Overcoming Scale-Up Challenges in Biological Methanation: A 300-Day Pilot TBR Study

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The combustion of fossil fuels is the leading cause of global carbon dioxide (CO_2) emissions. To meet necessary CO_2 emission cuts, societies must shift consumption habits, enhance energy efficiency, and significantly expand renewable energy adoption—including wind, solar, and hydropower—to displace fossil fuel-based generation. Power-to-Gas (PtG) is a key technology for transitioning to a renewable energy system, offering long-term energy storage and reducing carbon emissions by replacing fossil fuels with renewable gas. The process involves using surplus renewable electricity to produce hydrogen (H_2) through electrolysis (*Power-to-Hydrogen*). This H_2 can then be combined with CO_2 from external sources to synthesize methane (CH_4) (*Power-to-Methane*). Both H_2 and renewable CH_4 can be integrated into existing natural gas infrastructure, leveraging its vast storage capacity—enough to meet months of energy demand in countries like Germany and Denmark. However, H_2 injection into gas grids faces technical and regulatory constraints, whereas synthetic natural gas (SNG or methane) can be stored and distributed without significant restrictions (Thema et al., 2019).

Methanation offers a promising pathway to reduce the reliance on natural gas. In biological methanation, anaerobic microbes convert H_2 and CO_2 into CH_4, creating a renewable gas substitute. Initially developed for upgrading biogas, biological methanation can also utilize CO_2 captured from industrial flue gases—a significant source of global emissions. By converting this CO_2 into renewable CH_4, the technology could play a key role in decarbonizing energy systems and supporting climate goals. Trickle bed reactors (TBRs) amid several reactor configurations have demonstrated the highest efficiency for biological methanation. These reactors operate by filling them with packing materials characterized by high-surface-area and substrate gases, in this case, H_2 and CO_2. The liquid nutrient solution is distributed at the top and trickles down through the packed bed, delivering essential nutrients to the microbial biofilm growing on the surface of carrier material. This design enables efficient conversion of gases while maintaining flexible operation (Feickert Fenske et al., 2023).

While TBRs have gained growing attention for biological methanation, most research to date has focused on small-scale systems. Scaling up to pilot experiment dimensions presents significant technical hurdles, particularly in maintaining consistent process performance and conversion efficiency. The complexity increases substantially when moving from laboratory to industrial-scale operations, with system stability becoming a critical concern. Thus, our study focused on scaling up a high-efficiency lab-scale biomethanation system to a 100 L pilot-scale trickle bed reactor (TBR). The evaluation of this system was assessed in terms of output gas composition, pH values and volatile fatty acid (VFA) levels.

Figure 1. Photo of the prototype pilot methanation unit; (1) pilot reactor, (2) nutrient sump, (3) gases mass flow controllers, (4) nutrient recirculation pump, (5) nutrient sump mixing pump, (6) water displacement gas-metering system.

More specifically a 100 L working volume TBR (Fig. 1), that operated for almost a year was fed continuously with independently inserted H_2 and CO_2 gases, in a portion of 80% H_2 and 20% CO_2. The overall target was to maintain stable operational conditions with GRT values of at least 2 hours initializing from 4 hours. Maintaining precise GRT control was critical for optimizing gas-liquid-microbe contact time, directly governing the system's methane conversion efficiency. The dual challenge involved sustaining these retention times while simultaneously achieving >95% CH_4 purity in the product gas - a benchmark for pipeline-quality natural gas. This methane threshold serves two key purposes: (1) it validates the biological process's high conversion efficiency, and (2) ensures the gas meets specifications for direct injection into

energy grids or use as transportation fuel. The liquid (nutrient) medium was supplied from a storage vessel (no.2 in Fig.1) to the top of the TBR using a peristaltic pump (Sci-Q 300, Watson Marlow, United Kingdom). The composition of output gas stream and the concentration of VFAs were qualified and quantified using gas chromatography.

The pilot scale system exhibited stable performance over the entire operational period, maintaining a consistent methane production rate of 4.65 $L_{CH4}/(L_R \cdot d)$ while achieving CH_4 purity exceeding 95% at a gas retention time of 1 hour (Fig. 2). This high conversion efficiency indicates effective substrate utilization and minimal process losses, highlighting the robustness of the bioconversion process under the tested conditions.

