Biochemical Engineering Journal 94 (2015) 39–44

Contents lists available at ScienceDirect

Biochemical Engineering Journal

journal homepage: www.elsevier.com/locate/bej

Regular article

Carbon nanotubes simultaneously as the anode and microbial carrier for up-flow fixed-bed microbial fuel cell

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ARTICLE INFO

Article history:
Received 1 July 2014
Received in revised form
25 September 2014
Accepted 7 November 2014
Available online 15 November 2014

Keywords:
Anaerobic processes
Biofilms
Carbon nanotubes
Fixed-bed bioreactors
Microbial fuel cell
Wastewater treatment

ABSTRACT

A novel up-flow fixed-bed microbial fuel cell (FBMFC) using carbon nanotubes (CNTs) as the anode and microbial carrier was developed for continuous treatment of wastewater and electricity generation. A maximal power density of 590 mW m^{-3} was achieved with a maximal chemical oxygen demand (COD) removal rate of 90% at an organic loading rate (OLR) of 3.94 g COD l^{-1} d^{-1}. An OLR of up to 10.27 g COD l^{-1} d^{-1} caused the overloading of FBMFC, accompanied with an unexpected decrease in voltage generation below 0.1 V and a sudden accumulation of volatile fatty acids (VFAs) up to 1.82 g l^{-1}. The overloading also led to a rapid decline in COD removal rate (72%) and a morphology change of microbial consortia confirmed by scanning electron microscope (SEM). These results demonstrated the feasibility of Carbon nanotubes simultaneously as the anode and microbial carrier for up-flow fixed-bed microbial fuel cell. The overloading of MFC suggesting that further researches are still needed on improving the performance of FBMFC for energy production and wastewater treatment.

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1. Introduction

Microbial fuel cell (MFC) is a device that harvests electricity through the oxidation of organic or inorganic compounds by microorganisms [1–3]. The anode material plays a critical role in electricity generation [4], as it is directly related to the electron transfer between the microorganism and the anode.

Carbon nanotubes (CNTs) have been implemented as the anode or the anodic modifier in MFC due to its unique physical/chemical characteristics, including excellent electrical conductivity, superior mechanical properties, high specific area and acceptable biocompatibility [5–8]. The power density has been greatly improved by using the CNTs anode with polytetrafluoroethylene (PTFE) as a binder, compared to the activated carbon or flexible graphite anodes [9]. CNTs could be also spray-coated on a carbon cloth as the electrode material to strengthen the electron transfer [10]. One study [8] used carbon felt-supported nano-molybdenum carbide (Mo_2C)/CNT composite as the anode. Pre-treatment of CNTs such as high-temperature ammonia to boost the power density by improving bacterial adhesion and acid treatment that could oxidize carbon atoms and form carboxyl groups on the surface of CNTs to modify them to be hydrophilic [11]. However, CNTs without any modifications has seldom been directly packed as both the anodic electrode and microbial carrier of MFC.

Another important factor that affects the performance of MFC is its configuration [12,13]. For example, a three-dimensional packed bed MFC can increase the surface area of the anode for bacteria adhesion [12]. One study [14] reported that the inexpensive semi-coke and granular activated carbon were implemented as the packing material in MFC to treat domestic wastewater. An up-flow tubular air-cathode MFC packed with granular activated carbon was developed for animal carcass wastewater treatment [15]. So far, there have been no reports on up-flow fixed-bed MFC packed with CNTs for continuous treatment of wastewater.

We developed CNTs based microbial carriers for biohydrogen production through a high-rate anaerobic bioreactor recently [16].

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http://dx.doi.org/10.1016/j.bej.2014.11.006
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The current study aimed to investigate the feasibility of unmodified CNTs that significantly simplify the modification process used as a combination of microbial carriers and anodic material for an up-flow fixed-bed MFC. The electrochemical and biochemical properties of fixed-bed MFC need further for continuous wastewater treatment and electricity generation.

2. Materials and methods

2.1. MFC configuration and system setup

The FBMFC (Fig. 1A) was made of poly-methyl-methacrylate plastic cylinder, which was consisted of two chambers, the side of each chamber with three inlets/outlets. The anode chamber had a working volume of 290 ml and was packed with multi-walled CNTs (average diameter 11 nm, length 1–10 μm, purity ≥99.5%, surface area (BET) ≥200 $m^2\,g^{-1}$, and tap density $0.25 \pm 0.05\,g\,cm^{-3}$). It was obtained from Cnano Technology Company (Beijing, China) and served as the anodic electrode and microbial carrier. The packing density of the CNT was $0.19\,g\,ml^{-1}$. Moreover, spiral titanium wire was placed inside the CNT as the current collector. In addition, the anode chamber was connected with a KCl solution containing sterile Ag/AgCl reference electrodes through a salt bridge for desirable experimental conditions (Fig. 1B). The cathode was made of a carbon fiber felt with a surface area of 42 cm^2. The two chambers were separated by a proton exchange membrane (DuPont Nafion-117 with a thickness of 183 μm, and an effective area of 54.26 cm^2), which was supported by two pieces of plastics with 25 mm holes and 27.5 mm hole spacing (center to center). The membrane was treated with 5% (v/v) sulfuric acid at 80 °C for 2 h, and then was cleaned with deionized water prior to use.

