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# Leachate treatment using anoxic/oxic-bioelectrochemical reactor with intermittent aeration



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#### 1. Introduction

Leachate is wastewater composing of complicated organic and inorganic matters with high levels of heavy metals. Treatments of complicated industrial leachates need intensive processes at high operational costs [1–6]. Microbial fuel cells are a device that can convert electron donors and electric acceptors to their corresponding oxidized and reduced forms with external electricity [7–9]. Bioelectrochemical system (BES) reactors were proposed promising for removing chemical oxygen demand (COD) and harvesting electricity from landfill leachates [10–12]. You et al. [10] first applied a singlechambered microbial fuel cell (MFC) of 40 mL and a dual-chambered MFC of 125 mL with proton exchange membrane (PEM) to treat diluted landfill leachate, giving a COD removal of 69.5–98% and 21.2% electron recovery.

Apart from the organic and hazardous compounds, leachate contains a high ammonium concentration that could reach a few thousand milligrams per liter [13]. BES can efficiently remove organic compounds from wastewater within a reasonable time; however, the anaerobic condition in the anode of a BES does not effectively facilitate nitrification reaction [14]. Most prior BES studies focused on simultaneous energy generation and COD removal from landfills leachate, but paid little attention to expend efforts to improve ni-

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#### ABSTRACT

A membraneless, 3.5-L bioelectrochemical reactor with biocathode and anoxic/oxic zones was established for treatment of raw landfill leachate. At 24 h hydraulic retention time and 3 kg chemical oxygen demand (COD)  $m^{-3} d^{-1}$  volume loading, the tested BES could remove 86.5% of 20100 mg L<sup>-1</sup> (COD) and 97.9% of 1330 mg L<sup>-1</sup> NH<sub>4</sub>+-N in raw leachate. The corresponding maximum power density ( $P_{max}$ ) was 2.85 Wm<sup>-3</sup> and internal resistance  $R_{int}$  was 47.5 $\Omega$  in aerating period, and was 2.70 Wm<sup>-3</sup> and 52.5 $\Omega$  in the non-aerating period. Owing to the efficient cathodic denitrification via BES reaction, the removal of total nitrogen can exceed 90%, much higher than those reported in the literature.

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trogen removal [15–22]. In these studies, the single-chambered MFC or double-chambered MFC consisting of proton exchange membrane (PEM), catalyst-doped cathode and solutions with electron acceptors were often used. The biological nitrogen removal involves aerobic nitrification and anoxic denitrification, while the leachate feed in the literature works only passed through the anaerobic chamber where the organic matters were hydrolyzed and fermented, and the NH<sub>4</sub><sup>+–</sup> N was released from protein-like matters [23]. Hence, the nitrification of NH<sub>4</sub><sup>+</sup>–N in the tested BES was not effectively nitrified, leading to low ammonium removal rate (23–43%) [15,20,21,24].

Overall, existing studies in the literature as discussed above are either used expensive membrane and noble metal cathodic catalyst and/or with a limited volume, making the reported data not practical in applications. Besides, the reported removals of COD and ammonium are generally not satisfactory. A cost-effective bioelectrochemical reactor at low construction and operational cost and with sufficiently high COD and ammonium removals is required. This study utilized liter-scale, membraneless BES with both bioanode and biocathode and anoxic and oxic (A/O) zones to treat raw landfill leachate. Intermittent aeration at cathodic compartment was adopted for reaching nitrification and denitrification with low operational cost.

#### 2. Experimental

#### 2.1. The reactor

The reactor used was the same as that applied in [25]. The working volume of the reactor was approximately 3.5 L total. Carbon fiber

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brushes (STS40 24 K, Toho Tenax Co., Ltd, Tokyo, Japan) embedded in graphite granules (Jiuxin Carbon Goods Co., Jilin, China) were the cathodes and anodes.

500 mL raw leachate was fed into the anodic compartment without dilution at the beginning a testing day. The cathode chamber was intermittently aerated at 500 mL min<sup>-1</sup> in cycles (30 min aeration+90 min no aeration). All the other testing procedures were the same as those adopted in [25].

