



In-situ biogas upgrading in thermophilic granular UASB reactor: key factors affecting the hydrogen mass transfer rate



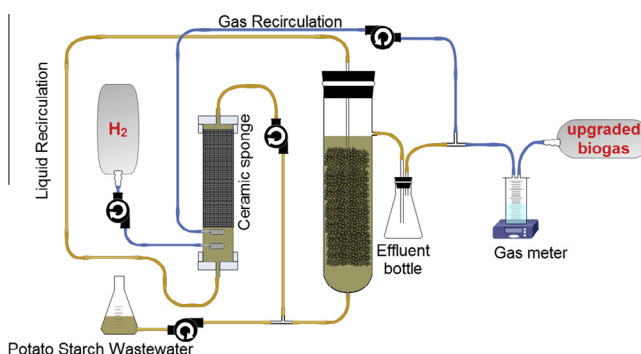
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HIGHLIGHTS

- Biogas upgrading to 82% CH₄ is feasible in a thermophilic granular UASB reactor.
- H₂ is introduced in a separate chamber having a volume of 25% of the reactor.
- H₂ low gas-liquid mass transfer rate limits the availability of H₂ for methanogens.
- H₂ distribution can be improved using porous inert devices, like ceramic sponge.
- Gas recirculation and chamber configuration help to maximize CO₂ conversion to CH₄.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 11 August 2016
Received in revised form 12 September 2016
Accepted 17 September 2016
Available online 20 September 2016

Keywords:

In-situ biogas upgrading
Hydrogen
Gas-liquid mass transfer rate
UASB
Granules
Anaerobic digestion

ABSTRACT

Biological biogas upgrading coupling CO₂ with external H₂ to form biomethane opens new avenues for sustainable biofuel production. For developing this technology, efficient H₂ to liquid transfer is fundamental. This study proposes an innovative setup for in-situ biogas upgrading converting the CO₂ in the biogas into CH₄ via hydrogenotrophic methanogenesis. The setup consisted of a granular reactor connected to a separate chamber, where H₂ was injected. Different packing materials (rashig rings and alumina ceramic sponge) were tested to increase gas-liquid mass transfer. This aspect was optimized by liquid and gas recirculation and chamber configuration. It was shown that by distributing H₂ through a metallic diffuser followed by ceramic sponge in a separate chamber, having a volume of 25% of the reactor, and by applying a mild gas recirculation, CO₂ content in the biogas dropped from 42 to 10% and the final biogas was upgraded from 58 to 82% CH₄ content.

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1. Introduction

Anaerobic digestion (AD) of organic waste is a promising technology for sustainable energy production (Weiland, 2010). The

potato-starch processing industry produces, as byproduct, up to 1 m³ of potato juice per ton of potatoes (Abeling and Seyfried, 1993). Potato-starch wastewater contains high concentration of biodegradable compounds, such as starch and proteins, suitable for biogas production via AD (Barampouti et al., 2005). Biogas typically contains ~50–70% CH₄ and 30–50% CO₂. Biogas upgrading to CH₄ content higher than 90% increases its heating value and its potential applications as alternative to natural gas (Deng and Hägg, 2010).

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Methods currently available for biogas upgrading are mainly based on physicochemical CO₂ removal. Nevertheless, these technologies require use of additional materials and chemicals considerably increasing the cost of the process and energy input. Alternatively, biogas can be upgraded by biologically coupling H₂, derived from water electrolysis, with CO₂ present in the biogas to convert them to CH₄. H₂ can be produced using the electricity generated by the surplus of energy from wind mills or photovoltaic facilities, which may result from variable weather conditions. This reaction is carried out by a group of microorganisms known as hydrogenotrophic methanogenic archaea that utilize CO₂, as carbon source, and H₂, as electron donor, to produce CH₄ via hydrogenotrophic methanogenesis (Muñoz et al., 2015). Previous studies demonstrated that the addition of H₂ to a conventional biogas reactor can lead to 20–40% increase in CH₄ production rate, as result of the conversion of the CO₂ present in the biogas to additional CH₄ (Luo and Angelidaki, 2013; Luo et al., 2012).

Although biological biogas upgrading offers economical and technical advantages compared to traditional methods (Nordberg et al., 2012), H₂ mediated biogas upgrading is still challenging. One of the main limitations is the low H₂ gas-liquid mass transfer rate (Bassani et al., 2015; Luo and Angelidaki, 2012; Luo et al., 2012).

H₂ gas-liquid mass transfer rate can be described by the following equation (1):

$$r_t = 22.4k_L a(H_{2gTh} - H_{2l}) \quad (1)$$

where r_t (L/(L.day)) is the H₂ gas-liquid mass transfer rate, 22.4 (L/mol) is the gas volume to mole ratio (1 mol gas corresponds to 22.4 L at STP), $k_L a$ (day⁻¹) is the gas transfer coefficient, H_{2gTh} (mol/L) represent the H₂ concentration in the gas phase while H_{2l} (mol/L) the H₂ dissolved in the liquid phase. One way to increase H₂ gas-liquid mass transfer rate is by increasing $k_L a$. This coefficient is specific for given reactor configuration and operating conditions (Pauss et al., 1990). Therefore, $k_L a$ can be modulated by changing parameters such as mixing speed (Bhattacharyya and Singh, 2010; Luo and Angelidaki, 2012), gas recirculation (Guiot et al., 2011) and H₂ diffusion device (Luo and Angelidaki, 2013; Díaz et al., 2015).

