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SNG production through fixed-bed methanation of biomass derived syngas with simplified warm gas cleaning

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Decentralised SNG production

The SNG production through gasification and subsequent catalytic methanation forms a possibility for an efficient and flexible utilization of solid fuels. In Europe small- to mid-scale decentralised SNG plants are the more viable concepts resulting in specific boundaries:

- A fully developed gas grid for SNG distribution
- Small- to mid-scale due to limited feed-in capacities of the gas grid
- Reduced process complexity

The presented SNG concept originates from the CO₂freeSNG2.0 project which was originally dedicated to lignite but can be easily applied for renewable biomass sources.

SNG production in project CO₂freeSNG2.0

The combination of CO₂ removal with removal of impurities in a single process step reduces the total process complexity. Hence, the proposed process consists of three main steps:

- Allothermal steam gasification in fluidized-bed
- Warm gas scrubbing with K₂CO₃ for integrated sulfur and CO₂ removal
- Two-stage methanation with intermediate water sequestration (Figure 1)

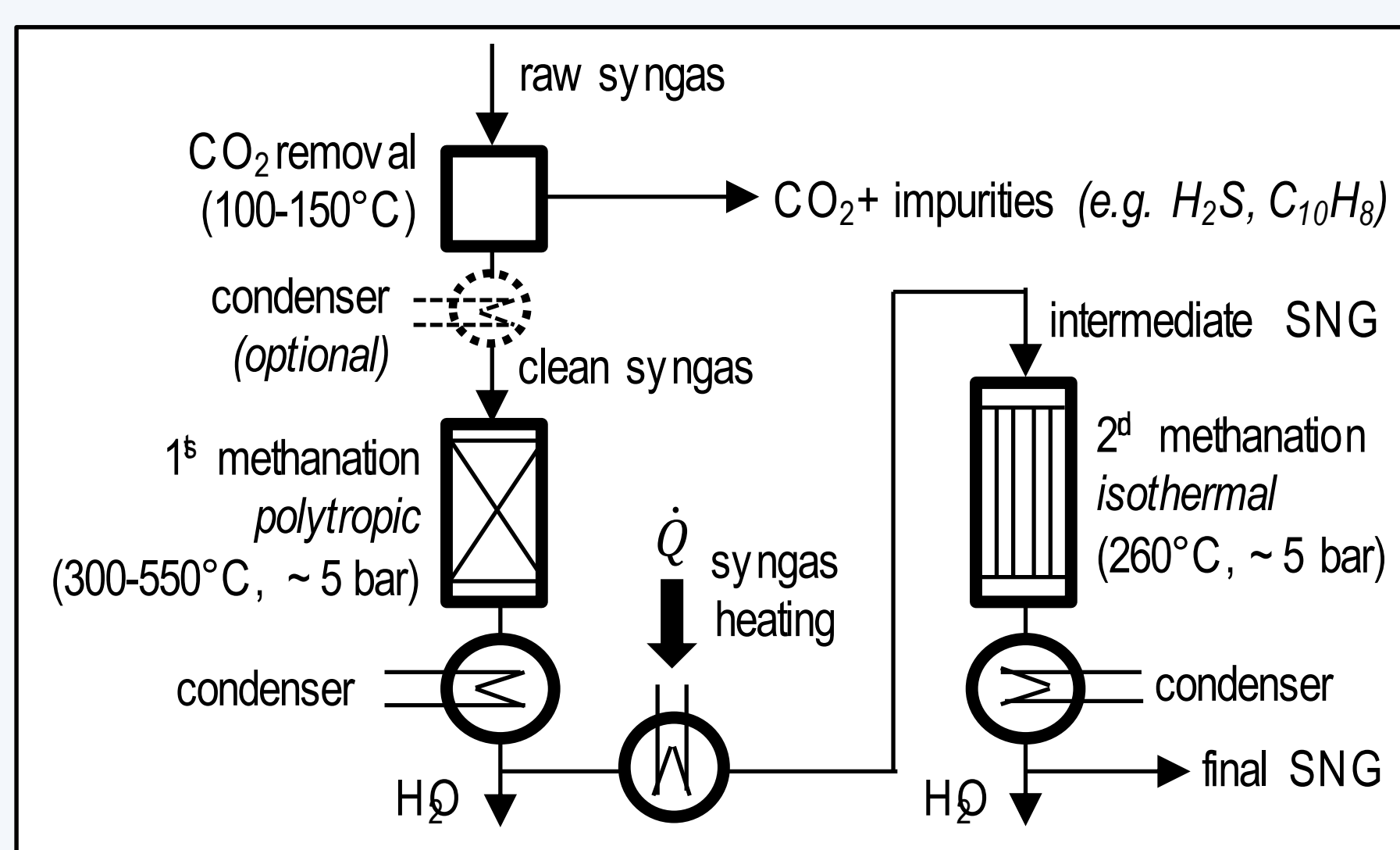


Fig. 1 – Two-stage methanation concept with intermediate water sequestration [1]

Thermodynamic equilibrium calculations show that a methane content higher than 90 vol.-% in final SNG is possible if upstream syngas treatment is operated in the optimum and as long as low outlet temperatures in the first methanation stage are assumed. The optimum methane content in final SNG depends, apart from temperature and pressure, mainly of stoichiometry of the feedgas, which is adapted by CO₂ removal. The optimum is calculated according to the following equation in dependence of raw syngas composition \hat{y}_i .

$$\eta_{CO_2, optimum} = 1 - \frac{2\hat{y}_{H_2} - 6\hat{y}_{CO}}{8\hat{y}_{CO_2}}$$

Influence of warm gas scrubbing on methanation step

The integrated CO₂ removal upstream of the methanation unit results in a major impact on the fixed bed methanation due to modified C/H/O ratio.

