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Electricity generation from sediment microbial fuel cells with algae-assisted cathodes

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ABSTRACT

One major limiting factor for sediment microbial fuel cells (SMFC) is the low oxygen reduction rate in the cathode. The use of the photosynthetic process of the algae is an effective strategy to increase the oxygen availability to the cathode. In this study, SMFCs were constructed by introducing the algae (*Chlorella vulgaris*) to the cathode, in order to generate oxygen *in situ*. Cyclic voltammetry and dissolved oxygen analysis confirmed that *C. vulgaris* in the cathode can increase the dissolved oxygen concentration and the oxygen reduction rate. We showed that power generation of SMFC with algae-assisted cathode was 21 mW m^{-2} and was further increased to 38 mW m^{-2} with additional carbon nanotube coating in the cathode, which was 2.4 fold higher than that of the SMFC with bare cathode. This relatively simple method increases the oxygen reduction rate at a low cost and can be applied to improve the performance of SMFCs.

Introduction

A sediment microbial fuel cell (SMFC), a device used to harvest electricity by locating the anode in the anaerobic sediment phase and suspending the cathode in overlying aerobic water, is an adaptation of the reactor-type MFCs [1,2]. As a power source, SMFCs have the advantages of maintenance-free operation, long-term power generation and powering devices in remote areas [3,4]. In addition, SMFCs could be applied to *in situ* sediment remediation; they do not require dosing with chemical compounds as electron acceptors and use electrodes as continuous long-term electron acceptors for removing organic matter in the sediment [5–7].

One of the drawbacks of SMFCs, which prevents their widespread usage, in practice, is the limitation on the output current. Several methods were implemented to improve the mass transfer rate between electron donors and the anodes of SMFCs, such as plant rhizodeposits [8,9], the addition of biomass [10,11] and anode modification [12]. For most SMFCs, oxygen is ubiquitous in the overlying

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water, and cathodes complete the circuit by reducing oxygen to water. Therefore the oxygen reduction rate in cathodes is another important limiting factor of SMFCs. One common method is the application of a catalyst at the cathode to improve the oxygen reduction rate [13,14]. Another effective method would be to increase the oxygen availability to the cathode. He et al. [15] applied a rotating cathode resulting in a higher power production (98 mW m^{-2} based on anodic electrode footprint area (EFA)) compared to a nonrotating cathode system (58 mW m^{-2} EFA). However, a rotating cathode consumes extra energy, which makes it unsuitable for application in the field.

Chlorella vulgaris is one of the fastest growing algae and can fix CO_2 from the environment during photosynthesis to accumulate lipid [16]. In addition, the photosynthetic process of *C. vulgaris* can release oxygen as a byproduct. Thus, *C. vulgaris* can be added into the cathodic chamber for improving the performance of MFC [17–19]. Furthermore, the graphite cathodes that are commonly used in SMFCs are unable to fully exploit oxygen, while carbon nanotube could be used as electrode materials to promote electron transfer reaction due to its high electrical conductivity and large surface area. Therefore, the immobilization of *C. vulgaris* on carbon nanotube offers a new strategy to fabricate cathode structure for improving the performance of SMFC.

In this study, we used a simple dipping-drying method to construct carbon nanotube modified cathode. The algae-assisted cathodes were constructed by adsorbing *C. vulgaris* on both carbon nanotube modified and bare electrodes. Furthermore, the differences in performance for SMFCs using different cathodes are explained through in-depth analyses.

Materials and methods

Electrode preparation

The graphite felt (GF, Shanghai Q-carbon Material Co., Ltd), with a size of 200 mm × 50 mm × 30 mm (length × width × thickness), served as electrode material. Multi-walled carbon nanotubes (MWNT, diameter of 20–40 nm, special surface area of 90–120 m^2/g, Shenzhen Nanotech Port Co., Ltd., China) were refluxed in a mixed concentrated solution of 18 mol L^{-1} H_2SO_4 and 16 mol L^{-1} HNO_3 (with a volume ratio of 3:1) at 80 °C for 1 h. The acid-treated MWNT was then filtrated using a 0.45 μm hydrophilic polytetrafluoroethylene membrane and washed with deionized water, until the pH of the filtrate became 7, prior to being dried in vacuum.

