Bioresource Technology 185 (2015) 426–430

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Short Communication

Carbon filtration cathode in microbial fuel cell to enhance wastewater treatment

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HIGHLIGHTS

- A homogeneous carbon filtration membrane was fabricated without using noble metals.
- Excellent effluent quality and power generation were achieved in the filtration MFC.
- The performance of filtration MFC kept relative stable during 20 days operation.

ARTICLE INFO

Article history:
Received 3 January 2015
Received in revised form 25 February 2015
Accepted 27 February 2015
Available online 6 March 2015

Keywords:
Carbon filtration membrane
Microbial fuel cell
Advanced wastewater treatment
Energy recovery

ABSTRACT

A homogeneous carbon membrane with multi-functions of microfiltration, electron conduction, and oxygen reduction catalysis was fabricated without using noble metals. The produced carbon membrane has a pore size of 553 nm, a resistance of $6.0 \pm 0.4\ \Omega\ cm^2/cm$, and a specific surface area of $32.2\ m^2/g$. After it was assembled in microbial fuel cell (MFC) as filtration air cathode, a power density of $581.5\ mW/m^2$ and a current density of $1671.4\ mA/m^2$ were achieved, comparable with previous Pt air cathode MFCs. The filtration MFC was continuously operated for 20 days and excellent wastewater treatment performance was also achieved with removal efficiencies of TOC (93.6%), NH_4^+–N (97.2%), and total nitrogen (91.6%). In addition, the carbon membrane was much cheaper than traditional microfiltration membrane, suggesting a promising multi-functional material in wastewater treatment field.

1. Introduction

Microbial fuel cell (MFC) is an electrochemical device that utilizes electroactive bacteria to recover energy in the form of electricity through oxidization of organic matter. As it couples electricity generation to wastewater treatment, MFC has drawn a great attention and received significant improvement in structure and performance in recent years. During the past decade, MFC has achieved a 10^5 times increase in power density from $0.01\ mW/m^2$ in 1999 (Kim et al., 1999) to $2.7\ W/m^2$ in 2007 (Fan et al., 2007) and various configuration with reactor size ranged from mL to >50 L (Liang et al., 2013) were reported. However, the present MFC is still impotent in achieving high quality effluent (Ge et al., 2013), due to the limited biomass retention (Logan, 2008) and low reaction rate under anaerobic condition in anode.

Integration of MFC with filtration technology may provide a potential solution to the existing problems. In a filtration bioelectrochemical system, energy could be ideally extracted in the form of electricity through oxidation of organic matter, while a high quality permeate could be obtained due to the filtration of membrane, achieving simultaneous energy recovery and advanced wastewater treatment. In previous studies, microfiltration (MF) or ultrafiltration (UF) membranes have been used as separation materials instead of expensive ion-exchange membranes in MFCs to intercept organic matter and biomass in anode (Kyoung Yeol et al., 2013). However, these traditional MF or UF membranes presented low mass transfer due to their low ion conductivity, resulting in a high internal resistance (Kim et al., 2007) of the integrated filtration MFC system. Another combination approach could be realized by using a conductive filtration membrane as air- or biocathode of a single-chamber MFC, thereby avoiding the requirements for separator or aeration in catholyte. Stainless steel mesh (Wang et al., 2011) and carbon-nanotube-deposited UF membrane (Malaeb et al., 2013) were employed as such conductive membranes to constitute a filtration cathode, and the feasibility and capacity of this configuration for simultaneous wastewater

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http://dx.doi.org/10.1016/j.biortech.2015.02.108
0960-8524/

treatment and energy recovery had been confirmed. Nevertheless, these fabricated filtration cathode materials had the following shortages: (I) nonuniform or much bigger pore size compared to MF or UF membrane (Wang et al., 2011, 2013); (II) utilization of noble metal (Pt) as catalyst (Malaeb et al., 2013); (III) relative high cost for large scale application (Malaeb et al., 2013; Wang et al., 2013).

In this study, conductive activated carbon powder, which possessed high specific surface area, was used as raw material for preparation of a homogeneous MF membrane. The obtained carbon MF membrane was then set as an air cathode to establish a filtration microbial fuel cell (F-MFC), which exhibited combined functions of MFC and membrane bioreactor (MBR). The physicochemical characteristics of the carbon filtration membrane were investigated before assembled in MFC. The power production and pollutants removal were evaluated during 20 days operation. And the performance of polluted filtration cathode in terms of power generation and flux were further investigated. In addition, the state-of-the-art progress in filtration MFC fabrication and performance was also summarized.

2. Methods

2.1. Fabrication of carbon filtration membrane

Carbon powder, originated from milling of activated carbon granule (Weishimei Environmental Technology Co., Ltd., Beijing, China), was screened with a 70-mesh sieve. Then 2.4 g screen-throughs were adequately mixed with 0.3 g sodium carboxymethyl cellulose (binder) and 1.2 mL H_2O (solvent) by stirring with a glass rod until a fluffy mixture was obtained. After that the fluffy mixture was transferred into a mould and pressed at 25 MPa for 2 min using a press machine (FW-4, Tianjin Optical Instrument Factory, China). The resulted carbon wafer was dewatered in a drying oven and at last carbonized in a vacuum tube furnace (GSL-1700X, Heifeikejing, China) at 900 °C for 90 min under argon protection. After cooling down, the obtained carbon membrane was stored at room temperature before use.

