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Ethanolamine degradation and energy recovery using a single air-cathode microbial fuel cell with various separators

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ABSTRACT

Ethanolamine (ETA) is commonly used for alkalinization to prevent corrosion of pipes in nuclear power plants. ETA, however, causes water to deteriorate, increasing the concentration of organic matter or nutrient salts in water systems. To generate power and degrade ETA at the same time, ETA was used as carbon source for microorganisms in a single air-cathode microbial fuel cell (MFC). Using a membrane as a separator in an MFC makes them costly to set-up. To reduce the expense and evaluate MFC performance, the experiments were conducted with three different separators: a proton exchange membrane (PEM), a cation exchange membrane (CEM), and polypropylene (PP) felt. The PP felt-MFC resulted in the most efficient COD (94%) and ammonium removal (52%). The CEM-MFC produced the highest power density of 583.7 mW m^{-2} at a current density of 0.15 mA cm^{-2}. The Coulombic efficiencies (CEs) were 25.1, 23.7, and 10.5% for PEM-, CEM-, and PP felt-MFCs, respectively. Although using PP felt decreased the power generation compared with membrane MFCs in terms of energy recovery, it increased ETA degradation and reduced the cost of initial set-up.

Introduction

In the 21st century, the supply of sustainable energy is one of the most important scientific challenges because of the global population growth (Lewis and Nocera, 2006). Therefore, the use of nuclear power has been steadily overtaking coal energy. Ethanolamine (ETA) is used to remove sour gases (H_2S and CO_2) in industrial sweetening processes and for alkalinization of water in steam cycles of nuclear power plants to control corrosion and scale formation in pipes (Ndegwa et al., 2004). Discharged water from nuclear power plants contains a high ETA concentration. ETA is difficult to degrade naturally, and its by-products can cause water to deteriorate, increasing the concentration organic matter or nutrient salts. To treat wastewater containing ETA, ion-exchange resin (Raught et al., 2005), biodegradation (Ndegwa et al., 2004), and oxidation (Puttaswamy et al., 2001) have been investigated. These methods, however, require a large amount of chemicals, high cost, and long time for the degradation reaction to proceed. Therefore, an alternative process is needed.

Microbial fuel cells (MFCs) are based on bio-electrochemical technology and are a promising technology to directly generate electricity from a substrate by attaching electrochemically active bacteria (EAB) to an anode. In MFC research, various substrates have been used to improve electricity production or treat wastewater, such as acetate, glucose, brewery wastewater, starch processing wastewater and dye wastewater (Pant et al., 2010). Although much wastewater and many substrates have been applied to MFCs for treatment and to generate power, wastewater containing ETA has not been investigated. The use of a membrane separator, such as a proton exchange membrane (PEM), a cation exchange membrane (CEM), or anion exchange membrane (AEM), increases the cost of installation and is one of the main problems in MFC research. Therefore, reducing the set-up cost is a key concern of this research. Polypropylene (PP) felt is affordable and not chemically reactive.

ETA can be biodegraded to ammonium and acetaldehyde by hydrolysis. Ammonium can be oxidized to nitrate under aerobic conditions, and nitrate can be converted to nitrogen gas under anaerobic conditions. Simultaneous nitrification and denitrification (SNdN) has been suggested to occur in the same floc (Abbasi and Adams, 2000). When a floc of microorganism forms, nitrification and denitrification can occur outside and inside of the floc. The acetaldehyde decomposes into acetate and ethanol by hydrolysis.

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The acetate and ethanol can act as electron donors for denitrification of nitrate and are degraded to CO_2 and H_2O under aerobic and anaerobic conditions (Ndegwa et al., 2004; Mrklas et al., 2004).

In this study, we demonstrated the possibility of utilization of ETA as a substrate and PP felt as a separator on a single chambered air-cathode MFC. The experiments to investigate application as a substrate and biodegradation of ETA were conducted by comparing with acetate and conventional anaerobic treatments. To evaluate PP felt as a separator in a MFC using ETA, an experiment was performed with PEM, CEM, and PP felt. Using PP felt for ETA degradation in a single air-cathode MFC was first investigated, and the performance of the fuel cell was discussed.

Material and methods

MFC reactor set up

A rectangular acrylic reactor which has a single chamber consists of anode and cathode (a working volume of 75 mL following dimension: length, 3 cm; width and height, 5 cm) as Fig. 1. Carbon cloth (1071 HCB, AvCarb®) was used as both the anode (without wet-proofing) and the cathode (30% wet-proofing). One side of the cathode contained 0.5 mg cm^{-2} of Pt catalyst from 10% Platinum/Carbon (10% Pt on Vulcan XC-72, Premetek) with Nafion solution as a binder. The air-facing surface of the electrode was coated with a carbon base and polytetrafluoroethylene (PTFE) in four diffusion layers. The cross-sectional area of the electrodes was 25 cm^2, and they were connected by titanium wire. The PEM (Nafion 117, Ion Power Inc.), CEM (CMI-7000, Membranes International Inc.), and PP felt (Oh-sung Corp, South Korea) were used as separators and placed between the electrodes to avoid a short circuit. The PP felt is a thin cloth with thickness of 0.3 mm. A thermometer was inserted in the reactor to maintain a consistent temperature for microorganism growth. To prevent a water leak, silicon gaskets were used between the end plates and chamber. All parts were combined by using rods and nuts.

