



Research article

A comparison of simultaneous organic carbon and nitrogen removal in microbial fuel cells and microbial electrolysis cells



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ABSTRACT

This study demonstrates simultaneous carbon and nitrogen removal in laboratory-scale continuous flow microbial fuel cell (MFC) and microbial electrolysis cell (MEC) and provides side-by-side comparison of these bioelectrochemical systems. The maximum organic carbon removal rates in MFC and MEC tests were similar at $5.1 \text{ g L}^{-1} \text{ d}^{-1}$ and $4.16 \text{ g L}^{-1} \text{ d}^{-1}$, respectively, with a near 100% carbon removal efficiency at an organic load of $3.3 \text{ g L}^{-1} \text{ d}^{-1}$. An ammonium removal efficiency of 30–55% with near-zero nitrite and nitrate concentrations was observed in the MFC operated at an optimal external resistance, while open-circuit MFC operation resulted in a reduced carbon and ammonium removal of 53% and 21%, respectively. In the MEC ammonium removal was limited to 7–12% under anaerobic conditions, while micro-aerobic conditions increased the removal efficiency to 31%. Also, at zero applied voltage both carbon and ammonium removal efficiencies were reduced to 42% and 4%, respectively. Based on the observed performance under different operating conditions, it was concluded that simultaneous carbon and nitrogen removal was facilitated by concurrent anaerobic and aerobic biotransformation pathways at the anode and cathode, which balanced bioelectrochemical nitrification and denitrification reactions.

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1. Introduction

Activated sludge wastewater treatment (WWT) process is one of the most commonly used biological technologies for removal of organic materials and nutrients from wastewater. In spite of advantages such as high efficiency in removing organic materials, nutrients, and pathogens from wastewater, the activated sludge process features significant energy consumption for aeration to provide adequate supply of dissolved oxygen to aerobic microorganisms. In addition, the disposal of secondary sludge produced during the wastewater treatment further adds to the treatment costs (Du et al., 2007). Extensive efforts are underway to develop technologies that reduce energy consumption for wastewater treatment or recover energy from wastewater.

In this regard, a particularly promising approach is the application of bioelectrochemical systems (BES) such as microbial fuel cells (MFCs) and microbial electrolysis cells (MECs) for wastewater treatment (Oh et al., 2010; Rozendal et al., 2008). BESs are based on the ability of anodophilic (electricigenic) microorganisms to oxidize

organic substrates while utilizing an electrically conductive material (anode) as a terminal electron acceptor (Lovley, 2008). BES applications for wastewater treatment have been widely tested, with studies primarily focussing on chemical oxygen demand (COD) removal and energy production (Logan, 2010; Lovley, 2008; Rozendal et al., 2008). However, besides organics wastewater contains nitrogen based compounds such as ammonium, nitrite, and nitrate that need to be removed before discharge. Traditionally this task is accomplished in a multi-step biotreatment process, which includes COD removal, nitrification, and denitrification phases. Simultaneous degradation of CODs, nitrification, and denitrification in a biofilm-based process has been previously demonstrated (e.g. Kotlar et al., 1996). However, unlike microbial electrochemical technologies, this process does not result in net energy production. Simultaneous carbon and nitrogen removal in MFCs and MECs is complicated by the impact of nitrogen on the design, performance, microbial kinetics, and operational conditions of BESs (Kelly and He, 2014). Nevertheless, successful adaptation of BESs for simultaneous carbon and nitrogen removal would reduce electricity consumption for aeration and offset energy use for wastewater treatment.

Ammonium as well as nitrite and nitrate removal has been demonstrated in MFCs (He et al., 2009; Virdis et al., 2010; Yan and

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Regan, 2013; Zhu et al., 2013). In MECs limited ammonium removal was observed with micro-aerobic conditions increasing the nitrification rate (Villano et al., 2013; Zhan et al., 2012). However, activity of the anodophilic and denitrifying microorganisms might be negatively affected by the presence of oxygen. Accordingly, to combine COD and ammonium removal the anodic and cathodic compartment configuration of MFCs and MECs has been extensively modified resulting in a multi-stage process design. This is contrary to real world applications, which might require simpler reactor configuration (Kelly and He, 2014). In addition, to confer a distinct advantage of BES-based WWT over conventional aerobic wastewater treatment, it is preferable to avoid active aeration and consider passive oxygen supply. Although both MFCs and MECs were shown to be capable of simultaneous carbon and nitrogen removal, previous studies have generally focused on carbon and nitrogen removal in only one system (MFC or MEC). The difference in reactor design, operating conditions and auxiliary parameters makes it difficult to compare the organic carbon and nitrogen removal potential of such systems resulting in a lack of comparative data.

This study reports the development and operation of continuous flow MFC and MEC for simultaneous carbon and nitrogen removal in a single-stage design instead of a multi-stage design. For a comparative analysis, the MFC and MEC were constructed using similar design and were simultaneously operated under similar carbon and nitrogen loads. To simplify reactor design both systems lacked proton exchange membrane and consisted of a single compartment housing anode and cathode electrodes. In both systems experiments with synthetic wastewater containing acetate and ammonium were followed by reactor operation on municipal wastewater.

