Chemical Engineering Journal 349 (2018) 800–807

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Fe/Fe_2O_3 nanoparticles as anode catalyst for exclusive power generation and degradation of organic compounds using microbial fuel cell

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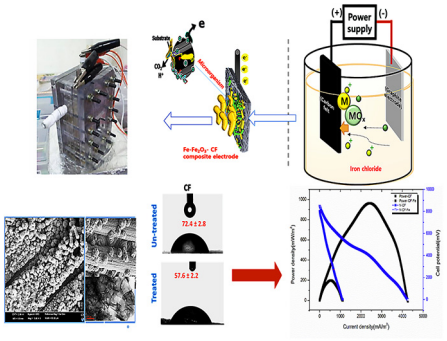
HIGHLIGHTS

- Fe/Fe_2O_3 nanoparticles as catalyst layer were fabricated by electro deposition.
- Catalyst layer enhances the surface wettability of carbonaceous anode materials.
- Significantly increasing in produced power was achieved based on modified anodes.
- The modified anodes were used in MFC to treat the wastewater and produce energy.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Microbial fuel cell
Anode materials
Metal deposition
Fe/Fe_2O_3 nanoparticles
Industrial wastewater treatment

ABSTRACT

Iron/iron oxide (Fe/Fe_2O_3) nanoparticles were deposited on the surface of different carbonaceous anode materials: carbon felt (CF), carbon cloth (CC), and graphite (G) as an effective catalyst to improve the anode performance of microbial fuel cell (MFC) based on the real industrial wastewater. Interestingly, the results of the characterization indicated the novel structure of the iron nanoparticles enveloped with a thin layer of iron oxide formed on the anode surfaces. This novel structure enhances the surface wettability of the electrode, the degradation reactions rate of organic compounds, and the microorganism adhesion on the electrode surface, and decreases the electron transfer resistance. Therefore, the generated power and current were considerable improved, where, the generated power was increased by 385%, 170%, and 130%, for the CF, CC, and G electrodes, respectively. Moreover, the MFC based on the modified electrodes achieved the excellent removal percentage (more than 80%) of organic compounds from wastewaters: This study presents a new approach for MFC application on a large scale based on low-cost and high-efficiency anodes for simultaneous power generation and wastewater treatment.

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https://doi.org/10.1016/j.cej.2018.05.138
Received 15 March 2018; Received in revised form 21 May 2018; Accepted 23 May 2018
Available online 24 May 2018

1. Introduction

The lack of renewable energy sources concurrently with the permanent increase of wastewater sources are the main challenges facing world progress and development [1]. The production of wastewater streams from both industrial and municipal sources is over 126 billion liters per day, which requires an annual treatment cost greater than 25 billion dollars [2]. Utilization of these wastewater as an energy source is considered a promising approach to meet the increasing energy needs as a substitute for fossil fuels. Considerable attention has recently been paid to the use microbial fuel cells (MFCs) as a promising technique for simultaneously production of energy and degradation of organic pollutants [3,4]. This system has the potential to provide clean, renewable, and sustainable energy using exo-electrogenic active bacteria as the biocatalyst [5–7]. This active bacteria decomposes the organic compounds presented in wastewater to the simplest structure while producing protons and electrons [8]. The electrons are transferred from the bacteria to the anode surface and then through the external circuit to the cathode in order to generate electricity [9]. Although, the several advantages of MFCs, its large-scale application is still limited owing to the high cost of its components as well as the low power output compared to other types of fuel cells [10].

Many factors affect the MFCs performance, such as microorganism culture, internal resistance, organic substrate concentration, reactor configuration, and efficiency of the electron transfer process [11]. Among these factors, the electron transfer rate at the biofilm/anode interface is considered the key factor determining the MFCs current and power generation [12]. It is therefore critical to adjust the anode surface morphology and chemistry to enhance the electron transfer process [13]. This objective can be achieved by using electrode materials with specific properties, such as large surface area, super hydrophilicity, high electrical conductivity, excellent chemical stability, high porosity, and biocompatibility [14,15].