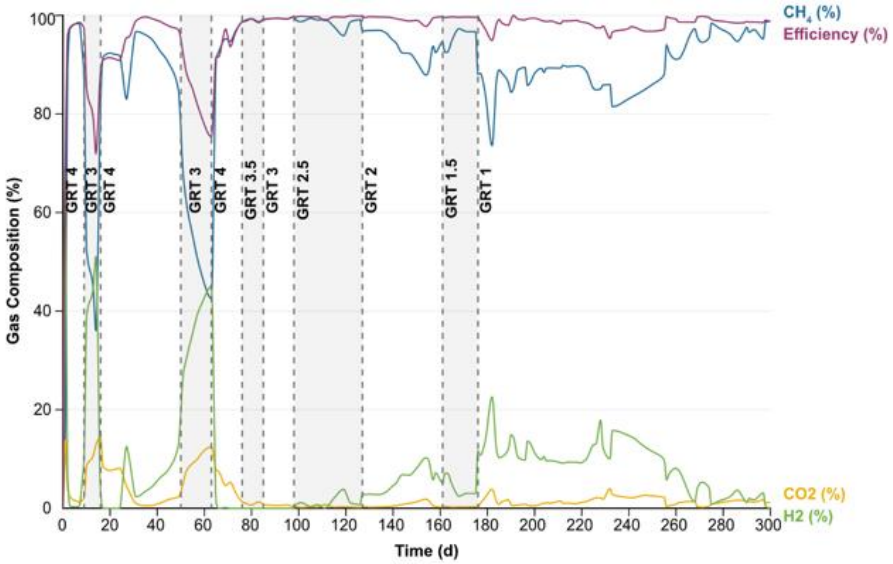

Figure 2. Evolution of output gas composition (H_2, CO_2, CH_4) during operational phases with varying gas retention times, with phase transitions indicated by dashed lines.

Notably, VFA concentrations remained low across all tested GRTs (Fig. 3), despite observable pH fluctuations, suggesting that operational challenges at reduced GRTs were primarily driven by physicochemical factors rather than biological limitations or shifts in metabolic pathways. The strong correlation between pH dynamics and methanation efficiency underscores the critical key role of pH control in optimizing the biomethanation process. Overall, these findings demonstrate the system's robustness under demanding operational conditions and mark a significant advancement toward the real-world scenario of scaling biological methanation technologies.

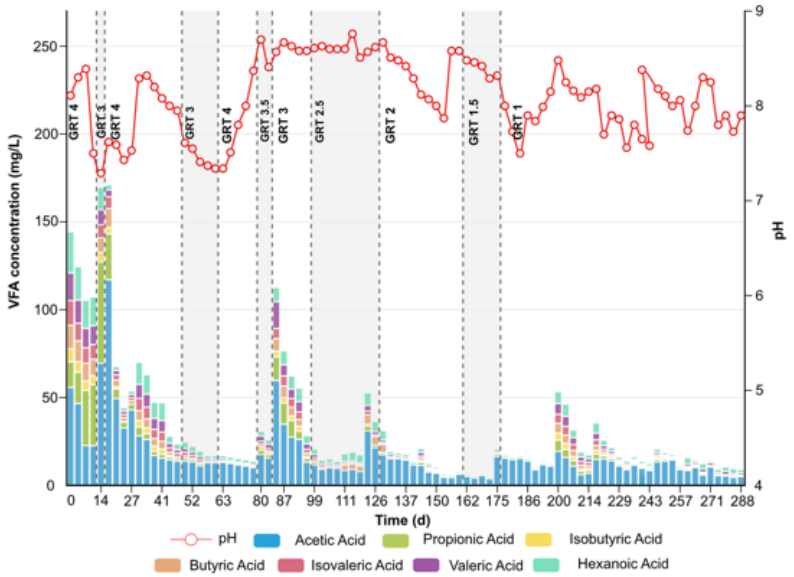

Figure 3. Stacked bar plot showing volatile fatty acid concentrations with corresponding pH variation (red line) across different gas retention times. Vertical dashed lines denote GRT phase transitions.

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