Besides, high-rate anaerobic treatment using up-flow anaerobic sludge blanket (UASB) reactors is commonly applied in industrial wastewater treatment plants (Gomec, 2010; Sevilla-Espinosa et al., 2010). Moreover, typically a UASB process is expected to provide higher methane content in the biogas than a CSTR process (Nizami et al., 2012).

UASB reactors' technology is based on the presence of granular sludge comprised of microorganisms responsible for catalyzing the biological conversion of organic matter to biogas. High recirculation flow rates and consequent high up-flow velocities have an important role for the hydraulic mixing improving the wastewater to granules contact (Powar et al., 2013; Zheng et al., 2012). It has been previously reported that carbohydrate degraders and hydrogenotrophic methanogens are predominant in starch-grown granules, likely due to their role in the interspecies H₂ transfer with syntrophic bacteria (Lu et al., 2015). Moreover, previous studies on H₂ mediated biogas upgrading demonstrated that H₂ affected the microbial community composition enhancing the hydrogenotrophic methanogenic pathway and the syntrophic relationship between bacteria and hydrogenotrophic methanogens (Bassani et al., 2015).

In this study an innovative setup consisting of a UASB granular reactor connected to a separate chamber, where the H₂ was injected, was designed to mediate efficient H₂ transfer to liquid phase for biological conversion of H₂ and CO₂ to CH₄. Key factors affecting the H₂ gas-liquid mass transfer rate were evaluated. More

specifically, the effect of different operating conditions aiming in increasing $k_L a$ of H₂ to gas, and thereby increase the gas to liquid transfer, were studied to elucidate their role in improving CO₂ and H₂ conversion to CH₄. Parameters examined were liquid and gas recirculation and configuration of diffusion devices. Moreover, the addition of packing materials as a mean to minimize the gas bubble size and thus increase the gas dissolution in the liquid was tested. Finally, the effect of gas retention time was evaluated using single or serial chamber configurations with different working volumes.

2. Materials and methods

2.1. Substrate characteristics and feedstock preparation

Potato-starch wastewater substrate was obtained from Karup Kartoffelmelfabrik potato-starch processing factory, Denmark. Because potato-starch processing involves an up-concentration step, the provided substrate was diluted 10 times with water and Basal Anaerobic (BA) medium, to adjust the volatile solids (VS) content to the required operation conditions. Successively, the substrate was stored at −20 °C, in 5 L bottles and thawed at 4 °C for 2–3 days, before usage. BA medium was prepared as described in Supplementary Information (SI). The diluted substrate had a pH of 6.05, chemical oxygen demand (COD) of 21.76 ± 0.15 g/L, total solids (TS) and VS content of 26.14 ± 0.17 and 18.73 ± 0.12 g/L, respectively. The concentration of total volatile fatty acids (VFA) was 49.29 ± 4.94 mg/L. Total Kjeldahl Nitrogen (TKN) and ammonium nitrogen NH₄⁺ (NH₄-N) were 1.24 ± 0.01 and 0.30 ± 0.01 g-N/L, respectively.

2.2. Setup and operation of the reactors

Each setup was composed of a UASB reactor with a working volume of 1.4 L, connected to a separate H₂-injection chamber with a working volume of 0.2 L. The feeding was introduced from the bottom of the UASB. The reactors were inoculated with 550 g of mesophilic granules, obtained from Colsen wastewater treatment plant treating potato starch wastewater (The Netherlands) and BA medium. The granules were adapted to thermophilic conditions for 25 days by feeding the reactors with diluted potato starch wastewater at hydraulic retention time (HRT) of 7 days and organic loading rate (OLR) of 2.79 gVS/L.day. A double net-separator was located in the upper part of each UASB to prevent the wash out of granules. One setup (R1) was used as upgrading reactor, while the other (R2) was utilized as control reactor operated throughout the experiment without H₂ injection. Both reactors were maintained at thermophilic conditions (55 ± 1 °C) by circulating hot water through a water jacket around the UASB reactors glass walls.

After the startup phase, the whole experiment was divided in 8 periods. During period I the OLR was increased to 3.73 gVS/L day shortening the HRT to 5 days (Pre H₂ phase). The recirculation flow rate was set to 4 L/h. From period II, H₂ was continuously injected to R1 through a diffuser placed at the bottom of the H₂-injection chamber (In-situ phase). Rashig rings (5 mm internal diameter) were inserted into the separate chamber of both reactors to maximize the H₂ gas-liquid mass transfer rate in case of R1. The volumetric H₂ flow rate was set to 4 times the CO₂ production rate (in the gas phase) recorded before the H₂ addition, according to Luo and Angelidaki (2013), i.e. 3.5 L/L.day, and then reduced to improve the H₂ consumption. In period III, the recirculation flow rate of both reactors was increased to 7 L/h. Successively, in period IV, rashig rings were replaced by an inert alumina ceramic sponge, while in periods V and VI different gas recirculation flow were applied. In order to evaluate the effect of the gas retention time,

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