#### 2.2. Leachate treatment tests

Landfill leachates were collected from a landfill site at Taiyuan, China, and the quality remained stable throughout the whole experiment. The leachates had a mean COD of 20100 mg L<sup>-3</sup>, biochemical oxygen demand (BOD) of 9040 mg L<sup>-3</sup>, total organic carbon (TOC) of 7450 mg L<sup>-3</sup>, NH<sub>4</sub><sup>+</sup>–N of 1330 mg L<sup>-3</sup>, total nitrogen (TN) of 1500 mg L<sup>-3</sup>, alkalinity of 6720 mg L<sup>-3</sup>, and pH 7.4.

The BES was started up by filling raw leachate to anodic compartment and 1:100 leachate-water mix at 1 kg COD·m<sup>-3</sup> d<sup>-1</sup> for 20 d. After the 20 d acclimation period the BES could stably generate current. Then the loadings of feed were increased incrementally from 1 to 2–4 kg COD·m<sup>-3</sup> d<sup>-1</sup>, each with a testing period of 10 d. The reactor was fed in batch at hydraulic retention time of 24 h. All experiments were conducted at room temperatures.

#### 2.3. Analytics and computations

A 32-channel data acquisition system (PISO-813, ICPDAS, Co., Ltd., Beijing, China) was applied to record the voltage difference between the cathode and anode across  $100\Omega$ . The potentials of the cathodic and the anodic electrodes were monitored with Ag/AgCl reference electrodes (+0.197 V vs. standard hydrogen electrode (SHE)) (model RE-5B, BASi, Ningbo, China). The polarization curves for the BES were established by linear potential decrease at  $10^{-3}$  V s<sup>-1</sup> (Parstat 263A, Princeton Applied Research, Oak Ridge, TN, USA). The power density obtained from the BES was calculated by potential and resistance values normalized by anodic liquid volume. The slope of the polarization curve determined the internal resistance ( $R_{int}$ ) of the cell.

When fresh land fill were fed to the anode chamber in batch model, the equal quantity effluent was discharged from the cathode chamber and used to analyze the water quality index. The TOC in the filtrate was analyzed using TOC-5000 Total Organic Carbon Analyzer (Shimadzu, Kyoto, Japan). Analyses on pH, concentrations of alkalinity, COD, TN,  $NH_4^+$ –N,  $NO_2^-$ –N and  $NO_3^-$ –N were performed according to Standard Methods [26].

#### 3. Results and discussion

#### 3.1. BES performance

The polarization curves and power density vs. current density curves for BES operating at 1–4 kg·COD m<sup>-3</sup> d<sup>-1</sup> feed are shown in Fig. 1, measured in the aerating period and in the non-aerating period. At 1 kg COD m<sup>-3</sup> d<sup>-1</sup>, the maximum power density ( $P_{max}$ ) and  $R_{int}$  were 2.53 Wm<sup>-3</sup> and 67.2 $\Omega$ , respectively, in the aerating period, and were 2.45 Wm<sup>-3</sup> and 70.8 $\Omega$  in non-aerating period. When the loading was increased to 2 or 3 kg COD m<sup>-3</sup> d<sup>-1</sup>, the corresponding  $P_{max}$  and  $R_{int}$  were respectively increased and decreased. At 3 kg COD m<sup>-3</sup> d<sup>-1</sup>,  $P_{max}$  and  $R_{int}$  were 2.85 Wm<sup>-3</sup> and 47.5 $\Omega$ , respectively, in the aerating period, and were 2.70 Wm<sup>-3</sup> and 52.5 $\Omega$  in non-aerating period. As the loading was further increased to 4 kg COD m<sup>-3</sup> d<sup>-1</sup>,  $P_{max}$  was declined while  $R_{int}$  was increased. The  $P_{max}$  in aerating period was always higher than that in non-aerating period, since in the former O<sub>2</sub> was the electron acceptor while in no-aerating period, the NO<sub>3</sub><sup>-</sup> was the electron acceptor, with O<sub>2</sub> having higher redox potential than NO<sub>3</sub><sup>-</sup> [27].



Fig. 1. Polarization curves and power output vs. current plots of BES using landfill leachate as substrate at of 1 (A), 2 (B), 3 (C), and 4 kg COD·m<sup>-3</sup> d<sup>-1</sup> (D).

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