The GF-MWNT cathodes were prepared by the dipping-drying method [20] in aqueous MWNT ink. Aqueous MWNT ink was prepared by ultrasonically dispersing acid-treated MWNT in deionized water with sodium dodecylbenzene sulfonate (SDBS) as a surfactant for 0.5 h. The concentration was 0.18% for MWNT and 1% for SDBS by weight. The GF was immersed into the MWNT ink, removed and dried at 100 °C in order to obtain a MWNT-modified electrode. In order to increase the MWNT loading in GF, the dipping-drying process was repeated for 3 times.

The *C. vulgaris* was purchased from FACHB-Collection (FACHB 1068, China). The BG11 medium for cultivating *C. vulgaris* contained the following compounds (g L^{-1}):$NaNO_3$, 1.5; $K_2HPO_4 \cdot 3H_2O$, 0.04; $MgSO_4 \cdot 7H_2O$, 0.075; $CaCl_2 \cdot 2H_2O$, 0.036; Na_2CO_3, 0.02; citric acid H_2O, 0.006; ammonium ferric citrate, 0.006; $Na_2 \cdot EDTA$, 0.001; and 1 mL of trace mineral solution at pH = 7.0. The trace mineral solution composition was listed as follows (g L^{-1}): H_3BO_3, 2.86; $MnCl_2 \cdot H_2O$, 1.81; $ZnSO_4 \cdot 7H_2O$, 0.222; $CuSO_4 \cdot 5H_2O$, 0.079; $Na_2MoO_4 \cdot 2H_2O$, 0.39; and $Co(NO_3)_2 \cdot 6H_2O$, 0.049. *C. vulgaris* was pre-cultured in an illuminated autoclaved Erlenmeyer flask on a 12:12 h light/dark cycle, using 2,000 lx fluorescent lamps. After *C. vulgaris* was continuously transferred three times, the GF-MWNT and GF electrodes were placed in the logarithmic phase culture of *C. vulgaris*. *C. vulgaris* was continuously cultivated for nine days to form a layer of biofilm on the electrodes. Finally, GF-MWNT-C (*C. vulgaris* adhesion on GF-MWNT) and GF-C (*C. vulgaris* adhesion on GF) cathodes were constructed.

SMFC construction and operation

Sediments (0–10 cm below the sediment–water interface) were collected from Jinchuan stream in Nanjing, China. The collected sediment was homogenized. The loss on ignition (LOI) of the sediment was 3.2% (w/w). SMFCs were constructed in glass beakers, 11 cm in diameter and 15 cm in height (Fig. 1). Each SMFC contained 700 g wet sediment and 650 ml simulated lake water. The simulated lake water (g L^{-1}) consisted of $K_2HPO_4 \cdot 3H_2O$, 0.0001; KH_2PO_4, 0.0002; NH_4Cl, 0.0115; $MgCl_2 \cdot 6H_20$, 0.1; $CaCl_2 \cdot 2H_2O$, 0.1; and $FeCl_2 \cdot 4H_2O$, 0.02. Each anode (graphite felt) was crimped along a polyvinyl chloride cylindrical holder (31.8 cm^2, electrode footprint area) to minimize the mass transfer limitation within the sediment. Four SMFCs with GF, GF-C, GF-MWNT and GF-MWNT-C cathodes were operated with an external circuit resistance of 1000 Ω at 25 °C. The cathode regions in the SMFCs were illuminated during the experiments by using 2,000 lx fluorescent lamps on a 12:12 h light/dark cycle. Water loss via

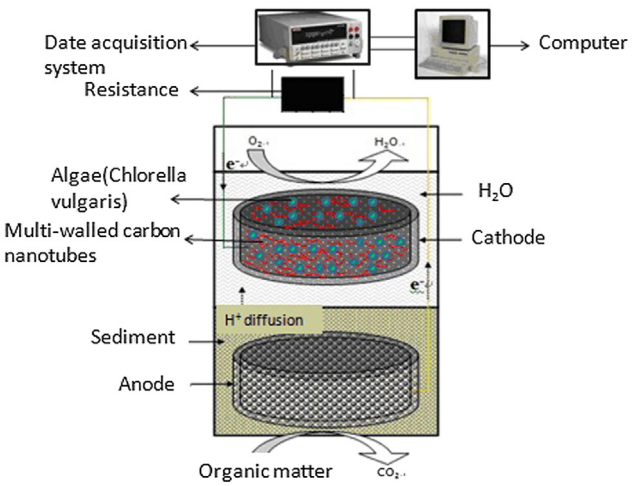

Fig. 1 – Schematic detail of configure of SMFC in this study.

evaporation during operation was routinely replenished with the simulated lake water.

