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*Published in:* Proceedings of 2nd international conference on renewable energy gas technology

Publication date: 2015

Document Version Publisher's PDF, also known as Version of record

#### Link back to DTU Orbit

Citation (APA):

Zhang, Y., & Ángelidaki, I. (2015). Innovative bioelectrochemical-anaerobic-digestion coupled system for ammonia recovery and biomethane production from ammonia-rich residues. In Proceedings of 2nd international conference on renewable energy gas technology

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## Innovative bioelectrochemical-anaerobic-digestion coupled system for ammonia recovery and biomethane production from ammonia-rich residues.

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#### 1. Introduction

Anaerobic digestion (AD) is an attractive technology widely used for organic waste treatment and simultaneously energy production in the form of biogas [1]. Livestock manures together with organic industrial wastes are widely applied as substrate, however, their high ammonia content can inhibit the digestion process, resulting in serious economic losses to the biogas plant [2-6]. Both biotic and abiotic technologies have been developed to counteract the ammonia inhibition by means of removal and/or recovery [7-13]. However, most of the methods are uneconomical, requiring high energy input, have low efficiency, and need addition of chemicals, and thus, fail for practical application.

Recently, the bioelectrochemical system (BES) has drawn attention as a renewable technology producing energy or products from wastes [14-17]. Microbial fuel cell (MFC) and microbial electrolysis cell (MEC) as typical BES have been demonstrated as promising technologies to recover ammonia from urine or reject water with net energy production and no additional chemical costs [18-20]. An MFC-based system has also been recently developed to remove/recover both ammonium and phosphorus from wastewater with power supply [21]. MFC has even been employed to remove ammonia from Arthrospira maxima biomass before AD process [22].

MFC or MEC could be a potential new approach to prevent ammonia toxicity during AD process by ammonia recovery. Nevertheless, the exoelectrogenic activity in the anode of MFC or MEC may also be inhibited at high ammonia levels (>4 g NH<sub>4</sub><sup>+</sup>-N/L) due to direct exposure of anodic biofilm to the ammonia-rich streams [16,23]. Moreover, the processes may reduce the organic contents of the waste for the following AD process. All these challenges need to be addressed before practical application. Operation of MEC as an abiotic electrochemical reactor has also been attempted, but the electric energy cost is much higher compared to biotic operation [24]. One of the most recent which BES is SO called microbial desalination cell (MDC) may offer a new solution [25-28]. MDC was initially designed for water desalination, but it shares several common characteristics with MFC, such as high pH in the cathode due to OH<sup>-</sup> accumulation [29]. Moreover,  $NH_4^+$  as a cation can also pass through CEM [16-19]. Therefore, we hypothesize that MDC could be a new method to recover ammonia and thereby preventing ammonia toxicity, by integrating AD of residues ammonia-rich into the desalination chamber. The MDC could not only inherit the advantages of MFC or MEC for ammonia recovery, but also avoid the risks of ammonia toxicity on the anodic biofilm and the reduction of organic contents. Besides, this new method involves a unique integration of AD with ammonia recovery instead of simply connecting the two processes in series, leading to an alternative way for both nutrient recovery and bioenergy production. However, the feasibility of MDC for ammonia recovery has never been reported. Furthermore, in order to reduce the costs upon the integration of the MDC and AD process, an in-situ applicable MDC design that can utilize existing AD infrastructures such as continuous stirred tank reactors (CSTR) should be pursued.

Both AD and BES are complex processes involving various biochemical reactions number catalyzed by а large of microorganisms [15,30]. It is crucial to understand the microbial ecology especially when these two processes are integrated. However, most of the BES studies were focusing on the ammonia recovery, while the impact of the recovery process on the microbial communities enriched in the both systems has never been reported.

#### 2. Technology description

A novel hybrid system consists of a submersible MDC (SMDC) and a CSTR reactor was developed in this work as an alternative approach to counteract ammonia toxicity during anaerobic digestion by means of in-situ ammonia recovery and electricity production. The SMDC contains only two chambers, i.e. anode and cathode chamber, and the conventional desalination chamber is not required (Figure 1). This special design makes it in-situ applicable to existing biogas infrastructures (e.g., CSTR), and thereby may lower the construction, operation and maintenance costs upon the integration. Such reactor design has never been applied for ammonia recovery or integrated with AD process. This work provided proof-of-concept а

demonstration of the integrated SMDC-CSTR system for ammonia recovery and biogas enhancement. The effects of ammonia concentration, external resistance and ionic concentrations on the system performance were investigated. The microbial community structure and diversity in such new system were studied using Ion Torrent PGM sequencing based 16S rRNA genes analysis.

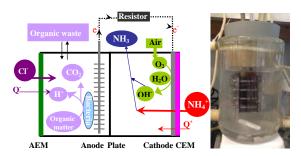
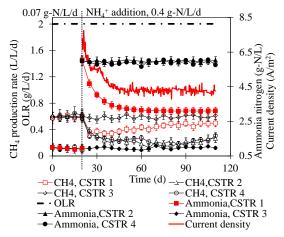


Figure 1 Schematic of the SMDC (A) and the image of the integrated SMDC-CSTR (B).  $Q^+$  and  $Q^-$  are unspecified cations and anions, respectively.

#### 3. Results

As shown in Figure 2, the CH<sub>4</sub> production rate decreased immediately by about 47% in the CSTR 1, CSTR 2 and CSTR 4 after elevating the total ammonia concentration to 6 g-N/L and the NLR to 0.4 g-N/L/d. After another 55 days of operation, the CH<sub>4</sub> production rate in the CSTR 1 gradually increased back to 83% of the value that was observed in the CSTR 3. Comparatively, no significant increase in CH<sub>4</sub> production rate was observed in CSTR 2 and CSTR 4. At this condition, 112% extra biogas was produced due to alleviation of the ammonia inhibition as the ammonia level in the reactor was reduced. In the CSTR 1, the ammonia concentration decreased with time and reached to a stable level of 3.1±0.1 g-N/L from day 60, resulting in a recovery rate of 86 g-N/ $m^2$ /d (based on the area of CEM). To the contrary, there was no significant change of ammonia concentration in the CSTR 2 and CSTR 4. It was also observed that the total VFA in the CSTR 1, CSTR 2 and CSTR 4 increased immediately after ammonia elevation (approx. 2.5 g-HAc/L), indicating ammonia inhibition. Unlike to the other reactors, the total VFA in the CSTR 1 decreased gradually with the ammonia level.



# *Figure 2 The system performance during continuous reactor operation.*

The mass balance analysis on nitrogen showed that 40.8% of the input ammonia was recovered by the SMDC. The preliminary energy balance analysis indicated that net energy (8.77 kWh/kgN, data not shown) was produced during continuous operation. It must be noted that this preliminary analysis was made to compare different recovery methods. The organic carbon requirement in the anode for the ammonia recovery is about 1 g-COD/g-N. The acetate modified buffer was used here to mimic waste streams in the anode, but wastewater or the supernatant effluent from anaerobic digester (e.g., CSTR) could be used as anode carbon source for the full scale implementation, giving synergistic advantages for both process simplification and energy savings.

#### 4. Conclusions

This study demonstrated a novel technology for counteracting ammonia toxicity during anaerobic digestion by means of in-situ ammonia recovery and electricity production using integrated SMDC-CSTR system. Furthermore, for the first time, we applied the newly launched Ion Torrent PGM high-throughput sequencing technology to reveal the microbial community diversity and structure that enriched in such new system.

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