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Simultaneous electricity generation and pollutant removal in microbial fuel cell with denitrifying biocathode over nitrite

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HIGHLIGHTS

- An MFC was successfully started up using nitrite as cathodic electron acceptor.
- The optimal HRT was deemed to be 8 h in this study.
- The suitable temperature for power generation was found to be 20 °C.
- The suitable temperature for pollutant removal was found to be 25 °C.
- Free buffer led to 50% decrease of TN removal rate and power generation.

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ABSTRACT

The influences of hydraulic retention time, temperature and free buffer on the performance of short-cut denitrifying microbial fuel cell were investigated after it was successfully started up using nitrite as the cathodic electron acceptor. The results revealed that a power density of 8.3 ± 0.5 W m^{-3} NC was obtained after 15 days operation. The desirable hydraulic retention time was found in this study to be 8 h, with a COD removal rate of 2.117 ± 0.006 kg m^{-3} NC d^{-1} and a total nitrogen removal rate of 0.041 ± 0.002 kg m^{-3} NC d^{-1}, respectively. It demonstrated that temperature had different effects on the electricity generation and pollutant removal performance of microbial fuel cell. The suitable temperature for power generation and pollutant removal was found to be 20 °C and 25 °C, respectively. Free buffer led to 50% decrease of both total nitrogen removal rate and power density of microbial fuel cell compared to that with phosphate buffer solution addition. The optimal total nitrogen removal rate obtained in the case with sodium azide addition (0.075 ± 0.008 kg m^{-3} NC d^{-1}) increased by 50% as compared to that without sodium azide addition. It suggested that abolishing oxygen or inhibiting nitrite oxidizing bacteria would favor nitrogen removal.

1. Introduction

Recently, microbial fuel cells (MFCs) attracted increasing attention as an economical pathway for sustainable electricity recovery [1]. In an MFC, the anode and cathode are linked by a conductive material containing a resistor. The organic substrates are oxidized by exoelectrogenic bacteria in the anode compartment to produce electrons. The produced electrons are transferred to anode electrode and then flow to cathode, eventually utilized to reduce the reducible compound in cathode compartment. Although experiencing significant development in recent years, the power generation of MFC was still insufficient for the practical applications. But it provided a sustainable technology for wastewater treatment since it can generate electricity and remove pollutant simultaneously from wastewater [2,3]. MFC being applied for various organic matter removal from wastewater had been widely reported [1]. In fact, MFC can not only convert organic matter to electricity but also utilize substances such as nitrate and sulfate as biocathodic electron acceptors [4], which shows a promise of the application of MFC for simultaneous removal of organic carbon and nitrate from wastewater.

Clauwaert et al. [5] firstly performed a complete cathodic denitrification without any power input in a denitrifying MFC (MFC equipped with a denitrifying biocathode), which demonstrated the feasibility of MFC being applied for simultaneous nitrate removal in cathode and organic matter removal in anode from wastewater. Afterwards, several configurations of denitrifying

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MFC were explored to accomplish complete nitrogen removal (converting ammonium to N_2 via nitrification/denitrification). Double-chamber MFCs coupled with a nitrifying bioreactor [2] or introduced with oxygen in the cathode were demonstrated to be effective configurations for complete nitrogen removal from wastewater [6,7]. A dual-cathode MFC, consisting of an aerobic cathode and an anoxic cathode sharing same anode, was recently reported to accomplish nitrification and denitrification [8]; but the nitrogen removal efficiency needs further improvement. Aside from configurations, the effects of temperature, pH, external resistance, substrate concentration, etc. on the performance of denitrifying MFC have been reported in many papers [9].

Nitrogen, present predominantly as ammonium in wastewater, should be oxidized aerobically to nitrate prior to being removed in denitrifying MFC. Compared to the nitrogen removal process that nitrification to nitrate followed by a reduction to dinitrogen gas, that nitrification to nitrite (i.e. partial nitrification) followed by a reduction to dinitrogen gas decreases the energy consumption (aeration) and the demand of electron donors. With respect to nitrogen removal from wastewater, denitrifying MFC could provide an opportunity of reducing the carbon demand for heterotrophic denitrification, and short-cut denitrifying MFC via nitrite reduction in cathode would further reduce the carbon demand [2]. This shows an advantage of the application of short-cut denitrifying MFC for nitrogen removal from wastewater with low C/N ratio.

Nitrogen removal via nitrate reduction in denitrifying MFCs has been reported in many papers, but little information exists in the literature about nitrogen removal via nitrite reduction in short-cut denitrifying MFCs. Moreover, those reported MFCs, in which nitrite removal was accomplished in the cathode, were formerly started up using a galvanostatic start-up strategy where a fixed current was imposed [2] or using nitrate as the cathodic sole electron acceptor [10]. In this experiment, a short-cut denitrifying MFC was started up using nitrite as the cathodic sole electron acceptor. Afterward the impacts of temperature, hydraulic retention time (HRT) and free buffer on the electricity generation characteristics and pollutant removal performance of short-cut denitrifying MFC were investigated.

