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Applying multiple bio-cathodes in constructed wetland-microbial fuel cell for promoting energy production and bioelectrical derived nitrification-denitrification process

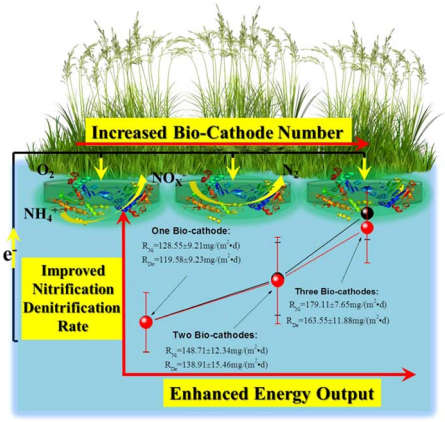
Lei Xu[a,b], Yaqian Zhao[a,b,c,*], Xiaodi Wang[b], Wenzheng Yu[d]

[a] State Key Laboratory of Eco-Hydraulic Engineering in Arid Area, Xi'an University of Technology, Xi'an 710048, PR China
[b] UCD Dooge Centre for Water Resources Research, School of Civil Engineering, University College Dublin, Belfield, Dublin 4, Ireland
[c] Key Laboratory of Subsurface Hydrology and Ecology in Arid Areas (Ministry of Education), School of Environmental Science and Engineering, Chang'an University, Xi'an 710054, PR China
[d] Department of Civil and Environmental Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, UK

HIGHLIGHTS

- Multiple bio-cathodes were applied into CW-MFC for enhanced energy production.
- The increased bio-cathode number helped to decrease both ΔV_{ohmic} and $\Delta V_{capacitive}$.
- The engagement of bio-cathode improved the nitrogen removal in CW.
- Energy production in CW-MFC directly influenced both r_{Ni} and r_{De}.

GRAPHICAL ABSTRACT



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ABSTRACT

A multiple bio-cathodes constructed wetland-microbial fuel cell (CW-MFC) system, aiming for higher power production and therefore the improved N removal, was investigated in this study. As the bio-cathode number increased from 1 to 3, the energy losses on both anode and cathode showed a significant decrement (from 97.85 mV to 46.09 mV for anode and from 221.5 mV to 45.89 mV for cathode, respectively). Accordingly, the maximum power density of the system showed a notable increase from 12.56 to 26.16 mW/m^2. In addition to the improved electrical performance, enhanced simultaneous nitrification & denitrification process was triggered due to the influence of the bioelectrical derived interaction between power production and systematic nitrification rate (r_{Ni}) and denitrification rate (r_{De}). Insight into the nitrification & denitrification process has been given that r_{Ni} increased from 98.59 ± 4.53 mg/(m^2·d) of the control system to 179.11 ± 7.65 mg/(m^2·d) of three bio-cathodes system while r_{De} increased from 89.64 ± 4.57 mg/(m^2·d) to 163.55 ± 11.88 mg/(m^2·d). Correction analysis showed that the amount of electrical related nitrogen removal is almost

* Corresponding author at: State Key Laboratory of Eco-Hydraulic Engineering in Arid Area, Xi'an University of Technology, Xi'an 710048, PR China.
E-mail address: yaqian.zhao@ucd.ie (Y. Zhao).

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linearly correlated to the produced electricity. Overall, this study presented a promising strategy and provided insight for higher energy production and the enhanced nitrogen removal from the newly established CW-MFC system.

1. Introduction

Evidence is mounting that water and energy are two major challenges in 21st century. Therefore, it is urgent to explore some self-sufficient wastewater treatment processes to confront the future scenario. Microbial fuel cell (MFC) utilizes bioelectrochemistry to treat the wastewater and concomitantly achieves power generation, presenting a promising option to establish a potential energy-neutral or even energy-positive platform for wastewater treatment [1,2]. Additionally, in order to further expand its practical application and commercialization, many integrated processes based on the core of MFC were proposed and verified in recent years [3–5]. Among them, constructed wetland-microbial fuel cell (CW-MFC), working under the synergy between CW and MFC, shows the prospect for traditional CW's updating. Since this integration owns the unique two-prong features of treatment efficiency enhancement and extra energy/electricity production [6–10].

In terms of CW-MFC, one of the main targets during its development is to improve the electricity production. To reach this goal, many attempts have been made during the last several years. Liu et al. (2014) optimized the cathode configuration and found that the granular activated carbon coupled stainless steel mesh (SSM) achieved the highest "maximum power density" (MPD) compared to only SSM or carbon cloth coupled SSM [11]. Furthermore, Doherty et al. (2015) revealed that a simultaneous upflow-downflow feeding regime contributed to a 70% higher MPD compared to the solely upflow regime [12]. In addition, the aeration rate and macrophytes in CW-MFC were also found to be the important issues determining the performance of the output power [13]. Though multiple cathodes have been utilized in pure MFC studies, they were connected separately rather than parallel connection [14,15]. More significantly, the multiple bio-cathodes system has never been tested in the newly emerged CW-MFC system before, for the purpose of achieving higher electricity production.

