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Investigation of different configurations of microbial fuel cells for the treatment of oilfield produced water

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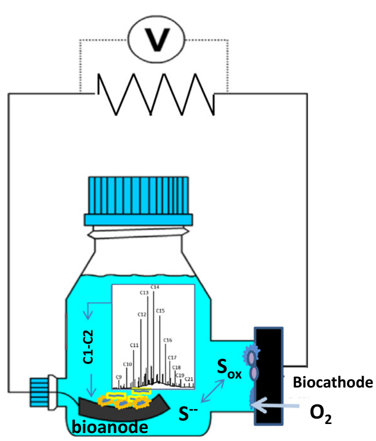
HIGHLIGHTS

- A simple MFC design has been investigated to improve oilfield produced water degradation process.
- Degradation of hydrocarbons was substantially complete and rapid as with acetate in MFCs.
- Power and Coulombic efficiency better than current literature were obtained, in spite of low values.
- Anaerobic conditions were slowly established on the anode due to hydrocarbons.
- Persistent aerobic condition on the biocathode made possible the rapid degradation of hydrocarbons.

GRAPHICAL ABSTRACT



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ABSTRACT

Produced water (PW) is the largest waste stream in the oil production process: it contains light polar and aliphatic hydrocarbons, production process compounds, dissolved gases, anions and cations.

Disposal of PW is subjected to strict legislations. Oil producing countries are focused on finding effective and economic methods for its treatment. Some physical and chemical methods have been used for treatment of PW and biological treatments have been proven to efficiently remove dissolved hydrocarbon compounds. Coupling of anaerobic biological treatment with electrochemical technology in microbial fuel cells (MFCs) can in principle lead to the production of clean water and electric energy.

The suitability of MFCs for improving the treatment of PW was investigated in the present work. For the first time, the simple design of single chamber MFCs fed with real PW (PW-MFCs) was studied, in different configurations: (i) with and without membrane; (ii) with and without Pt cathodic catalyst.

The results demonstrate the effectiveness of the membraneless configuration without chemical catalyst at the cathode. Even though the electrical output of PW-MFCs was very low (3 mW m^{-2}), it is currently the best reported performance. Furthermore, almost complete hydrocarbon degradation was achieved for each fed-cycle (96.6 ± 1.94%). The Coulombic efficiency was limited by the difficulty to obtain strict anaerobic condition at the anode, since the biocathode of PW-MFCs remained more permeable to oxygen than in acetate-fed MFCs.

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The DNA sequencing of operating anodic biofilm detected electroactive *Desulfobulbaceae* mixed to aerobic biodegraders (*Burkholderiales*) likely through cycling sulfur compounds, which enriched from the PW initial pool in the hypersaline environment.

Above all, the results pointed to the practical possibility of using a MFC to enhance and monitor the PW biodegradation process. In fact, the MFC electrical output indicated the occurrence of anaerobic degradation, while the electrochemical parameters of cathode (Tafel slope) resulted correlated to aerobic degradation, suggesting the possibility to design an on-line sensor of the biotechnological industrial treatments of PW.

1. Introduction

Although renewable energy sources have been intensively exploited in the last decades, oil and gas are still the most common conventional energy sources in the industrialized world. According to tremendous expansion in oil and gas production technologies, especially those using water injection, vast quantities of process water has been consumed and consequently vast amounts of wastewater has been generated. Produced water (PW or oilfield wastewater) is the largest waste stream generated in oil and gas industries and it is considered as the major source of pollution in the oil and gas fields. It is generated upon the exploitation of oil and gas fields and it is a mixture of injected water, formation water, hydrocarbon and treating chemicals and is extracted with the crude oil or natural gas. Physical and chemical properties of oilfield wastewater depend on the geological location, the lifetime and the operating conditions of the fields, the hydrocarbons produced and the chemicals employed. Although the exact composition can considerably vary from one field to another, the composition is similar to oil or gas that has been retrieved from the field [1]. It is usually rich in dissolved and dispersed oil (mainly aliphatic hydrocarbons, but also organic acids, benzenes, phenols and polycyclic aromatic hydrocarbons) and also contains very high amounts of inorganic dissolved compounds, heavy metals, radioactive substances and dissolved gases [2]. The content of salts may range from a few ppm to 300 g L^{-1} [3]. In the past, the discharge of PW has caused pollution to soil, waterways and underground water. Due to the increasing volume of this wastewater all over the world, the outcome of directly discharging on the environment has lately become a significant issue of environmental concern [4]. Moreover, many water-stressed countries with oil-fields are increasingly looking for ways to supplement their fresh water resources, focusing on efficient and cost-effective methods to treat oily saline produced water.

