



A fluidized bed membrane bioelectrochemical reactor for energy-efficient wastewater treatment

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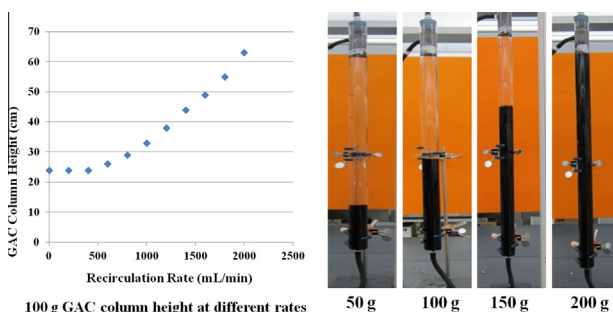
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HIGHLIGHTS

- Fluidized granular activated carbon (GAC) helps control membrane fouling in a MBER.
- Fluidized GAC plays a minor role in direct contribution to electricity generation.
- Fluidized bed MBER can be coupled to a microbial fuel cell for improved treatment.
- The couple system could achieve theoretically energy neutral.

GRAPHICAL ABSTRACT



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ABSTRACT

A fluidized bed membrane bioelectrochemical reactor (MBER) was investigated using fluidized granular activated carbon (GAC) as a mean of membrane fouling control. During the 150-day operation, the MBER generated electricity with contaminant removal from either synthetic solution or actual wastewater, as a standalone or a coupled system. It was found that fluidized GAC could significantly reduce transmembrane pressure (TMP), although its function as a part of the anode electrode was minor. When the MBER was linked to a regular microbial fuel cell (MFC) for treating a wastewater from a cheese factory, the MFC acted as a major process for energy recovery and contaminant removal, and the coupled system removed more than 90% of chemical oxygen demand and >80% of suspended solids. The analysis showed that the ratio of energy recovery and consumption was slightly larger than one, indicating that the coupled system could be theoretically energy neutral.

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1. Introduction

Sustainable wastewater treatment is of great importance to maintain a sustainable societal development, and its key features include high quality of treated effluent and energy-efficient treatment process. Those features can be realized separately using technologies such as membrane bioreactors (MBR) and microbial fuel cells (MFCs). MBR technology has been applied to treat both

municipal and industrial wastewaters. It has several advantages over conventional activated sludge system (Judd, 2008); however excessive energy consumption due to aeration and antifouling control is still a hurdle for its application especially in some energy-shortage areas. MFC technology is an emerging concept and has been intensively studied as an alternative method for energy-efficient wastewater treatment (Wang and Ren, 2013). Comparing with conventional activated sludge technology, MFCs have less or no demand for aeration and produce much less sludge due to anaerobic treatment (Rabaey and Verstraete, 2005). Research has demonstrated that MFC treatment of domestic wastewater could be energy-neutral (Zhang et al., 2013).

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Integration of MFCs with a membrane filtration process such as MBRs may provide an ideal solution to achieve high-quality effluent, with less energy requirement (than aerobic treatment systems). An early effort used the biofilm formed on the stainless steel as filter materials to achieve low effluent turbidity and high removal of both organic matter and ammonium nitrogen (Wang et al., 2011, 2012). The use of micro/ultra filtration membranes as filtration media in MFCs was reported in a membrane bioelectrochemical reactor (MBER), in which the commercially available hollow fiber membranes were installed in the anodic chamber of a tubular MFC (Ge et al., 2013b). This MBER system effectively treated both synthetic and domestic wastewater but membrane fouling was a serious issue. To facilitate the application of proper fouling control and minimize its effects on the anode microbial activity, hollow-fiber membranes were installed in the cathode compartment of an MFC with either an aerobic or anoxic cathode (Li et al., 2014). In addition to internally installed membrane, MFCs were also directly linked to an MBR (Malaeb et al., 2013).

In a treatment system containing membrane, fouling/scaling is always a great challenge. A new method for fouling control was developed by using fluidized granular activated carbon (GAC) in an anaerobic membrane bioreactor (AnMBR) (Kim et al., 2011). This method required little physical or chemical cleaning of membrane module during 120-day operation, and it also had energy benefits for fouling control comparing to conventional MBR. This fluidized AnMBR has been advanced to pilot test that achieved satisfactory performance (Shin et al., 2014). A similar fluidized AnMBR was linked to a single-chamber MFC as a post-treatment for improving effluent quality (Ren et al., 2014). The concept of fluidized particle bed was also applied to prevent inorganic scaling deposit on the surface of cathode electrode in a microbial electrolysis cell (MEC) (Cusick et al., 2014).

Intrigued by the fluidized AnMBR concept, a fluidized bed membrane bioelectrochemical reactor (MBER) was developed here for energy-efficient wastewater treatment. This MBER aimed to take advantage of fluidized GAC as both fouling control media and partial anode electrode. The objectives of this study were: (1) to examine the feasibility of electricity generation and wastewater treatment in this fluidized bed MBER; (2) to investigate membrane fouling affected by the operating conditions; and (3) to formulate a treatment system by linking the MBER to an MFC for treating actual wastewater.