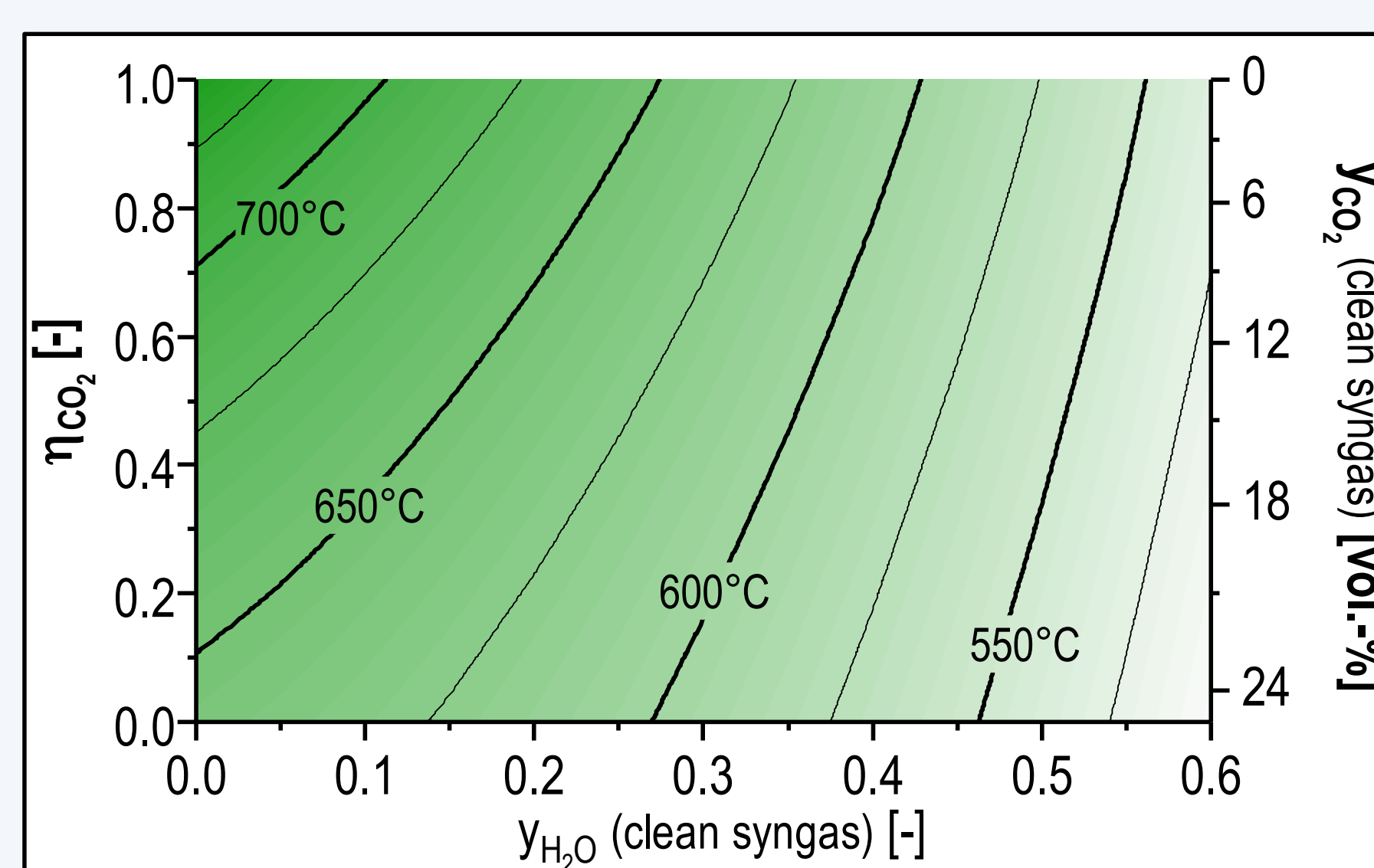


Fig. 2 - Operating map of adiabatic temperature depending on CO₂ removal and water content in clean syngas (dry) 45.3 % H₂, 18.8 % CO, 25.0 % CO₂, 11.0 % CH₄ ($T_{in} = 300^\circ\text{C}$, $p = 5$ bar)

As a consequence, the adiabatic temperature as theoretical maximum of occurring temperatures in 1st stage of methanation varies in dependency of upstream CO₂ removal and water content as shown in Figure 2. Depending on the applied methanation catalyst the synthesis temperature could gain catalyst deactivation by sintering. The axial temperature profiles in the fixed-bed were considered for further analysis of the impact of upstream warm gas cleaning on the 1st methanation stage. High resolution of axial temperature profiles were achieved by a fully automated device shifting a thermocouple quasi-stationary through the catalytic fixed-bed. The profiles under steady-state conditions with bottle-mixed syngas before and after a real-syngas experiment are compared and the global deactivation of the catalytic fixed-bed is calculated (see Figure 3).

