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Ammonia inhibition on hydrogen enriched anaerobic digestion of manure under mesophilic and thermophilic conditions

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ABSTRACT

Capturing of carbon dioxide by hydrogen derived from excess renewable energy (e.g., wind mills) to methane in a microbially catalyzed process offers an attractive technology for biogas production and upgrading. This bioconversion process is catalyzed by hydrogenotrophic methanogens, which are known to be sensitive to ammonia. In this study, the tolerance of the biogas process under supply of hydrogen, to ammonia toxicity was studied under mesophilic and thermophilic conditions. When the initial hydrogen partial pressure was 0.5 atm, the methane yield at high ammonia load (7 g NH_4^+-N L^{-1}) was 41.0% and 22.3% lower than that at low ammonia load (1 g NH_4^+-N L^{-1}) in mesophilic and thermophilic condition, respectively. Meanwhile no significant effect on the biogas composition was observed. Moreover, we found that hydrogentrophic methanogens were more tolerant to the ammonia toxicity than acetoclastic methanogens in the hydrogen enriched biogas production and upgrading processes. The highest methane production yield was achieved under 0.5 atm hydrogen partial pressure in batch reactors at all the tested ammonia levels. Furthermore, the thermophilic methanogens at 0.5 atm of hydrogen partial pressure were more tolerant to high ammonia levels ($\geq$5 g NH_4^+-N L^{-1}), compared with mesophilic methanogens. The present study offers insight in developing resistant hydrogen enriched biogas production and upgrading processes treating ammonia-rich waste streams.

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1. Introduction

Anaerobic digestion (AD) is a sustainable technology that has been used for the treatment of various waste streams such as animal manure, food waste and sludge. However, AD treatment of the substrates containing high total ammonia (ammonium ion and free ammonia) concentration can be seriously inhibited by the ammonia which is produced during the biodegradation of proteins, urea and nucleic acids (Angelidaki and Ahring, 1994). There are two principal forms of inorganic ammonia nitrogen in aqueous solution: Ammonium ion (NH_4^+) and free ammonia (NH_3). NH_3 has been considered to be the main inhibitor (Rajagopal et al., 2013; Yenigün and Demirel, 2013). NH_3 molecules diffuse into the microbes' cells freely which can cause proton imbalance, increase maintenance energy requirements, change intracellular pH and inhibit specific enzyme reactions (Gallert et al., 1998; Sprott and Patel, 1986). NH_3 concentration mainly depends on temperature, pH and total ammonia concentration in anaerobic digestion process (Hafner and Bisogni, 2009). For example, the concentration of NH_3 increases with an increase in pH and/or temperature which causes the enhanced ammonia toxicity on the AD process (Nielsen and Angelidaki, 2008).

The AD process can be described by four distinctive steps namely: hydrolysis, acidogenesis, acetogenesis and methanogenesis. In detail, with the exception of the initial solubilisation of complex particulate material, methanogenesis seems to be the rate-limiting step. Moreover methanogens are the most vulnerable to ammonia compared to other groups of microorganisms involved in AD process (Angelidaki et al., 2011). There are two distinct methanogenic pathways for converting acetate to methane, which has been well described in previous studies (Fotidis et al., 2013; Stams and Plugge, 2009; Wang et al., 2015). There are many papers referring on the sensitivity of the methanogens to ammonia (Fotidis et al., 2013). It was reported that acetoclastic methanogens (i.e. *Methanosarcinaceae* spp. and *Methanosaetaceae* spp.) are more vulnerable to ammonia toxicity compared to hydrogenotrophic methanogens (i.e. *Methanomicrobiales* spp., *Methanococcales* spp., *Methanocellales* spp., *Methanobacteriales* spp. and *Methanopyrales* spp.) (Angelidaki and Ahring, 1993; Yenigün and Demirel, 2013).

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Recently, an innovative AD process, which introduces hydrogen produced by water electrolysis using excess electricity from wind mill into anaerobic digester and subsequently converts it together with carbon dioxide in biogas into methane has been developed for simultaneous H_2 utilization and in-situ biogas upgrading (mainly refers to reduction of CO_2 content), giving synergistic advantages for both wind mills and biogas plants. (Deng and Hagg, 2010; Luo and Angelidaki, 2012; Luo et al., 2012). Such process has several advantages over conventional AD process: (1) low cost for further biogas upgrading since CO_2 content was reduced; (2) increase of methane production; (3) fully use of the wind mill capacity. Though promising, the H_2 enriched AD process is just emerging from a technology perspective. There are several challenges to be addressed for being able to develop a sustainable feasible technology. One important aspect is the resistance of the process to ammonia inhibition, which is the very aspect that is unclear so far. Considering that most of the feedstocks (e.g., cattle manure) in biogas plants (especially in Denmark) contain high level of ammonia, it is of outmost important to reveal the sensitivity of the process to high level of ammonia in order to accelerate the wide application of the technology. The outcome of such investigation will also help to find suitable strategy to counteract the ammonia inhibition.

