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The enhancement of ammonium removal from ethanolamine wastewater using air-cathode microbial fuel cells coupled to ferric reduction

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HIGHLIGHTS

- Alcohol group of ETA was effectively degraded in an air-cathode single-chamber MFC.
- Adding goethite in MFC with acetate and ammonium increased the ammonium removal.
- The maximum power density of the MFC with goethite and ETA was 210 mW m^{-2} .

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ABSTRACT

A microbial fuel cell (MFC) with biological Fe(III) reduction was implemented for simultaneous ethanolamine (ETA) degradation and electrical energy generation. In the feasibility experiment using acetate as a substrate in a single-chamber MFC with goethite and ammonium at a ratio of 3.0 (mol/mol), up to 96.1% of the ammonium was removed through the novel process related to Fe(III). In addition, the highest voltage output (0.53 V) and maximum power density (0.49 W m^{-2}) were obtained. However, the ammonium removal and electrical performance decreased as acetate was replaced with ETA. In the long-term experiment, the electrical performance markedly decreased where the voltage loss increased due to Fe deposition on the membranes.

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1. Introduction

Because nitrogen can cause eutrophication and oxygen depletion in aquatic environments, its removal from wastewater has received worldwide attention (Ahn, 2006). Many industrial wastewaters contain nitrogen in the form of ammonia and nitrate. Ethanolamine (ETA) is one source that results in increased chemical oxygen demand (COD) and total nitrogen concentration in wastewater. ETA is used in various industrial applications due to the bifunctional properties of the primary alcohol and primary amine. On the other hand, wastewater containing ETA is difficult to treat due to this bifunctional property. Two typical technologies for ETA removal are electrochemical (Yeon et al., 2007) and biological (Kim et al., 2010) treatment processes. The alcohol group can be treated easily whereas ammonium is refractory in the presence of organic in both processes. These processes, in turn, require high

energy consumption to treat organics together with ammonium. Thus, to treat ETA and reduce energy consumption, a microbial fuel cell (MFC) was investigated in this study.

The MFC is an energy recovery system that can produce electricity from biodegradable compounds due to the metabolism of microorganisms. MFC technologies have been investigated to simultaneously treat wastewater and harvest energy. Studies of ammonium removal using MFCs with different structures have been conducted. Virdis et al. (2010) reported that carbon and ammonium were removed in two-chamber MFCs by intermittently providing air at the cathode. Dual-cathode MFCs were also examined to remove carbon and nitrogen. Nitrogen was removed using two different processes (nitrification and denitrification) in the same manner as a conventional process (Xie et al., 2011; Zhang and He, 2012). In addition, a single chamber MFC was pre-enriched to improve the ammonium removal performance (Yan et al., 2012). The processes in the studies still require energy for aeration or have the possibility of volatilization.

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In our previous research, the utilization of ETA as a fuel for a single-chamber MFC was investigated (Shin et al., 2014). ETA can be biologically degraded to ammonium and acetaldehyde, and then acetaldehyde can be hydrolyzed to ethanol and acetate (Mrklas et al., 2004). The COD removal of 90% was obtained within 3 days in closed circuit mode while COD of 83% was removed after 12 days in open circuit mode. However, there were difficulties in removing ammonium due to the anaerobic operating conditions in the anode, which is a problem in existing treatment processes as well. Processes which facilitate ammonium removal even under anaerobic conditions should be integrated with MFCs to solve this problem.

There are two short pathways for anaerobic ammonium oxidation: anaerobic ammonium oxidation (Anammox) and ferric ammonium oxidation (Feammox). The Anammox process occurs in the absence of oxygen and aerobic ammonium-oxidizing bacteria. The process involves ammonium oxidation with nitrite as the electron acceptor to yield nitrogen gas (Jetten et al., 2001; Thamdrup, 2012). Anammox is one of the suspected mechanisms of ammonium losses in an air-cathode single-chamber MFC. However, this process can occur in the presence of nitrite (NO_2^-) together with ammonium (NH_4^+). During ETA biodegradation, NH_4^+ is produced and accumulated without further reaction (Mrklas et al., 2004; Hawthorne et al., 2005) under anaerobic conditions. Thus, the process may not play a major role in ammonium removal in MFCs.

The Feammox process takes place under anaerobic and iron-rich conditions by iron reducing anaerobes. This process has been reported in wetland soil (Clément et al., 2005; Shrestha et al., 2009), tropical upland soil (Yang et al., 2012), paddy soil (Ding et al., 2014), and anaerobic sludge (Sawayama, 2006; Park et al., 2009). Through Feammox, N_2 , NO_2^- , or nitrate (NO_3^-) is produced. The conversion of NH_4^+ to N_2 is thermodynamically favorable compared to the production of NO_2^- or NO_3^- . Moreover, the reaction to produce N_2 is feasible over a wider range of pH values whereas the reaction to NO_2^- occurs only at a low pH (below 6.5) (Yang et al., 2012). The Feammox process is one of possible processes to enable NH_4^+ removal under anaerobic conditions. However, research of the occurrence of this process in MFCs has never been explored.

