

Energy recovery of ethanolamine in wastewater using an air-cathode microbial fuel cell



Ja-Won Shin, Young-Hyun Song, Byung-Min An, Seok-Ju Seo, Joo-Yang Park*

Department of Civil and Environmental Engineering, Hanyang University, 17 Haengdang-Dong, Seongdong-Gu, Seoul, Republic of Korea

ARTICLE INFO

Article history:

Received 17 January 2014

Received in revised form

14 May 2014

Accepted 20 May 2014

Available online 7 July 2014

Keywords:

Ethanolamine

Microbial fuel cells

Air-cathode

Ammonia volatilization

ABSTRACT

Ethanolamine (ETA) is widely used as a metal corrosion inhibitor and for CO₂ capture. The treatment of ETA in wastewater involves advanced oxidation or electrolysis, which requires excessive energy since it is a not readily biodegradable organic containing amine. In this study, a microbial fuel cell (MFC) for simultaneous ETA degradation and electricity generation was investigated. 80 mL of an air-cathode single-chamber MFC was designed to determine the degradation and by-products formation of ETA and the power density. The reactor was inoculated with return activated sludge taken from a local wastewater treatment plant. This system was able to remove 91% of chemical oxygen demand (COD) and 37% of ammonia when ETA was injected into the MFC. A maximum power density of 240 mW m⁻² (7.65 mW m⁻³) and a Coulombic efficiency of 18.2% were obtained with the ETA-fed MFC. From the results, although further research is required to treat ammonium derived from ETA degradation and to improve the performance of the MFC, it was found that the MFC would be a promising technology for ETA wastewater treatment, as well as electricity production.

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1. Introduction

Ethanolamine (ETA) is widely used for acid or sour gases removal, known as a sweetening process in the natural gas industry (Ndegwa et al., 2004), CO₂ capture from flue gases (Kim et al., 2010) and as pH adjustment reagents to inhibit the metal corrosion in steam cycles of power plants, especially nuclear power plants with pressurized water reactors (Nordmann, 2004). Consumption of ETA is expected to increase due to the increasing production of natural gas (Hawthorne et al., 2005), as well as increasing demand of CO₂ capture. Generation of wastewater containing ETA is also expected to grow. Treatment of ETA in wastewater involves removal of chemical oxygen demand (COD) and total nitrogen (T-N). Treatability of ETA has been investigated using various chemical processes: ozonation and catalytic decomposition (Parisheva and Demirev, 2000), photooxidation using TiO₂ (Horikoshi et al., 2001) and electro dialysis reversal with electrolysis (Yeon et al., 2007). Anotai et al. (2012) improved its biodegradability by integrating a biological process and the Fenton reaction. Furthermore, relatively more research on biological degradation has been conducted under aerobic and/or anaerobic conditions to degrade ETA

in soil and water (Knapp et al., 1996; Mrklas et al., 2004; Ndegwa et al., 2004; Hawthorne et al., 2005; Kim et al., 2010). Most of the processes in the preceding experimental studies were energy consuming. They were focused on the treatment of ETA as just waste, although ETA is a potential organic source that can be utilized in microbial fuel cells.

A microbial fuel cell (MFC) is a promising process that can remove contaminants from wastewater and recover electrical energy concurrently by microbial metabolism. Much research on MFCs has been conducted to generate electricity using various substrates from simple organic matter, such as acetate and glucose (Cheng et al., 2006; Borole et al., 2009; Chae et al., 2009), to complex organic materials in domestic wastewater (Min and Logan, 2004) and brewery wastewater (Feng et al., 2008). In addition, MFCs have also been applied to wastewaters containing not only organic compounds but also ammonia, such as landfill leachate (Puig et al., 2011), wastewater from the starch process (Lu et al., 2009) and swine wastewater (Min et al., 2005). Nevertheless, MFC application to wastewater containing ETA has not been investigated in spite of the increasing demand for its treatment.

The biodegradability of ETA varies depending on the experimental conditions while intermediates observed during the process are the same. The amine and ethyl alcohol of ETA is biologically degraded into ammonium and acetaldehyde, respectively

* Corresponding author. Tel.: +82 2 2220 0411; fax: +82 2 2220 1945.

E-mail address: jooypark@hanyang.ac.kr (J.-Y. Park).

(hydrolysis). Nitrification/denitrification is a typical biochemical process to remove ammonia. In nitrification, ammonium is oxidized to nitrite and then to nitrate under aerobic conditions. Then, nitrate is used as an electron acceptor and transformed into nitrogen gas under anoxic conditions in the denitrification stage. Acetaldehyde is hydrolyzed to ethanol and acetate, and then these two products are finally degraded to CO₂ and water under both aerobic and anaerobic conditions (Mrklas et al., 2004).

The objective of this study was to investigate the feasibility of ETA as a fuel source for MFCs. ETA is known to be not readily biodegradable and toxic to microorganisms at a high level (Bakalova et al., 2008). Furthermore, ammonia, produced as an intermediate, can affect microbial activity and electricity production (Nam et al., 2010). Therefore, the treatability of ETA in a MFC needs to first be evaluated. The MFC was started up with acetate for the enrichment of electrochemically active bacteria (EAB), and then a portion of ETA was incremented for adaptation of the EAB to ETA. ETA degradation and its by-products were monitored, and electricity generation was observed as well.

