Process Biochemistry 50 (2015) 1103–1109

Contents lists available at ScienceDirect

Process Biochemistry

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

Microbial electrolysis cell with spiral wound electrode for wastewater treatment and methane production

Yanping Hou[a], Renduo Zhang[a], Haiping Luo[a,*], Guangli Liu[a,*], Younggy Kim[b], Shuxian Yu[a], Jia Zeng[a]

[a] Guangdong Provincial Key Laboratory of Environmental Pollution Control and Remediation Technology, School of Environmental Science and Engineering, Sun Yat-sen University, Guangzhou 510275, China
[b] Department of Civil Engineering, McMaster University, 1280 Main Street West, Hamilton, ON, L8S 4L7, Canada

ARTICLE INFO

Article history:
Received 25 November 2014
Received in revised form 31 March 2015
Accepted 5 April 2015
Available online 9 April 2015

Keywords:
Microbial electrolysis cell
Spiral wound electrode
Methane production
Energy efficiency

ABSTRACT

The aim of this study was to develop a microbial electrolysis cell (MEC) constructed with spiral wound electrode and to evaluate its effectiveness for wastewater treatment and methane (CH_4) production. The spiral wound design can provide more than 60 m^2/m^3 of specific surface area of the electrode and low internal resistance. With acetate as the substrate and increasing applied voltages from 0.7 to 1.3 V, the average current density and CH_4 production rate increased from 46 to 132 A/m^3 and from 0.08 to 0.17 m^3/m^3 d, respectively. With the increasing applied voltages, the energy efficiencies decreased from 157% to 69%, while the COD removal rates increased from 0.31 to 0.69 kg COD/m^3 d. The optimal applied voltage of the spiral-wound-electrode MEC was about 0.95 V. Fed with dairy wastewater, the MEC also showed good performance with the average current density of 24 A/m^3, CH_4 production rate of 0.03 m^3/m^3 d, energy efficiency of 66%, and COD removal rate of 0.20 kg COD/m^3 d.

1. Introduction

The microbial electrolysis cell (MEC) is a promising bioelectrochemical technology for renewable and sustainable production of biogases or valuable chemicals through microbially catalyzed electrolysis of organic matters with an applied voltage [1–3]. It has been demonstrated that value-added products, such as hydrogen (H_2), methane (CH_4), caustics, and hydrogen peroxide, can be produced through wastewater treatment with the MEC [4,5].

MEC reactors with various structure types (e.g., cube-shaped, tubular-shaped, and multi-electrodes) have been developed to improve the performance of lab-scale reactors [6–8]. Prior to scaling up the MEC system, optimized reactor configuration should be designed. Rozendal et al. [9] showed that the minimum thickness of MEC reactors should be approximately 1 cm to provide sufficient space for wastewater pumping. Rader and Logan [8] found that the specific surface area of the electrodes (projected electrode surface area per volume of reactor) is a key parameter for the MEC scale-up. Escapa et al. [10] suggested that the inter-electrode distance should be smaller than 1 mm in the MEC to minimize the internal resistance associated with the low conductivity of domestic wastewater. To our knowledge, only two pilot-scale MECs have been successfully operated to treat real wastewater [11–13]. Nevertheless, the specific cathode surface areas of the 1000-L and 120-L pilot-scale reactors were 18.1 and 16.4 m^2/m^3, respectively [11–13], which were relatively small. For the previously reported MEC configurations, it is difficult to increase the size of MECs and to maintain high specific surface area. Therefore, it is necessary to develop MEC reactor configuration that allows the high specific area requirement. Recently, a spiral wound microbial fuel cell (MFC) has been proposed with high ratio of surface area to volume and low internal resistance [14,15]. The high power generation of the MFC is mainly attributable to the novel spiral wound electrodes. Therefore, it is expected that a MEC constructed with spiral wound electrode should improve the performance with the potential for scaling up.

