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Enhanced denitrification at biocathode facilitated with biohydrogen production in a three-chambered bioelectrochemical system (BES) reactor

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HIGHLIGHTS

- Denitrification was apparently enhanced in three-chambered BES reactor.
- Hydrogen content produced in CHBC reached the highest.
- Electrochemical performances were bettered in CHBC than HC and BC.
- Most abundant denitrifiers got enriched in CHBC.

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ABSTRACT

Three groups of bioelectrochemical system (BES), including BC, HBC and CHBC, were investigated for denitrification at biohydrogen production facilitated biocathode. It was found nitrate removal efficiency reached 95% during 6 days for the three-chambered CHBC reactor, while 8 and 12 days were required for HBC and BC, respectively. Besides, highest hydrogen content of 52.49% could be obtained at the biohydrogen cathode in CHBC. In addition, power density and coulombic efficiency (CE) of CHBC reached 118.43 mW m^3 and 79.1 ± 0.2%, respectively, which were better than the other two groups. Resistance indicating electron transfer of CHBC also showed to be the lowest among the three systems. Through Illumina MiSeq high-throughput sequencing, it was found that 5 core genus promoting denitrification got enriched in CHBC, including *Simplicispira*, *Thauera*, *Thermomonas*, *Azoarcus* and *Ottowia*, while only 1 core genus was enriched in HBC and BC, respectively, which suggested that the three-chambered BES structure could affect the microbial diversity as well as the community compositions, and then promoted the nitrate reduction. Moreover, as hydrogen consumers featured for nitrate reduction were found to be the most abundant at CHBC biocathode, hydrogenotrophic denitrification at biocathode could be enhanced in the three-chambered BES reactor.

1. Introduction

As a common form of nitrogenous compound in water, nitrate itself is not considered as a great threat to animals or humans. However, its reduced form of nitrite is well known to be toxic for aquatic life, and can cause diseases for human, like gastric cancer and methemoglobinemia, etc. [1]. Of the numerous chemical, physico-chemical and biological treatment methods proposed for the nitrate removal from wastewater, bioelectrochemical system (BES) based denitrification at biocathode has drawn extensive public attention featured for its environmental compatibility, low energy consumption, and high treatment efficiency [2–4].

Usually, denitrifiers could be classified into heterotrophs and autotrophs according to the utilized carbon and energy sources [1]. In contrast that heterotrophs could only use organic carbon compounds as carbon source, autotrophs can make use of reduced inorganic compounds, like ferrous iron, sulfur compound and molecular hydrogen as electron sources to obtain energy, and bicarbonate (or carbon dioxide) as the carbon source. Thus,

Abbreviations: BES, bioelectrochemical system; MEC, microbial electrolysis cell; MFC, microbial fuel cell; DGGE, denaturing gradient gel electrophoresis; EIS, electrochemical impedance spectroscopy; CEM, cationic exchange membrane; VS, volatile solids; HPLC, high performance liquid chromatography; TA, total acids; NCC, net cathode compartment; CE, coulombic efficiency; PCR, polymerase chain reaction; OUT, operational taxonomic unit; Rs, ohmic resistance; Rct, charge transfer resistance; Rd, diffusion resistance.

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autotrophic nitrate reduction would greatly reduce the denitrification cost [5]. Using hydrogen as electron donor, hydrogenotrophic denitrification might be more promising than other autotrophic methods. With water as the only resulted by-products, hydrogenotrophic denitrification would not lead to any secondary pollution as ferrous and sulfur related nitrate reduction [5,6]. Limited by the low solubility in the aqueous phase and the hazardous nature for storage and transportation, to supply hydrogen efficiently and securely should be crucial for the large scale application of hydrogenotrophic denitrification [6,7].

Compared with other traditional hydrogen production process, anaerobic biohydrogen is more moderate to provide an inexpensive and sufficient electronic source for nitrate removal, since it can fundamentally reduce the production cost and then improve the hydrogen utilization efficiency [8]. Moreover, bioelectrohydrogenesis, namely the biohydrogen through microbial electrolysis cell (MEC) obtains the distinct advantage over traditional fermentation methods of reaching a higher hydrogen yield, and over traditional water electrolysis of running at greater energy efficiencies. MEC has also been shown to operate steadily at variant temperatures from 4 to 19 °C [9]. Hence, MEC based biohydrogen would provide more electrons for stable denitrification in BES.

