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Integrated electrochemical treatment systems for facilitating the bioremediation of oil spill contaminated soil

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HIGHLIGHTS

- Microbial fuel cells (MFCs) is applied to accelerate the remediation of oil pollution.
- Diesel removal rate increase by four times in MFCs.
- A high power density (29.05 W/m^3) was achieved.
- Metabolites may act as emulsifying agents benefit oil dispersion and bioremediation.

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ABSTRACT

Bioremediation plays an important role in oil spill management and bio-electrochemical treatment systems are supposed to represent a new technology for both effective remediation and energy recovery. Diesel removal rate increased by four times in microbial fuel cells (MFCs) since the electrode served as an electron acceptor, and high power density (29.05 W m^{-3}) at current density 72.38 A m^{-3} was achieved using diesel (v/v 1%) as the sole substrate. As revealed by Scanning electron microscope images, carbon fibres in the anode electrode were covered with biofilm and the bacterial colloids which build the link between carbon fibres and enhance electron transmission. Trace metabolites produced during the anaerobic biodegradation were identified by gas chromatography–mass spectrometry. These metabolites may act as emulsifying agents that benefit oil dispersion and play a vital role in bioremediation of oil spills in field applications.

1. Introduction

With demands for energy increasing worldwide, the excess exploitation of energy resources, especially for oil, is generating numerous types of pollution (Minakov, 2004). Severe oil spills have created heavy pollution that has endangered both marine and terrestrial environments. The Gulf of Mexico oil spill (2010) was the largest accidental ocean spill in U.S. history, in which more than 3.19 million barrels' worth of oil leaked into the gulf (Team, 2014). Such toxic and recalcitrant hydrocarbon pollution is becoming a lethal threat to the diversity of life forms in marine ecosystems and human health (Pavitran et al., 2006). Therefore, it is important to find effective technologies for eliminating contaminants to control oil pollution.

Microbial removal has the benefits of low cost and avoiding recontamination with secondary contaminants that are produced in physical and chemical remediation processes (Hong et al., 2005; Liu et al., 2017a). However, heavier fuels like diesel (compounds ranging from C_8 to C_{25}) take more time to be naturally remediated, especially under anaerobic conditions with limited nutrients (Morris et al., 2009). The anaerobic degradation of hydrocarbons can be stimulated in microbial fuel cells (MFCs) without adding electron shuttles and nitrate or sulphate as electron acceptors, and energy in contaminants can be recycled at the same time with

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sustainable production of electricity (Rabaey et al., 2007; Lovley, 2008).

Many varieties of organic compounds used as substrates in MFCs for electricity generation have been studied. Most analyses focused on glucose, acetate, lactate and the mixture of organic matters in wastewater (Pant et al., 2010). MFCs used for the remediation of oil pollution has also been studied, but the power density still too low for energy recovery.

The voltages at 190 mV (2.16 W m^{-3}) were achieved in a sediment MFC with 24% total petroleum hydrocarbons degradation after 66 days (Morris and Jin, 2012). As demonstrated by other study, the energy output reached 0.85 W m^{-3} in 25 days and the rate of removing petroleum hydrocarbons was enhanced by 120% in U-tube microbial fuel cells with saline soil (Wang et al., 2012). The diesel removal rate was enhanced using enriched biofilm anodes and the generation of power can be improved from 15.04 mW m^{-2} to 90.81 mW m^{-2} in diesel-fed microbial fuel cells (Venkidusamy et al., 2016). The bioremediation of petroleum contaminants was enhanced by using MFC technology in groundwater under anaerobic conditions with a maximum power density of 120 mW m^{-2} utilizing a mixture of refinery waste (Morris and Jin, 2007). Besides, it was proved that the diesel biodegradation rate could be raised to 82% after 21 days at 30 °C in a MFC with diesel refinery and surfactant for power generation (31 mW m^{-2}) (Morris et al., 2009). However, inadequate studies have been reported for effective biodegradation of oil pollution with high power output.

In this paper, we aim to reveal: (1) the relationship between diesel biodegradation and electricity generation; (2) how these two processes emerged simultaneously and impact on each other in a mediator-less MFC with diesel as the only carbon source; and (3) more evidence that improves the possibility of applying this technology to oil spill remediation.