Meanwhile, another important issue of integrating MFC into CW was to strengthen the treatment efficiency. Fang et al. (2013) found that the MFC integration improved 12.6% removal of chemical oxygen demand (COD) (using a bio-recalcitrant azo dye (ABRX3) as the substrate (at COD of 180 mg/L)) [16], while this value was reported in the range of 12–20% (0.5–0.75 g/L glucose load) [17]. In addition, Doherty et al. (2015b) also revealed that 33% of the total COD is removed at the anode chamber which only occupies 13.6% of the liquid volume of the system [12]. These all implied that the produced electricity within CW-MFC could be used to stimulate the oxidation of the organics, and thus contributed to the improvement of the overall COD removal. In addition to COD, the removal of nitrogen (N) in wastewater is also an important target. It is well known that the transformations of N in treatment wetlands are generally consisted by nitrification, denitrification and sometimes anaerobic ammonium oxidation process [18–20]. Since the electrons in CW-MFC were artificially collected and accepted by embedding the electrodes, the electrons pathway must have a close relationship to the N transformations, which indicates the possibility of applying MFC into CW for enhanced N removal. However, reported studies on CW-MFC so far were not addressing the relationship between the produced electricity and the N removal, which should be a significant aspect to be considered for the practical application of CW-MFC.

Herein, a lab-scale CW-MFC system was set up to investigate the influences of multiple bio-cathodes system on power output. Thereafter, focuses were placed on the systematic evaluation of the relationship between the N removal and the bioelectricity generation. Interestingly, an enhanced simultaneous nitrification denitrification (SND) process has been identified in this system. Overall, this study presented a promising approach for possible large scale CW-MFC construction towards achieving both higher energy recovery and improved N removal.

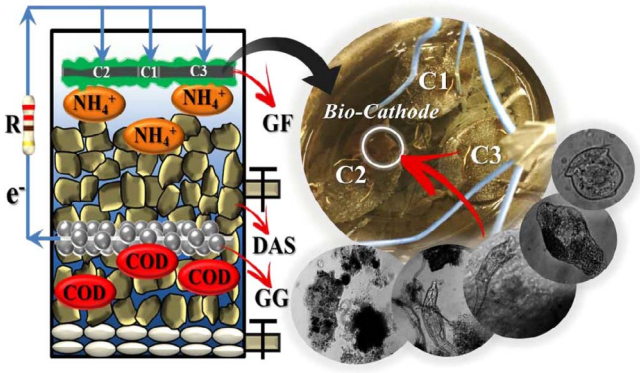

Fig. 1. Schematic diagram of multi bio-cathodes CW-MFC system; GF: graphite felt; GG: graphite gravel; C1, C2 and C3 represent three bio-cathodes (not in scale).

2. Materials and methods

2.1. CW-MFC construction and inoculation

A lab-scale CW, with dimensions of ϕ 0.15 m × 0.32 m, was set up as shown in Fig. 1. Dewatered alum sludge (DAS), collected from Ballymore Water Treatment Plant, Dublin, Ireland, treating reservoir water using aluminum sulphate as coagulant, was used as the main wetland substrate. It should be noted that DAS has been studied as a promising CW substrate [21], which has no potential risks in either aluminum or organics releases [22,23]. In addition, the existence of aluminum hydroxide or related hydrolysis products will not participate in the redox reactions in CW-MFC due to their lower reduction potentials:

$$Al(OH)_3 + 3e^- \rightarrow Al + 3OH^-,$$

$$-2.31\ \text{V vs SHE (standard hydrogen electrode)};$$

$$Al(OH)_4^- + 3e^- \rightarrow Al + 4OH^-, -2.328\ \text{V vs SHE}.$$

The average particle size of the DAS was 10–15 mm. The distance between DAS surface to the top of the reactor is 12 cm while the water surface is 3 cm higher than the DAS surface. The three carbon felt cathodes (diameter 70 mm) were located at the air-water interface. A stainless steel wire was interwoven through the felt to serve as the current collector. The anode consisted of a layer of graphite granules (30 mm height) buried under the cathode using a stainless steel mesh (SSM) as the current collector. The distance between the anode and cathode was around 100 mm. The boundary of cathodic and anodic compartment was recognized by the outlet port installed on the reactor, which located at 0.15 m below the top of the reactor. This arrangement resulted in a net liquid volume of cathodic compartment of around 0.65 L and anodic compartment of approximately 0.70 L, respectively. The anode and cathode were connected by insulated titanium wire through an external circuit with a load of 1000 Ω. At the very bottom of the system, a depth of 30 mm gravel with average diameter of 5 mm was filled to improve the distribution of the wastewater in the wetland. Prior to the start-up of the systems, a period of one month was used for the inoculation of the anodic compartment with anaerobic digestion sludge sourced from Ringsend Wastewater Treatment Plant, Dublin,