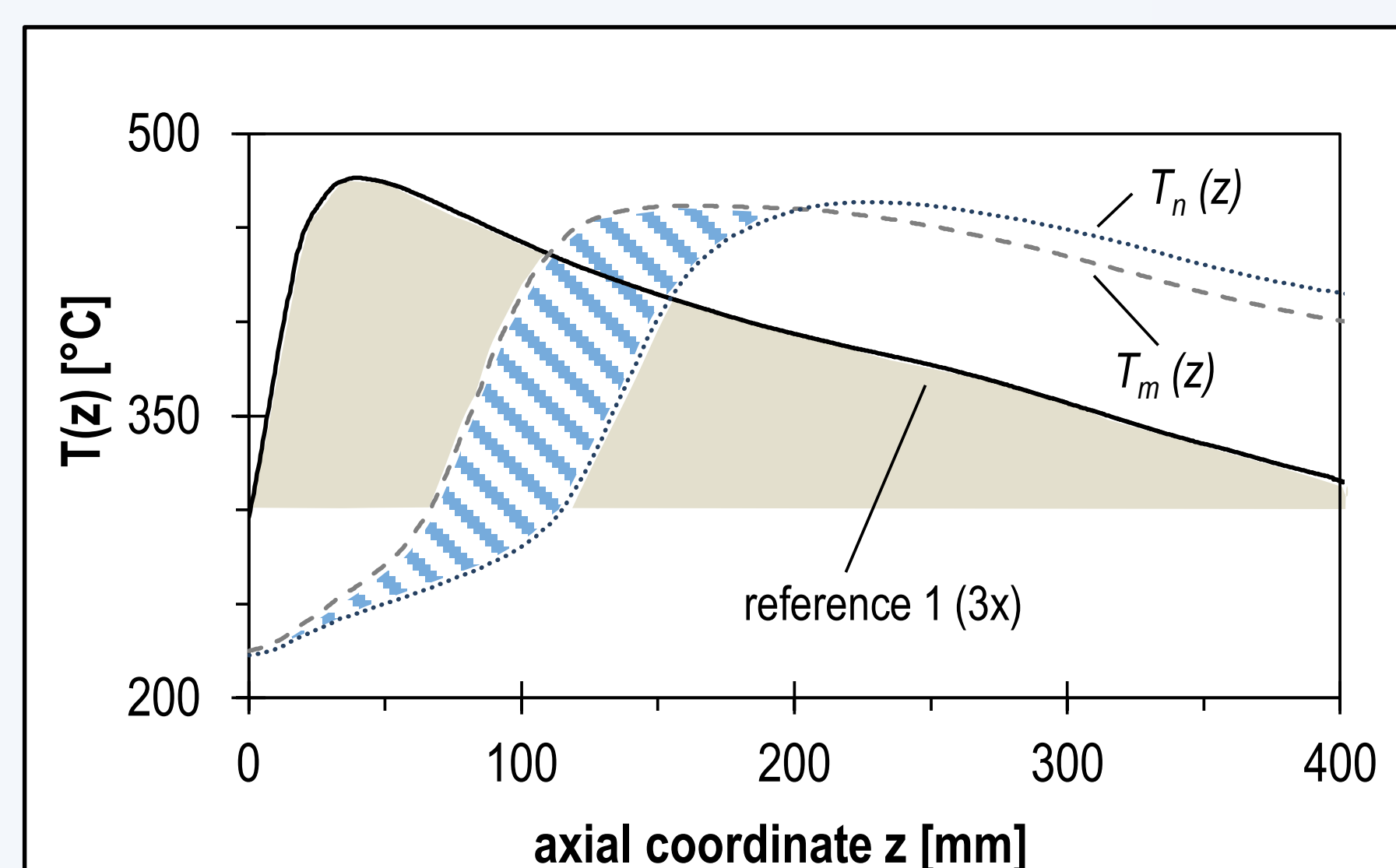


Fig. 3 - Axial temperature profiles for calculation of global deactivation of catalytic fixed-bed

This approach summarizes the deactivation effects of poisoning, sintering and coking which are all determined by the upstream synthesis gas treatment.

Experimental Results

The experimental campaign with 100 kW Heatpipe Reformer, pre-pilot K₂CO₃ scrubber and fixed-bed methanation in slip-stream of clean syngas revealed a high methane content after methanation though the CO₂ concentration was still very high due to insufficient CO₂ removal in the scrubbing step.

