



# Integration of microbial fuel cell techniques into activated sludge wastewater treatment processes to improve nitrogen removal and reduce sludge production



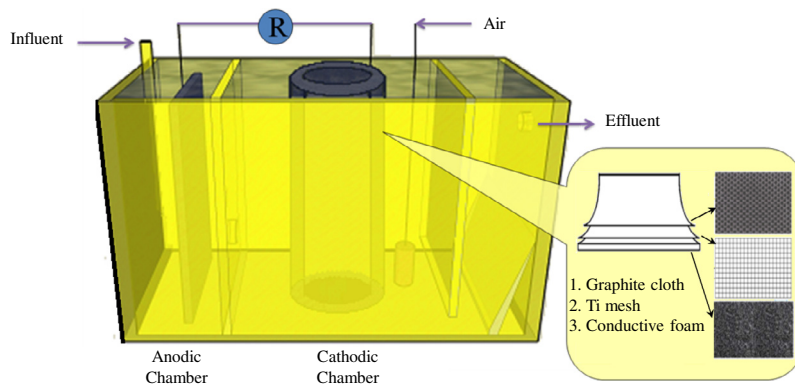
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## HIGHLIGHTS

- Microbial fuel cell technique was incorporated into the MLE and MBR process.
- No changes in sludge activity, settling property, and nitrifying community structure.
- Effluent COD concentration of the MLE–MFC was lower than that of MLE.
- MLE–MFC and MBR–MFC increased  $\text{NO}_3^-$ -N removal by 31% and 20% respectively.
- The MFC integrated systems reduced sludge production by 6–11%.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Bioelectrochemical systems are emerging for wastewater treatment, yet little is known about how well they can be integrated with current wastewater treatment processes. In this bench-scale study, the microbial fuel cell (MFC) technique was incorporated into the Modified Ludzack–Ettinger (MLE) process (phase I) and later with the membrane bioreactor (MBR) process (phase II) to evaluate the performance of MFC assisted wastewater treatment systems (i.e., MLE–MFC and MBR–MFC). There was no significant difference in the effluent  $\text{NH}_4^+$ -N concentration between the systems integrating MFC and the open circuit controls. The average effluent COD concentration was significantly lower in the MLE–MFC, but it did not change much in the MBR–MFC because of the already low COD concentrations in MBR operation. The MLE–MFC and MBR–MFC systems increased the  $\text{NO}_3^-$ -N removal efficiencies by 31% ( $\pm 12\%$ ) and 20% ( $\pm 12\%$ ), respectively, and reduced sludge production by 11% and 6%, respectively, while generating an average voltage of 0.13 ( $\pm 0.03$ ) V in both systems. Analysis of the bacterial specific oxygen uptake rate, the sludge volume index, and ammonia-oxidizing bacterial population (dominated by *Nitrosomonas* through terminal restriction fragment length polymorphism analysis) indicated that there was no significant difference in sludge activity, settling property, and nitrifying community structure between the MFC assisted systems and the open circuit controls. The results suggest that the wastewater treatment systems could achieve higher effluent water quality and lower sludge production if it is integrated well with MFC techniques.

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## 1. Introduction

Bioelectrochemical systems (BES) are emerging for wastewater treatment and energy production (Rozendal et al., 2008). Microbial fuel cells (MFC) are the archetype of BES and have been demonstrated successful in wastewater treatment (Ahn and Logan, 2013; Kim et al., 2013) although the power output is not high enough for the electricity production process (Logan and Rabaey, 2012). Research on MFC has mainly been focused on increasing power densities (Logan, 2009) although the ability to scale-up the process remains to be determined (Dewan et al., 2008). Numerous designs have been proposed to optimize MFC operations. There are two chamber systems, single chamber systems, up-flow mode systems, and stacked MFC systems (Du et al., 2007). The similarity in all these designs points to the presence of an anaerobic zone and an aerobic zone. Indeed, enhanced biological nutrient removal processes such as the Modified Ludzack–Ettinger (MLE) process, rely on the use of alternating aerobic and anoxic conditions (Yu et al., 2011). Furthermore, membrane bioreactor (MBR) technology can be used in these nutrient removal wastewater treatment plants (WWTPs) to produce higher quality effluent (Ge et al., 2013). By integrating MFC technology with these common wastewater treatment processes, many benefits could be gained including electricity production, lower sludge production (Logan et al., 2006) and therefore reduced sludge handling costs.

Taking MBR activated sludge process as an example, it was demonstrated that a small electric field ( $0.036\text{--}0.073\text{ V cm}^{-1}$ ) mitigated fouling of conductive membranes in the MBR (Liu et al., 2012b) but required novel materials for the membrane construction. Wang et al. (2011) and Ge et al. (2013) used a stainless steel mesh of  $40\text{ }\mu\text{m}$  pore size and hollow fiber membrane of  $0.02\text{ }\mu\text{m}$  pore size membrane respectively as the cathode for sludge filtration and higher effluent water quality. However these designs had to bear the cost associated with the use of cation exchange membrane (CEM), had limited nutrient removal due to the reactor design, and furthermore, the effect of bio-generated electricity in mitigating membrane fouling was not studied. A slightly modified design (Kim et al., 2013) used an ultrafiltration membrane with a molecular weight cutoff of  $1\text{ kDa}$  instead of a CEM. Although the cost associated with the use of CEM was avoided, the design called for a positive pressure system which posed scale-up issues and no nutrients were removed. While all the aforementioned demonstrated integrating MBR with MFC processes at lab scale they fall short of immediate potential for successful scale-up or integration with existing WWTPs.

