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Self-sufficient hydrogen production from biogas by reforming and CO combustion

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Motivation

Hydrogen is expected to play an increasingly important role in the future energy supply. However, the currently most important process for producing green hydrogen, the electrolysis of water, is very cost-intensive. It also requires large amounts of renewable electricity which is currently not available in sufficient quantities. Therefore, technologies are attractive that allow the production of hydrogen without large investment costs and with low energy input. One option for this is the reforming of biogas.

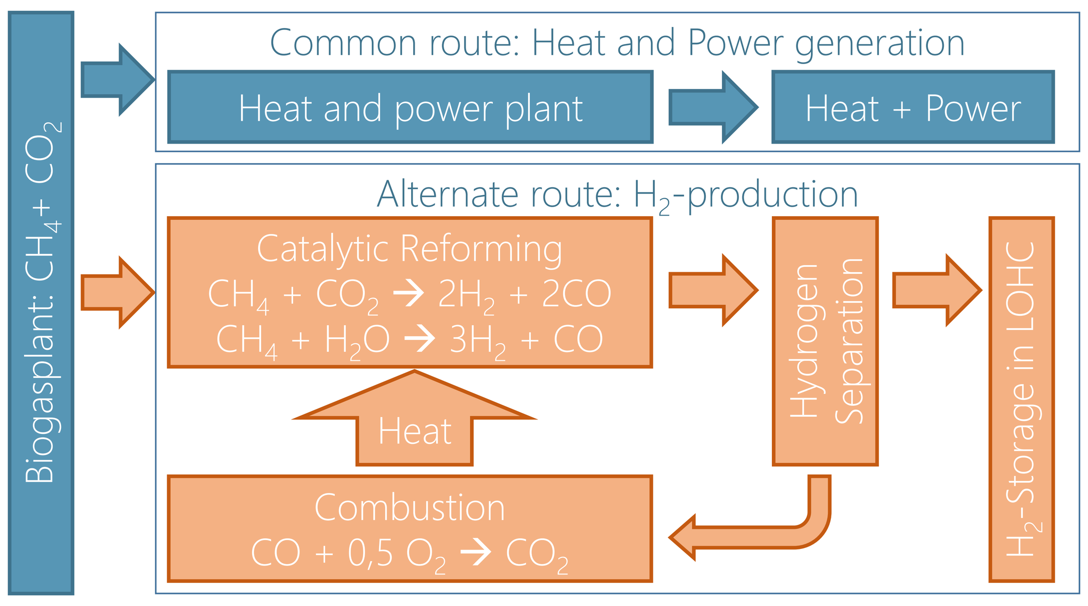

Fig. 1: Concept for self-sustaining hydrogen production from biogas as an alternative to the current route of generation of power and heat

The project BiogasGoesHydrogen

Under the current situation, the majority of biogas plants may not be economically profitable after the expiration of the EEG compensation. Production of the higher-priced hydrogen offers hereby an interesting alternative source of income. Instead of burning biogas in combined heat and power plants, the methane (CH_4) and carbon dioxide (CO_2) are converted into synthesis gas by catalytic reforming (Fig. 1). The hydrogen contained in the product is separated and the remaining residual gas rich in carbon monoxide (CO) is combusted next to the reforming to provide the heat of reaction. In this way, no additional energy is required for the endothermic reaction and thus the process is independent from the availability of electricity. The produced hydrogen can be stored and transported for instance by using liquid organic hydrogen carriers (LOHC). In this way the concept aims to enable biogas plants to contribute to the production of green hydrogen assuring a future economic mode of operation. At the same time, decentralized hydrogen production with the large number of small to medium-scale biogas plants offers great potential for opening up a wide range of different applications for the usage of hydrogen.