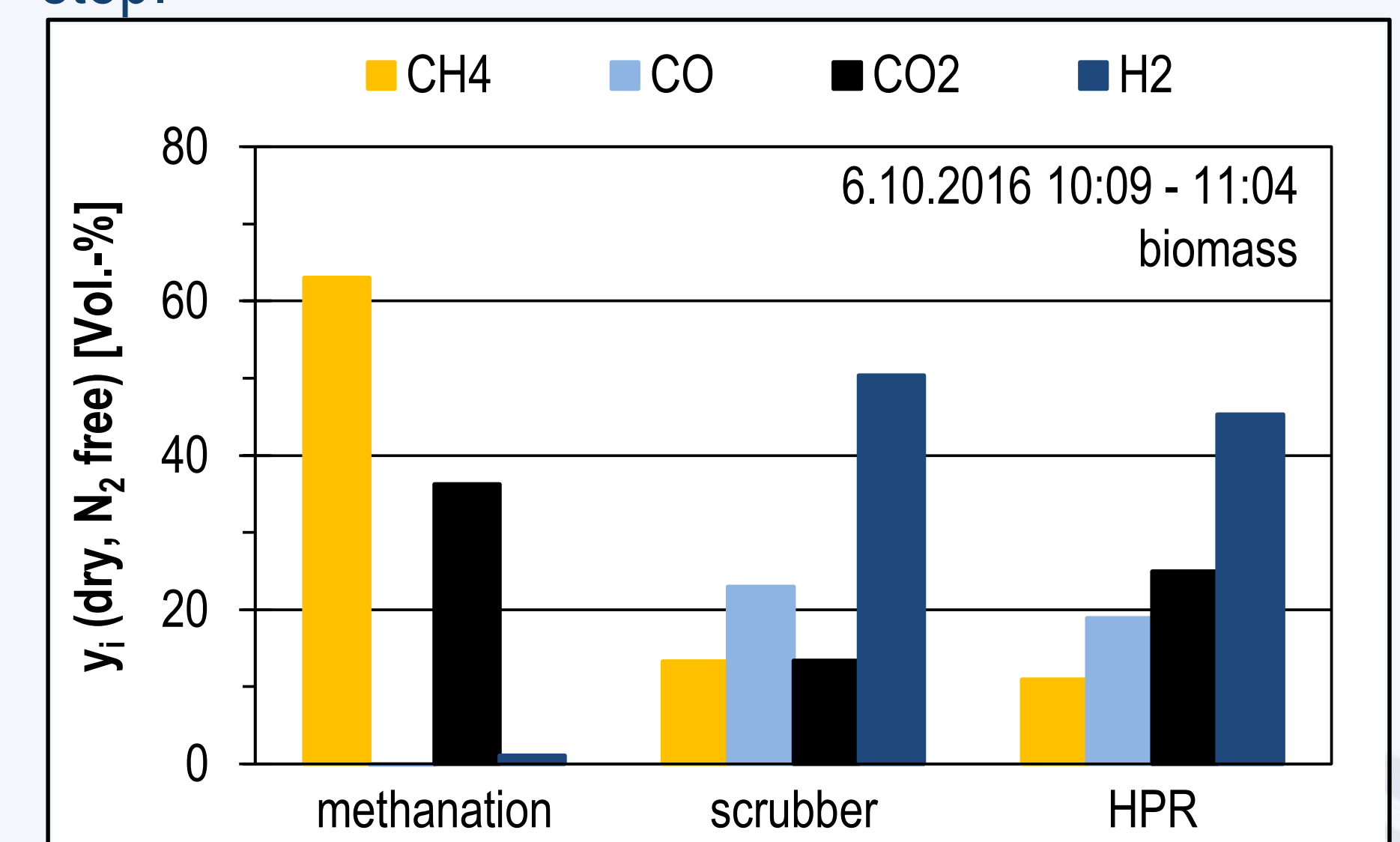


Fig. 4 - Gas composition of SNG process chain with 100 kW Heatpipe Reformer, pre-pilot scrubber and fixed-bed methanation in slipstream of clean syngas

Figure 5 summarizes the global deactivation of the fixed-bed within a series of experiments with one single catalyst batch.