MFC operation

Sludge was collected from the anaerobic zone in the Jungrang sewage treatment plant in Seoul, Republic of Korea. After the sludge was washed with phosphate buffer solution (PBS) to maintain neutral pH for microorganisms, the MFC reactor was inoculated with 50% sludge rinsed with PBS and 50% medium containing 0.5 g L^{-1} acetate. The medium and PBS solution consisted of (per liter) NH_4Cl, 0.31 g; KCl, 0.13 g; $NaH_2PO_4{\cdot}H_2O$, 6.6 g; Na_2HPO_4, 8.19 g; and mineral (12.5 mL) and vitamin (12.5 mL) solutions.

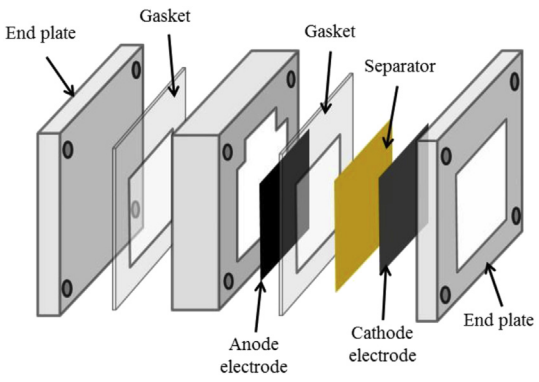

Fig. 1. Schematic diagram of the single air-cathode MFC configuration in this study.

The MFCs were operated in fed-batch mode by connecting to a fixed external resistance of 1000 Ω. For microorganism growth, nitrogen gas was bubbled to remove oxygen before inoculation. The substrate was replaced in an anaerobic glove box when the voltage decreased below 50 mV. All experiments were performed at 25 °C ± 2 and the electrolyte was stirred at 100 rpm with a magnetic bar to continuously mix the solution. After the MFCs were adapted to acetate with stable voltage, the ETA concentration was gradually raised to adjust the ETA reducing acetate ratio (5:0, 4:1, 3:2, 2:3, 1:4, 0:5). All reactors were operated for approximately one and a half months at least to promote the activity of exoelectrogen for the stable power generation.

Calculation and measurement

The cell voltages were measured every 10 min with a voltage recorder (VR-71, T&D Corporation) connected to a computer. Current was calculated according to Ohm's law and power was computed by $P = IV$, where V is the voltage (in millivolts; mV) and I is the current (in milliamperes; mA). Current and power density were normalized to the area of the anode. Polarization and power density curves were obtained by the multi-cycle method at different resistances (from 10 to 2000 Ω) as a function of current densities. Coulombic efficiency (CE) was estimated from chemical oxygen demand (COD) removal as $CE = M_s \int_{t_0}^{t_b} Idt/Fbv_{an}\Delta COD$, where M_s is the molecular weight of oxygen, F is Faraday's constant, b is the number of electrons per mole of oxygen, v_{an} is the volume of electrolyte and ΔCOD is the change in COD over time t_b.

All samples were centrifuged at 13,000 rpm for 15 min and filtered with a 0.2-μm syringe filter before analysis. Total COD, nitrate, nitrite and ammonium were measured at the beginning and end of each batch by a standard method using a Hach spectrophotometer (DR-2800, Hach Company). The pH and conductivity of the solution were analyzed with a pH meter (Orion 720A+, Thermo) and conductivity meter (Orion three star, Thermo), respectively.

Results and discussion

Effect of ETA as a substrate on a single air-cathode MFC

ETA was used and compared to acetate in a single air-cathode MFC to evaluate ETA as a substrate. Acetate is a well-known substrate in MFC research, because it can lead to high power density and CE (Pant et al., 2010). ETA was expected to generate higher electrochemical efficiency than acetate because the electrons from ETA can be produced more than that from acetate according to:

$$C_2H_7NO + 5H_2O \rightarrow 2HCO_3^- + 12H^+ + 10e^- + NH_3 \quad (1)$$

Fig. 2 shows the changes in voltage of the CEM-MFCs using 500 mg L^{-1} acetate and ETA as a substrate, respectively. The maximum cell voltage of the ETA-MFC was 0.502 V for about 4.3 days. In the acetate-MFC, the maximum cell voltage was 0.523 V for around 3 days. The voltage generation of the MFCs used acetate and ETA as an electron donor returned rapidly to the maximum levels. This indicates that the microorganisms attached to the anode surface can use acetate and ETA well as substrate for generating the voltage. The CEs of the ETA- and acetate-MFCs were 23.7% and 27.9%, respectively. Fig. 3 shows power density curves as a function of the current density for MFCs using ETA and acetate. The maximum power densities of the ETA- and acetate-MFCs were 583.7 mW m^{-2} and 476.1 mW m^{-2}, respectively. COD removal