However, the most common anode materials (carbon-based materials), used in MFCs, have hydrophobic nature which negatively affects the microorganism adhesion and increases the interfacial resistance for the electron transfer, due to the insufficient adhesion of microorganisms on the anode surface, which leads to decreased power and current density [16–18]. To overcome this obstacle and modify the surface properties of carbonaceous anode materials, various modification methods, such as chemical function group treatment, physical treatment, acid heat treatment, and transition metal coating techniques were introduced [19,20]. Transition metal nanoparticles are effective catalysts for surface modification because of their large surface area and unique combination of reactivity, stability, and selectivity [21]. Kim et al., examined the effect of Pd nanoparticle deposition on the anode performance of MFC, and the results showed that the generated current density was higher than that of the pristine by 1.5–2.5 times [22]. Moreover, a MnO_2/HNT nanocomposite was investigated as an anodic catalyst layer on the carbon cloth by Chen et al., in which the generated power was increased by 50% compared to the untreated electrode [23]. Lowy et al., reported that graphite modified by a graphite paste containing Fe_3O_4 achieved 1.5–2.2 times greater kinetic activity than pure graphite [24]. This enhancement can be attributed to the increased ability to motivate micro growth and accelerate the adhesion of microbe cells on the anode surface after metal deposition process [25]. Although, the great effect of transition metals layers on the anode performance of MFC, the reported coating techniques in the previous studies are costly, complicated, and suitable only for specific anode materials.

Herein, for the first time, the iron/iron oxide nanoparticles were synthesized as anode catalysts on different carbonaceous materials – carbon felt (CF), carbon cloth (CC), and graphite (G) – using an effective, simple, and low-cost electrodeposition (ED) technique. Furthermore, the effect of the transition metal nanoparticles on the chemical structure and the morphology for different carbonaceous anode materials were investigated using different analysis. Finally, the performance of different investigated anode materials before and after the modification process was tested in an air cathode MFC using real industrial wastewater.

2. Experimental

2.1. Materials and electrodes

Carbon cloth (EC-AC-cloth, Electro Chem. Inc., USA; CC), carbon paper (EC-TP1-120, Electro Chem. Inc., USA; CP), and carbon felt (Morgan, UK; CF) were investigated as different carbonaceous anode materials. The cathode material was carbon cloth loaded with Pt 0.5 mg cm^{-2} (EC-20-5, Electro Chem, Inc., USA). A cation exchange membrane (CEM, CMI-7000) purchased from Membrane International Inc., NJ, USA, was used as a proton exchange membrane between two electrodes. All utilized electrodes have the same dimensions (2.5 × 2.5 cm), with exposed surface area of 6.25 cm^2. Iron chloride was obtained from Sigma-Aldrich. A single chamber air cathode MFC based on industrial wastewater was used to investigate the performance of the modified anode materials.

2.2. Synthesis of Fe/Fe_2O_3 nanoparticle modified electrodes

First, different anode materials were cleaned in an ultrasonic bath with acetone for 24 h to remove any suspended impurities, followed by rinsing thoroughly with deionized water and dried overnight in dry oven at 70 °C. Then, the Fe/Fe_2O_3 nanoparticle catalyst layer was formed on the surface of different carbonaceous anode materials using the electro deposition technique (ED) with a simple two-electrode electrolytic cell under an ambient atmosphere. Typically, a rod graphite electrode was used as a counter electrode (anode), whereas the material to be modified (G, CC, and CF) was used as a working electrode (cathode), and the distance between them was fixed to be 2 cm. 100 ml of iron chloride aqueous solution (200 mM) was used as an electrolyte solution. Moreover, DC power supply was applied to support the cell with 20 V for 20 min. A schematic diagram of the electro deposition technique is shown in Fig. S1.

2.3. Microorganisms media

The MFCs were inoculated using real industrial wastewater collected from the Sooyoung sewage treatment plant in South Korea under oxygen-free conditions as a source of microorganisms without any pretreatment. The characteristics of the collected wastewater were analyzed at the Water Environmental Research Center, Busan, South Korea; and the results are summarized in Table S1.

2.4. MFC set up and operation

Single air-cathode MFCs, in which the cathode directly in contact with air, was used as shown in Fig. S2. The cell had a volumetric capacity of 84 cm^3, and the distance between the anode and cathode was kept at 4 cm. A cation exchange membrane (CEM) was used as a separator between the two electrodes. 316 Stainless steel plates of 1 mm thickness were used as current collectors. The current and power density were calculated based on the anode exposed surface area (6.25 cm^2).

80 ml of mixed culture sewage wastewater was purged with nitrogen gas for 20 min to remove the dissolved oxygen. It was then injected into the anode chamber, and the cell was operated in a batch mode until the open circuit voltage (OCV) stabilized. The potentiostat was used to measure OCV over time. Moreover, the anode and cathode potentials were measured with respect to Ag/AgCl reference electrode, which were placed in the anode chamber. After OCV stabilization (two half reactions (anode oxidation and cathode reduction) reached the