2. Methods

2.1. Reactor construction

2.1.1. MBER setup

The MBER was constructed as a tubular reactor (45 cm long and 5 cm in diameter) made of cation exchange membrane (CEM-Ultrex CMI 7000, Membrane International, Inc., Glen Rock, NJ, USA) (Fig. 1A). The main body of the anode electrode was a piece of carbon cloth supported by stainless steel mesh, which was installed inside the membrane tube (along the interior wall). Ten 38-cm PVDF hollow fiber membranes (15,000 Dalton, Litree Purifying Technology Co., China) were installed inside the membrane tube, which was then filled with 230 g of 8×30 mesh GAC (Calgon Carbon Corp., Pittsburgh, PA, USA), resulting in an anode liquid volume of 700 mL. The hollow fiber membranes had a pore size of $0.02 \mu\text{m}$ and the total membrane surface area was 0.021 m^2 . Before use, carbon cloth was soaked in acetone solution overnight and heated for 30 min at 450°C (Wang et al., 2009). The cathode electrode consisted of one layer of carbon cloth (Zoltek Corporation, St. Louis, MO, USA) coated with Pt/C powder (10%, Etek, Somerest, NJ, USA) with a loading rate $0.05 \text{ mg Pt cm}^{-2}$. The cathode electrode

wrapped the membrane tube and was exposed in the air for passive oxygen supply as described in the previous studies (Zhang et al., 2010). The anode and cathode electrodes were connected by using titanium wires to an external resistor of 48 ohm (which was determined by polarization tests for high power output).

2.1.2. MFC construction

A tubular MFC was constructed with a CEM tube (45 cm long and 5 cm diameter), which contained a one-meter long carbon brush folded as an anode electrode. The anode liquid volume was about 1000 mL. The cathode electrode was a piece of carbon cloth treated and coated with Pt catalyst similarly to that of the MBER. The electrodes were connected to an external circuit using titanium wires.

2.2. Operation conditions

2.2.1. MBER operation

The MBER anode was inoculated with anaerobic digester sludge from a wastewater treatment facility (South Shore, Oak Creek, WI, USA) and was operated at room temperature of $\sim 20^\circ\text{C}$. The synthetic anode solution contained (per L of tap water): sodium acetate 0.5 g; NH_4Cl 0.15 g; NaCl 0.5 g; MgSO_4 0.015 g; CaCl_2 0.02 g; KH_2PO_4 0.53 g; K_2HPO_4 1.07 g and 1 mL trace element (He et al., 2006). The anolyte was recirculated at 800 mL min^{-1} unless elsewhere stated. This rate was determined according to a test that examined the height of the fluidized GAC column affected by recirculation rate, and found that 800 mL min^{-1} could fully fluidize the GAC in the MBER. The hollow fiber membranes were operated under an intermittent mode that extracted water for 4 min and then relaxed for 1 min. Tap water was used as a catholyte to rinse the cathode electrode from top to bottom and additional tap water was added to compensate for evaporation. The effects of the anolyte recirculation and hydraulic retention time (HRT) were examined. Organic loading rates varied when adjusting HRT: $0.40 \text{ kg COD m}^{-3} \text{ d}^{-1}$ at 24 h, $0.80 \text{ kg COD m}^{-3} \text{ d}^{-1}$ at 12 h, and $1.31 \text{ kg COD m}^{-3} \text{ d}^{-1}$ at 8 h. The anode of the MBER was acclimated with the seed sludge for about one week with varying the external resistance from 2000 to 10Ω . After 100-day operation, the tubing for the anode feeding and the influent port of the CEM tube were clogged by GAC due to operating problems, and subsequently a new CEM tube was constructed (while other materials remained same) and used to the end of the study.

2.2.2. Integrated MFC + MBER operation

After four-month operation, the fluidized MBER was linked to an MFC to treat an industrial wastewater (Schreiber Foods, Inc., WI, USA). The wastewater collected from the effluent of DAF (dissolved air flotation) unit was first fed into the MFC anode and then the MFC effluent was supplied to the MBER (Fig. 1B). The coupled system was operated under an overall HRT 19.6 h (11.6 h in the MFC and 8 h in the MBER). The organic loading rate was about $1.00 \text{ kg COD m}^{-3} \text{ d}^{-1}$. The anolyte recirculation rates were 200 and 600 mL min^{-1} for the MFC and the MBER, respectively.

2.3. Measurement and analysis

The voltage was recorded every 3 min by a digital multimeter (2700, Keithley Instruments, Cleveland, OH). The pH was measured using a benchtop pH meter (Oakton Instruments, Vernon Hills, IL, USA). The concentration of Chemical Oxygen Demand (COD), ammonium, nitrite and nitrate concentration were measured according to the manufacture's procedures (Hach DR/890, Hach Company, Loveland, CO, USA). The trans-membrane pressure (TMP) was manually recorded 3 times daily and the average value was reported in this study. The turbidity was measured using a

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