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Continuous treatment of high strength wastewaters using air-cathode microbial fuel cells

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HIGHLIGHTS

- High strength wastewater was treated for 185 d in continuous flow mode MFCs.
- A maximum power density of 750 mW/m^2 was achieved after 12 d of operation.
- The power density decreased by 85% due to cathode fouling after 185 d.
- COD removal efficiency was improved by using higher current densities.
- COD removal rates were higher than those of low strength wastewaters.

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ABSTRACT

Treatment of low strength wastewaters using microbial fuel cells (MFCs) has been effective at hydraulic retention times (HRTs) similar to aerobic processes, but treatment of high strength wastewaters can require longer HRTs. The use of two air-cathode MFCs hydraulically connected in series was examined to continuously treat high strength swine wastewater (7–8 g/L of chemical oxygen demand) at an HRT of 16.7 h. The maximum power density of 750 ± 70 mW/m^2 was produced after 12 days of operation. However, power decreased by 85% after 185 d of operation due to serious cathode fouling. COD removal was improved by using a lower external resistance, and COD removal rates were substantially higher than those previously reported for a low strength wastewater. However, removal rates were inconsistent with first order kinetics as the calculated rate constant was an order of magnitude lower than rate constant for the low strength wastewater.

1. Introduction

Wastewaters generated in the US contain the equivalent of ~17 GW that if captured, could be used to power wastewater treatment plants that currently consume about 3% of electricity generated in the US (Logan and Rabaey, 2012; McCarty et al., 2011). Microbial fuel cells (MFCs) can be used to produce electricity from organic matter in wastewater by using exoelectrogenic bacteria on the anode coupled to oxygen reduction on an air-cathode (Rabaey and Verstraete, 2005; Rozendal et al., 2008; Wang et al., 2015). Power has been successfully produced using MFCs with many different types of wastewater (Pant et al., 2009). However, the focus of many of these studies has been power production, and not effective treatment in terms of overall chemical oxygen demand (COD) removal at hydraulic retention times (HRTs) that could make MFC treatment comparable to other technologies.

In order to obtain useful power production from an MFC at reasonable HRTs, they should be operated to reduce COD levels to 0.1–0.2 g/L, as power densities are greatly reduced at lower CODs (Zhang et al., 2015). For example, 65–69% COD removal (0.15 g/L in effluent) was obtained in a continuous flow MFC with a flat plate design, with 100–130 mW/m^2 maximum power densities at an HRT of 8.8 h with domestic wastewater (Kim et al., 2015). In a tubular MFC used for wastewater treatment, COD was reduced to 0.08–0.10 g/L but only 10–50 mW/m^2 was produced at an HRT of 11 h (Zhang et al., 2013). COD removal in the same reactor to an average of 0.03 g/L resulted in only 6–9 mW/m^2 at an HRT of 12 h (Ge et al., 2015). For low strength wastewaters, this limitation on the final COD suggests that MFCs should be operated to reduce

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the COD by 60–80% (0.5 g/L to 0.1–0.2 g/L). The residual COD in the wastewater can then be successfully removed to levels needed for wastewater discharge by using a second process, such as an anaerobic fluidized membrane bioreactor (AFMBR) that uses little electrical energy for treatment (Kim et al., 2016; Ren et al., 2014). For example, the effluent COD from a combined MFC-AFMBR system ranged from 16 to 36 mg/L, which would meet discharge standards in the US assuming a 1:2 ratio of BOD_5:COD (~10 h combined HRT) (Hays et al., 2011). In addition, MFCs need to have a high specific surface area (m^2/m^3, air cathode surface area per volume of reactor) to achieve a high power density based on both area and volume (Logan et al., 2015).

The use of high strength wastewaters (>1 g/L COD) in MFCs provides an opportunity for converting a greater percentage of the organic matter in a wastewater into electricity. Power densities of MFCs can be improved by using higher strength wastewaters, such as brewery or animal wastewaters. A maximum power density of 205 mW/m^2 was achieved using a cubic MFC with brewery wastewater (Feng et al., 2008), which was 1.4× higher than typical values using this type of reactor with lower strength domestic wastewater (Liu and Logan, 2004). Min et al. (2005) produced 1.8× greater maximum power densities with an animal wastewater than those obtained with a domestic wastewater. However, treatment times needed for high strength wastewaters can be longer than those for low strength wastewaters to achieve a reasonable COD removal efficiency. Several studies using high strength wastewaters in MFCs reported 66–88% COD removal, but HRTs of 19 h to 114 h were required (Kaewkannetra et al., 2011; Kim et al., 2014; Zhuang et al., 2012). Successful treatment of wastewaters using MFCs will also require stable power production and effective treatment over time. With acetate solutions, power declined by 40% over 12 months with activated carbon cathodes, and 22% over 16 months with carbon black mixed activated carbon cathodes (Zhang et al., 2011, 2014). However, power was restored to nearly the original levels by using a simple acid cleaning of the cathode (>85%). Cathode performance over time has not been well addressed in studies using high strength wastewaters.