Analysis

The voltages of the SMFCs were automatically measured at an interval of 10 min using a data acquisition system (Keithley Instruments 2700, USA). Polarization curves were measured by varying a variable external resistance in the range of 10 Ω–5000 Ω. The current density (I) and power density (P) were normalized to anodic electrode footprint area [1]. Internal resistance was calculated using the polarization slope method [21]. Cyclic voltammetry (CV) was performed using a potentiostat (CHI660D, Shanghai Chen hua Instrument Co, Ltd). The cathode was used as the working electrode, while the anode was used as the counter and reference electrode. The potentials were shifted from −600 mV to 600 mV at a scan rate of 20 mV s^{-1} while monitoring the current response. The electrode potentials were calculated as previously described [22], the saturated calomel electrode (SCE) as reference electrode was positioned near the cathode to measure the cathode potential. Anode potential was estimated by subtracting the voltage of MFC from cathode potential.

Dissolved oxygen (DO) was measured using a DO meter (TPSJ-605, Shanghai Leici) at a temperature around 25 °C. The LOI of the sediment was determined as previously described by weighing the sediment before and after combustion at 550 °C for 4 h [6]. The surface morphologies of the cathode surfaces were studied by a scanning electron microscope (SEM; JSM-5900, Japan). The samples were fixed in 2.5% paraformaldehyde for 3 h at 4 °C and then washed three times in a phosphate buffer solution (0.1 mol L^{-1}, pH 6.8). The samples were then washed twice by stepwise dehydration in a gradient series (50%, 70%, 80%, 90%, 100%) of water/ethanol solutions (V/V) and dried in vacuum. Finally, the samples were coated with Au/Pt before SEM observation.

Results

Electrode characterization

The surface morphology of the bare GF and modified GF cathodes were characterized by SEM (Fig. 2). The bare GF cathode had a relatively smooth and clean surface (Fig. 2A). In contrast, the surface of the MWNTs coated GF is much rougher (Fig. 2B). MWNTs were uniformly distributed over the surface of the GF, thus forming a three-dimensional network structure. Furthermore, the MWNT was measured about 4 wt % on modified GF electrode by weighing before and after the dipping-drying method. For the cathode with *C. vulgaris* adhesion, some algal cells can be clearly observed on the GF-C (Fig. 2C). Each single cell exhibits a spherical appearance, with μm level size. In contrast, a large number of spherical algal cells can be clearly observed on the GF-MWNT-C. This three-dimensional nanostructure on the GF-MWNT-C was expected to provide favorable conditions for *C. vulgaris* adhesion (Fig. 2D).

Electricity generation from the freshwater sediment

The voltages from all applied SMFCs decreased first and then sharply rose during the first 5 days of operation (Fig. 3). Afterward the voltages of SMFCs increased slowly and fluctuated in a small range. Around 12 days of operation, the maximum

