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Research paper

Development of an innovative two-stage fermentation process for high-calorific biogas at elevated pressure

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ABSTRACT

The two-stage high-pressure fermentation (HPF) process enables the production of methane at high operating pressure. Pressure significantly reduces the energy needed for injecting the produced biogas into the gas grid by 45–60%. It also allows for incorporating large parts of the necessary biogas upgrading process into the synthesis step. As a result, the two-stage HPF process provides pressurized biogas with methane volume fraction ranging from 0.75 to 0.94. The pressure is not generated by energy intensive gas compression, but in-situ by microbial gas production. In comparison to conventional biomethane production, the overall costs could be reduced up to 20%. HPF is most beneficial when its operating pressure is adapted to that of the gas grid.

The article presents briefly the development of the two-stage HPF beginning with tests in batch reactors, followed by experiments on gas solubility, and proof-of-concept in continuously operated methanogenesis reactors (MR) up to 9 bar. It also represents the effect of incorporating microfiltration (MF) of the feed stream, on improving the biogas quality and process stability of a continuously operated lab scale HPF process. By linking the MF with the HPF, methane volume fraction in the MR increases from 0.86 to 0.94 at 25 bar. Finally, the simulation and experimental results show good agreement with each other thereby making them a good basis for further optimization of the HPF process.

1. Introduction

In 2016, Germany had a 31.7% share of renewables in gross electricity consumption and biogas, as a carbon dioxide (CO_2) neutral energy source, accounted for 17.2% of the renewable-based electricity generation [1]. However, biogas with a methane (CH_4) volume fraction of 0.55–0.6 is predominantly used for combined heat and power (CHP) generation [2]. In rural areas, opportunities to use the heat is often difficult to find. To improve the energy efficiency of biogas utilization in such cases, purification of biogas to biomethane and its injection into the natural gas grid is a reasonable alternative, as it decouples gas production from its usage (both in time and space) [2,3]. Furthermore, the admixing of biomethane to natural gas provides the potential of further reducing GHG emissions by the greening-of-gas. By the end of 2016, at least 367 plants in Europe with a total biomethane production of 17,267 GWh were in operation whereas 210 biomethane plants in Germany produce 130,000 m^3h^{-1} by the end of 2017 [4,5].

In conventional anaerobic digestion processes, biogas is produced in a single digester at ambient pressure. The most common reactor types are continuously operated stirred tank reactors (CSTR) which are often used for wet substrates. Reactor volume varies between 1000 and 4000 m^3 (CSTR) [7,8]. If dry substrates (> 25% mass fraction of dry matter) are applied, horizontal or vertical plug flow reactors (PFR) with volumes of up to 700 m^3 can be used instead [7,8].

In case of biomethane injection into the gas grid, gas upgrading and gas compression is mandatory before injection into the gas grid. Operating the fermentation process at high pressure would allow for integrating the gas compression and the upgrading of biogas into the microbial production process itself. The pressure, which can be produced by the microorganisms in-situ, would also allow for incorporation of water scrubbing in the fermentation process and for providing a product gas with significantly higher methane volume fractions in the range of 0.75–0.9 and for coupling the process with any subsequent synthesis steps; like biological methanation. Unfortunately, increasing the pressure of state-of-the-art CSTR fermenters results in higher production costs as pressure equipment in such large dimensions is

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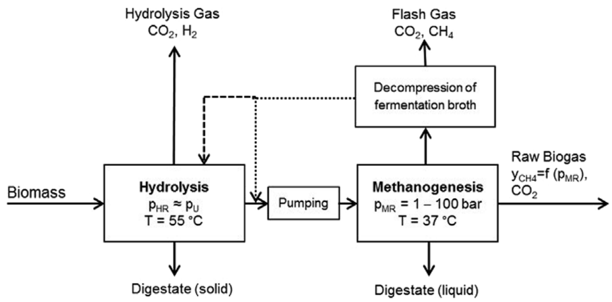

Fig. 1. HPF process conditions in the Hydrolysis reactor (HR) and the Methanogenesis reactor (MR).

relatively more expensive.

Biomass degradation involves several microorganisms and takes place in four steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis [9,10]. The rate of the individual degradation steps can be reduced via applying different process conditions in terms of pH value and temperature. In common single stage digesters, pH ranges between 6.5 and 8 and is one of the process variables which is hardly adjustable [11]. Obviously, single process values cannot be adapted when all four degradation steps take place in a single digester.

Consequently, in the two-stage high-pressure Fermentation (HPF), the degradation steps are separated into two individual reactors, the Hydrolysis (leach-bed or acidification reactor) and the Methanogenesis (also known as anaerobic filters) reactor.