Swine wastewater was examined here, as a typical high strength wastewater, for treatment in air-cathode MFCs. Two MFCs were connected hydraulically in series to separately study reactor performance at the influent COD of ~8 g/L, and at a lower COD following treatment in the first reactor. The performance of these MFCs was examined based on voltage generation over a period of approximately half a year (185 d), with maximum power densities assessed using polarization data after 12 d and 185 d of operation. COD removal was separately examined for both the upstream and downstream MFCs. COD removal in MFCs has been shown to have good agreement with first order kinetics using a low strength domestic wastewater (0.22 g-COD/L) or acetate solutions (0.26–0.84 g-COD/L) (Zhang et al., 2015). For example, a rate constant of $k = 0.18\ h^{-1}$ was achieved with filtered domestic wastewater, which was similar to that obtained in other MFC studies treating domestic wastewater under continuous flow mode operation (Ge et al., 2015; Kim et al., 2015). However, it is not known if such rate constants can be extended to wastewaters with much higher CODs. Therefore, to better understand COD removal in MFCs treating higher strength wastewaters, COD removal was examined on the basis of COD removal rate (g/L-d) and a first order rate constant derived from influent and effluent CODs. The removal rate is expected to increase with COD, but a lower rate constant at higher CODs would indicate that such removal was not in direct proportion to the increased COD. The impact of current on COD removal was examined by using different external loads by varying the external resistance (5 or 500 Ω).

2. Materials and methods

2.1. Swine wastewater

Swine wastewater was collected from the Penn State Swine Center located in Pennsylvania State University. The swine wastewater was initially screened through a stainless steel mesh (50 × 50 mesh, 0.23 mm wire) to remove large particles. The screened swine wastewater had 19 ± 2 g/L COD with conductivity of 17.8 ± 0.5 mS/cm. Without further modification or treatment, the screened swine wastewater was used as inoculum for bacteria acclimation of in MFCs.

For the MFC operation in continuous flow mode, the COD of the screened swine wastewater was adjusted to a range of 7–8 g/L by settling and dilution. Each new wastewater sample was first stored in a plastic container (at 4 °C) for at least one day without shaking, and solids that settled to the bottom of the container were discarded. If the COD after this step exceeded 8 g/L, the wastewater was diluted using deionized water to keep the COD in the desired range of 7–8 g/L, and then stored at 4 °C. The influent wastewater over the 185 d of operation had an average influent COD of 7.6 ± 0.7 g/L, total suspended solids (TSS) of 5.4 ± 0.7 g/L and a conductivity of 8.1 ± 1.7 mS/cm. Since this study focused on removal of organic compounds using air-cathode MFCs, the removals of inorganic chemical species such as ammonia nitrogen were not considered during operation.

2.2. MFC construction and operation

Two identical air-cathode MFC reactors (working volume of 100 mL) were constructed as previously described (Ahn and Logan, 2013). Anodes were graphite fiber brushes (Mill-Rose, Mentor, OH) with a wound titanium wire core (25 mm diameter by 35 mm long). Before use, the anodes were heat treated at 450 °C for 30 min, and the brushes were trimmed along their length to make a half cylinder shape. Three anode electrodes were placed directly next to an air-cathode (0.5 cm distance from the cathode) with the flat side placed on the separator. Cathodes (40 cm^2, projected surface area) were prepared by a phase inversion technique using a poly(vinylidenefluoride, PVDF) binder, made from a mixture of activated carbon (AC, Norit SX plus, Norit Americas Inc., TX), 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 separator (46% cellulose, 54% polyester; 0.3 mm thick; Amplitude Prozorb, Contec Inc.) were placed on the cathode surface (solution side) as separators.

The two swine wastewater fed MFCs were initially filled with screened swine wastewater as both fuel and inoculum, and the anode and cathode were connected with a 1000 Ω resistor for each MFC. After 30 d of acclimation, two MFCs were hydraulically connected in series, and the swine wastewater adjusted to a COD of 7–8 g/L was pumped into the first MFC reactor using peristaltic pump (Model No. 7523-90, Masterflex, Vernon Hills, IL). The first MFC was labeled the upstream (U) reactor and the second one the downstream (D) reactor (Fig. S1). The MFCs were initially operated in continuous flow mode for ~3 months at a flowrate of 0.05 mL/min (theoretical HRT of 2.8 d), and an external resistance $R_{ext} = 50\ \Omega$. After 74 days of operation, the separators were replaced, the cathode surfaces (solution side) were cleaned using DI water. The flowrate was then increased after 74 d to 0.2 mL/min, for a total HRT of 16.7 h. Solids that had settled in the reactor were removed on days 74, 121 and 142. Current, COD removal, and TSS removal by MFCs were measured (at least four times) at different external resistances (500 Ω, 50 Ω, and 5 Ω) for days 74–185, with values reported based on the average and standard deviation.