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Enhance performance of microbial fuel cell coupled surface flow constructed wetland by using submerged plants and enclosed anodes

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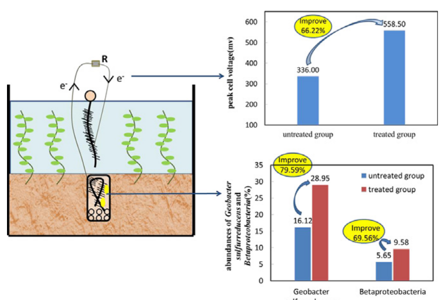
HIGHLIGHTS

- Usage of submerged plants and enclosed anodes significantly improved the peak cell voltage of CW-MFC system.
- NH_4^+-N removal rate of CW-MFC system with submerged plants was 31.25% higher than that without plants.
- Much higher abundances of electrogenic bacteria were observed on anode of treated CW-MFC system.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, submerged plants and enclosed anodes were used to improve the performance of surface flow constructed wetland coupled microbial fuel cell system (CW-MFC). The peak cell voltage produced from CW-MFC planted with *Hydrilla verticillata* and equipped with enclosed anode was 558.50 mV, 66.22% higher than that of the system without these factors. Positive correlation between cell voltage and abundances of *Geobacter sulfurreducens* and *Betaproteobacteria* was obtained. Submerged plants could overcome the shortage of immoderate elongation caused by emerged plants' roots which would ruin electrode materials, and thus enhance power generation of CW-MFC. More importantly, system with enclosed anodes showed higher cell voltage, mainly because of the higher rates of diffusion in sediments and better advection of porewater to anode materials. In addition, the planted system showed the highest NH_4^+-N removal rate, which was 88.92%, much higher than that of the unplanted system, which was 67.75%. In summary, submerged plants and enclosed anodes could improve the power generation of CW-MFC, along with better removal of NH_4^+-N.

1. Introduction

A microbial fuel cell system (MFC) is a type of fuel cell that generates power by the microbiological degradation of organic matter while removing pollutants. The MFC usually consists of an anode, a cathode, a separator and an external circuit. Anodes are put into an anaerobic medium to act as a soluble electron receptor. *Electrogenic* bacteria around the anodes can degrade organic matter and release electrons to the anodes. These electrons arrive at the cathodes through the external circuit, and the protons that are released during the oxidation of the organic matter come to the cathodes through the separator or the wastewater. At the cathodes, the final electron acceptor reacts with these electrons and protons to finish a reduction reaction [1–3]. For the MFC, the redox gradient of the anodes and cathodes is critical

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for power generation. Oxygen is widely used as the final electron acceptor due to its high availability and redox potential [4]. Thus, increasing the dissolved oxygen around the cathodes is one effective way to enhance the MFC's performance.

A constructed wetland (CW) is a type of artificial wastewater treatment system that can remove pollutants by physical, chemical and biological actions. Due to the advantages of easy operation and maintenance, low cost, and good purification capacity, CWs have been widely used [5]. The upper parts of the CW near the surface receive oxygen from the atmosphere, and the plant roots' radial oxygen loss contributes to the increase in oxygen in the inner zones near the roots. Thus, these parts have high oxidation-reduction potential [6]. The deeper parts and the areas that are far from the surface or plant rhizosphere have a low oxidation-reduction potential, indicating that the CW could provide a redox gradient, which is the necessary condition for the MFC to generate power [7].

The MFC inside the constructed wetland system (CW-MFC) can use the dissolved oxygen released by the photosynthesis of the wetland plants and the organic matter that comes from the wastewater, or the sediment of the wetlands, to finish the generation of power in the sites. Many previous studies have proved that plants play an important role in the CW-MFC system [8]. Plants provide oxygen and organic matter for the rhizosphere, in which these substances can be used by a wide range of microbes. They also do well in offering places to which microbes can attach [9]. Liu et al. [10] has found that the power produced in a planted system was 142% higher than that of an unplanted CW-MFC system, which was 5.13 mW/m^2.

Nonetheless, these CW-MFCs mainly focus on combining the MFC with a subsurface flow constructed wetland planted with emergent aquatic plants [11]. However, as the emerged plants grow, the immoderate elongation of the plant roots can ruin the anodes. In addition, the oxygen transferred to the anode zones by the roots is harmful to the anodes [12]. In contrast, the roots of submerged plants are not as flourishing as those of the emerged plants. Thus, the oxygen transferred by the roots and the possibility of ruining the anodes decrease. In addition, submerged plants can offer oxygen through the leaves and stems, which can add DO around the cathodes and dramatically increase the oxidation-reduction potential and power generation.

