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# Bioelectricity generation from treatment of petroleum refinery wastewater with simultaneous seawater desalination in microbial desalination cells

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ABSTRACT

Petroleum refinery wastewater (PRW) contains a high concentration of pollutants and may pose serious environmental risks. Conventional treatment methods of PRW are energy intensive, and thus there is an urgent need to develop sustainable technologies. Microbial desalination cells (MDCs) represent a new energy-efficient technology for simultaneous treatment of PRW and seawater. In this study, PRW was for the first time treated in an MDC and the effects of salt concentration and catholyte were studied. The maximum COD removal was achieved by the MDC using an initial salt concentration of 20 g/L NaCl solution, and the COD removal increased slightly from 64.0% to 70.5% when the catholyte was changed from phosphate buffer solution (PBS) to acidified water. The maximum desalination efficiency of 19.9% was obtained by an MDC operated with real seawater and PBS. Acidified water was found to be an efficient catholyte in terms of specific energy production. When desalinating real seawater, the highest total energy produced was 32.6 W h/kg COD with respect to COD removal or 9.5 W h/$m^3$ with respect to the total volume of the desalinated water. These results demonstrated that complex PRW could be used as an anodic substrate in MDCs for electricity generation and seawater desalination.

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## 1. Introduction

The global consumption of fossil fuels is increasing due to the high needs for energy. To meet this demand, various petroleum refineries have been established. Petroleum refining, including crude oil desalting, distillation, hydro-treating and water cooling systems, not only consumes a considerable amount of water but also generates large quantities of wastewater. It is estimated that 3.5–5 $m^3$ of petroleum refinery wastewater (PRW) is generated per ton of crude oil processed [1]. Direct discharge of PRW will pose serious risks to the environment as it contains a variety of contaminants. Typical PRW has a COD (chemical oxygen demand) concentration of 300–800 mg/L, a BOD (biological oxygen demand) concentration of 15–350 mg/L, a total petroleum hydrocarbon around 3000 mg/L, a suspended solid concentration >100 mg/L and a phenols level of 20–200 mg/L. In addition, ammonia, sulfide and heavy metals (e.g. Chrome, Lead, Nickel and Vanadium) are commonly found in PRW [1]. Conventional treatment methods of RPW are based on physiochemical methods such as oil water separation, coagulation, membrane-based technologies (e.g. reverse osmosis and membrane filtration), electrocoagulation and biological treatment [2]. These technologies are energy intensive and cannot meet the requirement of sustainability. Due to the large quantities of nutrients present in this wastewater, microalgae based treatment process was also used [3]. This process can be coupled with traditional biological process, but it is a relatively slow process [3].

In the past two decades, substantial effort has been devoted to search for more energy efficient wastewater treatment methods. Among them, biological treatment holds certain promise because of its relatively lower cost and less environmental impact. Bioelectrochemical systems (BES) have emerged as a new technology for energy-efficient water and wastewater treatment [4]. It has been proposed to treat various types of wastewater including petroleum refinery wastewater. An early attempt was to employ MFCs (microbial fuel cells) to treat diesel range organics (DRO), a serious contaminant of groundwater [5]. The results showed that MFCs could significantly improve DRO degradation compared to an anaerobic digester. The removal of phenol, a main component in PRW, has also been studied in MFCs, and a removal efficiency of 90% has been reported [6]. Another study used U-tube MFCs to remediate petroleum-contaminated soil and found that the MFC enhanced the degradation by 120% [7]. When sediment-type MFCs

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were used to remove petroleum compounds in contaminated sediments [8–10], the degradation was almost 12 times faster than that by a control group operated at an open circuit mode, indicating that MFCs might be applicable for *in situ* remediation of petroleum contamination in sediments of natural water bodies [11]. Ren et al. treated refinery wastewater by using microbial electrolysis cells (MECs) [12]. The results showed that the MECs achieved COD reduction up to 79% with a current density of 2.1 A/$m^2$. By comparison, an air–cathode MFC showed higher power production when treating refinery wastewater [13]. In MFCs, higher voltage and current can be generated by using in series and parallel cell connections [14]. The produced energy can thus be used to balance energy consumption from pumping [15].

Developed from the BES concept, microbial desalination cells (MDCs) can achieve simultaneous wastewater treatment and salt water desalination [16,17]. An upflow MDC with an anode volume of 1900 mL and a salt chamber volume of 850 mL could remove up to 99% NaCl, but the HRT (hydraulic retention time) of four days was too long for practical applications [18]. The advantage of upflow MDCs is that large-scale systems can be developed through hydraulic connection. In a recent study, a 105-L MDC system has been developed by connecting 30 upflow MDCs and achieved a salt removal rate of 9.2 kg/$m^3$/d and an electricity of 2000 mA, which provided insights into the design and applications of MDCs [19]. While synthetic wastewater was widely used as a model substrate in MDCs [20–25], limited studies focused on the treatment of real wastewater. When domestic wastewater was used as the anode substrate, the MDC performance degenerated over time due to the low BOD concentration and biofouling on the anion exchange membrane (AEM), which seriously inhibited electricity generation and ionic transfer [26]. Recently, microbial osmotic fuel cell (MDC coupled with FO membrane and ion exchange membrane) was used for simultaneous treatment of domestic wastewater, generation of bioelectricity and desalination of real oilfield produced water [27]. Oilfield produced water is the water that is produced from oilfield along with oil or gas during production process. Produced water is characterized by high TDS. The maximum COD removal of wastewater, the TDS removal of produced water and the maximum power density achieved were 92%, 80% and 48.52 mW/$m^2$ respectively. These results were obtained after 10 days of continuous operation. As mentioned above, PRW contains high concentrations of organic matters and thus may enhance current generation and desalination in MDCs [3]. To our knowledge, the treatment of PRW by MDCs has not been reported before. The simultaneous treatment of PRW and seawater by MDCs is potentially energy efficient and economic, and thus could be promising for practical applications.

