

## Applying fluidized bed methanation for Power-to-Gas: long duration test and upscaling

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### Highlights

- Fluidised bed methanation applied for biogas upgrading without prior CO<sub>2</sub> separation.
- Successful long duration test (1100h) validated gas cleaning and methanation step.
- Influence of operation conditions could be predicted by rate based model.
- Pilot scale hydrodynamic experiments support scale-up.

### 1. Introduction

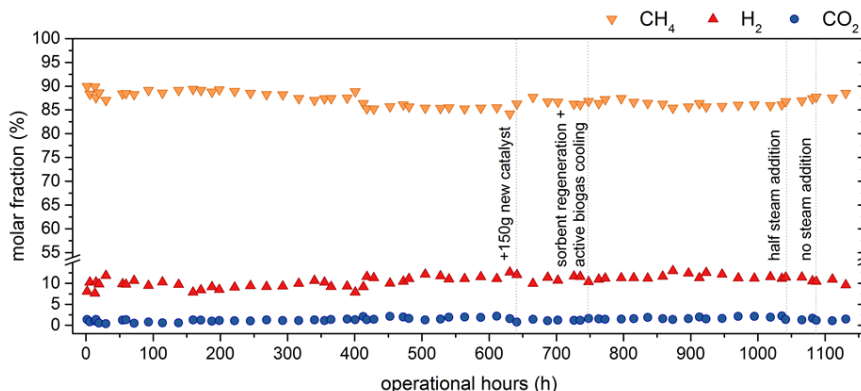
Under the different Power-to-Methane pathways, up-grading of biogas is the most promising approach for the near future. It has the lowest technical and financial hurdles to overcome due to the high methane content in biogas. This is especially true for technologies without prior separation of methane and carbon dioxide. Such process applying catalytic fluidized bed methanation was validated in collaboration with energie360°, Switzerland's largest gas supplier. By direct methanation of biogas, the CH<sub>4</sub> yield of biogas is considerably increased ( $\text{CO}_2 + 4 \text{H}_2 \leftrightarrow \text{CH}_4 + 2 \text{H}_2\text{O}$ ;  $\Delta H_r^0 = -165 \text{ kJ/mol}$ ). For this purpose, H<sub>2</sub> was added in a test facility to the biogas from the sewage treatment plant as well as a bio-waste digestion plant in Zurich-Werdhölzli.

### 2. Methods

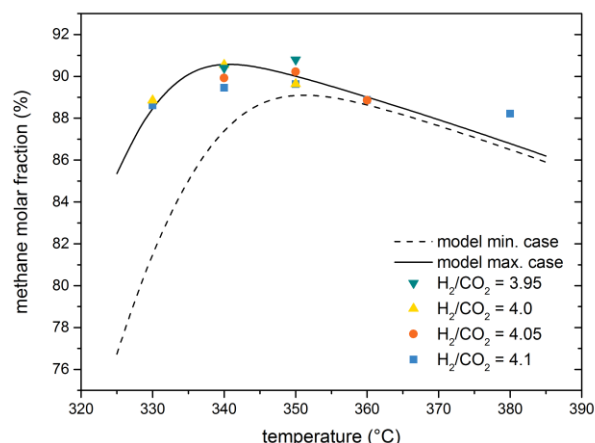
A pilot plant COSYMA (20 kW<sub>SN</sub>G) was built and a first series of methanation tests using bottled gas investigated the influence of important operation conditions (pressure, temperature, H<sub>2</sub>/CO<sub>2</sub> ratio). Then, the plant was installed in Zurich close to an existing biogas upgrading plant and connected to the gas grid. In 2017, a 1100 hour long-duration experiment was successfully conducted with a single catalyst charge. In parallel, an existing rate based model of a fluidized bed methanation reactor [1,2] was used to predict the influence of the operation conditions, and the predictions were compared to the modelling results. Further, up-scaling activities focus on pilot scale hydrodynamic experiments with optical probes in a vessel of 22 cm diameter and X-ray tomography. Determination of parameters such as bubble size distribution, bubble rise velocities [3] allows to further improve the rate based reactor model.

### 3. Results and discussion

The predicted gas quality of the methanation operating with real biogas was reached and the gas was injected into the natural gas grid, see figure 1.

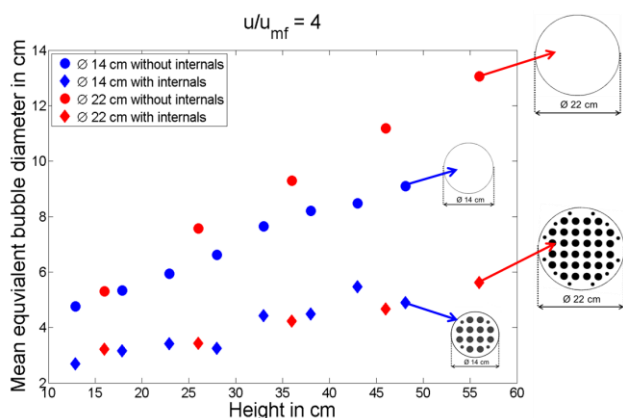


**Figure 1.** Long duration test of fluidized bed methanation using hydrogen for converting biogas to biomethane



**Figure 2.** Dry molar fraction of methane after the reactor as function of reactor temperature for different hydrogen-to-CO<sub>2</sub> ratios. Comparison between experiment and rated based model for minimum and maximum case at p=5.7 barg

While at temperatures lower than 330°C, the reactions are limited by kinetics, at higher temperatures, thermodynamic equilibrium leads to a characteristic maximum in the yield, see figure 2. The rate based model using kinetics that was determined for CO-methanation is satisfyingly able to proper predict this maximum. The hydrodynamic work shows that vertical internals lead to significantly smaller bubbles and simplify up-scaling, see figure 3.



**Figure 3.** Experimentally determined bubble diameters for fluidised beds with and without vertical internals: the latter lead to significantly smaller bubbles and simplify up-scaling

#### 4. Conclusions

A 1'100 hour long-time experiment validated successfully the fluidized bed methanation of biogas. The predicted gas quality was reached and the gas was injected into the natural gas grid. The experimental results of fluidised bed methanation could be predicted by the rate based model. Based on these results and the findings from pilot scale hydrodynamic experiments, the technology can be scaled up to industrial scale.

#### References

- [1] Kopyscinski J. Production of synthetic natural gas in a fluidized bed reactor - Understanding the hydrodynamic, mass transfer, and kinetic effects. Dissertation ETH Zürich Nr. 18800, 2009.
- [2] Teske SL. Integrating rate based models into a multi-objective process design & optimisation framework using surrogate models. Dissertation EPF Lausanne; 2014
- [3] F. Schillinger, S. Maurer, E.C. Wagner, J.R. van Ommen, R.F. Mudde, T.J. Schildhauer; Int. J. Multiphase Flow 97 (2017)46-59

#### Keywords

Fluidised bed methanation; rate based model; Power-to-Gas with biogas; pilot scale hydrodynamic experiments.