



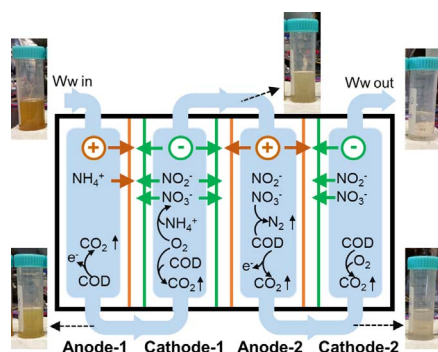
Enhanced organics removal and partial desalination of high strength industrial wastewater with a multi-stage microbial desalination cell



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GRAPHICAL ABSTRACT



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ABSTRACT

Conventional microbial desalination cells (MDCs) extract organic energy from wastewater for in situ desalination of saline water, but cannot treat industrial wastewater containing high strength organics that may cause blocking in the thin membrane stack. In this study, a multi-stage MDC (M-MDC) was fabricated and operated for simultaneous treatment and desalination of diluted industrial wastewater. With wastewater (COD: 8723 ± 456 mg/L, conductivity: $24,612 \pm 772$ μ S/cm) flowing serially from anode-1 \rightarrow cathode-1 \rightarrow anode-2 \rightarrow cathode-2, the maximum power density of the M-MDC reached 566.1 mW/m², with removal efficiencies of COD, TN, NH₄⁺-N, and conductivity reaching 97.8%, 90.6%, 98.4%, and 31.6% respectively with addition of 1 V external voltage. The high strength organics were significantly removed due to alternative anaerobic/oxic conditions, the nitrogen removal was enhanced by simultaneous electrical migration and biological nitrification/denitrification. Moreover, the addition of external voltage enhanced current generation (67.4 ± 3.9 mA), desalination efficiency, coulombic efficiency (14.1%), and organics removal in two anodes (47.3%), producing an effluent (COD < 500 mg/L, TN < 15 mg/L) qualified to be discharged into municipal wastewater pipe systems. These results indicated great potential of M-MDC to work as a pretreatment technology for high strength industrial wastewater.

1. Introduction

Industrial wastewater often contains high strength organics, nitrogen, and salinity that need to be removed (or partially removed)

before being reused or discharged into municipal sewage systems and natural water bodies. Traditional biological treatment processes such as anaerobic/oxic activated sludge were often utilized to remove organics and nitrogen due to their low cost and high efficiency [1], and

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membrane-based desalination technologies, such as electro dialysis [2] and reverse osmosis [3] were often utilized to remove salinity. However, traditional biological processes could not remove salt, and their performance on nitrogen removal was often insufficient to meet the national discharge standard [4]. As for desalination, reverse osmosis and electro dialysis also could not be directly utilized for desalination of industrial wastewater containing high strength of organics, which may cause fouling on their nano-pores [5] or blocking of millimeter-thick membrane stack [6]. Therefore, developing a technology that can simultaneously realize organics removal and desalination would be of great significance for the treatment of high strength industrial wastewater.

Microbial desalination cells (MDCs) are newly developed technology that can treat wastewater in anode chamber and desalinate salt water in the middle membrane stack with electric field generated by oxidation of organics contained in wastewater [7]. Since MDCs realize desalination without extra energy input, they have drawn great attention and experienced considerable developments in the past several years. Stacked MDCs were invented to improve the desalination rate and current efficiency [8,9]. Ion exchange resins were packed in the desalination chamber to decrease internal resistance for treatment of low salinity water [10–13]. Some extended functions, such as MDCs for nutrients recovery [14–16] and heavy metal removal [17], filtration cathode MDC for integrated organics removal and desalination [18], forward osmosis combined MDCs for seawater desalination [19,20], and magnification to 10 L for practical application [12] etc., were also reported. These studies expanded the function of MDC systems, and improved their performance in terms of current generation, desalination efficiency, operation stability, and practical feasibility etc. However, most of these MDCs utilized wastewater in the anode chamber to produce electricity desalinating salt water in the middle membrane stack [21–23]. It was difficult for them to realize simultaneous organics removal and desalination for the same stream of wastewater. In addition, traditional MDCs also faced the limitation of insufficient organics and nitrogen removal in anode effluent (< 85% [24]), and they were not capable for desalination of salt water containing high organics etc., which greatly limited their application in wastewater treatment field.

To address these disadvantages, a multistage MDC was invented in our previous study based on the conception of multistage anaerobic/oxic biological process and electro dialysis technology [25]. It realized significant organics removal due to alternative anaerobic/oxic conditions, advanced nitrogen removal by co-effect of electrical migration and biological nitrification/denitrification, and self-driven desalination with energy produced by organics contained in wastewater itself, without any blocking in desalination chambers. In this study, a same configuration of M-MDC was continued, but the M-MDC was utilized for treatment of high strength industrial wastewater to take its advantage of efficient organics removal. During operation, a diluted industrial essence wastewater was utilized as feed water; it flowed serially from anode-1 chamber to cathode-1, anode-2 and cathode-2 chambers. The influence of hydraulic retention time (HRT) on removal efficiency of organics, nitrogen, and salinity was investigated. Detailed analysis on wastewater quality was conducted to determine the contribution of each chamber. Various external voltages were also added on the M-MDC to enhance its performance including current generation, desalination, organics removal, coulombic efficiency etc. In addition, microbial community structure on different electrodes of the M-MDC was also characterized, to help understand their functions in the M-MDC system.