2. Methods and materials

2.1. Configuration of the MFC system

Dual-chamber MFCs, in which each chamber has an effective volume of 28 mL as shown in Scheme 1, were used in this study. Chambers were separated by a proton exchange membrane (PEM). Carbon brush electrodes were washed in 1 M HCl and NaOH to remove possible metal and biomass contamination before applying them in both chambers, and the electrodes were connected to a fixed external resistance of 1000 Ω to close the electrical circuit.

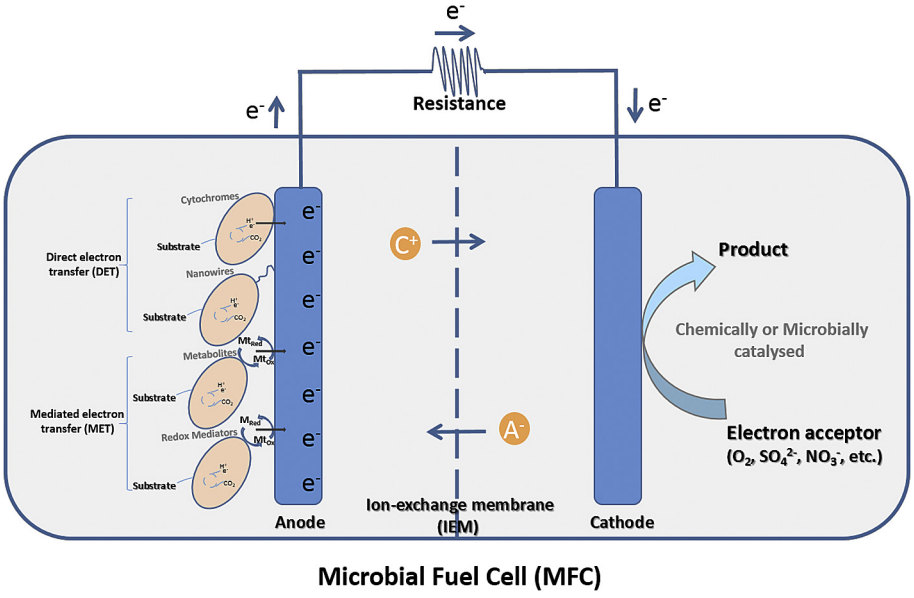
2.2. System inoculation and operation

The mixed cultures enriched from hydrocarbon contaminated soil samples were used for anodic inoculum. Commercially available diesel was used which contains 76.5% of aliphatic hydrocarbon, 23.50% of aromatic hydrocarbon, and 0.003% of sulphur in weight percent. The anodic medium consisted of the following: diesel; NH_4Cl 0.03 g L^{-1}; $NaH_2PO_4.2H_2O$ 0.92 g L^{-1}; $CaCl_2.2H_2O$ 0.0056 g L^{-1}; $MgSO_4.7H_2O$ 0.035 g L^{-1}; KCl 0.0052 g L^{-1}; $NaNO_3$ 0.044 g L^{-1}; and 0.1 mL L^{-1} microelement solution (Logan et al., 2006). The medium in the cathode contained: $NaH_2PO_4 \cdot 2H_2O$ 5.54 g L^{-1}; $Na_2HPO_4 \cdot 12H_2O$ 23.08 g L^{-1}; and $K_3[Fe(CN)_6]$ 32.93 g L^{-1}. Anolyte was sparged with nitrogen to keep anaerobic condition.

Two control groups were set. The first group was sterile controls using the same dual-chamber MFCs without inoculation. The second one was consist of single anode chamber with inoculation. For each cycle, the anode and cathode solution was replaced when the voltage output fell below 50 mV. All test were triplicated and operated at a constant room temperature of 23 ± 3 °C.

2.3. Analytical methods

The output (E_{cell}) was continuously monitored across the resistor using a digital multimeter (Keithley Instruments, Inc., Cleveland, Ohio, USA) connected to a data acquisition system. The current I in Amperes was calculated using Ohm's law, $I = E_{cell}/R_{ext}$, where E_{cell} is the measured voltage in volts and R_{ext} is the known value of the external load resistor in Ohms. Coulombic efficiency (CE) were calculated as previously described (Logan et al., 2006), and the power density (P, W m^{-3}) was calculated and normalized to the anode volume. The maximum power density and polarization curve were determined by adjusting the external resistance from 100000 Ω to 5 Ω. During this process the external resistance changed when a stable output voltage was recorded. All the tests



Scheme 1. Principal sketch of the microbial fuel cell.