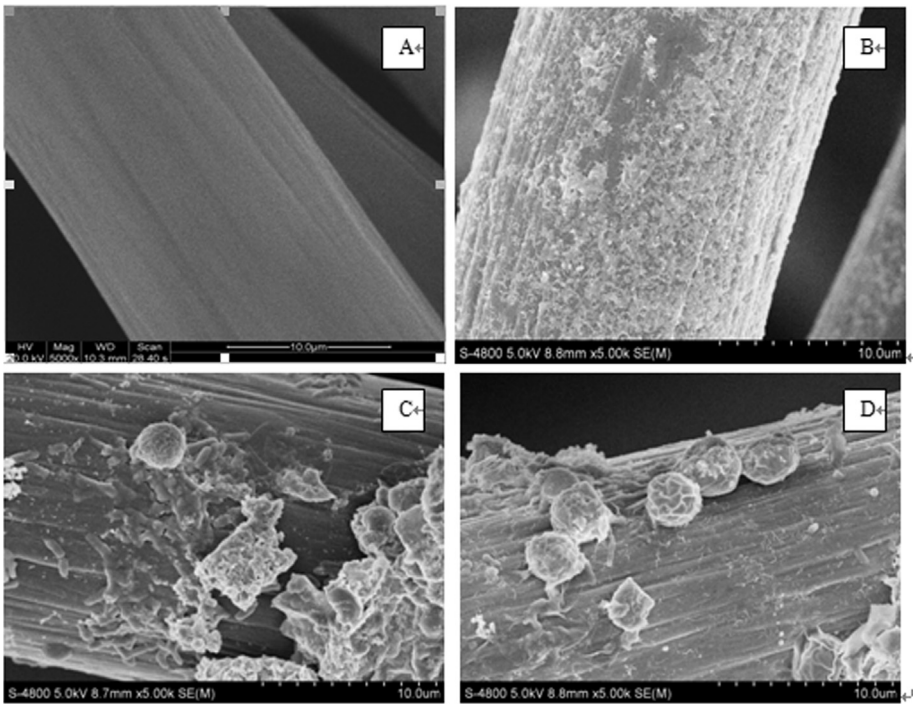

Fig. 2 – SEM of different cathode (A) graphite felt (B) MWNT modified graphite felt (C) *C. vulgaris* adhesion on graphite felt (D) *C. vulgaris* adhesion on MWNT modified graphite felt.

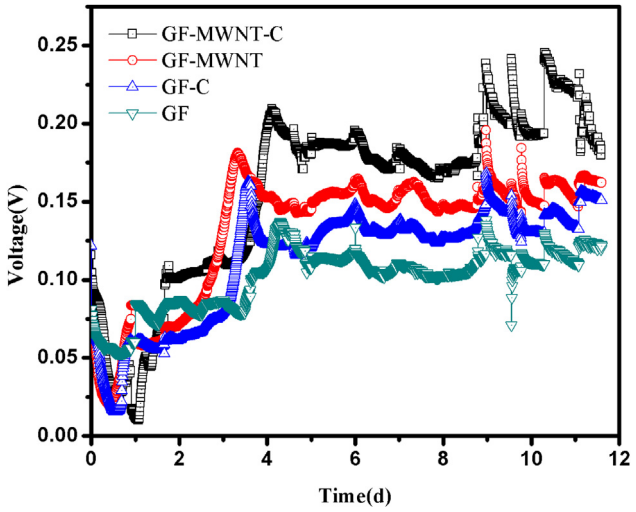

Fig. 3 – Voltage generation of SMFCs produced by different electrode configurations during the 12 days of operation.

voltage of 246 mV was produced from SMFC with GF-MWNT-C cathode, followed by SMFC with GF-MWNT cathode (193 mV) and then SMFC with GF-C cathode (166 mV). The SMFCs with bare GF cathode produced the lowest voltage (136 mV).

The maximum power density (P_{max}) and polarization curves as a function of current density of the different SMFCs were determined at the end of the experiment (Fig. 4). SMFC with GF-MWNT-C cathode generated the highest P_{max} (38 mW m^{-2}), followed by that of SMFC with GF-MWNT cathode (25 mW m^{-2}) and SMFC with GF-C cathode (21 mW m^{-2}), SMFC with bare GF generated the lowest P_{max} (16 mW m^{-2}). The P_{max} of SMFC with GF-MWNT-C cathode was 2.4 times that of the SMFC with bare GF cathode. Internal resistance was estimated from the slope of the plot of voltage versus current (Table 1).

Higher internal resistance (1823 Ω) was observed in SMFC with GF cathode, MWNT can increase electron transfer reaction and *C. vulgaris* increase the oxygen availability to the cathode, so SMFC with GF-MWNT cathode and GF-C cathode can both decrease internal resistance. Furthermore, SMFC with GF-MWNT-C cathode can obtain the lowest internal resistance (489 Ω), which is the same as synergistic effect of MWNT and *C. vulgaris* that can increase the reduction reaction from *C. vulgaris* release oxygen. The result demonstrated that a combination of algae and MWNT modified cathode can generate the highest P_{max} and the lowest internal resistance.