2. Materials and methods

2.1. Analytical methods and media composition

Acetate concentration was measured using an Agilent 6890 gas chromatograph (Agilent Technologies Inc, Santa Clara, CA, USA) equipped with a flame ionization detector. The COD and BOD₅ concentrations were determined following procedures outlined in the Standard Methods (APHA et al., 1995). Ammonium concentrations were analysed using a Thermo HPLC system (Thermo scientific, MA, USA) equipped with a Waters conductivity detector (Mississauga, ON, Canada) and a Hamilton PRP-X200 (250 × 4.6 mm) cation exchange column using 30% methanol in 4 mM nitric acid at a flow rate of 0.75 mL min⁻¹. Nitrate and nitrite were quantified using Dionex ICS-3000 HPLC equipped with an ionic conductivity detector (Dionex Corporation, Sunnyvale, California, USA). The separation was carried out on a DIONEX IonPac AS15 column (2 × 250 mm) at 35 °C. The mobile phase was a gradient of KOH at a flow rate of 0.3 mL min⁻¹. The off-gas flow rate was measured using the MilliGascounter™ (Ritter Apparatus, Bochum, Germany). The off-gas composition was measured using an HP 6890 gas chromatograph (Hewlett Packard, Palo Alto, CA, USA) equipped with a thermal conductivity detector and a 5 m × 2.1 mm Carboxen-1000 column (Supelco, Bellefonte, PA, USA) with argon as the carrier gas. Further details of the analytical methods are provided in Tartakovsky et al. (2008). An average of at least three analytical measurements was used for each value reported in Tables 2 and 3.

A concentrated stock solution of acetate and nutrients contained (in g L⁻¹): KCl (74), K₂HPO₄(32), KH₂PO₄(20), and yeast extract (0.8). To obtain different influent concentrations of acetate and ammonium during MFC and MEC operation phases outlined in Tables 1A and B the stock solution was supplemented with the

Table 1A
Phases of MFC operation.

Phase	MFC		
	Influent COD mg L ⁻¹	Influent NH ₄ ⁺ mg L ⁻¹	Duration day
1	1067	200	36
2	2067	200	7
3	0	200	8
4	1067	100	16
5 ^a	1067	100	7
6 ^a	0	100	8
7 ^a	1067	100	32
8 ^b	1067	100	19
9 ^c	120–140	30–40	14

^a MFC in OCP mode.

^b Low conductivity influent.

^c Municipal wastewater.

Table 1B
Phases of MEC operation.

Phase	MEC			
	Influent COD mg L ⁻¹	Influent NH ₄ ⁺ mg L ⁻¹	Influent H ₂ O ₂ mg d ⁻¹	Duration day
1	1067	200	0	36
2	2134	200	0	7
3	0	200	0	8
4	1067	100	0	9
5	1067	100	5	9
6	1067	100	10	8
7 ^a	1067	100	0	42
8 ^b	120–140	30–40	0	15

^a No applied voltage.

^b Municipal wastewater.

desired amounts of sodium acetate and ammonium chloride. The trace metal stock solution was prepared according to Tartakovsky et al. (2008). All chemicals were of analytical grade. The stock solutions were filter-sterilized with a 0.22 μm filter (Fisher Scientific, Ottawa, ON, Canada) and maintained at 4 °C until use. The acetate-based feed had a conductivity of 15–16 mS cm⁻¹. For phase 8 of MFC operation at low conductivity conditions (Table 1A), the concentration of KCl was reduced to a half, while the concentration of K₂HPO₄ and KH₂PO₄ were reduced to a fourth of the initial concentration in the stock solution. Accordingly, the feed solution had a conductivity of 3–5 mS cm⁻¹. For MFC (phase 9) and MEC (phase 8) operation on real wastewater, the reactors were fed with municipal wastewater collected from the Ste-Catherine (QC, Canada) wastewater treatment plant. To avoid carbon felt blockage, solids were decanted resulting in a total COD concentration of 120–140 mg L⁻¹ and NH₄⁺ concentration of 30–40 mg L⁻¹. Wastewater conductivity was 1.5–3.0 mS cm⁻¹.

2.2. MFC and MEC design, operation, and characterisation

The continuous flow MFC and MEC used in this study were constructed from custom-made nylon plates. The single chamber air cathode MFC shown in Fig. 1A consisted of a 50 mL anodic compartment that contained a single carbon felt anode with dimensions of 10 cm × 5 cm × 0.5 cm (SGL Canada, Kitchener, ON, Canada). Titanium rods with Ir-MMO coating (Magneto Special Anodes, B.V, Netherlands) inserted into the felt were used as current collectors. Commercially available gas diffusion cathodes containing MnO₂ – based catalyst (Electric Fuel Ltd., Bet Shemesh, Israel) were used. The cathode measuring 10 cm × 5 cm in dimension was placed next to the anode. The cathode was

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