRT, and 49.1% of COD removal was reported by Ren et al. (2014) using four MFCs (two separated flow line, two MFCs in series) with a combined HRT = 8 h.

3.5. Energy production by MFCs

Current and power generation were different for the two types of MFCs, and upstream and downstream reactors. The single cathode, downstream MFC (N1C-D) MFC showed the highest current density (maximum of 0.22 A/m^2) in Phase I, while the S2C-U MFC produced a higher current density (0.13 A/m^2) than the S2C-D MFC (0.09 A/m^2) (Fig. 5). After reactor cleaning, the highest current density was observed for the N1C-U MFC (maximum of 0.21 A/m^2 on day 90), although it had produced the lowest and most inconsistent current (0.06 A/m^2) in Phases I to IV.

Total energy production by the four MFCs was initially $0.012 \pm 0.003 \text{ kWh/m}^3$ (Phase I), which decreased to $0.003\text{--}0.004 \text{ kWh/m}^3$ from Phase II to Phase IV as current densities decreased. Total energy production was restored to $0.012 \pm 0.005 \text{ kWh/m}^3$ (the same as that in Phase I), after reactor cleaning prior to Phase V. Total energy production obtained by the MFCs in this study (0.012 kWh/m^3) is about 40% lower than the previous study (0.0197 kWh/m^3) (Ren et al., 2014), likely due to the decrease in cathode performance over time. Assuming that total electrical energy requirement for pumping is 0.0186 kWh/m^3 , as reported in the previous study, total electrical energy generated by the MFCs would be insufficient to provide the needed energy for pumping. In addition, a doubling of the HRT of the AFMBR from 1 h to 2 h, would double that energy requirement. However, based on the findings in this study, increasing the HRT of the AFMBR is not needed since COD removal efficiency of AFMBR would not be changed even with increasing HRTs.

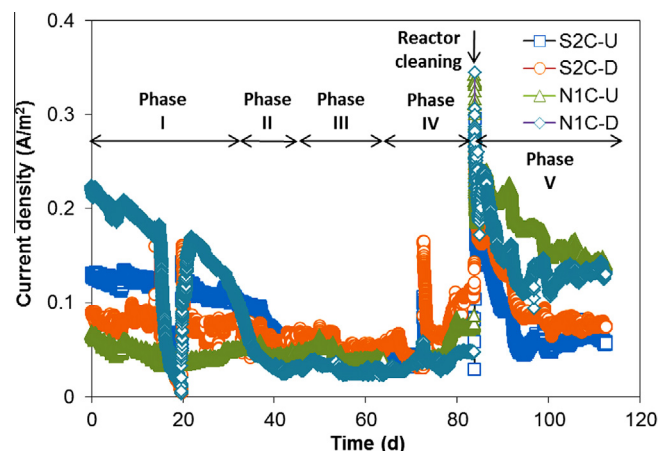


Fig. 5. Current generation (external resistance: 200Ω) of the 4 MFCs (S2C-U, S2C-D, N1C-U, N2C-D) for 112 days operation. The reactor cleaning includes removal of solids in the MFC reactors and biofilm on cathode electrode and separator using DI water. No wastewater supply at the break time (day 18).

Although the current densities were drastically increased after reactor cleaning, similar or lower current densities were observed after 95 days compared to those obtained in Phase I. This lower current was likely due to the low influent COD during Phase V ($315 \pm 83 \text{ mg/L}$) compared to Phase I ($469 \pm 78 \text{ mg/L}$). Analysis of possible relationships between influent COD and energy production based on data obtained here (Fig. S1) showed that the energy production (kWh/m^3) by upstream MFCs was in better agreement with influent CODs than downstream MFCs, likely because the performance of the downstream was dependent on the upstream MFCs (Fig. S1). These results show that COD concentrations are important for power generation, and that proper cleaning of the cathodes will be needed to ensure good performance relative to power production, but not COD removal, as COD removal efficiencies were not greatly impacted before and after cathode cleaning.