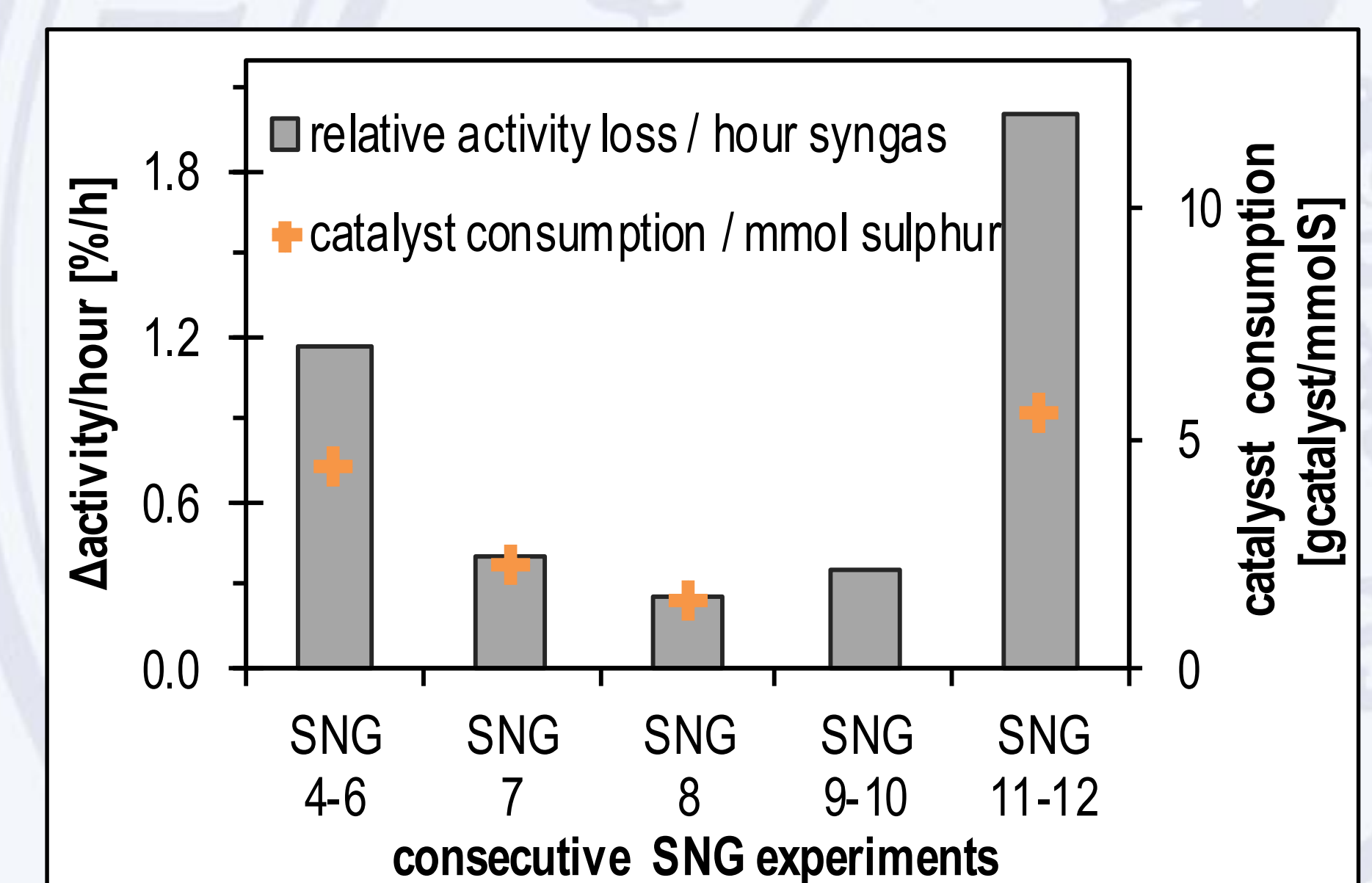


Fig. 5 - Global deactivation of catalytic fixed-bed in a series of experiments with real syngas (lignite and biomass derived)

Conclusion

The proposed simplified SNG concept is suitable to reach a high methane content, whereas the C/H/O conditioning has to be accomplished within a narrow range in order to reduce catalyst consumption.

[1] Neubert et. al, Simulation-based evaluation of a two-stage small-scale methanation unit for decentralized applications; Energy & Fuels (2017), 31, p. 2076-2086



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