Electrode potential

To understand the difference in electricity generation between different SMFCs, the anode and cathode potentials versus the SCE reference were determined (Fig. 5). The anode potentials were almost the same. In contrast, there were obvious changes in cathode potential. The initial cathode potentials were negative in all SMFCs. The fastest increase in cathode potential was exhibited by the SMFC with GF-MWNT-C cathode, followed by the SMFC with GF-MWNT cathode and the SMFC with GF-C cathode. The cathode

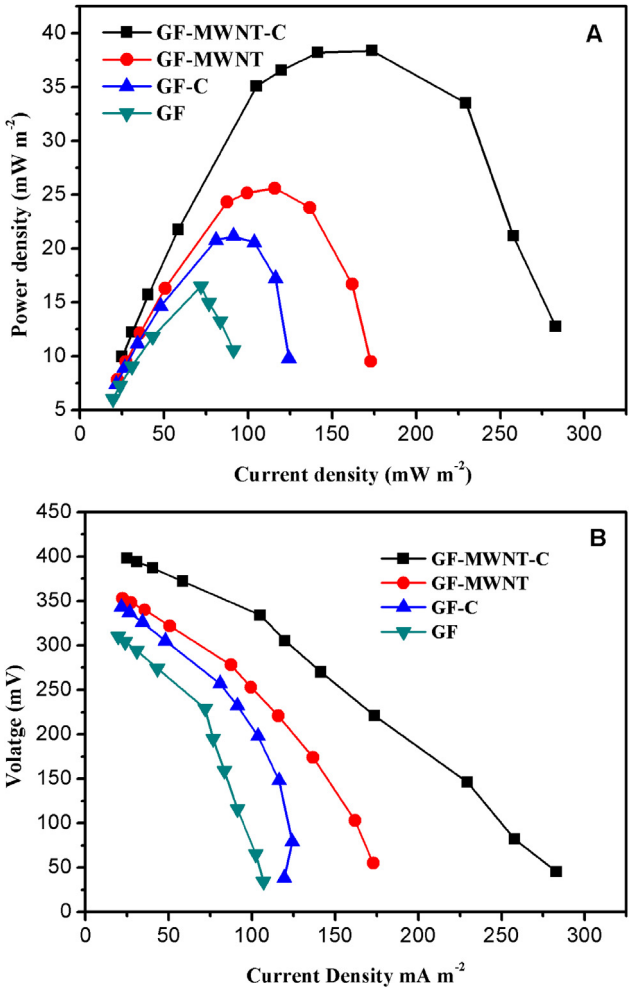

Fig. 4 – (A) power density and (B) polarization curve as a function of current density of the different SMFCs.

potential of the SMFC with bare GF increased slowly and still had low (high positive) cathode potential. Compared to the anode, the cathode potential variations were more significant. This phenomenon indicates that the differences in SMFC performance mostly come from the differences in the cathodes.

Dissolved oxygen concentration at the cathode

Algae can capture CO_2 to produce oxygen during the light phase and increase DO in water. For SMFCs, oxygen can

Table 1 – Electrochemical properties of SMFCs with different cathode.

SMFC reactors	Maximum voltage (mV)	Maximum power density (mW/m^2)	Calculated internal resistance (Ω)
GF-MWNT-C	246	38	489
GF-MWNT	193	25	667
GF-C	166	21	809
GF	136	16	1823

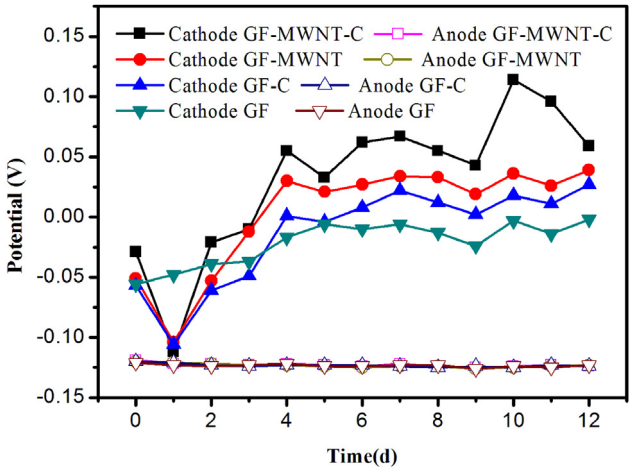

Fig. 5 – Working potential of electrodes of SMFC with different cathode during the 12 days operation.

