Bioresource Technology 146 (2013) 161–168

Contents lists available at SciVerse ScienceDirect

Bioresource Technology

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

Removal of ammonia nitrogen from wastewater using an aerobic cathode microbial fuel cell

Xiaoyan Zhang, Feng Zhu, Li Chen, Qin Zhao, Guanhong Tao *

College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou, Jiangsu 215123, China

HIGHLIGHTS

- Removal of NH_4^+-N was achieved by microbial fuel cells integrated with a bioreactor.
- Up to 90.2% of total NH_4^+-N removed with an initial concentration of 100 mg/L.
- The system was successfully applied to the treatment of brewery wastewater.

ARTICLE INFO

Article history:
Received 2 May 2013
Received in revised form 3 July 2013
Accepted 6 July 2013
Available online 12 July 2013

Keywords:
Microbial fuel cell
Ammonia nitrogen
Removal
Wastewater
Biological denitrification

ABSTRACT

A new system for removing ammonia nitrogen was developed, which integrated a microbial fuel cell (MFC) with an aerobic bioreactor. A three-chamber reactor consisted of an anode chamber, a middle chamber and a cathode chamber. The chambers were separated by an anion exchange membrane and a cation exchange membrane (CEM), respectively. Driven by the power generated by the MFC, NH_4^+ in the middle chamber could migrate through CEM into the cathode chamber. The migrated NH_4^+ further removed via biological denitrification in the cathode chamber. Up to 90.2% of total NH_4^+-N could be removed with an initial concentration of 100 mg/L in 98 h. Affecting factors were investigated on the removal efficiency including cathode surface area, electrode spacing, chemical oxygen demand concentration, dissolved oxygen concentration, and NH_4^+-N concentration. The system was characterized by simple configuration and high efficiency, and was successfully applied to the treatment of brewery wastewater.

1. Introduction

In recent years, with the expansion of industrial scale and intensification of human activities, emissions of domestic sewage and industrial wastewater containing ammonia nitrogen have been increased rapidly (Camargo and Alonso, 2006). The wastewater containing ammonia nitrogen may cause eutrophication (Nixon, 1995) and produce toxic substances if discharged into the aquatic eco-system (Yamamoto, 2003). Ammonia also increases chlorine consumption for water disinfection and industrial circulating water sterilization treatment. It is therefore imperative to remove ammonia nitrogen during the wastewater treatment, which could add to the difficulty and cost of the treatment (Frink, 1967).

The practice and research on the removal of ammonia nitrogen in wastewater has been carried out extensively in recent years. They can be divided into two categories: one is physicochemical treatment, including air stripping (or steam stripping) (Bonmati and Flotats, 2003), distillation, sedimentation (Battistoni et al., 1998), reverse osmosis, membrane absorption, wet air oxidation (Deiber et al., 1997), break point chlorination, electrochemical treatment, catalytic cracking, etc.; the other is biological nitrogen removal technology (Li and Zhao, 2001), including nitrification and algae farming methods. Conventional biological nitrogen removal technology has some shortcomings, such as sludge bulking (Wanner and Grau, 1988), long process, and difficulties in maintaining sufficient nitrifying biomass (Eikelboom and Grovenstein, 1998), although the theory is sound and the technology is mature. It is thus required to develop new biological techniques for effective removal of ammonia nitrogen with low cost.

Recently microbial fuel cells (MFCs) have drawn much attention for simultaneous wastewater treatment and electricity generation. MFCs are the devices that use microorganisms to convert chemical energy from biodegradable substrates to electrical energy (Logan et al., 2006). Electrons and protons are generated during the oxidization of substrates by microorganisms. The generated electrons flow from the anode via an external circuit to the cathode, where they typically combine with protons and oxygen to form water. Electricity was thus produced and harnessed by inserting a load between the two electrodes. This provides MFCs promising

* Corresponding author. Tel.: +86 512 65226517; fax: +86 512 65223409.
E-mail address: taogh@suda.edu.cn (G. Tao).

0960-8524/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.biortech.2013.07.024

potentials to generate renewable electricity while accomplishing the biodegradation of organic matters or wastes when they are assembled and integrated in wastewater treatment process. Therefore, the application of MFCs has attracted great attentions recently, especially in the field of wastewater treatment.

It has been reported that MFCs can be used for refractory wastewater treatment, such as corn stover biomass (Wang et al., 2009), steroidal drug production wastewater (Liu et al., 2012), and phenol simulated wastewater (Zhu et al., 2011b). Another important application of MFCs is desalination. In the report by Cao et al. (2009), two membranes were placed between the anode and cathode, creating a middle chamber for water desalination. When current was produced by the MFCs, ionic species in the middle chamber were transferred into the two electrode chambers, which desalinated the water in the middle chamber. MFCs also can be used for heavy metal wastewater treatment. Huang et al. (2010) achieved Cr (VI) reduction from Cr (VI)-contaminated site using indigenous bacteria as inoculum.

As a new technology, MFCs have drawn much attention for the reduction of nitrate compounds, which are the familiar pollutants in agricultural, industrial and domestic wastewater. The earliest biological denitrification using MFCs was reported in 2004 (Gregory et al., 2004). Researchers found that microorganisms catalyzed the reduction of nitrate to nitrite with cathode as the electron donor. Clauwaert et al. (2007) firstly demonstrated the electrochemical denitrification in a bio-cathode MFC. Xie et al. (2011) constructed a coupled MFC system comprising of an oxic-biocathode MFC (O-MFC) and an anoxic-biocathode MFC (A-MFC), which was utilized for simultaneous removal of carbon and nitrogen from a synthetic wastewater. O-MFC and A-MFC were used for nitrification and denitrification, respectively. An additional nitrification reactor was added in the MFC system by Virdis et al. (2008). Zhang and He (2012) constructed a unique dual-cathode (aerobic cathode and anoxic cathode) MFC device. Nitrification and denitrification were occurred in two different reaction chambers, respectively. These devices were quite complicated although effective removal of nitrate could be achieved.