In this study, we demonstrated for the first time the feasibility of using PRW as an anodic substrate in an MDC for simultaneous PRW treatment, power generation and desalination. Two different NaCl concentrations (5 g/L, 20 g/L) and real seawater were used in the desalination chamber, with acidified water or phosphate buffer solution (PBS) serving as the catholyte. The effects of the salt concentration and the catholyte on COD removal, power generation, and salt removal were examined. To further understand the effects, electrochemical impedance spectrometry (EIS) was conducted to characterize the internal resistance of the membranes at different salt concentrations in the desalination chamber.

## 2. Materials and methods

### 2.1. MDC configuration and operation

An MDC was constructed using plexiglass cubes (Fig. 1). The dimension of each chamber was 14 cm × 9 cm × 9 cm. A carbon brush (2.5 cm diameter × 3.0 cm length) was used as the anode

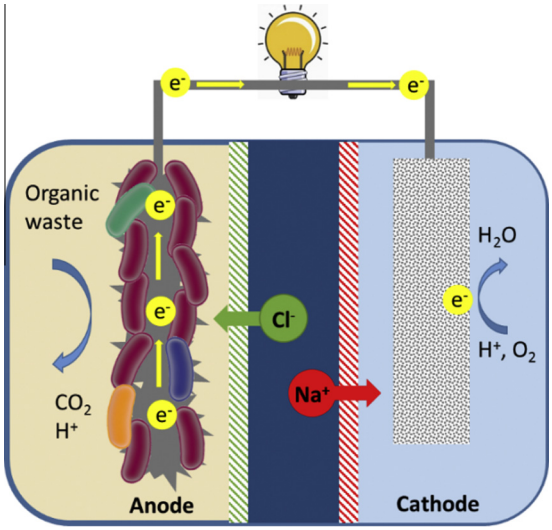


**Fig. 1.** Schematic of the MDC.

electrode (Gordon Brush Mfg. Co., Inc.). A piece of carbon cloth (5 cm × 5 cm) was used as the cathode electrode (Zoltech Companies, Inc., USA). A titanium wire was used to connect the electrodes. Anion exchange membrane (AEM, AMI-7001, Membrane International, Inc., Ringwood, USA) and cation exchange membrane (CEM, CMI-7000, MembraneInternational, USA) were used to separate the anode, salt and cathode chambers. Anaerobic microbes were collected from a local wastewater treatment plant in Doha, Qatar and incubated for 3 months in a 5 L batch reactor with glucose medium (Glucose 5 g/L, $MnCl_2$ 10 mg/L, $ZnCl_2$ 10 mg/L, $CaCl_2$ 20 mg/L, $MgCl_2$ 0.25 g/L, $NH_4Cl$ 0.5 g/L, $KH_2PO_4$ 0.2 g/L and $K_2HPO_4$). This acclimated culture was used to inoculate the anodic chamber of the MDC.

### 2.2. Characteristics of the PRW and seawater

The PRW used in this study was collected from a local petroleum refinery wastewater treatment plant in Doha, Qatar (Feed to the wastewater treatment plant). This wastewater is resulted from industrial activities only and not mixed with sewage system in the refinery. The PRW has a conductivity of 2.3 mS/cm, a COD of 806 mg/L, a pH value of 8.0, a TDS 1.1 ppt and a salinity 1.2 psu. The PRW was stored at 4 °C before use. NaCl solution (5 g/L or 20 g/L) or real seawater was used as the salt solution. The two NaCl concentrations were selected to mimic brackish water [28]. The seawater collected from Corniche, Doha has a salinity of 42.8 psu, which is higher than the average seawater salinity of 35.5 psu. The initial conductivity was 7.8, 28.0 and 64.1 mS/cm for the 5 g/L NaCl, 20 g/L NaCl and seawater, respectively. The cathode chamber was fed with PBS (50 mM) or acidic water (pH 2). According to the Nernst equation, oxygen reduction reaction has higher potential under an acidic condition, and thereby enhancing current generation and desalination. Previous studies have also demonstrated that acid solution costed only 3% of PBS, and thus may be more economically viable for practical applications [29].

### 2.3. Analytical measurements and calculations

The voltage was recorded every 10 min by a digital multimeter (Fluke -289). Polarization curve was measured by changing the external resistance from 10,000 Ω to 1 Ω with a resistance decade box (Microteknik, India). The pH, total dissolved solids (TDS) and conductivity were measured by a benchtop pH/conductivity meter

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