As can be seen in Fig. 1, the Hydrolysis reactor (HR) is operated at ambient pressure and 55 °C, the Methanogenesis reactor (MR) at pressures of up to 100 bar and 37 °C [12,13]. Reactor pressure in the MR can be adapted to the pressure levels of the gas grid according to the type of grid system e.g. flowlines, gathering lines or transmission pipelines. Additionally, in the HPF process, the methane formation takes place in a fixed-bed reactor. The microbes are immobilized on a support material, which enables to operate the process at very high organic loading rates and short retention times, thus enabling a significant reduction of the required reactor size in comparison to conventional single-stage CSTR fermenter systems [12,13].

2. Development of high-pressure fermentation technology

Many previous studies showed the advantages of two-stage anaerobic digestion systems [12–19] operated at ambient pressure. Although there is microbiological activity in the deep sea at pressures of up to 100 bar [20], it was not clear if the microbial consortia relevant for methane production would allow methane production under these conditions in a technical reactor.

2.1. Preliminary examinations on the microbial activity at high pressure

For first fundamental experiments on the influence of pressure of up to 100 bar on the microbial activity, pressurized batch MRs were implemented at the University of Hohenheim. The design of these batch systems were already published [13,21].

The experimental results (see Fig. 2) showed no negative effects on methane yield and production rate at the applied pressure levels of 1, 50 and 100 bar. All organic acids were degraded by the end of the experiments and specific methane yields did not differ from the calculated potentials at any pressure level. These findings lead to the construction of a continuously operated high-pressure MR.

2.2. Gas solubility and evolution of pH value at high pressure

Due to the fact that high reactor pressure and different solubility of CO_2 and CH_4 lead to a high-calorific biogas, high operating pressure is

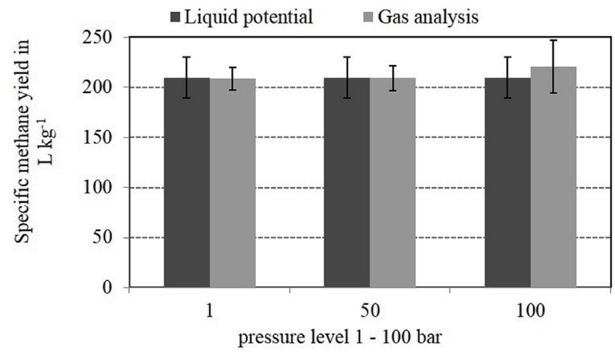

Fig. 2. Specific methane yield per kg chemical oxygen demand (COD) input at T = 37 °C calculated from the liquid and gas analysis for three different initial pressures [13].

also a big challenge for the pH-probes and its calibration. Theoretical and experimental study at DVGW-EBI in order to calculate the pH value via gas composition and gas/liquid equilibrium resulted in a technical application instead of using expensive high-pressure pH-probes. At DVGW-EBI, experimental investigations on gas solubility in fermentation liquids at different pressure and temperatures were already conducted [21–23]. Furthermore, the effect of dissolved CO_2 and dissociated carbonic acid on pH value was determined experimentally for various test media containing acids, alcohols and various cations and anions. The data were used for developing a correlation for prediction of gas solubility of CO_2, based on the Henry coefficient of the individual gases but extended by the effect of volatile organic acids etc. in the solution on the dissociation of CO_2.

As shown in Fig. 3 and according to recent publications, the calculation of the pH value, either performed by single analysis of each acids, alcohols, and cations or by sum parameters show a good agreement with experimental results [21]. Without considering the dissolved CO_2 (see LIQ in Fig. 3), the calculated pH value deviates strongly from measured pH value. The theoretical and fundamental findings on gas solubility are incorporated in the modelling of the process (see Sec. 5).

2.3. Proof-of-concept in methanogenesis reactor

For proof-of-concept of the HPF, the high-pressure approach was demonstrated in former projects with a stand-alone Methanogenesis reactor at pressures up to 9 bar, using maize silage as a substrate [21–24,26,27]. The feedstock was hydrolyzed in a batch hydrolysis unit and transferred to a buffer tank, supplying the high-pressure MR with liquid acids. The studies demonstrated a continuous operation of the high-pressure part of the HPF system and showed that biogas

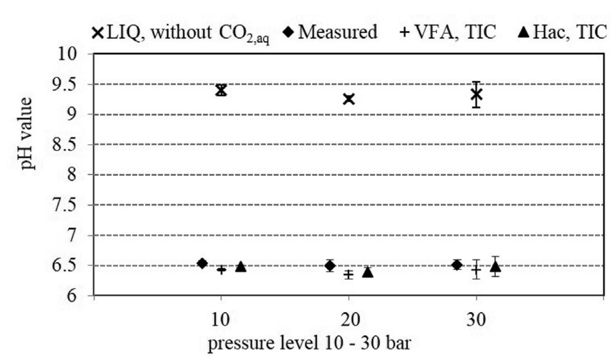

Fig. 3. Measured and calculated pH values for three different initial pressures. The pH value calculated either only by liquid analysis (LIQ), volatile fatty acids (VFA) and total alkalinity (TIC) or by acetic acid equivalent (HAc) and total alkalinity (TIC) [21].