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Cathodic fluidized granular activated carbon assisted-membrane bioelectrochemical reactor for wastewater treatment

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ABSTRACT

A membrane bioelectrochemical reactor (MBER) is a system integrating membrane filtration into microbial fuel cells. To control membrane fouling, fluidized granular activated carbon (GAC) was applied to the cathodic compartment of an MBER, which was examined for contaminant removal and electricity generation with low or medium strength synthetic wastewater. The MBER was operated for more than 160 days and achieved nearly 100% removal of organic compounds, regardless of presence/absence of GAC. However, fluidized GAC alleviated the membrane fouling issue and maintained transmembrane pressure (TMP) between 10 and 15 kPa with 24 g of GAC. The presence of GAC also enhanced current generation from 200.3 to 256.0 A m^{-3}, because some GAC might have functioned as a part of the cathodic electrode through physical contact with the electrode during fluidization. A higher aeration intensity could benefit both membrane fouling control (via scouring effect) and electricity generation (via oxygen supply), but required a higher energy demand. The energy consumption of the MBER including pumping and aeration was estimated to be 0.38 kWh m^{-3} or 0.25 kWh $kgCOD^{-1}$, lower than that of conventional membrane bioreactors (MBRs). Those results encourage further investigation and development of the MBER technology to treat wastewater in an energy efficient way.

1. Introduction

Microbial fuel cell (MFC) technology offers a viable option for energy-efficient wastewater treatment with energy recovery [1,2]. In conventional MFCs, organics are biologically degraded by electrochemically-active bacteria (EAB), which are colonizing on an anodic electrode, and the generated electrons are migrating to a cathodic electrode via an external circuit [3,4]. Because of potential advantages in minimal aeration demand, less sludge production, and energy production, MFCs have been considered as an emerging approach for treating municipal or industrial waste streams [5–7]. In addition to organics removal and energy recovery, high-quality treated effluent could be achieved by integrating membrane separation with MFCs in either internal or external configurations [8]. The feasibility studies have been conducted by using either stainless steel or commercial hollow fiber polymeric membranes as an effective separating method for treating the MFC effluent, forming new systems termed as membrane bioelectrochemical reactors (MBERs) [9–11].

Like conventional membrane bioreactors (MBRs), membrane fouling always poses a great challenge for MBER application and would result in high operational cost on fouling controls (e.g., off-line cleaning and periodic backwashing) [12]. To address the fouling issue, several strategies have been examined, such as modifying membrane surface property by coating polydopamine to enhance its hydrophobicity [13] and adjusting particle zeta-potential to increase electro-repulsion force [14]. Those modified membranes would need long-term examination with actual wastewater for fouling situation. Less fouling could be achieved by connecting a membrane filtration process via an external connection with MFCs [15,16]. For example, a bench-scale air cathode MFC was connected with an anaerobic fluidized membrane bioreactor (AFMBR) to achieve a good quality of the final effluent [15]. In another study, a tubular MFC coupled with a membrane module had been operated for more than 100 days and in the absence of proper fouling control, the TMP could increase to 30 kPa within only three days under the operation/relaxation mode [16]. Installing hollow fiber membranes in a cathodic compartment could be another way to minimize membrane fouling by taking advantage of organic removal in the anode and constant *in-situ* aeration in the cathode. It has been demonstrated that an MBER, which contained membrane bundle in its cathodic compartment, could treat both synthetic solution and cheese wastewater, but it was found

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that the TMP increased from 15 to 24 kPa within nine days with aeration as the only fouling control method [17]. Therefore, aeration alone still has limited effects on fouling control and intensive aeration (to improve its fouling control) will result in increased energy consumption.

Granular activated carbon (GAC) has been applied in water and wastewater treatment for its superior characteristics of absorption, filterability, regeneration ability and low cost. A fluidized GAC bed can be effective to mitigate membrane fouling issue via a physical contact between GAC and membrane surface. For example, fluidized GAC bed was incorporated in an anaerobic membrane bioreactors (AnMBR), resulting in little need for physical and chemical cleaning of membrane module during its 120-day operation [18]. Such integration also gains more energy benefits comparing to conventional MBRs. Likewise, fluidized bed GAC was also applied to the anodic compartment of a tubular MBER to address membrane fouling issue, and it could significantly slow down the increase in transmembrane pressure (TMP), although its function as a partial anodic electrode was insignificant [19]. Therefore, applying fluidized bed GAC seems to be a promising solution to control membrane fouling in MBERs.