4. Conclusions

The COD removal efficiency of the AFMBR (65–70%), which treated effluent from the MFCs, was not significantly impacted by different HRTs. As a result, COD concentration in the AFMBR effluent was directly proportional to the influent COD. An average effluent COD of $36 \pm 6 \text{ mg/L}$ over the 112 days of the study. Membrane fouling was successfully controlled by GAC fluidization, and TMP was adversely affected by solids build up but not by changes in HRTs. Although MFC cathode cleaning greatly impacted on the power generation of MFCs, there was no significant effect of cathode cleaning on the COD removal efficiencies.

Acknowledgements

This research was supported by the Grant from the Strategic Environmental Research and Development Program (SERDP), and Award OSR-2015-SEED-2450-01 from the King Abdullah University of Science and Technology (KAUST).

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.biortech.2016.02.067>.

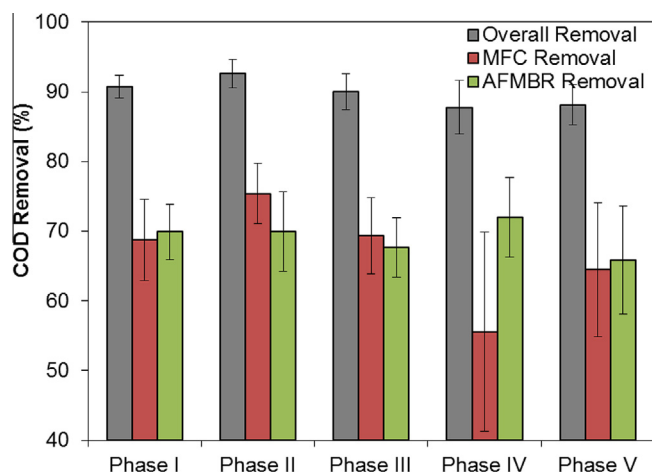


Fig. 4. Overall COD removal (%) by the combined process (MFC–AFMBR) and COD removal by the MFCs and the AFMBR at each phase.

