



# Submersible microbial desalination cell for simultaneous ammonia recovery and electricity production from anaerobic reactors containing high levels of ammonia



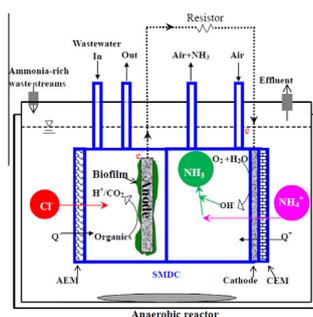
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## HIGHLIGHTS

- Novel submersible microbial desalination cell as a new method for ammonia recovery.
- In situ lowering ammonia level in anaerobic reactor by ammonia recovery.
- Current driven  $\text{NH}_4^+$  migration and free  $\text{NH}_3$  diffusion for ammonia transportation.
- Initial ammonia level and external resistance affected the ammonia recovery.
- Coexistence of other cations had no negative effect on the ammonia transportation.

## GRAPHICAL ABSTRACT



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## ABSTRACT

High ammonia concentration in anaerobic reactors can seriously inhibit the anaerobic digestion process. In this study, a submersible microbial desalination cell (SMDC) was developed as an innovative method to lower the ammonia level in a continuous stirred tank reactor (CSTR) by in situ ammonia recovery and electricity production. In batch experiment, the ammonia concentration in the CSTR decreased from 6 to 0.7 g-N/L during 30 days, resulting in an average recovery rate of 80 g-N/m<sup>2</sup>/d. Meanwhile, a maximum power density of 0.71 ± 0.5 W/m<sup>2</sup> was generated at 2.85 A/m<sup>2</sup>. Both current driven  $\text{NH}_4^+$  migration and free  $\text{NH}_3$  diffusion were identified as the mechanisms responsible for the ammonia transportation. With an increase in initial ammonia concentration and a decrease in external resistance, the SMDC performance was enhanced. In addition, the coexistence of other cations in CSTR or cathode had no negative effect on the ammonia transportation.

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## 1. Introduction

Anaerobic digestion of waste streams containing urea, protein and organic nitrogen is always accompanied with the release of high level of ammonia in the anaerobic reactor which in return

can significantly inhibit the process (Angelidaki and Ahring, 1993; Hansen et al., 1998; Nielsen and Ahring, 2007; Nielsen and Angelidaki, 2008; Smith, 2002). Ammonia is defined here as the sum of free ammonia ( $\text{NH}_3$ ) and ionic ammonia ( $\text{NH}_4^+$ ). Inhibition has been reported to start at a total ammonia level of 1.5 g-N/L and is especially distinct when digesting swine or poultry manure, which often has total ammonia concentration higher than 4 g-N/L (Nielsen and Angelidaki, 2008). Thus, in order to re-stabilize or

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rescue the anaerobic digestion process, the excess ammonia in the anaerobic reactors need to be removed. Both ammonia-based fertilizer production and ammonia removal from wastes are energy intensive processes, and thus, direct ammonia recovery from anaerobic reactors may bring the synergistic benefits to both nitrogen recycling and anaerobic digestion process.

Recently, microbial fuel cell (MFC) and microbial electrolysis cell (MEC) as typical bioelectrochemical systems (BESs) have been demonstrated as promising technologies for ammonia removal or recovery (Ieropoulos et al., 2011; Kelly and He, 2014; Kuntke et al., 2012; Wu and Modin, 2013). The ammonia removal in a BES is mainly through biological processes such as nitrification and electrochemical denitrification (Kelly and He, 2014; Zhang and Angelidaki, 2013). In respect to alternative ammonia removal technologies, bioelectrochemical recovery of the ammonia via cathode stripping may improve the environmental and economic sustainability of wastewater treatment systems (Kelly and He, 2014; Kuntke et al., 2012). An electricity assisted MFC-based system has also been recently developed for recovery of ammonium and phosphorus (Zhang et al., 2014). MFC has even been employed to remove ammonia from *Arthrospira maxima* biomass before anaerobic digestion (Inglesby and Fisher, 2012). Abiotic operation of MEC has also been attempted for ammonia recovery, but the electric energy cost is much higher than that of biotic operation (Desloover et al., 2012). Compared to conventional technologies such as  $\text{NH}_3$  stripping, the BESs for ammonia recovery require much less energy (or even produce net energy) and no need of additional chemicals (Kuntke et al., 2012; Wu and Modin, 2013). Though promising, there are still several challenges need to be addressed before field application. For example, direct exposure of anodic biofilm to the ammonia-rich streams may inhibit the exoelectrogenic activity and therefore deteriorate the recovery process (Kuntke et al., 2011; Nam et al., 2010). In addition, the anode or cathode process may alter the characteristics of the waste streams (e.g., pH), and thereby affecting the reuse or further treatment of the waste streams after the recovery. One of the most recent BESs termed microbial desalination cell (MDC) might offer a new solution (Cao et al., 2009; Chen et al., 2011; Jacobson et al., 2011; Mehanna et al., 2010). MDC was initially developed for water desalination, but it has several common characteristics with MFC, such as high cathode pH due to  $\text{OH}^-$  accumulation (Chen et al., 2012; Pavlostathis and Giraldo Gomez, 1991). Moreover,  $\text{NH}_4^+$  like other cations (e.g.,  $\text{Na}^+$ ) in the desalination chamber can also pass through CEM to the cathode (Kuntke et al., 2011, 2012). Therefore, we hypothesize that MDC could be a new method to recover the excess ammonia from the anaerobic reactors suffering ammonia inhibition. The MDC could not only inherit the advantages of MFC or MEC for ammonia recovery, but also avoid the risk of ammonia toxicity on the anodic biofilm. In addition, it would not affect the stream properties such as pH during recovery, as the waste streams will be fed into the desalination chamber instead of anode or cathode. Nevertheless, the possibility of such new method has never been demonstrated. In addition, to achieve the goal of cost-effective recovery of ammonia without disturbing the anaerobic digestion process, a novel MDC design that can be in situ applied to the existing anaerobic digestion reactors such as continuous stirred tank reactor (CSTR) should be pursued.

