



Performance of two-stage anaerobic fluidized membrane bioreactors using effluents of microbial fuel cells for treating domestic wastewater

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ABSTRACT

Anaerobic fluidized membrane bioreactors(AFMBRs) have been substantially developed as a post-treatment process to produce high quality effluent with veritably low energy consumption. The performance of an AFMBR was examined using the effluent from a microbial energy cell(MFC) treating domestic wastewater, as a function of AFMBR hydraulic retention times(HRTs) and organic matter loading rates. The MFC AFMBR achieved 90 ± 3 % reduction of the chemical oxygen demand(COD), with an effluent of 35 ± 6 mg- COD/ L over 112 days operation. The AFMBR had veritably stable operation, with no significant changes in COD loading rate, 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 bars through solids loading. This study proved that the AFMBR could be operated with a short HRT but a low COD loading rate was needed to achieve low effluent COD.

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. 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. 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, 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 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. 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. 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 test, a 90 L stackable MFC produced a relatively high power density for brewery wastewater of 171 ± 8 mW/m² on the basis of cathode projected area but it only produced 1.1 W/m³ 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. In a recent multi-electrode MFC test, a maximum of 400 ± 8 mW/m^2 (12 W/m^3) was produced using domestic wastewater by using a reactor with 29 m^2/m^3 of cathode area. 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. 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; adding a membrane module into the MFC reactor; 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. 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 MFCAFMBR 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 L/m^2 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 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.

1.1 Wastewater strength

The higher the concentration of organic matter in a wastewater, the stronger it is said to be. Wastewater strength is often judged by its BOD₅ or COD (Table 1.2). The strength of the wastewater from a community is governed to a very large degree by its water consumption. Thus, in the US where water consumption is high (350400 l/person day) the wastewater is weak (BOD₅ = 200250 mg/l), whereas in tropical countries the wastewater is strong (BOD₅ = 300700 mg/l) as the water consumption is typically much lower (40100 l/person day). The other factor determining the strength of domestic wastewater is the BOD (= amount of organic waste) produced per person per day. This varies from country to country and the differences are largely due to differences in the quantity and quality of sillage rather than of body wastes, although variations in diet are important. A good value to use in developing countries is 40 g BOD₅ per person per day

2. METHODS

2.1. AFMBR CONSTRUCTION

The AFMBR reactor (65 mL) was constructed from a transparent polyvinyl chloride (PVC) tube (300 mm long by 16 mm diameter). Granular activated carbon (GAC) (10 g wet of 10 – 30 mesh; was used as the fluidized particles for scouring 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 μ m pore size) that were added to the reactor after the GAC was acclimated as a fluidized bed reactor. A Hungate tube (10 mL) 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;). A gas sampling bag was connected using a needle through the rubber stopper to collect gas. A vacuum pressure gauge was installed in the liquid effluent tube to monitor transmembrane pressure (TMP) of the membrane module. Single-chamber, air cathode MFCs was constructed and used to provide partially treated wastewater to the AFMBR.

MFC CONSTRUCTION

The MFC contained 3 anodes (25 mm diameter, 35 mm long) made from graphite fiber brushes with a titanium core. Cathodes (40 cm² projected surface area) were made from a mixture of activated carbon, carbon black, and a PVDF binder (8.8 mg/cm², 30:3:10). Two layers of a textile cloth (46% cellulose, 54% polyester; 0.3 mm thick;) 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. 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. Here, the main function of the MFCs was to provide a partially treated feed to the AFMBR.

AFMBR

Domestic wastewater was collected from the sewage pit, 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. \ MFCs were connected and then operated in two separate parallel flow paths to provide a combined feed to the AFMBR.

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 flow rate 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. Current was calculated using Ohms 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. All COD samples were analyzed following standard methods using HACH COD analyzer kits. Soluble COD samples were filtered through syringe filters (0.45 μ m pore size, PVDF, 20 mm diameter.)

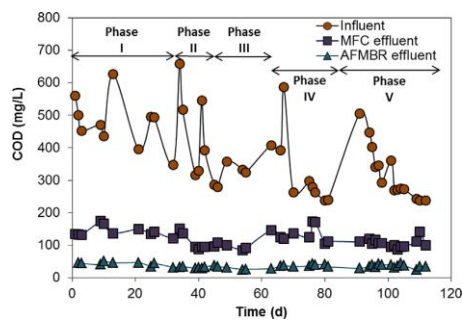


Fig. 1.