One more important benefit in the MFC systems to be explored is the enhanced nitrogen removal from wastewater (Virdis et al., 2010; Yu et al., 2011; Zhang and Angelidaki, 2012). In one study, MFC with a reactor volume of  $0.336\text{ L}$  was coupled with an external aerobic nitrification reactor to convert ammonia in the feed solution to nitrate before it was circulated through the MFC cathodic chamber where bacteria on biocathode play a role for nitrogen removal (Virdis et al., 2008). However, the set-up of an additional external nitrifying bioreactor makes it difficult to integrate with existing wastewater treatment systems. Furthermore, the cost associated with proton exchange membrane (PEM) and high recirculation flow to support *in-situ* nitrification (Clauwaert et al., 2007; Virdis et al., 2010) can often be prohibitive for wastewater treatment. Similarly, the complexity of the dual-cathode MFCs would not allow to easily integrate MFC techniques into traditional wastewater treatment operations, although such a system was capable of generating electricity and removing nitrogen more efficiently (Zhang and He, 2012).

The objective of this study was to evaluate the performance of MFC assisted wastewater treatment systems, to improve

wastewater treatment and sludge management practices. Unlike the previous systems where small standalone MFC modules were used in sequence (Kim et al., 2013) or immersed in bioreactors (Zhang and Angelidaki, 2012; Ahn and Logan, 2013), we operated the MFC assisted activated sludge wastewater treatment systems ( $7.2\text{ L}$  each) in consistence with the use of alternating aerobic and anoxic conditions in the MLE or MBR process. These processes were selected because MLE is one of the most commonly used processes for enhanced nitrogen removal (USEPA, 2000) and MBR becomes increasingly used for higher effluent water quality and water reuse applications.

## 2. Materials and methods

### 2.1. Bioreactor design and operation

Two identical bioreactors were constructed with glass, each having a working volume of  $7.2\text{ L}$ . A schematic of the bioreactors and the flow pattern is shown in Fig. 1. The bioreactors were divided into three zones: anaerobic/anoxic chamber (far left), aerobic chamber (middle) and an internal settling chamber (far right) separated by plastic baffles. The effective volumes of the resulting chambers were  $2.1$ ,  $3.3$ , and  $1.8\text{ L}$ , respectively. An array of three horizontal openings, each  $0.5\text{ cm}$  in diameter was made in the baffle wall separating the anaerobic and aerobic chamber.

An MFC module was integrated into one of the bioreactors by placing the anode in the anaerobic/anoxic chamber and the cathode in the aerobic chamber (Fig. 1). The other bioreactor served as the control, which included a similar electrode module, but operated on open circuit. The anode was made of  $120\text{ cm}^2$  ( $8\text{ cm} \times 15\text{ cm}$ ) of commercially available graphite cloth (Plantraco, Saskatoon, SK, Canada). The cloth was in plain weave (checkerboard pattern) and had a specific weight of  $0.08\text{ kg m}^{-3}$  with a resistivity of  $1 \times 10^{-5}\text{ }\Omega\text{ m}$ . The cathode was designed to be a hollow cylinder so that it could accommodate the membrane module during the MBR study (Fig. 1). The hollow cylindrical core was made out of a rigid polypropylene mesh tube (catalog number RN1900),  $15\text{ cm}$  in length,  $6\text{ cm}$  in diameter and an open area of  $35\%$  (Industrial netting, Minneapolis, MN, USA). The rigid hollow tube was covered with a single layer of the following materials in the following order:

- (1) Carbon fiber/graphite cloth, which served as a primary electrode material.
- (2) Titanium mesh of  $0.28\text{ mm}$  diameter wire in a  $12 \times 12$  wires per  $2.54\text{ cm}$  plain weave (Unique wire weaving, Hillside, NJ, USA) for electrical connection, and
- (3) Conductive low-density black polyurethane foam (All-spec industries, Wilmington, NC, USA), which served as a primary filter to support biofilm growth on its large surface area and control membrane fouling in the MBR study.

To construct the MFC module, the carbon fiber/graphite cloth and the titanium mesh were soaked in  $50\%$  ethanol for about  $1\text{ h}$  and rinsed with tap water to wash away any impurities before use. The anode and cathode were connected together with a  $1000\text{ }\Omega$  resistor and spaced approximately  $5\text{ cm}$  apart from each side of the baffle plate. PEM was not used due to cost constraints.

The bioreactors were inoculated with return activated sludge from the Columbia Wastewater Treatment Plant (Columbia, MO, USA). The reactors were fed with synthetic wastewater that was mainly composed of nonfat milk powder with a COD of approximately  $500\text{ mg L}^{-1}$  (Liu et al., 2012b). Other components of the synthetic wastewater included  $51.7\text{ mg L}^{-1}$  total nitrogen (TN),

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