be used as an electron acceptor for the cathodic reaction and the DO amount can affect the performance of the SMFC. Fig. 6 shows the DO change in the overlying water in SMFCs using different cathodes. DO was the same at the beginning of the experiment (3 mg L^{-1}). As the reaction proceeded, the DO in the SMFCs without *C. vulgaris* adhesion (GF and GF-MWNT) decreased and maintained in a relatively stable range (1.1–1.8 mg L^{-1} for the SMFC with GF-MWNT and 0.8–1.3 mg L^{-1} for the SMFC with GF). In contrast, the DO in the SMFCs with *C. vulgaris* adhesion (GF-C and GF-MWNT-C) continuously increased and then maintained in a relatively stable range (3.7–4.1 mg L^{-1} for the SMFC with GF-MWNT-C, 3.2–3.7 mg L^{-1} for the SMFC with GF-C). This result implies that the oxygen reduction reaction at the cathode can consume DO, while *C. vulgaris* can increase DO in water.

CV study

The oxygen reduction characterizations of the different electrodes were analyzed by CV (Fig. 7). SMFC with GF-MWNT-C cathode visualized maximum current in both forward scan (13.7 mA) and reverse scan (−17.6 mA), followed by SMFC with GF-MWNT (7.7 mA, −9.3 mA) and GF-C (6.5 mA, −14.1 mA). Relatively lower current output (2.8 mA, −0.8 mA) was recorded in SMFC with bare GF cathode. This result indicated that the material of the electrode played a key role in the catalytic behavior of the oxygen reduction reaction, which was consistent with the power generation. More interestingly, by providing a better oxygen supply, the adhesion of *C. vulgaris* promoted the oxygen reduction reaction, when the same cathode material was used.

Discussion

The algae-assisted cathode significantly promoted the P_{max} of the SMFC. The P_{max} of the SMFC with GF-C cathode was 1.3 times that of the SMFC with bare GF cathode. This result demonstrates the importance of oxygen in the performance of SMFC. The released oxygen (Fig. 6) from the photosynthetic process of the *C. vulgaris* can promote the oxygen reduction reaction of the SMFC. Furthermore, this effect of the oxygen availability strengthened, when the cathode was coated with MWNT. The P_{max} of the SMFC with GF-MWNT-C cathode was 2.4 times that of the SMFC with bare GF cathode. As shown in the DO change (Fig. 6), the oxygen consumption rate of the SMFC with MWNT was higher than that of the SMFC without MWNT. In addition, the three-dimensional nanostructure on the GF-MWNT-C (Fig. 2D) has a synergistic effect on the reduction reaction of the cathode. The algae layer benefits the oxygen generation and transportation, while the microporous MWNT layer catalyzes the oxygen reduction. In freshwater environments, the P_{max} in SMFC with GF [23] as cathode were 4 mW m^{-2}. The P_{max} of SMFC can significantly increase to

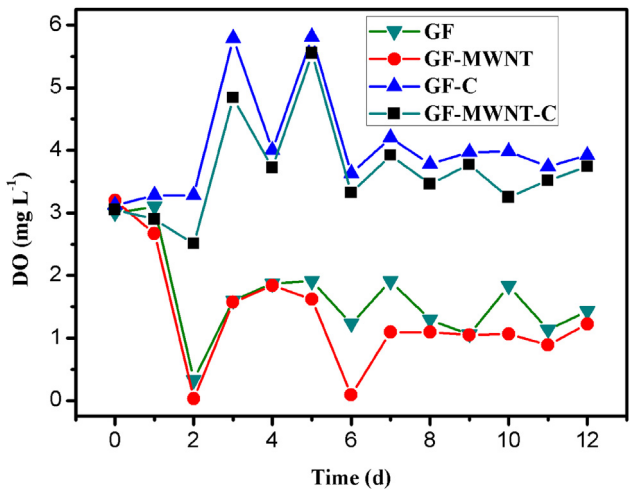

Fig. 6 – DO changes in overlying water of the SMFC with different cathode.