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.

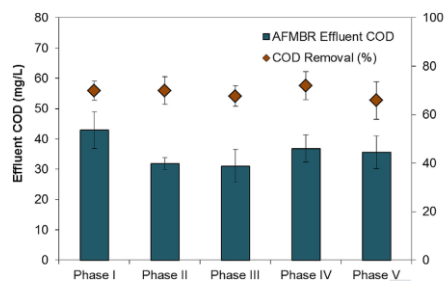


Fig. 2. Effluent COD level (mg/L) and COD removal efficiencies (%) in the AFMBR at

each Phase (with different HRTs and COD loading rates).

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). Gas production from the AFMBR was minimal (little observed in the gas bag), consistent with previous study AFMBR tests where we found little gas production, 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 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 similar COD removal efficiencies for the AFMBR at different HRTs ($65 \pm 10\%$ at 3.4 h; $64 \pm 9\%$ at 2.3 h), at organic loading rates (1.0_1.2 kg COD/m³-d) comparable to those used here. 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 1.21.4 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 MFCAFMBR was on average $76 \pm 4\%$ (effluent COD: 35 ± 6 mg/L), with $51 \pm 7\%$ of soluble COD in the wastewater (influent) removed by the MFCs, and $50 \pm 8\%$ removed by the AFMBR (Table 1). A higher COD effluent (35 ± 6 mg/L) was observed in this study compared to 16 ± 3 mg/L in a previous MFCAFMBR study. This difference might be due to the different amount of soluble COD in the AFMBR 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. There are also abiotic alternatives such as electrochemical, precipitation and coagulation processes. For example, nutrient removal using air cathode electro coagulation (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.

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 0.090.12 bar in Phase V (Fig. 3). The TMP variation until day 62 in this study was similar to that reported by R 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, 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 and periodically removed excess suspended solids by withdrawing reactor fluid from the recirculation line or reactor, and 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.

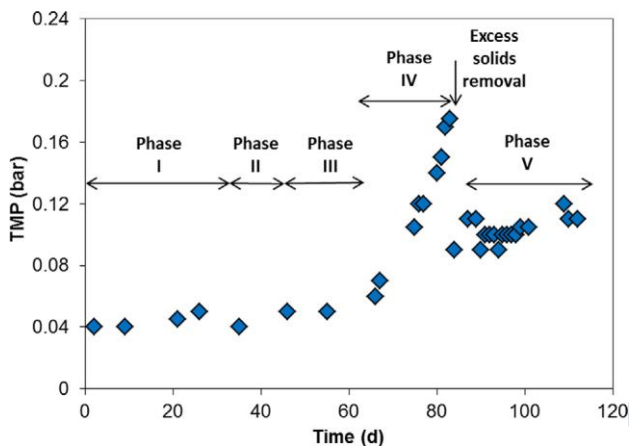


Fig. 3.

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.

3.4. MFC performance

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

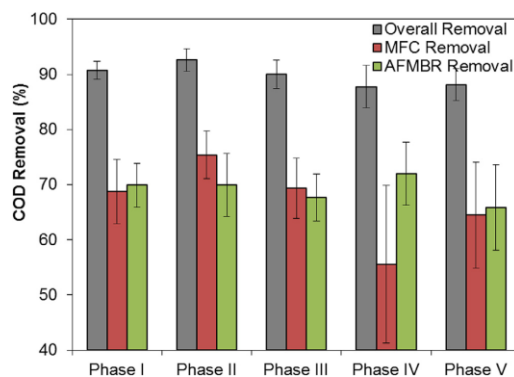


Fig. 4.

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

3.5. Energy production by MFCs

Total energy production by the MFCs was initially 0.012 ± 0.003 kWh/m³ (Phase I), which decreased to $0.003_0.004$ kWh/m³ from Phase II to Phase IV as current densities decreased. Total energy production was restored to 0.012 ± 0.005 kWh/m³ (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/m³) is about 40% likely due to the decrease in cathode performance over time. Assuming that total electrical energy requirement for pumping is 0.0186 kWh/ m³, 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. 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 ± 83 mg/L) compared to Phase I (469 ± 78 mg/L). 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 ± 6 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.