During this process, enrichment of hydrogenotrophic methanogenic cultures in anaerobic biogas reactors is occurring. In Luo and Angelidaki (2012)'s study, hydrogen was injected into anaerobic reactors to achieve a hydrogen partial pressure of 0.8 atm. After two months cultivation with H_2, the hydrogenotrophic methanogenic activities increased to 198 mL CH_4 (g VSS h)$^{-1}$ under mesophilic and 320 mL CH_4 (g VSS h)$^{-1}$ under thermophilic condition, from around 10 mL CH_4 (g VSS h)$^{-1}$ of the original inoculum. This indicated that hydrogenotrophic methanogens were successfully enriched by long term injection of hydrogen. Thus, it would be obvious to assume that this process would be more resistant or tolerant to ammonia toxicity due to the enrichment of hydrogenotrophic methanogenesis compared to the conventional AD processes (Luo & Angelidaki, 2012, 2013b; Luo et al., 2012). So far, information about the effect of ammonia toxicity on this innovative AD process is still lacking. Therefore, in this study, the effect of different ammonia levels on hydrogen enriched biogas upgrading process (different hydrogen partial pressure were included in the current study) in anaerobic reactors at both mesophilic and thermophilic temperature was explored.

2. Materials and methods

2.1. Inoculum and feedstock

The mesophilic and thermophilic inoculum were obtained from mesophilic and thermophilic anaerobic reactors in Hashøj Biogas plant (Denmark) and Snertinge Biogas Plant (Denmark), respectively. Both biogas plants use a mixture of manure (pig and cattle) and organic waste (fat and flotation sludge from food industries) as feedstock. As feedstock, dairy manure taken from Hashøj municipality (Denmark) was used in this study. The dairy manure was mixed in one plastic barrel and was sieved, in order to remove the large solid particles, and then kept at −18 °C. Before use as substrate in the batch experiment, the frozen manure was thawed and stored at 4 °C for 2–3 days. The basic characteristics of the inoculum and feedstock were analyzed and shown in Table 1.

2.2. Experimental setup

Both mesophilic and thermophilic inocula were incubated under four different ammonia concentrations (1, 3, 5 and 7 g NH_4^+-N L^{-1}) with NH_4Cl as ammonia source. As batch reactors, vials with 118 mL total and 40 mL working volume, respectively were used. The working volume contained 10 mL inoculum, 10 mL dairy manure and 20 mL distilled water. After filling the content into the vials, butyl rubber stoppers and aluminum crimps were used to seal them. Then all the batch reactors were flushed with nitrogen (flow rate 290 ml/s) for 10 min. Before the hydrogen injection, the same volumes as the injected hydrogen of gas were extracted from the batch reactors to make sure the total pressure of all the batch reactors was the same. After that, 19.5, 39 and 78 mL of hydrogen were introduced with syringes into batch reactors to obtain different hydrogen partial pressure (0.25, 0.5, and 1 atm) for each ammonia level. Moreover, batch reactors without hydrogen addition, were also included. Additionally, reactors only with inoculum were used as blanks to evaluate the residual methane production. Two shaking incubators (37 ± 1 °C and 55 ± 1 °C, 180 rpm) were used for mesophilic and thermophilic batch reactors respectively and each condition was evaluated in triplicates (n = 3).

2.3. Analytical methods

Total solids (TS), volatile solids (VS), pH, total ammonia and total Kjeldahl nitrogen (TKN) were measured according to APHA's Standard Methods (Federation and Association, 2005). The pH level of the batch reactors was determined by using PHM99 LAB pH meter which was connected to the Gel pH electrode (pHC3105-8, Radiometer analytical). The electrode was filled with a gel containing KCl. Before measuring samples, the pH meter was calibrated at the temperature of the corresponding batch reactors. Shimadzu-14A gas chromatograph (GC) equipped with a thermal FID detector with hydrogen as a carrier gas (Shimadzu, Kyoto, Japan) was used to measure methane accumulation in the headspace of batch reactors. Hydrogen concentration in batch reactors was measured by using GC-TCD fitted with a 4.5 m × 3 mms-m stainless column packed with Molsieve SA (10/80). Moreover, a gas-chromatograph (GCTCD) equipped with a column of 1.1 m × 3/16 "Molsieve 137 and 0.7 m × 1/4" chromosorb 108 (MGC 82-12, Mikrolab A/S, Denmark) was used to determine the biogas composition in the headspace of batch reactors. The bottles were not vented during the whole experiment. The methane concentration (in percentage) in the headspace was measured by GC with pressure. Thus, the accumulated methane was obtained by multiplying headspace volume of the batch reactors (78 ml) and the methane concentrations measured by GC. Additionally, the accumulated volatile fatty acids (VFA) concentration of the batch reactors were determined by using a gas-chromatograph (HP5890 series II) equipped with a flame ionization detector and a FFAP fused silica capillary column, (30 m × 0.53 mm i.d., film thickness 1.5 μm), which uses nitrogen as carrier gas.

2.4. Calculations

2.4.1. Calculation of methane production

The hydrogen injected into the batch reactors was consumed by hydrogenotrophic methanogens to produce methane. Thus, the reactors with hydrogen addition had higher average methane yield compared to the reactors without hydrogen injection. Therefore, the calculation of subtracting the theoretical methane production from the introduced hydrogen in the batch reactors was made.

2.4.2. Statistical analysis

OriginLab program (OriginLab Corporation, Northampton, Massachusetts) was used for all the statistical analyses. For statistical analysis, one way Analysis of Variance (ANOVA) at 0.05 level was used. The effects of two factors (ammonia concentrations and

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