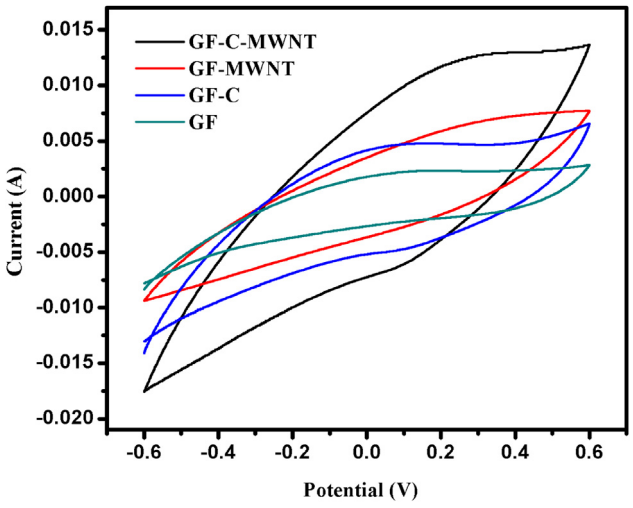

Fig. 7 – Cyclic voltammetry curve of the different cathode at the end of the experiment.

98 mW m^{-2} by using a rotating cathode [15]. This showed that the cathode oxygen reduction reaction is an important limiting factor of SMFCs. In the present study, relatively high power densities (38 mW m^{-2}) were obtained from SMFCs with GF-MWNT-C cathode. Compared to a previous reported, oxygen reduction rate in cathodes can be increased without consuming extra energy. Because the LOI in sediment was low and maintained high (low negative) anode potential throughout the experiment, the P_{max} of the SMFC (38 mW m^{-2}) in this experiment was lower by an order of magnitude than the P_{max} of the SMFC (195 mW m^{-2}) with the addition of the biomass as described previously [11]. This result implies that the transfer rate of organic matter in the anode region impacts the performance of the SMFC. Therefore, it can be expected that more power might be obtained from SMFCs, if the transfer rate of biomass was optimized in the sediment of the SMFC.

Oxygen is ubiquitous and has a relatively high reduction potential, so it is an ideal terminal electron acceptor. Aqueous cathodes were used in MFCs where water was bubbled with air to provide dissolved oxygen to the electrode. To reduce the cost of MFCs, air-cathode was installed in the MFC [24,25] and obtained the oxygen at a low cost. However, SMFC has special structures and is usually used in natural water; so air-cathode is not suitable for providing low-cost oxygen to the SMFC, therefore, other methods need to be considered for increasing the oxygen availability. This study indicates that *C. vulgaris* can increase the oxygen availability to the cathode and improve the performance of the SMFC. In addition, *C. vulgaris* absorbed on cathode in SMFC, which was different from the dual MFC with algae-assisted cathode [17–19], in which *C. vulgaris* was freely added into the cathode chamber. The free *C. vulgaris* in the SMFC might reach the anode region, releasing oxygen which will destroy the anaerobic conditions of the sediment, thus reducing SMFC output power; therefore the immobilization of *C. vulgaris* on the cathode not only provides oxygen on the cathode surface but also reduces the risk of disruption of the anode region, which was well suited to SMFC system. Furthermore, *C. vulgaris* can be cultivated in SMFC without any further addition of CO_2, as the CO_2 can be generated from the oxidation of organic matter in the sediment [26]. Therefore, this method is relatively simple and provides oxygen at a low cost. Before applying this technology in field conditions, further efforts are needed to study the effect of culture conditions and long time running on the performance of SMFCs.

Conclusions

This study shows that the introduction of *C. vulgaris* to the cathode can improve the output power of SMFC. Carbon nanotubes are an efficient catalyst, strengthening the oxygen reduction rate from *C. vulgaris*' oxygen release. The highest power production in SMFC with GF-MWNT-C cathode was 38 mW m^{-2}, which was 2.4 times that of the SMFC with bare GF cathode. Therefore, an algae-assisted cathode is an efficient *in situ* oxygenator and facilitator of the cathodic reaction in SMFCs.

Acknowledgments

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