The removal grade of contaminants and the ultimate quality depend strongly on the final usage of this water and are strictly governed by regulations. Many physical, chemical, biological methods, and combination of them, have been attempted for PW treatment [3,5]. The more effective removal of dissolved oil is usually accomplished using biological processes, though they potentially fail when the salinity of water is very high. Microbial electrochemical technology has been proposed to address environmental problems, simultaneously producing electricity and treating wastewater by anaerobic oxidation of contaminants using microorganisms as biocatalysts. Several kinds of contaminant can be also bio-reduced at cathode, including metals [6] and nitrogen compounds [7]. The treatment of various kind of wastewater in microbial fuel cells (MFCs) has been extensively investigated [8–10]. Several industrial wastewaters have been used for feeding MFCs including municipal wastewater [11], wastewater from diary industry [12,13], potato processing factory [14], paper recycling [15], chemical industries [16], and distillery wastewater [17]. Recent works refer also to high saline wastes and related issues [18,19]. However, only few works focused on the removal of recalcitrant compounds like petroleum hydrocarbons. The treatment of synthetic wastewater containing single or mixed pollutants (i.e. phenantrene, benzene, terephtalic acid) were investigated taking into account the effect of temperature, salinity, pH and organic load [20,21]. Some specific applications were also tailored to treat real field petroleum sludge and diesel contaminated sediments and ground water [22–24]. In this respect, the application of MFC technology to untreated real PW has seldom been reported [25] and it has been addressed as especially challenging. Its high salinity, in fact, might negatively affect the physiology of anaerobic microbial population [19].

In present work, MFCs with a simple design and cost effective carbon based electrodes were used to study the suitability of MFC technology for the treatment of real, high saline PW from Iranian oil fields. Membraneless single-chamber MFCs without cathodic catalyst were previously applied to treat municipal wastewater, with the simultaneous generation of electric power [26]. However, the complex nature of pollutants in PW and the extremely high salinity suggested detailed investigation of the best configuration of MFC. Different configurations were investigated: i) with and without membrane; ii) with and without Pt catalyst on the cathode. A group of MFCs (PW-MFCs) was filled with real produced water collected from western oilfields of Iran. Another group of MFCs (SW-MFCs), used as control, was filled with synthetic wastewater and sodium acetate, having the same high salinity (conductivity of 132 ± 8 mS cm^{-1}) and COD content (2000 mg L^{-1}) of the real PW. The MFCs were operated in batch mode for several cycles.

2. Materials and methods

2.1. MFC configuration and setup

Simple Pyrex® bottles (125 ml volume) with two lateral openings were used as lab-scale MFCs, as described in [27] (Fig. 1). The bigger opening accommodated the cathode and the smaller one was used to place the anode on the bottom of the cell.

Both anodes and cathodes were made of the same carbon cloth (Fuel Cell Earth). Anodes were built from 3 × 5 cm carbon cloth electrically connected to a copper wire. The connection between copper and carbon cloth was insulated by at least three layers of non-conductive high viscosity epoxy resin (Mapei Epojet). Cathodes were made of 5 × 5 cm carbon cloth modified by the addition of a microporous layer (MPL) on the water side. MPL was prepared by coating four layers of a carbon-PTFE ink on untreated carbon cloth and subsequent heating in a furnace at 340 °C after each layer deposition. The ink consisted of a suspension of conductive carbon black (ENSACO, TIMCAL carbon black), PTFE (60% dispersion in water, Sigma Aldrich) and Triton (Triton™ X-100, Sigma Aldrich) in Milli-Q water. Carbon black, PTFE and Triton were in the ratio 3:3:2. The geometric cathodic surface area exposed to the solution was 3 cm^2. Anode and cathode were then connected through an external circuit with a resistance of 100 Ω.

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