2.2. Inoculum, electrolyte, and substrates

The inoculum was collected from the Xiaohongmen Municipal Wastewater Treatment Plant (MWTP, Beijing, China). The municipal wastewater is separated into the liquid supernatant and the sewage sludge. The liquid supernatant is treated by the Anaeroxic–Anoxic–Oxic (A2O) process, whereas the sewage sludge is degraded through anaerobic digestion. The inoculum was collected from an anaerobic digester of Xiaohongmen MWTP, in order to enrich anaerobic electroactive bacteria. The synthetic wastewater was used as the substrate to enrich electroactive microbial consortia in the anode: peptone $16\,g\,l^{-1}$, meat extract $11\,g\,l^{-1}$, urea $3\,g\,l^{-1}$, glucose $3.6\,g\,l^{-1}$, NaCl $2.9\,g\,l^{-1}$, $CaCl_2$ $0.4\,g\,l^{-1}$, $MgSO_4{\cdot}7H_2O$ $0.3\,g\,l^{-1}$, K_2HPO_4 $2.8\,g\,l^{-1}$, vitamin solution ($10\,ml\,l^{-1}$), mineral solution ($10\,ml\,l^{-1}$) and tap water (pH 7.0) [17]. The stock solution had a total COD of $36.3\,g\,l^{-1}$ and was diluted for the desired experiment. The catholyte consisted of a 50 mM phosphate buffer ($0.31\,g\,NH_4Cl\,l^{-1}$, $0.13\,g\,KCl\,l^{-1}$, $2.45\,g\,NaH_2PO_4{\cdot}H_2O\,l^{-1}$, $4.58\,g\,Na_2HPO_4\,l^{-1}$, pH 6.8–7.1).

2.3. Experimental procedure

The FBMFC system (Fig. 1B) consisted of FBMFC, gas–liquid separation and collection, feedstock delivery and data acquisition units. Dissolved oxygen was removed from the anodic medium by purging with nitrogen gas for 15 min before use. The FBMFC started up with an inoculum to substrate ratio of 1:1 (v/v), and it was operated in continuous mode at room temperature (25 ± 2 °C). In contrast to continuous flow mode, electricity cannot be continuously obtained through MFC operated in batch mode, which is not conducive to practical applications [18]. Wastewater was fed up-flow at a flow rate of $0.3\,ml\,min^{-1}$. The effluent in the anode chamber was separated through the gas–liquid separator, whereas the effluent in the cathode chamber was recycled with an oxygen-saturated phosphate buffer. The feedstock at OLRs of 0.22–10.27 g COD l^{-1} d^{-1} was fed into the anode chamber of the FBMFC. Note that HRT was set as 16 h throughout the experiments. FBMFC was run for around two months during experiments. The analysis of electrochemical properties and metabolites was performed when the electricity generation reached the steady state after more than one HRT at each OLR.

2.4. Analytical methods

2.4.1. Electrochemical analysis

The voltage between the anode and the cathode was automatically collected every 60 s. The power density, P ($W\,m^{-3}$), was calculated using the formula $P = U^2/(Rv)$, where U (V) was the voltage of the external resistance (R_{ext}), and v (m^3) was the volume of the anode chamber. The value of R_{ext} was 1000 Ω except during the polarization experiments where R_{ext} was changed over a range of 200–10,000 Ω, the runtime with each external resistance was half an hour (a relative stability of voltage could reached in half an hour), 30 data of voltage were obtained, then cut the maximum and minimum data, calculated the mean of the other 28 data as the voltage of this external resistance. After a polarization experiment, an electrochemical workstation (CHI660E, Shanghai Chenghua, China) was used for the analysis of electrochemical impedance spectroscopy (EIS). In order to measure the internal resistance (R_{in}) of the FBMFC, EIS test was carried out at a frequency range of 0.01 Hz–100 kHz and an alternating current signal of 0.01 V amplitude. The analysis of EIS was performed after the circuit of a FBMFC was open for one hour. The MFC anode served as the working electrode, whereas the cathode was used as the counter and the reference electrode, in order to record the impedance spectra of the FBMFC through the electrochemical station. The Nyquist plots were obtained after EIS test and R_{in} was calculated based on the sum of the resistances due to the ohmic polarization, activation polarization, and concentration polarization of PBMFC [12]. Each test was repeated three times.

2.4.2. The analysis of gas and liquid products

The gas products were collected and analyzed using a gas chromatograph (GC1490, Agilent Technologies). The GC was equipped with a capillary column (TXD-01, 0.25 mm inner diameter, 0.15 μm film thickness). The measured gas components included nitrogen, carbon monoxide, methane, hydrogen, and carbon dioxide. COD was measured using a rapid spectrophotometric method (HACHI, the U.S.). VFAs were analyzed by High Performance Liquid Chromatography (HPLC) (LC-10 AVP Shimadzu, Japan) using an ultraviolet detector and a synergi 4u Hydro-RP column. 5 mM H_2SO_4 was used as the mobile phase at a flow rate of $1\,ml\,min^{-1}$. When the electricity generation reached the steady state with each OLR, the COD of influent and effluent and the VFAs of effluent were measured, each sample was measured for three times, the mean of the results was accepted. Before testing, the samples were centrifuged at 25 °C for 10 min and then filtered with a 0.45 μm filter.

2.4.3. Microbial morphology

Microbial morphology was observed using scanning electron microscope (SEM, FEI Quanta 200). The microbial samples from FBMFC were fixed with 2.5% (v/v) glutaraldehyde in 0.1 M cacodylate, pH 7.2 at 4 °C, washed with PBS (0.1 M, pH 7.2) for three times (10 min/time) and then dehydrated stepwise using a gradient series of ethanol solutions (50%, 70%, 80%, 90%, 100%, v/v). The ethanol was then replaced with tertiary butanol and dried at 20 °C for 20 min. Samples were finally coated with Au and observed using SEM.

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