Journal of Photochemistry and Photobiology A: Chemistry xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Journal of Photochemistry and Photobiology A: Chemistry

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

Wastewater treatment and electricity generation from a sunlight-powered single chamber microbial fuel cell

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ARTICLE INFO

Article history:
Received 30 June 2017
Received in revised form 1 October 2017
Accepted 18 October 2017
Available online xxx

Keywords:
TiO_2 nanotube arrays
Photoanode
Microbial fuel cell
Methylene blue
Electricity generation

ABSTRACT

A novel hybrid single chamber microbial fuel cell is described in which a TiO_2 nanotube array (TNT) photoanode is coupled with a conventional bioanode to achieve simultaneous degradation of methylene blue (MB) dye with improved power generation. As compared to a conventional microbial fuel cell (MFC), the described hybrid-MFC exhibits enhanced power density (14%), current density (33%), and voltage (4%) while simultaneously degrading MB dye, 82.79% after 3.5 h of operation under simulated solar light illumination. The key factor attributed to the enhanced performance is the addition of photogenerated electrons to the MFC external circuit. The effect of various design configurations is also investigated, such as the presence of an air cathode, anode type, and illumination. The hybrid-MFC strategy provides new directions for productive and economical utilization of microbial fuel cells.

1. Introduction

Bacteria in microbial fuel cells (MFCs) oxidize organic compounds found in domestic and industrial wastewater to generate electricity [1]. The capability to simultaneously treat wastewater while generating electricity suggests MFC technology is an excellent tool for environmental remediation [2]. However, at the present time practical application of MFC technology is limited due to relatively low performance efficiencies [3,4]. The MFC anode [5,6] produces electrons via anaerobic respiration [1] which travel through an external circuit to reach the cathode, where the electrons react with protons and atmospheric oxygen forming water molecules [7].

Previous studies have focused on both the underlying biological reactions as well as effect of MFC architecture [8], electrode materials [9–11], and solution chemistry [8,12,13]. MFC performance is determined by factors that include types and characteristics of microbial inoculum [14,15,25], chemical substrate [16], internal and external resistances [17], electrolyte medium [18], electrode materials [19], and interfacial charge transfer between electrode and bacteria [20]. The cathode is a key MFC design element with a slow oxygen reduction reaction (ORR) on its surface commonly limiting overall performance [20]. ORR induced cathode resistance manifests itself in electron depletion which hinders biofilm formation on the anode, in turn lowering the overall MFC performance [21]. Research efforts focused on enhancing MFC performance have studied air cathode structure [22], development of cathode catalysts for improved ORR [23,24], MFC redox mediators [26,27], and various MFC configurations with minimal internal and/or external resistances [28,29].

MFCs have recently been utilized for the degradation of organic dyes [30–44] in wastewater, with studies investigating the effect of redox mediators [30,31], modified cathode materials [33,34], anodic inoculum [35], redox mediators modified anode [30], cathodic biofilms [36], and various MFC configurations such as stacked MFC-biofilm reactor coupled system [37], integrated MFC and aerobic two stage bioreactor system [38], and MFC coupled constructed wetland [39]. All of these studies are aimed towards the simultaneous degradation of toxic dyes and electricity generation, with the dye degradation being proceeded typically by biodecolorization [40,41] and/or reductive decolorization [34,42].

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https://doi.org/10.1016/j.jphotochem.2017.10.030
1010-6030/

Herein, we present an innovative hybrid MFC design (h-MFC) and investigated it for degradation of methylene blue (MB) dye, our target pollutant present in wastewater, while seeking to improve overall MFC performance. The h-MFC assembly is comprised of a TiO_2 nanotube (TNT) array photoanode paired with a conventional carbon felt bioanode both sharing a common air cathode, inserted into a mixture of MFC-medium/MB dye within the MFC chamber. Under simulated solar light, the additional contribution of photogenerated electrons from the TNT photoanode are found to enhance the rate of MB dye degradation. Further, the additional flow of photogenerated electrons from the TNT photoanode appear to enhance the ORR resulting in improved power output. Fig. 1 presents a schematic illustration of the proposed mechanism for MB dye degradation and improved h-MFC performance. To elucidate the underlying mechanisms of MB dye degradation and improved h-MFC performance various control experiments were conducted (see Table I) under similar experimental conditions.

2. Experimental section

2.1. Chemicals and materials

Wet proof carbon cloth (30 wt.% PTFE, Fuel Cell Earth LLC, USA), Pt on Vulcan XC-72 (10%, Premetek Co., USA), 2-propanol (99.5%, Sigma-Aldrich, USA), Nafion perfluorinated resin solution (15–20%, Sigma-Aldrich, USA), carbon vulcan powder and polytetrafluoroethylene (PTFE, 60 wt.% dispersion in H_2O, Sigma-Aldrich, USA) were used for air cathode preparation. Carbon felt (Samjungcng Co., Republic of Korea) was used as the anode. Di sodium phosphate (anhydrous, for molecular biology, Applichem, Germany), ammonium chloride (for molecular biology, suitable for cell culture, ≥99.5%, Sigma-Aldrich, USA), potassium chloride (Sigma-Aldrich, USA), sodium phosphate monobasic monohydrate (ACS reagent, ≥98%, Sigma-Aldrich, USA) and glucose (bacteriological grade, Oxoid, UK) were used for MFC medium preparation. All chemicals were used as received without further purification. Titanium mesh (10.0 mesh, 0.5 mm diameter) and titanium wire (1.0 mm diameter, 99.5%) were purchased from Nilaco Corporation, Japan and used to connect the cathode and bioanode to the external electrical circuit.

