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Influence of packing material characteristics on the performance of microbial fuel cells using petroleum refinery wastewater as fuel

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ABSTRACT

The present study investigated the influence of packing material characteristics on the electricity generation performance, treatment efficiency and degradation mechanism of petroleum pollutants over microbial fuel cells (MFCs) using petroleum refinery wastewater (PRW) as fuel. The granule graphite (GG) and granule-activated carbon (GAC) were used to MFCs as packing materials, respectively. GG-packed MFC showed the highest electricity generation and PRW treatment performance, power density (330.4 $mWcm^{-3}$) and oil removal (84 ± 3%) were both better than GAC-packed and non-packed MFCs due to the excellent electrical conductivity. GAC-packed MFC showed the most stable voltage output (higher than 200 mV for 576 h) and lowest mass transfer resistance than GG-packed and non-packed MFCs owing to the stronger adsorption ability. The properties of the packing materials affected the degradation mechanism of petroleum pollutants. Chain hydrocarbons were preferentially degraded in non-packed MFC; volatile phenols and benzene were preferentially removed in GAC-packed MFC; and all petroleum pollutants were simultaneously decomposed in GG-packed MFC.

1. Introduction

Petroleum refining industries converts crude oil into more than 2500 petroleum products including gasoline, kerosene, aviation fuel, diesel fuel, lubricating oils, etc., as well as feed stocks for petrochemical industries. The Wastewater generated from Petroleum Refinery (PRW) contains many chemicals, such as benzene, volatile phenol, sulfides, ammonia, suspended solids, cyanides, nitrogen compounds and heavy metals [1]. PRW is characterized with high organic pollution, high production, acute toxicity and poor biodegradability [2]. The discharge of PRW arguably causes increasing damage to ecological system and human health due to rapid development of petroleum refining industries. Currently, traditional treatment processes such as oil separation, flotation and biological treatments are used to the disposal of PRW, and biological treatment is an indispensable process that is generally used to remove dissolved petroleum pollutants in wastewaters [3]. However, the high energy consumption limits its further application particularly in the context of energy shortage. A great deal of chemical energy is stored in the chemical bonds of organics contained in PRW (>7.6 kJ L^{-1}) [4]. The chemical energy recovery along with wastewater treatment has enabled the renovation of traditional biological treatment by simultaneously solving energy shortage and environmental pollution concerns.

Microbial fuel cells (MFCs) are the emerging sustainable wastewater treatment technologies [5]. They use electricigens as cheap catalyzers to break down chemicals in the anolyte and directly transform chemical energy into electricity. MFCs allow for the recovery of electric energy and value-added products, meanwhile offer qualified effluent and low environmental impact due to effective combination of biological and electrochemical processes. The chemical energy stored in chemical bonds in petroleum pollutants can be transformed into electricity by using MFCs to treat PRW. Additionally, the energy consumption is low and energy conversion efficiency is high compared to traditional biological treatment. Therefore, use of MFCs in PRW treatment is one of the most effective pathways for the regeneration of wastewater resources.

Biomass-contained wastewaters including the livestock wastewater [6], brewing wastewater [7], rice mill wastewater [8],

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municipal wastewater [9] and landfill leachate [10] have been used as substrates for MFCs. However, few studies using bio-refractory wastewaters as substrates were carried out in MFCs. Morris et al. [11] first applied MFCs to the bioremediation of hydrocarbons in groundwater. And the potential of using MFCs to enhance the anaerobic biodegradation of diesel was proposed by them in the next year [12]. The diesel removal in MFCs was up to 82% in 21 days, which was far higher than that in traditional anaerobic biological treatment (31%). The studies of MFCs using PRW as fuel, however, have not been reported.

The low power density output of MFCs limits their applications [13–16]. Various studies were carried out to improve power density output of MFCs. They included optimizing structures of MFCs to reduce internal resistance [17], modifying electrodes to promote electron transfer rate [18], continuous injection of substrates to accelerate reactant transfer [19], decreasing sizes of MFCs to increase power density [20], stacking a plurality of MFCs to increase voltage or current outputs [21,22], etc. The packing materials in the anode chamber can extend the superficial anode area and increase microbial enrichment on the anode surface to improve power density output of MFCs.

Granule graphite (GG) was the primary packing material used in MFCs because of its ability to promote electron transfer [23–25]. Sun et al. [26] generated MFCs using carbon felt, GG, GAC and granule semi-coke as packing materials in the anode compartment. The results indicated that the type of packing materials directly affected microbial species and abundances in the anode and then influence electricity generation performance of MFCs. GAC-packed MFC showed the highest power density among the four MFCs because GAC was more favorable for adhesion and growth of *Geobacter*. In granular semi-coke, microorganisms such as *Synergistes*, *Bacteroidetes* and *Castellaniella* may jointly contribute to electricity generation. Huggins el al [27] used wood-based biochars as electrodes of MFCs. Biochars showed comparable performance to other electrode materials owing to their economic and environmental benefits. However, the influence of packing material characteristics on electricity generation performance and wastewater treatment efficiency of MFCs has not been investigated.