2.2. MFC assembly and operation

As illustrated in Fig. S1, the filtration MFC is a cylindrical bioreactor (total volume: 28 mL) comprising a carbon brush anode and a carbon filtration air cathode. The anode brushes are 3 cm in length and 3 cm in diameter, which were inoculated with anolyte of a mature MFC that has been operated for several years in our laboratory. The effective area of the cathode is 7 cm^2 and the shortest distance between cathode and anode carbon is 1 cm. An O-ring titanium sheet was used as electricity-collecting material for the filtration cathode, and an external resistance of 2000 or 200 Ω was connected between cathode and anode to simulate an electric appliance.

After assembling the MFCs, PBS-buffered acetate was used as synthetic wastewater, which contained (per liter of deionized water) 0.5 g CH_3COONa (Chemical oxygen demand, COD: 390.2 mg/L; Total organic carbon, TOC: 146.3 mg/L), 0.15 g NH_4Cl (NH_4^+–N: 39.3 mg/L), 1.04 g KH_2PO_4, and 2.16 g K_2HPO_4, all chemicals were reagent grade and purchased from Sigma-Aldrich (St. Louis, MO). During operation, one air cathode MFC was operated intermittently with synthetic wastewater replaced every 48 h as traditional batch mode MFC, therefore it was nominated as traditional MFC (T-MFC). In contrast, the other MFC was continuously operated with synthetic wastewater firstly pumped into the anode and further go through the filtration cathode at a flow rate of 0.58 mL/h (hydraulic retention time (HRT) of 48 h), it was nominated as filtration MFC (F-MFC, Fig. S1).

2.3. Measurements and analysis

Physicochemical characteristics including morphology and elemental composition were evaluated with scanning electron microscopy (SEM, TESCAN, Czech Republic) and X-ray photoelectron spectroscopy (XPS, Perkin-Elmer Phi 5300, USA) respectively. Porosity, pore size distribution, and specific surface area were completed with a mercury porosimeter (Autopore IV 9510, USA). Capacitance of the carbon filtration membrane was measured by galvanostatic charge-discharge method at both anodic and cathodic current of 10 mA in 5 M KOH solution using an electrochemical workstation (Autolab, Metrohm, Switzerland). Catalytic performance of carbon filtration membrane was evaluated by linear sweep voltammetry (LSV), with platinum wire used as control electrode and saturated calomel electrode (SCE, 0.242 V vs. standard hydrogen electrode, Leici, China) as reference electrode, and potential swept from +0.6 to −0.4 V at a scan rate of 1 mV/s. The LSV was conducted in a 20 mM PBS solution with three conditions: Ar aeration, O_2 aeration, and exposed in air (Fig. S2).

After the fabrication of the two MFCs, the cell voltages (U) across the external resistor (R_{ex}) were measured automatically in 1 min interval with a data acquisition system (DAQ2213, ADLINK, China). Current (I) was determined according to ohm's law: $I = U/R_{ex}$. Power density (P) and internal resistance (R_{in}) were calculated according to polarization curve based on the anode chamber volume (28 mL). The coulombic efficiency (CE) was introduced to evaluate the proportion of recovered energy from organic matter, which was calculated according to the following equation:

$$CE = \frac{3\int I dt}{FV(C_i - C_e)} \quad (1)$$

where F is Faraday constant (96,485 C/mol), V is the volume of treated wastewater (L), t is operation time (s), C_i and C_e are the TOC concentrations of influent and effluent (g/L), I is produced current (A).

During operation, the quality of synthetic wastewater and effluent from the two MFCs were evaluated including TOC, NH_4^+, NO_2^-, and NO_3^-. TOC was measured by a TOC analyser (Shimadzu TOC-VW, Japan). NH_4^+, NO_2^-, and NO_3^- were quantified by ion chromatography (ICS-1100, DIONEX, USA). Flux of the carbon filtration membrane was measured at different pressures to assess the development of membrane fouling. During the flux measurements, nitrogen gas was utilized to provide a trans-membrane pressure (TMP) for the F-MFC influent, TMP and effluent weight were continuously detected using a pressure sensor and an electric balance (Fig. S3). The calculated flux was normalized to cathode projected area (7 cm^2).

3. Results and discussion

3.1. Physicochemical properties of carbon filtration membrane

The filtration membrane is a carbon wafer which has a diameter of 38.0 ± 0.2 mm and thickness of 2 ± 0.1 mm (Fig. S4A). It possesses a microporous structure with porosity of 43.4% and pore size of 553 nm (Fig. S4B), and it can hold a water pressure of >50 kPa with an area of 7 cm^2. The specific flux is 6.9 ± 0.5 L/(h m^2 kPa), which is comparable with traditional ceramic membranes (Nandi et al., 2008). X-ray photoelectron spectroscopy (Fig. S4C) illustrated that the carbon filtration membrane is mainly comprised of C (94.14%), O (3.91%), N (1.49%) and a small amount of metals including Na (0.24%) and Ca (0.13%). The resistivity was 6.0 ± 0.4 Ω cm^2/cm and the specific surface area was 32.2 m^2/g.