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## Bioelectrochemical desalination and electricity generation in microbial desalination cell with dewatered sludge as fuel



Fanyu Meng<sup>a</sup>, Junqiu Jiang<sup>a</sup>, Qingliang Zhao<sup>a,b,\*</sup>, Kun Wang<sup>a,b</sup>, Guodong Zhang<sup>c</sup>, Qingxin Fan<sup>a</sup>, Liangliang Wei<sup>a</sup>, Jing Ding<sup>a</sup>, Zhen Zheng<sup>d</sup>

<sup>a</sup>School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China

<sup>b</sup>State Key Laboratory of Urban Water Resources and Environments (SKLUWRE), Harbin Institute of Technology, Harbin 150090, China

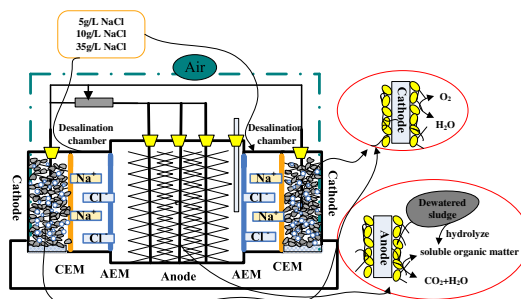
<sup>c</sup>Institute of Resources and Environment Engineering, Shanxi University, Taiyuan 030006, China

<sup>d</sup>School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China

### HIGHLIGHTS

- Simultaneous desalination and dewatered sludge stabilization were achieved.
- Desalination rate for low concentration salt water was more than 40% in 24 h.
- The  $P_{max}$  of 3.178 W/m<sup>3</sup> and open circuit voltage of 1.118 V occurred on 130 d.
- Removal of organic matter was up to 25.71 ± 0.15% after operation for 300 d.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Microbial desalination cells (MDCs) with common liquid anodic substrate exhibit a slow startup and destructive pH drop, and abiotic cathodes have high cost and low sustainability. A biocathode MDC with dewatered sludge as fuel was developed for synergistic desalination, electricity generation and sludge stabilization. Experimental results indicated that the startup period was reduced to 3 d, anodic pH was maintained between 6.6 and 7.6, and high stability was shown under long-term operation (300 d). When initial NaCl concentrations were 5 and 10 g/L, the desalination rates during stable operation were 46.37 ± 1.14% and 40.74 ± 0.89%, respectively. The maximum power output of 3.178 W/m<sup>3</sup> with open circuit voltage (OCV) of 1.118 V was produced on 130 d. After 300 d, 25.71 ± 0.15% of organic matter was removed. These results demonstrated that dewatered sludge was an appropriate anodic substrate to enhance MDC stability for desalination and electricity generation.

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## 1. Introduction

Freshwater is the most essential renewable resource for life, food production and industrial development, with its demand being tripled over the last fifty years. However, the over-exploitation of

aquifer results in the depression of underground water. The difficulty to purify water and the high cost to transport aggravates the problem of water deficits. Therefore, the exploitation of seawater and brackish-water desalination has drawn unprecedented attention (Shannon et al., 2008). A drawback of the conventional desalination technologies is the intensive consumption of energy, such as thermal distillation, membrane separation, freezing, electro dialysis, etc. (Busch and Mickols, 2004). Fortunately, the recent emerging microbial desalination cells (MDCs) have a great

\* Corresponding author at: School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China. Tel./fax: +86 (451) 86283017.  
 E-mail address: [qlzhao@hit.edu.cn](mailto:qlzhao@hit.edu.cn) (Q. L. Zhao).

potential as a low cost and energy savings desalination process with significant environmental benefits (Mohan et al., 2008), having such advantages as operation under room temperature, normal pressure, and neutral pH. Specifically, MDC utilizes the bio-electricity to accomplish salt removal. It is a variant of microbial fuel cells (MFCs), in which an additional middle chamber is integrated between the anode and the cathode. Exoelectrogenic bacteria oxidizing the organics in the anode chamber promotes the electron transfer and results in the migration of anions and cations from the middle chamber to the anode and cathode chambers (Kim and Logan, 2013).

The first introduction of conceptual three-chambers MDC by Cao et al. (2009) demonstrated that the desalination ratio reached 90% after 24 h operation with ferricyanide as catholyte. Mehanna et al. (2010a) recognized that the designation of an air-cathode MDC would make the MDC more practical, as well as avoid the pollution of ferricyanide usage. However, the usage of expensive catalyst (such as Pt) or catholyte (such as ferricyanide) make the MDC with common abiotic cathodes costly and unsustainable, and the inexpensive and environmental friendly biocathode which uses microbial catalyst to facilitate dissolved oxygen reduction in MDC is uncommon and practically urgent. Wen et al. (2012) applied a biocathode MDC producing a maximum voltage of 609 mV, and the salinity of salt water (35 g/L NaCl) was reduced by 92% in a single desalination cycle (within 480 h). In addition, the behaviors of multiple ions and their effects on membrane scaling and fouling have attracted considerable attention. Luo et al. (2012a) demonstrated that the sparingly soluble cations in saltwater negatively affected the performance of MDC due to the scaling on the membrane surface.