In this study, double-chambered packing-type MFCs using PRW as fuel were constructed to explore the potential to synchronously remove petroleum pollutants and recover energy. GG and GAC, which have excellent electrical conductivity and adsorption ability, respectively, were employed in MFCs to investigate the influence of packing material characteristics on the electricity generation performance, treatment efficiency and degradation mechanism of petroleum pollutants.

2. Methods

2.1. MFC construction

The MFCs used were constructed with two cylindrical glass compartments of equal dimensions according to the previous study [28]. The working volume of each compartment was 400 mL. The two compartments were connected by a Nafion proton-exchange membrane which was first pretreated in 5% hydrogen peroxide solution and then 0.5 mol L^{-1} sulfuric acid solutions to remove organics and metal ions. The compartment that functioned as the anode chamber was sealed with a rubber stopper to maintain anaerobic conditions. A carbon rod (0.6 cm × 18 cm) was used as the anode electrode. The other compartment, which was used as a cathode chamber, was filled with 20 mmolL^{-1} Fe(III)-EDTA solution as the catholyte, and the cathode electrode was made of graphite flake (0.5 cm × 2.5 cm × 17 cm). A small air pump (ACO-5503, Haili Group, Inc., Guangzhou, China) was used to introduce compressed air into the cathode chamber for aeration. If not specified, the external resistance was set to 1000 Ω, and the operational temperature was 30 °C. The MFC in non-packed conditions was named as MFC I. 100 mL GAC (average grain diameter 0.5–2.0 mm, specific surface area 779 m^2 g^{-1}, pore volume 0.123 ccg^{-1}, water absorption 50%, hydrophilic) and 100 mL GG (average grain diameter 0.5–2.0 mm, specific surface area 2.482 m^2 g^{-1}, pore volume 0.0143 ccg^{-1}, contact angle 90° for 30 min, hydrophobic) were used as packing materials for MFC II and MFC III. The electrical resistivity of the GAC and GG were 2 Ωm^{-1} and 1.1 × 10^{-3} Ωm^{-1}, respectively, according to previous measurement. It suggested that GG had higher conductivity than GAC.

2.2. Wastewater and inoculum

The PRW was an effluent of a flotation process of wastewater treatment plant in Beijing Yansan Petroleum Refinery, China. The chemical oxygen demand (COD), oil content and pH value were 450 ± 50 mgL^{-1}, 50 ± 5 mgL^{-1} and 8.5 ± 0.5, respectively. The anolyte of MFCs was made of PRW and growth medium at a 1:1 ratio [12]. The COD, oil content and pH value of the mixed anolyte were 250 ± 40 mgL^{-1}, 17 ± 1 mgL^{-1} and 7.1 ± 0.1, respectively. Prior to inject into the anode chamber, N_2 was bubbled into the anolyte for 30 min to maintain an anaerobic environment. The microbial inoculum was the activated sludge collected from a PRW treatment plant in Dagang Petrochemical Company, China. When the MFCs were started up, 5 mL of this activated sludge was added to 100 mL anolyte in the anode chamber.

2.3. Calculations

The voltage output of MFCs was recorded automatically with a data logger (e-corder, ED401, eDAQ Pty. Ltd., Australia), and the time interval was 1 min. The current density and power density were calculated with the formulas below:

$$I = UR^{-1}V^{-1} \quad (1)$$

$$P = UI \quad (2)$$

where I is the current density ($mAcm^{-3}$), U is the voltage (mV), R is the external resistance (Ω), V is the working volume of anode chamber (cm^3), and P is the power density ($mWcm^{-3}$).

The potential of electrodes was measured using the anode or cathode as the working electrode with a SCE (saturated calomel electrode) reference, and the value was recorded with a multimeter (BM857, Brymen, Shenzhen, China). The apparent internal resistance and ohmic resistance were determined by the steady state discharge method [29] and current interruption method [30].

2.4. Analytics

2.4.1. Conventional water quality analysis

The COD was measured with a 5B-6 COD speed meter (Lian Hua Tech, Lanzhou, China), the oil content was determined with an infrared oil analyzer (MC-OIL420, Huaxia Kechuang, Inc., China) after carbon tetrachloride extraction. The pH value was recorded by a pH monitor (PHSJ-4, Leici Instrument, Inc., Shanghai, China).

2.4.2. Scanning electron microscopy (SEM)

The packing materials in the anodic chamber were taken out using sterile tweezers and fixed in glutaraldehyde solution (volume fraction was 2.5%). The fixed samples were rinsed for 15 min with 0.1 mol L^{-1} sterile phosphate buffer saline (PBS, pH 7.2) 3 times and