To date, a variety of BES reactor configurations have been adopted for denitrification. Yan et al. studied nitrogen removal in a single-chamber microbial fuel cell (MFC) with nitrifying biofilm enriched at the air cathode [10]. A two-chambered MFC was established for nitrogen removal by a nitrifying-denitrifying microbial community on an intermittent aerated cathode [11]. Previously, Liu et al. studied reductive denitrification in two-chambered BES (Fig. 1A) and confirmed that nitrate reductive removal could be further promoted with the combination of exogenous hydrogen producers [3]. Taking into account the in-situ utilization of the electric energy generated by an MFC for hydrogen production in an MEC without external power supply [12], a MFC coupled MEC (Fig. 1B), and a three-chambered BES reactor, namely one anode coupling with a hydrogen biocathode and a denitrification biocathode (Fig. 1C), were proposed for hydrogenotrophic denitrification in this study, respectively.

To further demonstrate the biochemical denitrification process, many molecular biology methods were adopted for microbial diversity investigation. Using denaturing gradient gel electrophoresis (DGGE), Liu et al. indicated an obvious community shift after the introduction of hydrogen producers into the denitrification biocathode [3]. However, as traditional molecular techniques like DGGE lacks sufficient sequences to obtain comprehensive and systematic information on microbial populations in large scale, high-throughput sequencing were better to discover diverse microbial communities [13,14]. Based on 454 high-throughput pyrosequencing, Kondaveeti et al. investigated that *Proteobacteria* and *Firmicutes* could be enriched in the BES cathode for denitrification with NO_3^--N and NO_2^--N as electron acceptors, respectively [15], while Sotres et al. revealed that the simultaneous nitrifying-denitrifying microbial community at the BES cathode could be due to the dominance of *Nitrosomonas* sp. for nitrification and *Comamonadaceae* phylotypes for denitrification, respectively [11]. As the most powerful and efficient high-throughput sequencing system for metagenomics exploration, Illumina has been adopted to research microbial community structure for denitrification in a solid-phase denitrifying biofilter and a partial nitrification SBR, respectively [16,17]. However, few literatures could be tracked about the microbial diversities for denitrification at BES biocathode using Illumina based large scale sequencing.

In this study, hydrogenotrophic denitrification were conducted at the biocathodes with hydrogen from external fermenter (BC),

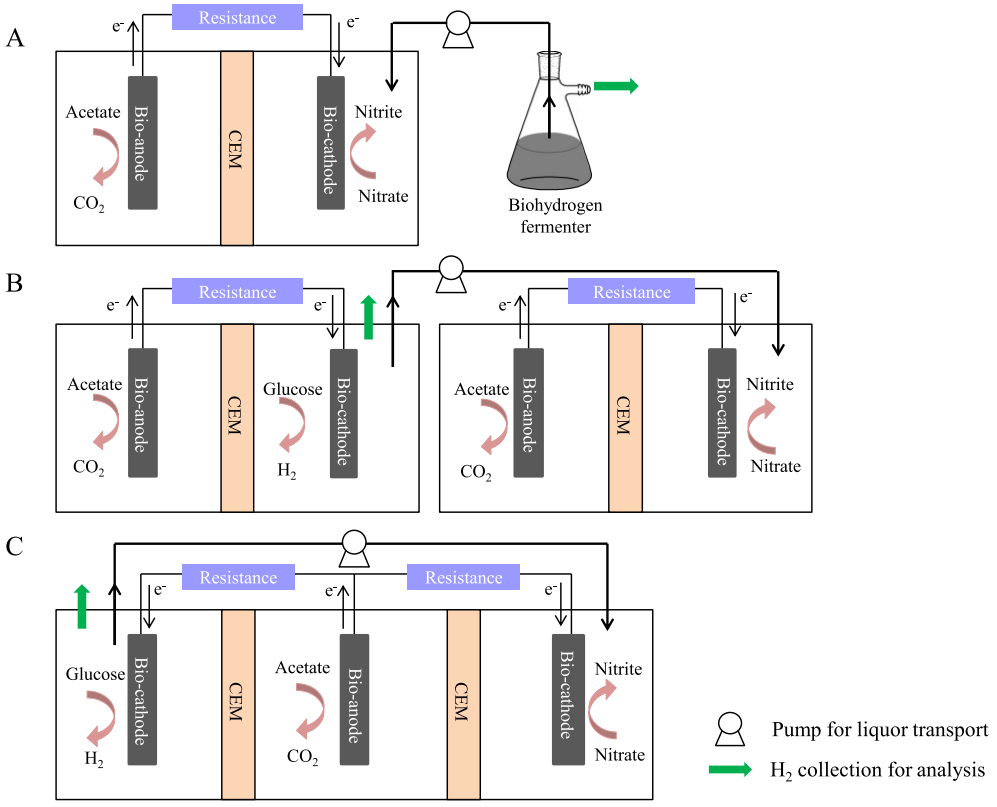

Fig. 1. Schematic diagram of BES reactor for denitrification: (A) BC was double-chambered; (B) HBC was consist of two double-chambered reactors; (C) CHBC was a three-chambered system; and all electrode were made of graphite fiber fixed on titanium wire.