Therefore, in this study, the feasibility of MFCs combined with ferric reduction was investigated to completely treat ETA by improving the NH_4^+ removal. First, acetate and NH_4^+ , which are the main by-products of ETA biodegradation, were supplied as the substrate to the anodic bacteria in order to determine the occurrence of the Feammox process in a MFC depending on the Fe(III) source (soluble and insoluble). Then, ETA was introduced into the MFCs. Secondly, three MFCs, which were a control MFC, a pre-enriched MFC with Fe(III) injection, and a newly prepared MFC with Fe(III) injection, were operated to compare the start-up and nitrogen removal efficiency.

2. Methods

2.1. MFC configurations

Transparent rectangular acrylic reactors with an empty volume of 80 mL were prepared as shown in Fig. 1. Non wet-proofed type carbon cloth (1071 HCB, AvCarb[®]) and 30% wet-proofed type carbon cloth (1071 HCB, AvCarb[®]) were used as the anode and the cathode electrodes, respectively. The cathode electrode was coated with a carbon base layer and four polytetrafluoroethylene (PTFE) diffusion layers on the air-facing side. Then, Nafion (5 wt%, Sigma-Aldrich) binder mixed with Pt (10% Pt on Vulcan XC 72, Premetek) of 0.35 mg cm^{-2} was applied to the solution-facing side as a catalyst layer. The projected surface area of all electrodes was

25 cm^2 . The anode and cathode were separated by a cation exchange membrane (CEM, CMI-7000, Membranes International Inc.) and these electrodes were spaced 2 mm apart from the membrane. The CEM was pretreated in a 5% NaCl solution at 40°C for 24 h and immersed in deionized water prior to use. Titanium wires were fitted and then sealed with a conductive silver epoxy to the body of the MFCs and an end plate to collect electrons. The wires were connected to a fixed external circuit resistance of 1000Ω . The temperature of the MFCs was monitored with a thermometer embedded in the reactors.

2.2. MFCs Operation

The experiments were conducted in two parts. First, four MFCs were inoculated with different phosphate buffered medium solutions (PBSs) and return activated sludge obtained from the Jungnang sewage treatment plant (Seoul, Republic of Korea). The sludge was washed with phosphate buffered medium (0.1 M) three times before the inoculation. Four different PBSs were prepared as follows: without any Fe(III) source (MFC-C), with goethite (FeOOH) (MFC-G) and hematite (Fe_2O_3) (MFC-H) as insoluble Fe(III) sources, and with FeCl_3 (MFC-S) as a soluble Fe(III) source for the feasibility test. The medium solution initially contained acetate and NH_4^+ for acclimation and then, ETA was injected to the medium in all of the MFCs instead of acetate and NH_4^+ after adaptation (Fig. 1(b)).

The second part of the experiments involved long-term investigation and comparison of the start-up. Two pre-enriched MFCs and one new MFC were prepared. The pre-enriched MFCs were operated for about one year to enrich the electrochemically active bacteria (EAB) and to adapt the bacteria to ETA. One was for a control test and this was operated without Fe(III) addition (WO-MFC) and the other MFC was operated with goethite (WF-MFC1) to compare with a newly inoculated MFC (WF-MFC2). The new MFC was inoculated with return activated sludge and goethite at the same time (Fig. 1(c)). The MFCs were operated with the solution containing 1000 mg L^{-1} ETA.

In the experiment with Fe(III) addition, the Fe(III) sources (goethite and FeCl_3) were injected into the medium solution at a ratio of $\text{Fe(III)}:\text{NH}_4^+$ or $\text{ETA} = 3:1$ (mol/mol; 1 mol of ETA generates 1 mol of NH_4^+). The chemicals used for the phosphate buffered medium solution were added as follows (grams per liter of deionized water, g L^{-1}): 2.69 g L^{-1} $\text{NaH}_2\text{PO}_4 \cdot \text{H}_2\text{O}$, 4.33 g L^{-1} Na_2HPO_4 , and 0.13 g L^{-1} KCl. In addition, the medium was amended with the same amount (12.5 mL) of Wolfe's mineral and Wolfe's vitamin solutions.

The MFC experiments were carried out in a fed-batch mode. After observation the cell voltage at less than 100 mV, the solutions in the MFCs were completely replenished with a new feed solution. The prepared solution was purged with nitrogen gas to remove oxygen, and during the experiments, the solutions in the MFCs were continuously mixed with a magnetic stirrer at 100 rpm. The MFC operation was conducted at room temperature ($26 \pm 2^\circ \text{C}$).

2.3. Analyses of chemicals and calculations

Before the chemical analysis, all samples were filtered through a syringe filter ($0.45 \mu\text{m}$ pore diameter). The concentration of soluble COD and inorganic nitrogen compounds were measured using a HACH spectrophotometer according to the standard methods as follows: COD (closed flux colorimetric method), ammonia (including both $\text{NH}_4^+\text{-N}$ and $\text{NH}_3\text{-N}$, Salicylate method), nitrite (diazotization method), and nitrate (chromotropic acid method). The pH of the solution before and after the batch cycle was measured by a pH meter.

During the tests, the cell voltages across the external resistor were measured and recorded automatically at an interval of

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