In many CW-MFC systems, the anodes were buried into the anaerobic environment of CW particularly wetland sediment [13]. The organic matter that comes from the sediment and the deposition of organic compounds in the rhizosphere of living plants can be used by the microbes around the anodes. There are many shortages, such as low rates of diffusion in sediments, low concentrations of labile organic matter, and passivation of anode surfaces, that occur by putting the anodes directly into the sediments [14,15]. Nielsen et al. [16] reported that in a benthic microbial fuel cell system, it is effective to enclose the anodes in a benthic chamber. It can realize higher rates of diffusion in sediments and better advection of reductant-rich porewater to anode materials. In the CW-MFC system, this may also be an effective way to reduce the limitation of slow diffusion in sediments and enhance electricity.

In this study, we examined the role of submerged plants and the placement of the anodes. These two factors were combined in different ways to achieve different performances of both the power generation and the pollutant removal. The removal rates of the COD, NH_4^+-N, NO_3^--N, and TP were measured to study the removal efficiencies of this novel CW-MFC system. Tests on the power generation and other electrochemical indices were conducted to better study the MFC performance. In addition, the abundances of *Geobacter sulfurreducens* and *Betaproteobacteria* in the anode biofilm were estimated using the fluorescence in situ hybridization (FISH) technique.

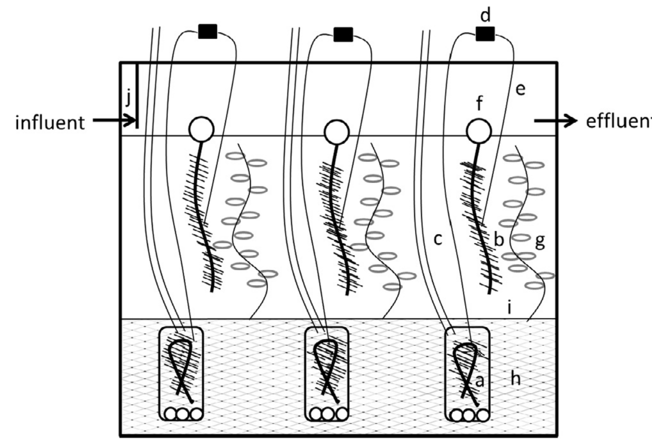

Fig. 1. The structure of the CW-MFC. (a) anode; (b) cathode; (c) gas vent; (d) external resistance; (e) tire wire; (f) floater; (g) *Hydrilla verticillata*; (h) sediment; (i) wastewater; (j) water baffle.

2. Materials and methods

2.1. CW-MFCs configuration and electrode preparation

As shown in Fig. 1, the CW-MFC reactors were made of organic glass (40 cm in height, 100 cm in length and 80 cm in width). Six anodes were placed in each CW installation. As one type of submerged plants, *Hydrilla verticillata* was picked because it is inexpensive and effective in terms of survival. Well-grown *Hydrilla verticillata* with a length of approximately twelve centimetres were selected to be planted in a configuration. For one installation, *Hydrilla verticillata* was planted; for the other reactor, no plant was planted, and it was covered by black-out cloth to prevent the growth of plants. The two reactors were operated under the same conditions of continuous flow.

The CW was designed as a surface flow. The sediment was collected from the Xiaomei Wetland in Shandong, China (116.1°E, 36.5°N). The sludge below the surface at approximately 10 cm was collected. The clumps in the sludge were picked out. The sludge was left alone for more than ten minutes. Then, the supernatant was drained off. Last, the sludge was sifted by a 200-mesh sieve. Each reactor was filled with 100 L of collected sludge.

The electrode material was a carbon fibre brush consisting of fine carbon fibres densely packed on twisted–pair titanium wires [17]. These materials had a surface area of 26.3 m^2 per metre [18] and were pre-treated before use. Deionized water was used to wash the surface of these materials several times until the water was clean. Next, the materials were submerged in 5% ethanol for 24 h. Then, they were dried in a drying oven at 100 °C. Lastly, these materials were put into a muffle at 450 °C for one hour. The anode chamber was fabricated from polyethylene material with a net volume of 565.3 cm^3. Thirty small holes with a diameter of 1 cm were made in the bottle of the anode chamber to make the sediment porewater flow in. In the top of the anode chamber, a rubber tubing was fixed to connect the inside of the bottle to the air. The anodes had two different methods of placement to study the influence of the enclosed anodes. In one method, the anodes were placed into an enclosed chamber; in the other method, the anodes were placed directly into the sediment. Each type of MFC had three parallel tests.

2.2. Inoculation and operation of CW–MFCs

Before operation of the CW-MFCs, the anode materials were inoculated by anaerobic sludge collected from the *Everbright Water Wastewater Treatment Plant*, located in Jinan, Shandong (117.0°E, 36.7°N), China. The anodes were put into an organic glass cylinder with