2. Material and methods

2.1. MFC configurations

The MFC consisted of a single chamber (an empty volume of 80 mL) made of a rectangular acrylic reactor. Carbon cloth (1071 HCB, AvCarb[®]) was used for both the anode (non wet-proofed type) and the cathode (30% wet-proofed type). The air-facing side of the cathode electrode was coated with a carbon base layer and four polytetrafluoroethylene (PTFE) diffusion layers, and a Pt catalyst layer of 0.35 mg cm⁻² (10% Pt on Vulcan XC 72, Premetek) with Nafion binder was applied to the solution-facing side as previously described (Cheng et al., 2006). The projected surface area of both electrodes was 25 cm². A cation exchange membrane (CEM, CMI-7000, Membranes International Inc.) was emplaced between the two electrodes and these electrodes were spaced 2 mm apart. For expansion of the membrane, the CEM was immersed in a 5% NaCl solution at 40 °C for 24 h and soaked in deionized water prior to use. Titanium wire was fitted to the body of the MFC and the end plate with a hole for air contact. The wire was then sealed with a conductive silver epoxy for connection to a fixed external circuit resistance at 1000 Ω. A thermometer was embedded in the reactor to monitor the temperature of the anode chamber. Silicon gaskets were placed between the end plates and the reactor body as well as

the electrode and membrane in order to prevent water leakage. All parts were fabricated with rods and nuts as shown in Fig. 1.

2.2. MFC operation

Return activated sludge was obtained from the Jungnang sewage treatment plant in Seoul, Republic of Korea as the inoculum. The sludge was rinsed with a phosphate buffer solution three times. The pretreated sludge and a medium solution containing 0.5 g L⁻¹ acetate were injected into the MFC at a ratio of 1:1 for the start-up stage. The chemicals used for the phosphate buffered medium were as follows (grams per liter of deionized water, g L⁻¹): NaH₂PO₄·H₂O, 2.69; Na₂HPO₄, 4.33; NH₄Cl, 0.31; KCl, 0.13, as reported by Kim et al. (2007). Also, the medium was amended with the same amount (12.5 mL) of Wolfe's mineral solution and Wolfe's vitamin solution.

The MFC experiments were conducted in fed-batch mode. After observation of a stable voltage, the solution in the anode compartment was completely replaced with a feed solution. The solutions containing the nutrient medium were prepared with various concentration ratios of acetate to ETA (5:0, 3:2, 1:4, 0:5) and the prepared solution was flushed with nitrogen gas before replacement to remove oxygen. The refill procedure was repeated when the cell voltage decreased to less than 40 mV and was carried out in an anaerobic glove box. The solution in the reactor was continuously mixed with a magnetic stirrer at 100 rpm during the tests. MFC operation was carried out in a temperature-controlled room at 26 ± 2 °C.

2.3. Analyses and calculations

All samples were centrifuged at 4500 rpm for 5 min and filtered through a syringe filter (0.45-μm pore diameter) before chemical analysis. Soluble COD and three inorganic nitrogen concentrations were analyzed using a HACH spectrophotometer according to standard methods as follows; COD (closed flux colorimetric method), ammonia (including both NH₄⁺-N and NH₃-N, Salicylate method), nitrite (Diazotization method) and nitrate (Chromotropic acid method). The electrical conductivity and pH of the solution were measured by a conductivity meter (Orion three star, Thermo) and a pH meter (Orion 720 A+, Thermo), respectively.

Cell voltages across the external resistor were automatically measured and recorded every 10 min using a data logger (VR-71; T&D Co.) during the experiments. These data was used to calculate

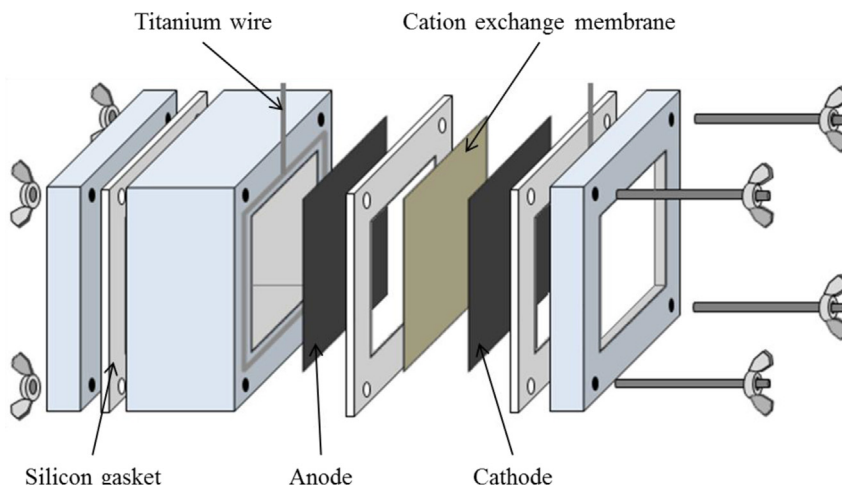


Fig. 1. A schematic of the single-chamber MFC used in this study.

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