Considering the above, a submersible microbial desalination cell (SMDC) was developed in this work as an alternative approach to recover ammonia from anaerobic reactors. The SMDC contains only two chambers and can be easily submerged into existing anaerobic digestion reactors such as CSTR for in situ ammonia recovery, which could greatly reduce the construction, operation and maintenance costs. This work provided a proof-of-concept demonstration of SMDC as a new avenue to lower the ammonia

level in the anaerobic reactor by ammonia recovery and electricity production.

## 2. Methods

### 2.1. Experimental setup

The SMDC was a rectangular reactor made of nonconductive polycarbonate plates (Fig. 1), which contains anode and cathode chamber (inside dimensions  $3 \text{ cm} \times 3 \text{ cm} \times 1 \text{ cm}$  for each). An AEM (AMI 7001, Membrane international, NJ) and a CEM (CMI 7000, Membrane international, NJ) based membrane electrode assembly were placed on the end of anode and cathode chambers, respectively. The anode and cathode chambers were separated with the polycarbonate plate. The CEM and cathode made of carbon cloth ( $3 \text{ cm} \times 3 \text{ cm}$ , Quintech, Germany) were used to prepare membrane electrode assembly ( $0.5 \text{ mg Pt/cm}^2$ ) as previously described (Jacobson et al., 2011; Zhang and Angelidaki, 2012a). The anode was made of carbon paper ( $3 \text{ cm} \times 3 \text{ cm}$ , E-TEK division, USA) and was pre-colonized in an MFC inoculated with domestic wastewater and fed with acetate modified buffer solution (see detail below) over 6 months (Zhang and Angelidaki, 2012b). External resistance of  $10 \Omega$  was used except otherwise indicated. All the electrical connection and electrode pretreatment were performed according to previous study (Zhang et al., 2009).

### 2.2. Reactor operation

The SMDC (18 mL total compartment volume) was submerged about 1.5 cm below the liquid surface in a CSTR reactor (1 L, liquid volume of 0.4 L) which was used to mimic the anaerobic digester. Acetate modified nutrient buffer solution from feed bottle (2000 mL, Kimax®GL 45) was continuously recirculated through the anode chamber at a rate of 10 mL/min. The acetate modified nutrient buffer solution was prepared with deionized water containing (per liter, pH  $7.0 \pm 0.5$ ): 1.6 g  $\text{CH}_3\text{COONa}$ , 4.58 g  $\text{Na}_2\text{HPO}_4$ , 2.45 g  $\text{NaH}_2\text{PO}_4$ , 0.05 g  $\text{NH}_4\text{Cl}$ , 0.1 g  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ , 0.1 g  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ , 0.1 g KCl and 10 mL of trace mineral metals solution (Cao et al., 2009). The medium in the feeding bottle was replaced every 8 h to ensure sufficient organic matter supply and prevent pH drop

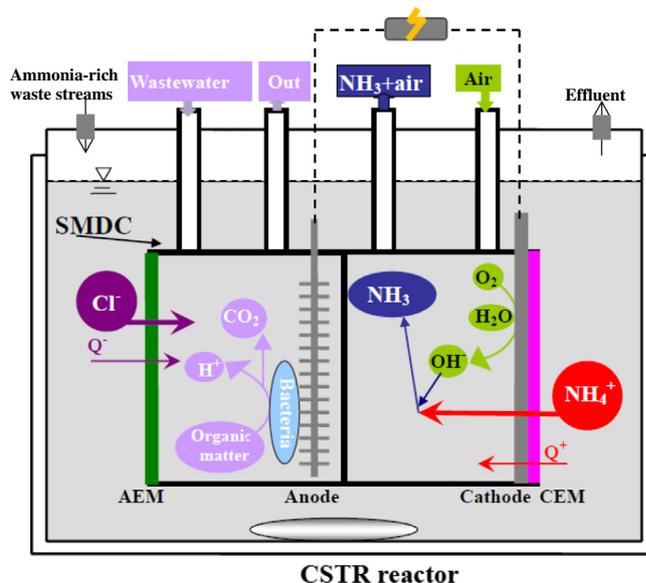


Fig. 1. Schematic diagram of the SMDC in an anaerobic reactor (CSTR).

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