In this research, a new MFC system was constructed for the removal of ammonia nitrogen from synthetic wastewater. It consisted of an anoxic-bioanode chamber, a middle chamber and an aerobic-biocathode chamber. NH_4^+ in the middle chamber migrated to the cathode by power generated by the MFC. NH_4^+ migrated to the cathode eventually became nitrogen via the further nitrification and denitrification reactions by the activated sludge in the aerobic-biocathode chamber. In the present MFC, nitrification and denitrification reactions were occurred in an aerobic cathode simultaneously. Ammonia nitrogen could thus be removed from wastewater effectively with a simple MFC construction without additional power supply. Wastewater collected from a local brewery was later used to evaluate the applicability of the developed system.

2. Methods

2.1. MFC construction

The scheme of the MFC is shown in Fig. 1. It consisted of three rectangular chambers. The chambers were separated by an anion exchange membrane (AEM) and a cation exchange membrane (CEM), and clamped together with gaskets that provide a water seal between the chambers. The membranes were purchased from Qianqiu Group Co. Ltd., Zhejiang, China. The total liquid volume of each chamber was about 1200 mL; the cross section of each ion exchange membrane was about 128 cm^2. Both anode and cathode were made from graphite plates (Wangnian Graphite Factory, Jiangsu, China) without catalyst-coating treatment. Each electrode was rectangular with a surface area of 55 cm^2. They were connected with copper wire to provide the connection points for the external circuit. All the connection points were sealed with epoxy resin. The anode and cathode were connected via an external variable resistor. The synthetic wastewater containing NH_4Cl was used in this study. In the process of ammonia nitrogen removal, Cl^- migrated to the anode chamber through the AEM and NH_4^+ migrated to the cathode chamber through the CEM with the help of the power generated by the MFC. NH_4^+ migrated to the cathode eventually became nitrogen through the nitrification and denitrification reactions by the activated sludge in the aerobic-biocathode chamber.

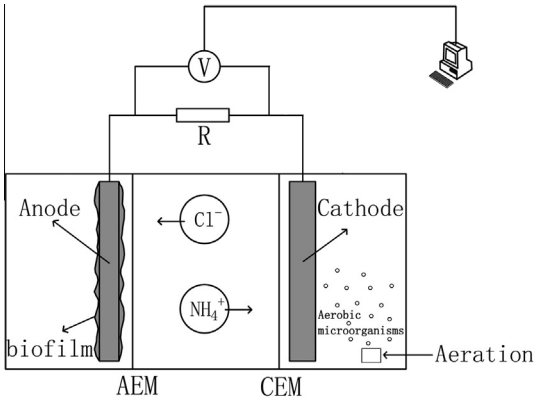

Fig. 1. Schematic diagram of the aerobic cathode MFC.

2.2. Inoculation and operation

The anode chamber of the MFC was fed by a solution of glucose (3.75 g/L) in a nutrient buffer containing (per liter in deionized water): 0.38 g NH_4Cl, 5.36 g NaCl, 0.015 g $FeSO_4{\cdot}7H_2O$, 0.026 g $MgSO_4$, 0.019 g $CaCl_2$, 0.76 mg $NiSO_4{\cdot}6H_2O$. A phosphate buffer solution (50 mM) was used to adjust its pH to 7.0. The anode of the MFC was inoculated with anaerobic activated sludge collected from the up-flow anaerobic sludge bed reactor in our laboratory (Zhu et al., 2011a), which has been run for over three years to treat synthetic glucose wastewater. The cathode chamber was fed by a solution of glucose (1.02 g/L) in a nutrient buffer (pH 7.0) containing (per liter in deionized water): 0.35 g $MgSO_4$, 0.05 g $CaCl_2$, 0.07 g NaCl, 0.07 g KCl, 1.10 g $NaHCO_3$, 1.10 g KH_2PO_4, 0.50 g Starch, 0.40 mL CH_3CH_2OH, 0.20 mL CH_3OH. The cathode of the MFC was inoculated with aerobic activated sludge. The middle chamber was filled with the synthetic wastewater containing NH_4Cl. The initial concentration of NH_4Cl was about 100 mg/L.

Aeration of the catholyte was provided by an air pump while the air-flow was manually adjusted by a flow rotameter to ensure dissolved oxygen (DO) concentration between 4.0 and 6.0 mg/L. The DO concentrations were monitored with a DO meter (SevenGo pro SG6, Shanghai, China). The external resistance (R_e) was fixed at 230 Ω, except as noted, using a resistance box (ZX21, Shanghai, China; range of 0.1–99 999.9 Ω). All experiments were conducted in a static mode. After the ammonia in the middle chamber was substantially removed, the solutions in the three reaction chambers were replaced with new ones. Unless specifically stated, the solution added in each reaction chamber volume was 1000 mL in each experiment. The MFC was thermostatically controlled at ambient temperature of 27 ± 3 °C.

Potential affecting factors on the nitrogen removal efficiency were investigated, which included cathode surface area, electrode spacing, chemical oxygen demand concentration, dissolved oxygen