



Interactive effect of electrode potential on pollutants conversion in denitrifying sulfide removal microbial fuel cells

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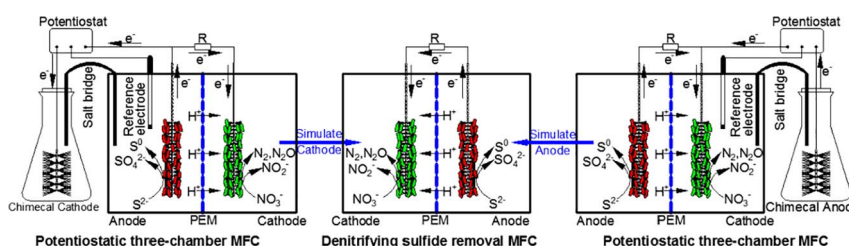
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HIGHLIGHTS

- The suitable anode potential for sulfur recovery was -188 mV to -146 mV.
- The suitable cathode potential for gaseous nitrogen formation was -181 mV to -125 mV.
- Varying external resistance achieved desirable sulfur and gaseous nitrogen formation.
- Potential effect on pollutant conversion can be studied in potentiostatic three-chamber MFC.

GRAPHICAL ABSTRACT



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ABSTRACT

The denitrifying sulfide removal microbial fuel cell (MFC) provided an approach for treatment of wastewater containing both sulfide and nitrogenous compounds, but the production of sulfate and nitrite need to be prevented. The interactive effect of electrode potential on sulfide and nitrate conversion was investigated using potentiostatic three-chamber MFC. Both abiotic and biotic processes involved in anodic sulfate formation. Rising anode potential led to an increase of sulfide removal rate and a decrease of sulfur formation percent. The suitable anode potential favoring both sulfide removal and sulfur recovery ranged from -188 mV to -146 mV. The nitrate removal rate and gaseous nitrogen formation percent showed an increase followed by a decrease along with the rise of cathode potential. The suitable cathode potential favoring both nitrate removal and gaseous nitrogen formation ranged from -181 mV to -125 mV. Regulating cathode potential through varying the external resistance was feasible, achieving the optimal sulfur formation percent ($32.4 \pm 1.9\%$) and gaseous nitrogen formation percent ($92.5 \pm 0.3\%$) at a cathode potential of -139 ± 37 mV. The potentiostatic three-chamber MFC is a suitable configuration for investigating the effect of electrode potential on pollutant conversion because its uncontrolled electrode could simulate the electron driving force situation occurring in a MFC.

1. Introduction

Microbial fuel cell (MFC) is a sustainable biotechnology to recover electricity from various inorganic or/and organic compounds, providing a novel approach for the integration of electricity generation with wastewater treatment. To date, the applications of MFC to remove various pollutants such as organics, sulfurous and nitrogenous

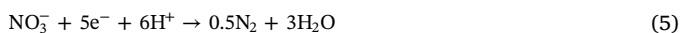
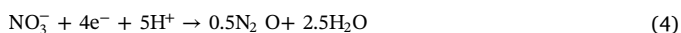
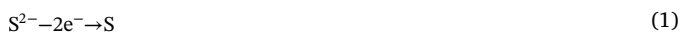
compounds from wastewater have been widely reported [1–4]. We recently explored a denitrifying sulfide removal MFC, using sulfide and nitrate separately as the anodic electron donor and the cathodic electron acceptor, to recover electricity coupled with sulfide and nitrate removal [5]. In this denitrifying sulfide removal MFC, sulfide was oxidized in the anode chamber to produce electrons, then the produced electrons was transferred through external circuit to the cathode and

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eventually utilized to reduce nitrate in cathode chamber. Some wastewaters, generated from tanneries, the mining industry, the chemical industry, swine and landfill, contained both sulfide and nitrogenous compounds at high levels [6]. This denitrifying sulfide removal MFC provided an approach for treatment of these wastewaters containing both sulfide and nitrogenous compounds. However more than 71.9% of sulfide was oxidized to sulfate in this denitrifying sulfide removal MFC, which risked second pollution due to sulfate could be biologically reduced to sulfide under specific condition.

In previous studies of MFC, attention has been focused on the pollutant removal efficiency (or rate) and electricity output. From the viewpoint of wastewater treatment, the end products of pollutants conversion should also be considered in MFCs for sulfide and nitrate removal. Sulfide oxidation involves sulfide oxidation to sulfur and sulfur further oxidation to thiosulfate, sulfite and sulfate. Sulfur and sulfate as the main end products of sulfide oxidation (Eqs. (1) and (2)) in sulfide removal MFCs have been extensively reported [3,7–9]. Sulfur is a useful resource while sulfate is a potential pollutant. Nitrate reduction involves four sequential steps in which nitrate is firstly reduced to nitrite then to gaseous nitrogen including nitric oxide, nitrous oxide, and dinitrogen gas. Besides dinitrogen gas, nitrite and nitrous oxide as the main end products of nitrate reduction (Eqs. (3)–(5)) have been frequently observed in denitrifying MFCs [10,11]. Nitrite is more toxic for human health than nitrate and nitrous oxide is a high-impact greenhouse gas [12]. Since both sulfate and nitrite risk water pollution, their production need to be prevented or controlled.