2. Materials and methods

2.1. MFC construction

A short-cut denitrifying MFC was constructed with two rectangular chambers served as anode and cathode chamber. The two chambers were separated by a proton exchange membrane (nafion 117, DuPont, USA). Each chamber was filled with granular graphite (diameter: 2–6 mm, porosity: ~54%) as electrode and inserted with a graphite rod, which led to the eventual cathodic (or anodic) net chamber (NC) volume of 192 cm^3. The granular graphite has rough surface and layered structure (see Fig. 2(a)), which would favor biofilm attachment. The cathode and anode rods were connected with a manual variable resistor to close the circuit. The pretreatment process of proton exchange membrane and granular graphite was stated previously [11]. Two different influent was continuously pumped into corresponding chambers at a flow rate of 0.4 mL min^{-1} unless specified, which resulted in an HRT of 8 h. Both anodic and cathodic liquids were recirculated at a rate of 20 mL min^{-1} for mixing. All experiments were performed at ~30 °C unless specified.

2.2. Inoculation and synthetic wastewater

The anode and cathode chambers of MFC were respectively inoculated with anaerobic and anoxic sludge from Longwangzui wastewater treatment plant of Wuhan, China. The synthetic wastewater for anodic influent contained CH_3COONa (COD = 300 mg L^{-1}) and basic substrate. The synthetic wastewater for cathodic influent contained $NaNO_2$ (NO_2^-–N = 28 mg L^{-1}), $NaHCO_3$ (1 g L^{-1}) and basic substrate. The basic substrate contained: NaCl (0.5 g L^{-1}), $MgSO_4 \cdot 7H_2O$ (0.1 g L^{-1}), $CaCl_2$ (0.015 g L^{-1}), 1 mL L^{-1} trace nutrient solution [12] and phosphate buffer solution (PBS). The concentration of PBS was maintained at 50 mM unless specified.

2.3. Experiment procedure

The short-cut denitrifying MFC, with influent COD loading of 885.6 ± 15.9 g m^{-3} NC $d^{-1}$and nitrite loading of 86.1 ± 2.1 g m^{-3} NC d^{-1}, was started up at an external resistance of 100 Ω. After the stable electricity production and pollutant removal was achieved, the short-cut denitrifying MFC was firstly run successively at HRT of 8, 6 and 4 h, respectively. Then it was in turn operated at a fixed HRT of 8 h and varied temperature (15, 20, 25, 30 and 35 °C). Afterward, the short-cut denitrifying MFC was operated without buffer at a fixed HRT of 8 h and a fixed temperature of 35 °C. At the end, it was run with introducing 24 μM sodium azide to the cathodic influent to inhibit biological nitrification at 35 °C and HRT = 8 h. Under each operation condition, the polarization curve and internal resistance were obtained under steady state, as evidenced by steady cell voltage being obtained [13]. Then the external resistance was set at levels close to its internal resistance and corresponding to limiting current respectively for at least 12 h, during which stable electrogenesis parameters and pollutant removal performance were tested.

2.4. Analysis and calculations

Chemical oxygen demand (COD), total nitrogen (TN), nitrate, nitrite and pH were determined according to standard methods [14]. All analyses were carried out in triplicate except in start-up stage. After successful start-up of the denitrifying MFC, several graphite granules were respectively sampled from anode and cathode chamber and observed with scanning electron microscope (JSM-5610LV, JEOL Ltd., Japan). The pretreatment process of

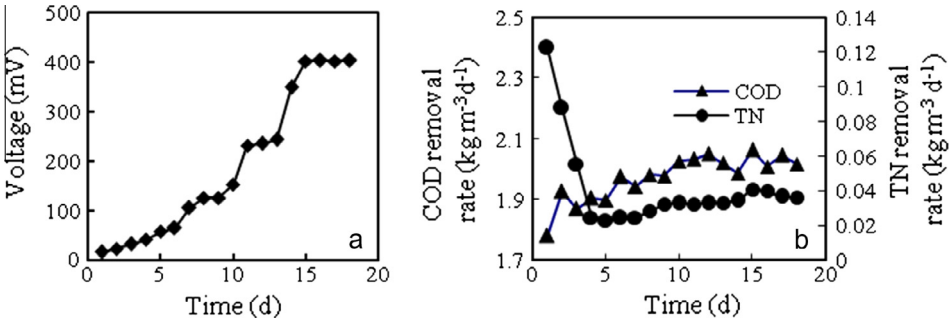

Fig. 1. Variation of cell voltage, COD and TN removal rates during start-up stage.