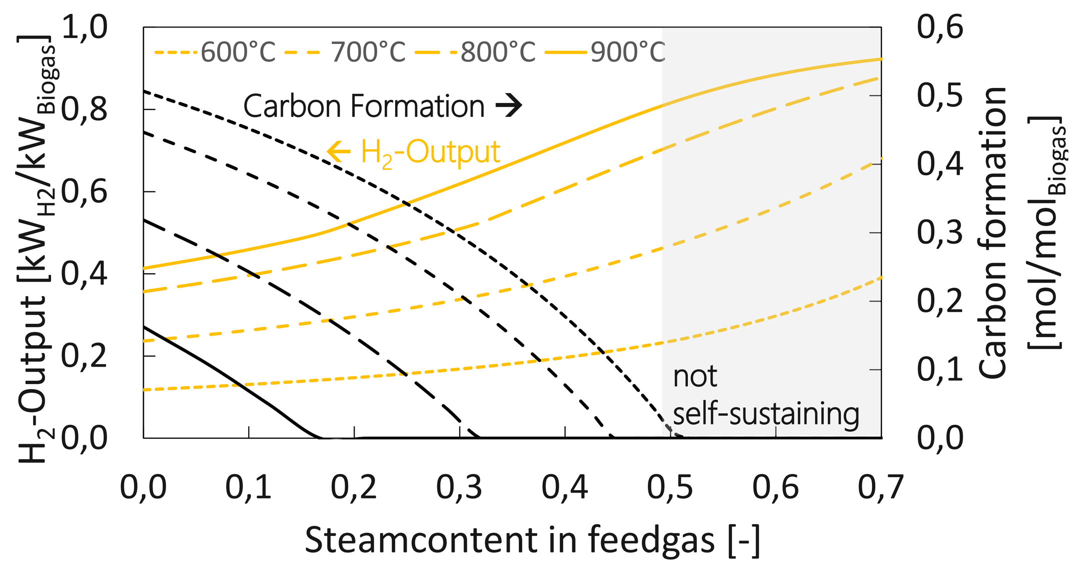

Fig. 2: H_2-output and carbon formation over reforming temperature and steam content in the biogas (simulation in AspenPlus: equilibrium at 10 bar_g and CO-shift reaction at 350°C)

Reforming of Biogas

The challenge in reforming biogas (near dry reforming) is the risk of carbon formation and the resulting catalyst deactivation. The addition of steam reduces this problem and also increases the hydrogen yield. However, the energy demand increases with rising steam content. For this purpose, simulations were carried out in AspenPlus with the aim of finding optimum operating conditions and a self-sufficient process (Fig. 2). Depending on the reaction temperature, hydrogen yields of approx. 0,6-0,8 kW_{H2}/kW_{Biogas} are possible at a maximum steam content of 50% (S/C=1). Carbon formation shows no problem at these operating conditions.

In a catalyst screening, three commercial available catalysts were investigated for their suitability under different operating conditions. All catalysts showed similar behavior concerning activity and stability (Fig. 3). CH_4 conversion increases with higher heat source temperature and reaches a level of less than 1% at 950°C (EVT10). Carbon formation could not be observed, despite in some cases the reaction temperature was significantly below the threshold of 680°C (Fig. 3).