The MEC is initially designed for H_2 production [16], which has great advantages over the H_2 process from fossil fuels because of the renewable feature, high H_2 yield, and high energy efficiency [1]. However, in the MEC for H_2 production, it is often observed that CH_4 can become the dominated biogas after a period of operation due to the cathodic methanogenesis [3,17–22]. Compared with H_2 production, CH_4 production in the MEC is more stable with various substrates. Therefore, it is worth to examine CH_4 production performance in the MEC.

* Corresponding authors. Tel.: +86 20 84110052.
E-mail addresses: luohp5@mail.sysu.edu.cn (H. Luo), liugl@mail.sysu.edu.cn (G. Liu).

http://dx.doi.org/10.1016/j.procbio.2015.04.001
1359-5113/

The objective of this study was to develop a MEC configuration with spiral wound electrode. A cost-effective catalysis material (Ni foam) was used as the cathode. The spiral-wound-electrode MEC was compared with others in terms of specific surface area and internal resistance. The performance of the spiral-wound-electrode MEC was evaluated based on average current generation, coulombic efficiency, CH_4 production, and energy balance between CH_4 production and COD removal.

2. Materials and methods

2.1. Reactor configuration

The single-chamber MEC reactor was made of plexiglass, with inner diameter of 7 cm and height of 15 cm (total volume of 577 mL), and working volume of 500 mL.The anode was carbon cloth (Toray, woven fabric, Toray Industries, Inc. Japan) and the cathode was Ni foam (Kunshan Bi Tai Xiang Electronics Co., Ltd, China). A schematic graph of the MEC is shown in Fig. 1. A membrane electrode assembly was made by putting an anion exchange membrane (AEM; Ultrex CMI17000, Membranes International) between the anode and cathode. The size of the anode and cathode was 30 cm × 10 cm (cross-sectional projected area of 300 cm^2) and the membrane size was 30 cm × 10.4 cm. The electrode assembly was rolled into a compact structure (i.e., the spiral wound electrode) and fitted into the reactor. Two titanium wires were used as current collectors. An external resistor of 1 Ω was used to measure the electric current in the MEC. A glass tube (1.6 cm inner diameter and total volume of 15 mL) sealed with a butyl rubber stopper and an aluminum crimp top was attached to the reactor top.

2.2. Startup and operation

The MEC was inoculated with a 50:50 mixture of the domestic wastewater (primary clarifier overflow from Liede Wastewater Treatment Plant, Guangzhou) and a medium. The medium contained a sodium acetate (1 g/L) solution, phosphate buffer solution (50 mM; 4.09 g/L Na_2HPO_4, 2.54 g/L NaH_2PO_4, 0.31 g/L NH_4Cl, 0.13 g/L KCl), trace mineral solution (12.5 mL/L), and vitamin solution (12.5 mL/L) [23]. During the start-up period, a constant voltage of 0.7 V was applied to the circuit using a D.C. power source (IT6720, Itech, China). After a startup period about 30 days and a steady operation period of 30 days, different applied voltage conditions (from 0.7 to 1.3 V) were examined. To be effective, fed-batch experiments were conducted for initial tests and characterization of the MEC reactor design, electrode materials, wastewater treatment, and energy recovery performance [24]. The first test of the spiral-wound-electrode MEC was operated in a fed batch mode at 25 ± 2 °C. Before each cycle of operation, the reactor medium was sparged with N_2 gas for 10 min.The medium fed in the MEC was replaced when the current was below 5 mA. After testing with acetate as the substrate, the spiral-wound-electrode MEC was fed with artificial dairy wastewater, which was with main organic components of carbohydrate, fat, and protein, chemical oxygen demand (COD) of 1120 mg/L, and pH of 6.94, under an applied voltage of 0.9 V. Biogas was collected using a sampling bag (Shanghai ELOR Co., Ltd, China) and gas volume was measured by withdrawing the gas using glass syringe.