In MFCs, anodic sulfide oxidation was mediated by both abiotic and biotic processes while cathodic nitrate reduction was mediated by the biotic process [3,13]. Many sulfur-oxidizing bacteria (such as *Paracoccus* sp., *Pseudomonas* sp. and *Rhodobacter* sp.) were found to play an important role in sulfide oxidation [3]. Bacteria from Proteobacteria, Bacteroidetes, Chloroflexi, and Planctomycetes were dominant members in nitrate reduction [13]. Sulfide and nitrate conversion in MFCs were performed through a series of electrochemical reactions involving electrons transferring among substrate, microbe and electrode. Many biochemical and operational factors related to electron transferring, such as microbial community, electrode potential and co-substrate, would affect sulfide and nitrate conversion [1,8,14]. The overpotential, difference between the electrode potential and the equilibrium potential of the electrode after open circuit stabilization [15], provides driving force for electrochemical reactions. Therefore the electrode potential poses important impact on pollutants conversion in MFCs [16]. It was reported that sulfide oxidation to sulfur was potential-dependent and increasing anode potential can facilitate further oxidation of sulfur [14]. Tests under poised cathode potential revealed that increasing cathode potential led to incomplete denitrification and decreasing cathode potential reduced the accumulation of nitrite and nitrous oxide [10,16]. However, nitrite accumulation increased when the cathode potential was decreased lower than -303 mV [12].

Former studies involving pollutants conversion at different electrode potentials were commonly conducted in a potentiostatic two-chamber experiment setup [10,14,16]. The results obtained from these studies cannot precisely guide the operation of real MFCs due to two defects as analyzed below. In a MFC, the anode and cathode are commonly connected through an external circuit, and the potentials of both electrodes are interactively affected and driven. Varying potential of one electrode would affect the potential of another electrode through

the external circuit. Therefore, the electrode potential might not only pose effect on pollutants conversion in the self-electrode chamber but also pose effect on that in the other electrode chamber. However, former studies mainly focused on phenomena occurring in the controlled electrode (self-electrode) chamber. This arise the first defect: the effect of sole electrode potential on pollutants conversion in both anode and cathode chambers was not considered. The second defect is that the potentiostatic two-chamber experiment setup could not precisely simulate the electron driving force situation occurring in MFCs. In a typical potentiostatic two-chamber MFC, the overpotential depends on both electrochemical activity of controlled electrode and electron demand of potentiostat, and the electron driving force from overpotential is sufficient due to sufficient electron demand of potentiostat. This is different from the situation in MFCs, where the electron driving force (overpotential) depends on both the electrochemical activity of self-electrode and the electron demand of the other electrode [12], and is not always sufficient due to limited electron demand from the other electrode. To overcome the second defect, an alternative configuration to potentiostatic two-chamber experiment setup need to be explored.

This work aimed to study the effect of electrode potential on performance of denitrifying sulfide removal MFC, concentrating on the interactive effect of electrode potential on pollutants conversion in both chambers. The sulfide conversion pathway was firstly investigated in the potentiostatic two-chamber MFC. Then the potentiostatic three-chamber MFC, where a complementary chamber being connected to the denitrifying sulfide removal MFC to divert part of the electrons from (or to) the controlled electrode, was used to investigate the interactive effect of electrode potential on pollutants conversion in both chambers. At the end of the study, regulating electrode potential for preferable gaseous nitrogen and sulfur formation through adjusting the external resistance was tested.

2. Materials and methods

2.1. Experiment setup and procedure

The denitrifying sulfide removal MFC was constructed with two semi-cylindrical chambers served as anode and cathode chamber (Fig. 1). The two chambers were separated by a proton exchange membrane (nafion 117, DuPont, USA). A graphite fiber brush prepared with graphite fiber tying on a platinum wire was inserted in each chamber as electrode, and the net liquid volume of each chamber was 180 mL. The anode and cathode were connected with a resistor of 20Ω to close the circuit unless otherwise specified. The pretreatment processes for proton exchange membrane and electrode were stated previously [2]. During potentiostatic experiments, an Ag/AgCl reference electrode ($+0.197$ V vs. standard hydrogen electrode, SHE) was always placed in the chamber where the electrode potential being controlled. A 22 h fed-batch mode was applied as most of sulfide were removed within that time (unless otherwise stated). At the end of each batch, the substrates concentrations were examined as effluent concentrations. Then the solution in each chamber was totally replaced by corresponding fresh synthetic wastewater. The temperature was maintained at 30 ± 0.5 °C. The experiment procedures are detailed below.

2.1.1. Sulfide conversion mediated by biotic and abiotic processes under poised anode potentials

These experiments were performed using the potentiostatic two-chamber MFC where the external resistance was replaced by a potentiostat (CHI1040, Chenhua Co., Ltd., China). The anode potential was in turn poised at -300 , -200 and -100 mV to investigate sulfide conversion pathway. To distinguish abiotic sulfide conversion from biotic sulfide conversion, an identical experiment setup except for the biotic anode being replaced by an abiotic anode was used as the control. It was reported that electrochemical nitrate reduction mediated by abiotic process was negligible at an applied cathode potential of

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