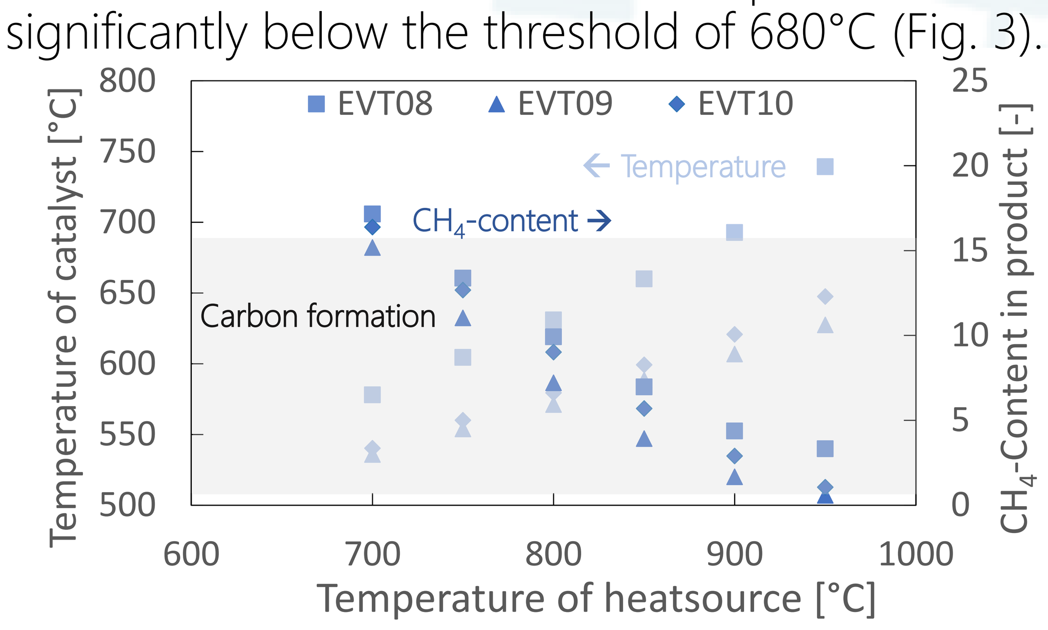

Fig. 3: CH_4-content and resulting reaction temperature over the heatsource temperature for different reforming catalysts (0 bar_g, GHSV=5000 h^{-1}, 1:1:1=CH_4:CO_2:Steam)

Conception of reactor

To simplify the subsequent application of the concept, the complexity of the overall process is reduced by designing a combined reformer unit. For this purpose, reforming, combustion and heat recovery take place directly next to each other in an easily scalable plate-reactor (Fig. 4).

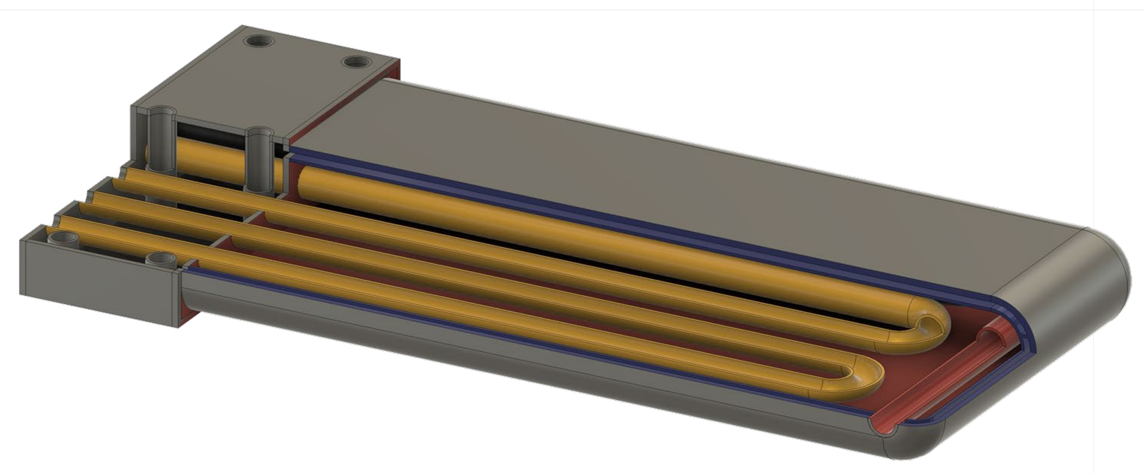

Fig. 4: Concept sketch of additively manufactured reforming reactor with internal heat supply by combustion and heat recovery

In order to reach the highest possible overall efficiency, it is necessary for the reforming to take place at temperatures around 800°C. This is achieved by efficient preheating and heat recovery from the hot product gases. Heat transfer is thus maximized by small wall strengths and large transfer surfaces due to microstructures. For this purpose, combustion must also be as efficient, stable and flexible as possible. To address this, various options are being investigated, such as combustion in microstructures like pores or microchannels. Furthermore, additional options, such as air staging or fluidic adaptations, are considered.

Since the geometries required for this cannot be realized using conventional methods, the reactor is manufactured additively. This also allows a flexible selection of the needed high-temperature materials such as nickel-based alloys or ceramics. Furthermore, this ensures low-cost and fast production.

Conclusion

The concept aims for a simple and cheap production of hydrogen from biogas. Reforming at low vapor contents does not show any problems in respect to catalyst deactivation. In further experiments, the combustion characteristics in microstructures is investigated. Subsequently, the design of the reformer is finalized, and experimentally analyzed in a high-temperature test rig.

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