References

- Ahn, Y., Logan, B., 2013. Domestic wastewater treatment using multi-electrode continuous flow MFCs with a separator electrode assembly design. *Appl. Microbiol. Biotechnol.* 97, 409–416.
- Bae, J., Shin, C., Lee, E., Kim, J., McCarty, P.L., 2014. Anaerobic treatment of low-strength wastewater: a comparison between single and staged anaerobic fluidized bed membrane bioreactors. *Bioresour. Technol.* 165, 75–80.
- Delgado Vela, J., Stadler, L.B., Martin, K.J., Raskin, L., Bott, C.B., Love, N.G., 2015. Prospects for biological nitrogen removal from anaerobic effluents during mainstream wastewater treatment. *Environ. Sci. Technol. Lett.* 2 (9), 234–244.
- Dong, Y., Qu, Y., He, W., Du, Y., Liu, J., Han, X., Feng, Y., 2015. A 90-liter stackable baffled microbial fuel cell for brewery wastewater treatment based on energy self-sufficient mode. *Bioresour. Technol.* 195, 66–72.
- US EPA., 2010. National Pollutant Discharge Elimination System (NPDES) Permit Writers' Manual. United States Environmental Protection Agency. EPA-833-K-10-001.
- Ge, Z., Li, J., Xiao, L., Tong, Y., He, Z., 2013a. Recovery of electrical energy in microbial fuel cells: brief review. *Environ. Sci. Technol. Lett.* 1 (2), 137–141.
- Ge, Z., Ping, Q., He, Z., 2013b. Hollow-fiber membrane bioelectrochemical reactor for domestic wastewater treatment. *J. Chem. Technol. Biotechnol.* 88 (8), 1584–1590.
- Hays, S., Zhang, F., Logan, B.E., 2011. Performance of two different types of anodes in membrane electrode assembly microbial fuel cells for power generation from domestic wastewater. *J. Power Sources* 196 (20), 8293–8300.
- He, W., Zhang, X., Liu, J., Zhu, X., Feng, Y., Logan, B.E., 2016. Microbial fuel cells with an integrated spacer and separate anode and cathode modules. *Environ. Sci. Water Res. Technol.* 2, 186–195.
- Kim, J., Kim, K., Ye, H., Lee, E., Shin, C., McCarty, P.L., Bae, J., 2011. Anaerobic fluidized bed membrane bioreactor for wastewater treatment. *Environ. Sci. Technol.* 45 (2), 576–581.
- Kim, K.-Y., Yang, E., Lee, M.-Y., Chae, K.-J., Kim, C.-M., Kim, I.S., 2014. Polydopamine coating effects on ultrafiltration membrane to enhance power density and mitigate biofouling of ultrafiltration microbial fuel cells (UF-MFCs). *Water Res.* 54, 62–68.
- Kim, K.-Y., Yang, W., Logan, B.E., 2015. Impact of electrode configurations on retention time and domestic wastewater treatment efficiency using microbial fuel cells. *Water Res.* 80, 41–46.
- Liao, B.-Q., Kraemer, J.T., Bagley, D.M., 2006. Anaerobic membrane bioreactors: applications and research directions. *Crit. Rev. Environ. Sci. Technol.* 36 (6), 489–530.
- Logan, B.E., Rabaey, K., 2012. Conversion of wastes into bioelectricity and chemicals using microbial electrochemical technologies. *Science* 337, 686–690.
- Logan, B.E., Wallack, M.J., Kim, K.-Y., He, W., Feng, Y., Saikaly, P.E., 2015. Assessment of microbial fuel cell configurations and power densities. *Environ. Sci. Technol. Lett.* 2 (8), 206–214.
- Malaeb, L., Katuri, K.P., Logan, B.E., Maab, H., Nunes, S., Saikaly, P.E., 2013. A hybrid microbial fuel cell membrane bioreactor with a conductive ultrafiltration membrane biocathode for wastewater treatment. *Environ. Sci. Technol.* 47 (20), 11821–11828.
- Martin, I., Pidou, M., Soares, A., Judd, S., Jefferson, B., 2011. Modelling the energy demands of aerobic and anaerobic membrane bioreactors for wastewater treatment. *Environ. Technol.* 32 (9), 921–932.
- Ren, L., Ahn, Y., Logan, B.E., 2014. A two-stage microbial fuel cell and anaerobic fluidized bed membrane bioreactor (MFC-AFMBR) system for effective domestic wastewater treatment. *Environ. Sci. Technol.* 48 (7), 4199–4206.
- Rozendal, R.A., Hamelers, H.V.M., Rabaey, K., Keller, J., Buisman, C.J.N., 2008. Towards practical implementation of bioelectrochemical wastewater treatment. *Trends Biotechnol.* 26 (8), 450–459.
- Shin, C., McCarty, P.L., Kim, J., Bae, J., 2014. Pilot-scale temperate-climate treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR). *Bioresour. Technol.* 159, 95–103.
- Smith, A.L., Skerlos, S.J., Raskin, L., 2013. Psychrophilic anaerobic membrane bioreactor treatment of domestic wastewater. *Water Res.* 47 (4), 1655–1665.
- Tian, Y., He, W., Zhu, X., Yang, W., Ren, N., Logan, B.E., 2016. Energy efficient electrocoagulation using an air-breathing cathode to remove nutrients from wastewater. *Chem. Eng. J.* <http://dx.doi.org/10.1016/j.cej.2016.02.004>.
- Wang, H., Park, J.-D., Ren, Z.J., 2015. Practical energy harvesting for microbial fuel cells: a review. *Environ. Sci. Technol.* 49 (6), 3267–3277.
- Yang, W., He, W., Zhang, F., Hickner, M.A., Logan, B.E., 2014. Single-step fabrication using a phase inversion method of poly (vinylidene fluoride) (PVDF) activated carbon air cathodes for microbial fuel cells. *Environ. Sci. Technol. Lett.* 1 (10), 416–420.
- Yoo, R., Kim, J., McCarty, P.L., Bae, J., 2012. Anaerobic treatment of municipal wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR) system. *Bioresour. Technol.* 120, 133–139.
- Zhang, F., Brastad, K.S., He, Z., 2011. Integrating forward osmosis into microbial fuel cells for wastewater treatment, water extraction and bioelectricity generation. *Environ. Sci. Technol.* 45 (15), 6690–6696.
- Zhang, X., He, W., Ren, L., Stager, J., Evans, P.J., Logan, B.E., 2015. COD removal characteristics in air-cathode microbial fuel cells. *Bioresour. Technol.* 176, 23–31.