Currently, sodium acetate and domestic wastewater are widely used as anodic fuels (Forrestal et al., 2012; Yuan et al., 2012). Luo et al. (2012b) compared the performance of MDC (using raw wastewater as the sole anolyte) with a traditional MFC, and concluded that the power output (8.01 W/m<sup>3</sup>) of MDC was enhanced fourfold. Besides, the organic matters in sewage sludge and manure as anodic substrates in bioelectrochemical system can also be degraded efficiently and lead to a stable electricity generation (Jiang et al., 2010; Scott and Murano, 2007). Jiang et al. (2009) reported a maximum power density of 8.5 W/m<sup>3</sup> and an average stable voltage (0.687 V) during the treatment of sewage sludge by MFC with potassium ferricyanide as catholyte, and the total chemical oxygen demand (TCOD) of sludge was reduced by 46.4% after 250 h operation. Zhang et al. (2012) reported a higher maximum power output of 13.2 ± 1.7 W/m<sup>3</sup> with an open circuit potential (OCV) of 0.94 V using biocathode MFC reactor fed with sewage sludge. Although several studies have attempted to clarify the behavior of MFCs on sludge treatment, to the authors' knowledge no research has been performed to examine the efficiency of MDC on sewage sludge, especially on the desalination with dewatered sludge as fuel.

The primary objectives of this study are to (1) construct a five-chambers biocathode MDC to desalinate salt water using dewatered sludge as anode fuel; (2) evaluate the desalination and electricity generation performance, and electrochemical characteristics of electrodes during MDC operation; (3) clarify the biodegradation mechanism of sludge organic matter during the whole operation.

## 2. Methods

### 2.1. MDC construction

The MDC reactor (Fig. 1) consisted of five plexiglas chambers with a symmetric construction, i.e. 2 cathode chambers (cube, 90 mm × 90 mm × 30 mm, V = 500 mL), 2 desalination chambers

(cube, 90 mm × 90 mm × 30 mm, V = 500 mL), and 1 anode chamber (cylinder,  $\Phi = 12$  cm,  $h = 13$  cm, V = 1400 mL). Each of the two chambers was connected by square window (90 mm × 90 mm) to facilitate ionic migration. Specifically, the anion-exchange membranes (AEM, Ultrex AMI-7001, 90 mm × 90 mm) were used to separate the anode chamber and desalination chambers, and cation-exchange membranes (CEM, Ultrex CMI-7000, 90 mm × 90 mm) were used to separate cathode chambers and desalination chambers. The graphite fiber brush (GFB) made of carbon fibers (STS40 24 K, 650 ± 17 m<sup>2</sup>/m<sup>3</sup>, 1.7  $\Omega$ /cm, average fiber diameter of 7.0  $\mu$ m, Toho Tenax) was cut to a fixed length and twisted by two titanium wires. Three GFBs ( $\Phi 55$  mm × 120 mm) were connected as the anode, and one graphite brush ( $\Phi 30$  mm × 85 mm) embedded in graphite granules (GGs) (diameter of 1–5 mm, 0.5–0.6  $\Omega$ /granule, 55 m<sup>2</sup>/m<sup>3</sup>, Jiuxin Carbon Goods Co., Jilin, China) was the cathode. AEM, CEM and electrodes were prepared according to the previous report (Mehanna et al., 2010b). Standard copper wires sealed with epoxy resin (2 M) were used to connect the anodes and the cathodes to form a closed circuit, in which a fixed resistance ( $R_{ex} = 1000 \Omega$ ) was applied as an external load.

### 2.2. MDC operation

The dewatered sludge collected from Taiping wastewater treatment plant (Harbin, China) and stored at 4 °C prior to experiments was directly used as anodic inocula and substrate. The sludge had a pH (w/v: 1/10) of 5.88 ± 0.13, water content of 80.27 ± 1.45%, organic matter of 65.32 ± 3.04% (dry basis), total carbon (TC) of 407.6 ± 18.9 mg/g, total nitrogen (TN) of 43.8 ± 1.5 mg/g, NH<sub>4</sub><sup>+</sup>-N of 1.2 ± 0.4 mg/g, total phosphorus (TP) of 14.5 ± 2.7 mg/g, TCOD of 1068.4 ± 22.3 mg/g, and soluble chemical oxygen demand (SCOD) of 14.6 ± 3.1 mg/g. Top soil obtained from the turf at Harbin Institute of Technology was used for catholyte inoculation, which contained abundant hydrogen-oxidizing bacteria (Zhang et al., 2011). The cathode chambers were fed with a solution containing (per liter of deionized water) 1.0 g NH<sub>4</sub>Cl, 1.2 g K<sub>2</sub>HPO<sub>4</sub>, 0.5 g MgSO<sub>4</sub>, 0.5 g KCl, 0.14 g KH<sub>2</sub>PO<sub>4</sub>, 0.01 g Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·H<sub>2</sub>O and 10 mL trace elements as previously described (Rabaey et al., 2005). The cathode chambers were continuously aerated at 300 mL/min to maintain a dissolved oxygen level of 4 mg/L.

Different concentrations of NaCl solution (5, 10 and 35 g/L) were added in the desalination chambers of MDC. During the start-up period, the desalination chambers were filled with 35 g/L of NaCl solution. When the reactors operated stably, desalination rates of different NaCl concentrations were investigated. The desalination rate of 35 g/L NaCl solution was firstly investigated, and the NaCl solution and catholyte were changed every 4 d during the 12 d operation. Afterwards the MDC was operated for 42 d and daily desalination rates were analyzed in triplicate. The experimental procedures for the 10 g/L and 5 g/L NaCl solution were similar to that of 35 g/L NaCl solution. The MDC desalination performance was again examined at 35 g/L NaCl solution during the efficiency-reduction period.

All experiments were conducted at room temperature of 25 ± 2 °C. The anode substrate of fresh dewatered sludge was replenished when the voltage of MDC was below 0.3 V. Two MDC reactors were operated simultaneously, one of which was operated in open circuit as a control.

### 2.3. Analyses and computations

Voltage was directly recorded every minute using a 32-channel data acquisition system (PISO-813, ICP DAS, Co., Ltd, Beijing, China), and the average voltage was calculated every 24 h. The anode and cathode potentials were measured against a reference

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