



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



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## 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.

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## 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  $36 \pm 6$  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.

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## 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

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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  $1.1 \text{ W/m}^3$  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;  $\text{m}^2/\text{m}^3$ ) as the cathode typically limits power production (Logan et al., 2015). In a recent multi-electrode MFC test, a maximum of  $400 \pm 8 \text{ mW/m}^2$  ( $12 \text{ W/m}^3$ ) 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 ( $\sim 100\text{--}200 \text{ mg/L}$ ), 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  $0.0197 \text{ kWh/m}^3$ , 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  $6\text{--}10 \text{ L/m}^2 \text{ h}$  (LMH) with an initial transmembrane pressure of  $0.03\text{--}0.06 \text{ bar}$  that increased over time to  $0.1 \text{ bar}$ . In contrast, the AFMBR (1 h HRT) operated following an MFC produced a flux of  $16 \text{ LMH}$ , with  $0.02\text{--}0.04 \text{ bar}$  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,  $10 \times 30 \text{ mesh}$ ; 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 ( $40 \text{ cm}^2$  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 ( $8.8 \text{ mg/cm}^2$ , 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 ( $4 \text{ }^\circ\text{C}$ ) 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 ( $31.8 \text{ mL/h}$ , S2Cs;  $22.8 \text{ mL/h}$ , 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

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