2.3. Analysis and calculations

The voltage across the resistor (1 Ω) was measured using a multimeter data acquisition system (Model 2700, Keithley Instruments, Inc., Cleveland, OH, USA) to calculate the current. Concentrations of H_2, CO_2, and CH_4 were analyzed using gas chromatography (GC 2014, Shimadzu. Co., Japan) with a thermal conductivity detector. The carrier gas was high purity argon at a flow rate of 10 mL/min.Chemical oxygen demand (COD) of the influent and effluent was measured according to the standard method (Method 5220, APHA et al., 1995; HACH COD system; Hach Co., Loveland, CO).

The average current over the time period to accumulate 90% of the total charge was used to evaluate the current generation in the MEC and calculated as follows [24]:

$$I_{avg,90} = \frac{Q_{total}}{t_{90}} \tag{1}$$

where Q_{total} is the total charge (number of Coulombs transferred).

The COD removal rate was calculated as follows:

$$r_{\mathrm{COD}} = \frac{\mathrm{COD_{In}} - \mathrm{COD_{Out}}}{t} \tag{2}$$

where $\mathrm{COD_{In}}$ and $\mathrm{COD_{Out}}$ are the COD values of the influent and effluent; t is the operation time of one cycle.

To examine the biogas production performance, average CH_4 production rate was calculated as follows:

$$r_{\mathrm{CH_4}} = \frac{V_{\mathrm{CH_4}}}{Vt} \tag{3}$$

where $V_{\mathrm{CH_4}}$ (m^3) is the volume of CH_4 collected from one cycle; V (m^3) is the liquid volume of the single chamber reactor; t (d) is the cycle time.

The coulombic efficiency (CE, %) was calculated based on total coulombs recovered as current to original coulombs in the degraded substrate[1]:

$$CE = \frac{8\int_0^t I\ \mathrm{d}t}{FV\,\Delta\mathrm{COD}} \times 100\% \tag{4}$$

where I is the current, F is Faraday's constant (96,485 C/mol), V is the liquid volume of the single chamber reactor, ΔCOD is the COD removed.

The amount of energy added to the circuit (W_{E}, kWh) was calculated by [1,19]:

$$W_{\mathrm{E}} = \sum_{i=1}^{n} (I_i E_{\mathrm{ap}}\,\Delta t - I_i^2 R_{ex}\ \Delta t) \tag{5}$$

where E_{ap} (V) is the voltage applied using the power source, I_i (A) is the current, Δt (h) is the time increment for n data points measured during a batch cycle, and R_{ex} = 1 Ω is the external resistor.

Since energetic biogases, among which CH_4, accounts for more than 95%, are simultaneously recovered while removing organic matter in the MEC, the energy consumption per COD removal is estimated by [21]:

$$W_{\mathrm{E,COD}}{}' = \frac{W_{\mathrm{E}} - 0.247 n_{\mathrm{CH_4}}}{\Delta\mathrm{COD}} \tag{6}$$

where $n_{\mathrm{CH_4}}$ is the number of moles of CH_4 recovered during a batch cycle and 0.247 is the energy content of one mole of CH_4 (in kWh/mol). The energy efficiency (η_{E}) relative to the electrical input was the ratio of energy for the CH_4 recovery to the input electrical energy, i.e.,

$$\eta_{\mathrm{E}} = \frac{0.247 n_{\mathrm{CH_4}}}{W_{\mathrm{E}}} \tag{7}$$

Electrochemical impedance spectroscopy (EIS) measurement was conducted to examine resistance of the spiral MEC system. EIS was carried out using an electrochemical workstation (CHI660C, CH Instruments, Inc. Shanghai, China) over frequencies varying from 100 kHz to 0.01 Hz with a sinusoidal perturbation of 10 mV amplitude. During the measurements of MEC impedance spectra, the anode was served as the working electrode, and the cathode was