TiO_2 nanotube (TNT) array photoanodes were prepared using titanium foil (0.1 mm thickness, 99.5%, Nilaco Corporation, Japan).

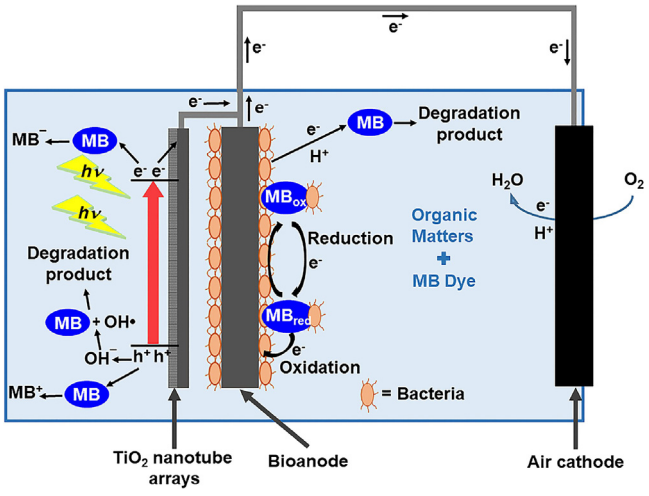

Fig. 1. Schematic illustration of the hybrid-MFC, with the depiction of possible reactions at the coupled TNT array photoanode/bioanode within the microbial fuel cell.

Ethylene glycol (Spectrophotometric grade, 99%, Alfa Aesar, USA) and ammonium fluoride (98.0%, Alfa Aesar, USA) were used for the preparation of the anodization electrolyte.

2.2. Synthesis of TiO_2 nanotube (TNT) photoanodes

The TNT photoanodes were prepared by electrochemical anodization of Ti foil (6.0 cm × 4.0 cm). Prior to anodization, the Ti foil was sequentially cleaned by sonication in acetone, ethanol and deionized (DI) water, for 10 min each step. The electrochemical anodization was carried out in a two-electrode electrochemical cell comprised of Ti foil as a working electrode (anode) and carbon paper as the counter electrode (cathode). The electrolyte used for anodization was comprised of 0.5 wt.% NH_4F and 2 vol.% DI water in ethylene glycol. The anodization was performed at 40 V for 0.5 h with the two electrodes separated by a distance of 2.0 cm. The anodized Ti foils were sonicated in ethanol for 120 s to remove any debris and then annealed at 450 °C for 2 h under static air in a box furnace with a ramp rate of 2 °C·min^{-1}.

2.3. Characterization of TNT photoanodes

TNT photoanode crystallinity was investigated by X-ray diffraction spectroscopy (XRD, Miniflex, Rigaku), operating at 40 kV and 30 mA with Cu Kα radiation (λ = 1.54 Å) as an X-ray source, scanned at 2.5°·min^{-1} in the range of 2θ = 20–90°. Morphological analysis was conducted using a field emission scanning electron microscope (FE-SEM, Hitachi S-4800) with an accelerating voltage of 3 kV. Surface composition and oxidation states were determined by X-ray photoelectron spectroscopy (XPS, ESCALAB 250×i, Thermo VG) using Al Kα line (148606 eV) as the X-ray source.

2.4. MFC configuration

The air cathode of an MFC consisted of a single cylindrical chamber (4.0 cm length, 28.0 mL), prepared using wet-proofed carbon cloth with platinum (Pt) catalyst (0.5 mg·cm^{-2}) layer. Pt catalyst (0.5 mg·cm^{-2}) was coated on the side of the carbon cloth in contact with MFC medium, while the air side of the cathode was protected with a layer of 40 wt.% carbon black powder and four layers of 60 wt.% PTFE [22]. The bioanode was carbon felt (2.0 cm × 2.1 cm × 1.0 cm) connected, through a Ti wire, to a Ti mesh serving as the current collector; prior to use, the carbon felt was washed with DI water to remove contaminants.

The MFCs were operated under fed batch conditions at 30.0 ± 1.0 °C. Prior to performance evaluation, the MFCs were operated for more than 2 months for stabilization of the generated voltage, after which, one of the MFCs was connected to a 7.0 cm^2 TNT photoanode using copper wire. To help elucidate the effect of the TNT photoanode, five different MFC configurations were fabricated and tested. The MFC configurations are tabulated in Table 1 with a schematic illustration of each configuration portrayed in Fig. S1.

2.5. MFC inoculum

Hybrid and conventional MFC bioanodes were inoculated with wastewater from Hyeonpung sewage treatment plant (Daegu, Republic of Korea). The medium contained glucose (2 g·L^{-1}) as a substrate and a 50 mM phosphate buffer solution (PBS) containing: 4.58 g·L^{-1} Na_2HPO_4; 2.45 g·L^{-1} $NaH_2PO_4·H_2O$; 0.31 g·L^{-1} NH_4Cl; 0.13 g·L^{-1} KCl; trace vitamins (10 mL·L^{-1}) and minerals (10 mL·L^{-1}) stock solutions [45]. The initial pH was adjusted to 7.1, reactors were kept at 30 °C in an incubator and were refilled when the

Please cite this article in press as: S.H. Lee, et al., Wastewater treatment and electricity generation from a sunlight-powered single chamber microbial fuel cell, J. Photochem. Photobiol. A: Chem. (2017), https://doi.org/10.1016/j.jphotochem.2017.10.030