Intrigued by the energy advantage of MBER and the effectiveness of fluidized bed GAC on membrane fouling control, we proposed a two chambered MBER with fluidized GAC in the cathodic compartment for minimizing membrane fouling issue. Such an arrangement was based on the facts that the cathodic membrane module could have less fouling and the cathodic aeration may also be used to fluidize the GAC. The proposed MBER system would have some potential advantages including less GAC mass due to more turbulent movement with air bubbles, long retention time of oxygen molecules facilitated by GAC, a high surface area for aerobic treatment of high strength waste stream, and less effect of membrane cleaning on the anodic microbial activity. The specific objectives of this study were: (1) to demonstrate the feasibility of cathodic fluidized GAC in the MBER and its effects on membrane fouling control; (2) to examine the MBER treatment of the enhanced-strength wastewater; and (3) to investigate the impact of varied GAC mass and aeration intensity on the MBER performance.

2. Materials and methods

2.1. MBER setup

The MBER system was constructed as a tubular reactor (23 cm long and 3.8 cm in diameter) made of anion exchange membrane (AEM-Ultrex AMI 7001, Membrane International. Inc, Glen Rock, New Jersey USA), as shown in Fig. 1. Carbon cloth (Zoltek Corporation, St. Louis, MO, USA) was used as the material for both the anodic and the cathodic electrodes. Before use, the carbon cloth was soaked in acetone solvent overnight and then heated for 30 min at 450 °C [20]. The cathodic electrode was coated with Pt/C powder (10% Etek, Somerest, NJ, USA) with a loading rate of 0.5 mg Pt cm^{-2}. The finished cathodic electrode was installed along with inner surface of the AEM tube and supported by a plastic mesh (Industrial Netting, MN, USA), resulting in a net cathodic liquid volume of 270 mL. The anodic electrode was plain carbon cloth and wrapped the AEM tube. A segment of PVC tube (an inner diameter of 5.1 cm) was used as an outer cover, resulting in a net anodic volume of 75 mL. The anodic and cathodic electrodes were connected to a 10 Ω resistor (otherwise stated). Four 7-cm PVDF hollow fiber ultrafiltration membranes (15,000 Da, Litree Purifying Technology Co. China) were glued by using epoxy to be a bundle and installed in the cathodic compartment. Twenty-four grams (otherwise stated) of granular activated carbon (8 × 30 mesh Calgon Carbon Corp, Pittsburgh, PA, USA) were added into the cathodic compartment as the fluidizing media.

2.2. Operating conditions

The MBER was operated at room temperature of ~20 °C. Its anodic compartment was inoculated with the anaerobic sludge from a local wastewater treatment plant (Radford, VA, USA) and fed with a synthetic solution containing (per L of tap water): glucose 0.5 g or 1.5 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 elements [21]. The anolyte was recirculated at 40 mL min^{-1}. The MBER was operated in a full loop mode, in which the synthetic wastewater was fed into the anodic compartment first and then the anodic effluent flowed into the cathodic compartment, before the final effluent was extracted from the membrane module. The flowrate was controlled by a peristaltic pump to achieve a desired hydraulic retention time (HRT). The aeration was supplied by using an aquarium fishing pump, and the intensity of aeration was controlled by a mass flow controller (Aalborg. Inc, Orangeburg, NY, USA).

2.3. Measurements and analysis

The MBER voltage was recorded every 5 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 conductivity was measured by a benchtop conductivity meter (Mettler-Toledo, Columbus, OH, USA). The concentration of chemical oxygen demand (COD) was measured by using a colorimeter according to the manufacturer's procedure (Hach DR/890, Hach Company, Loveland, CO, USA). Transmembrane pressure (TMP) was recorded manually and the average daily value was reported in this study. Turbidity was measured using a turbidimeter (DRT 100B, HF Scientific, Inc, Fort Meyers, FL, USA). The comparison of experimental data was analyzed by using two sample *t*-test.

2.4. Energy balance

Energy recovery was evaluated by normalized energy recovery (NER) in kWh m^{-3} or kWh $kgCOD^{-1}$ [22,23], which is a key parameter to assess the amount of energy that could be generated for treating one m^3 of wastewater or one kg COD removed. The energy consumption by the pumping system (for feeding, recirculating, and membrane extracting) was estimated by using the following equation [18]:

$$P = \frac{Q\gamma E}{1000}$$

where P is power requirement (kW), Q is flowrate (m^3 s^{-1}), γ is 9800 (N m^{-3}) and E is head loss (m H_2O). The energy consumption by aeration was estimated according to a previous study [24]:

$$p_b = \frac{p_1 T \lambda}{273000\varsigma(\lambda - 1)} \times \left[\left(\frac{p_2}{p_1}\right)^{(1-(1/\lambda))} - 1\right] \times Q_A$$

where p_b is power consumption of a blower; p_1 and p_2 are the inlet (normally atmospheric) and outlet absolute pressures (Pa) respectively; ς is blower efficiency (~0.8); λ is aerator constant (~1.4); T is the inlet temperature (K); and Q_A is the volumetric flow rate of air (m^3 s^{-1}).