2. Materials and methods

2.1. Construction of M-MDC

The structure and size of the M-MDC is the same with that reported in our previous study [25]. The M-MDC is made of polycarbonate. It has

2 anode chambers, 2 biocathode chambers, and 3 concentrate chambers (Fig. S1A & B). The dimension of the anode, biocathode, and concentrate chambers are $3.5 \times 8 \times 20$ cm (560 mL), $4.5 \times 8 \times 20$ cm (720 mL), and $0.5 \times 8 \times 20$ cm (80 mL), respectively. The total volume of the M-MDC is 2800 mL, and the sectional area is 160 cm^2 . In anode and biocathode chambers, conductive activated carbon granules (diameter of c. 5 mm, length of c. 10 mm) were packed as electrode material as well as carrier for the attachment of bioelectrochemical active bacteria, and two titanium meshes (screen mesh size of c. 2×2 mm) were assembled in each electrode chamber as current collectors (Fig. S1A). The anode, biocathode, and concentrate chambers were separated by cation exchange membranes (CEMs, 2.0 mol/kg, Shanghai, China) and anion exchange membranes (AEMs, 1.8 mol/kg, Shanghai, China). The CEMs were set adjacent to anode chambers, and the AEMs were set adjacent to cathode chambers (Fig. S1A). Mixed cation exchange resin (Na form, 4.2 mmol/g, Sinopharm, China) and anion exchange resin (Cl form, 3.0 mmol/g, Sinopharm, China) were packed into concentrate chambers (with a ratio, by weight, 1.0: 1.4) as ion conductor to decrease internal resistance [10–13]. An aeration diffuser was embedded in the bottom of each biocathode chamber to supply O_2 (Fig. S1A). The external resistance of each separate MDC was 2Ω at all conditions.

2.2. Solution and operation

Wastewater taken from an essence processing company in Tengzhou, Shandong province of China was adopted as influent of the M-MDC. The wastewater had a chemical oxygen demand (COD) of $133,700 \pm 4600$ mg/L, conductivity of $247,300 \pm 6700$ $\mu\text{S/cm}$, pH of 6.6 ± 0.4 , a strong irritant smell and a deep yellow color. Domestic wastewater taken from Tsinghua campus was employed to dilute the industrial wastewater for bioelectrochemical treatment, which had a COD of 393.8 ± 80.0 mg/L, $\text{NH}_4^+ \text{-N}$ of 102.4 ± 21.6 mg/L, total nitrogen (TN) of 129.2 ± 12.7 mg/L, conductivity of 1673.3 ± 39.2 $\mu\text{S/cm}$, and pH of 7.66 ± 0.02 . Deionized water was applied as feed water of concentrate chamber.

During operation, the industrial wastewater was first diluted by 15 times utilizing domestic wastewater taken from Tsinghua campus. The diluted industrial wastewater had a COD of 8723 ± 456 mg/L, TN of 94.6 ± 8.2 mg/L, $\text{NH}_4^+ \text{-N}$ of 83.7 ± 5.8 mg/L and conductivity of $24,612 \pm 772$ $\mu\text{S/cm}$. Then the diluted wastewater flowed successively from anode-1 (anaerobic) to cathode-1 (oxic), anode-2 (anaerobic), and cathode-2 (oxic) (Fig. S1A), to realize simultaneous desalination and removal of organics and nitrogen. Meanwhile, deionized water was circulated at a rate of 12 mL/min in the three parallel concentrate chambers and an external reservoir, working as concentrate. The concentrate had a total volume of 560 mL, and its replacement cycle kept the same with HRT of wastewater in M-MDC, maintaining a 20% volume production of concentration at all conditions.

Since CEMs and AEMs were adjacent to anode and cathode chambers respectively in this configuration, cations and anions would migrate respectively from anode and cathode chambers to the concentrate chambers under electric field, realizing simultaneous desalination and organics removal in electrode chambers. Therefore, the M-MDC was expected to have the following advantages: (i) simultaneous desalination and biological treatment of high-strength wastewater; (ii) enhanced organics and nitrogen removal due to the multi-stage anaerobic/oxic biochemical reaction; (iii) no clogging in the diluate and concentrate chamber (due to high organic wastewater flowed in the big spacing diluate chamber (electrode chamber) and DI water flowed in the small spacing concentrate chamber). In addition, the conductivity of wastewater decreased along the flow direction, which was also conducive to the growth and activity of electrode microorganisms.

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