EXPERIMNTAL AND NUMERICAL ANALYSIS OF A MICROSTRUCTURED PACKED BED REACTOR FOR METHANATION OF CO/CO_2 MIXTURES

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Abstract

The storage of electrical excess energy using the Power-to-Gas concept can be regarded as one of the most promising ways of storing large amounts of energy by converting electrical to chemical energy. The methanation step is very challenging regarding temperature control due to high reaction heat release. This contribution presents the ideas behind an innovative microstructured reactor concept for providing decentralized plants by enabling compact methanation units. The methanation reactor was manufactured and experiments using syngas throughputs of up to 1.4 Nm^3/h (10% CO, 7% CO_2, $H_2/C=4$) were conducted while examining the cooling potential of different cooling fluids e.g. air, steam and water. The manufactured device was analyzed in detail using a 3D CFD model for better understanding of the heat and mass transfer in the cooling zone and the micro packed bed. The numerical results were validated against experimental data and allowed further optimization for scale-up in high-throughput applications.

Keywords

Methanation, micro reactor, CFD reactor simulation

Introduction

The production of energy using renewable energy sources like e.g. sun or wind can be regarded as the most promising way of future energy supply. The use of fossil resources can be decreased and the ecological sustainability can be improved by the stepwise integration of the renewable energy. However, storage and release of excess energy in times of energy demand makes this integration process difficult and the storage options are thus subject of intensive research.

Power-to-Gas (PtG) is one of the promising concepts able to deal with efficient storage of large amounts of excess energy. It converts electrical to chemical energy and storable compounds like methane are preferred options as final gas product. Methane can be effectively used in heating and mobility applications or can be injected in the gas grid.

The PtG process consists of two steps: “green” energy is initially used for hydrogen production by electrolysing water in an electrolysis cell (EC). Waste CO_2 can either be converted with the hydrogen from the EC or can be reduced electrochemically to CO in parallel in the EC before it is further converted with the hydrogen to methane. The latter option demands for a high temperature Solid Oxide EC which is regarded as preferred system in terms of overall process efficiency.

Mixtures of CO_2 and CO are finally obtained as feed into the methanation process. The control over the process conditions in such mixtures is, however, crucial with regard to the process control.

Experimental

In this study the attention is paid to methanation of CO and CO_2 using new process technology i.e. microchannel reactors for intensification purposes.

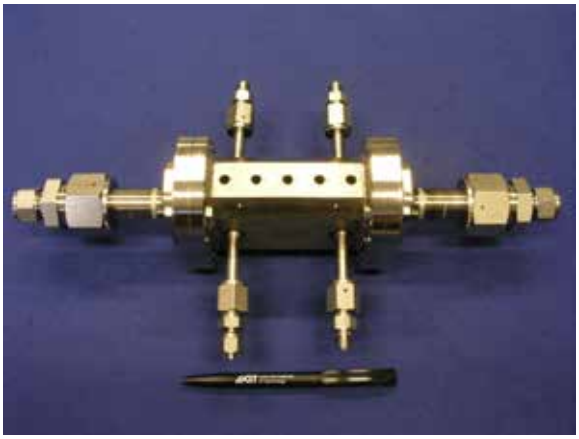

Figure 1. Microreactor prototype for mixed CO/CO_2 methanation.

The microreactor prototype of 14 cm length, 7 cm width and only 2 cm basic stack height has been designed for 1 Nm^3/h of synthesis gas with initial pre-heat by heating cartridges. The potential of indirect cooling with air, steam and liquid water was examined during methanation of syngas containing CO and CO_2 at the total flow of 15 and 23.2 Nl/min with concentrations 10% CO, 7% CO_2, 68% H_2, rest N_2. The reactor was filled with 5 g of commercial Ni-catalyst. The reactor performance has been evaluated with a Fluent® model including literature kinetics [1] for detailed understanding of the heat transfer in the micro packed bed, i.e. in order to determine the effective bed heat conductivity λ_{eff}.

Results

According to our study, methanation of the CO/CO_2 mixtures can provide an extremely high volumetric heat flux due to the exothermic and very fast reaction, easily limited by equilibrium at temperatures above 500°C, and catalysts can be easily damaged by coke formation and sintering, accelerated by moving hot-spots under load changes.

We were able to show that the microreactor prototype is capable to fully convert 1.4 Nm^3/h of a CO/CO_2 mixture ($H_2/C=4$) in the range of 300-490°C. The process heat, approximately 520 W, was extracted to a satisfying extent by air as cooling gas (see Figure 2), steam and water from the reaction zone. No deactivation of the catalyst was observed during several 100 h of testing.

From CFD simulations in Fluent® it was possible to describe the reactor behavior regarding its temperature distribution and the conversion by carefully looking at heat transfer on both, the cooling and reaction side. The issue of a thin bed with particle diameter to slit height less than 10 has been evaluated carefully and a method different to theory has been found to determine the effective heat transfer coefficient λ_{eff} while having a close look on parameter dependence of pressure and gas velocity.

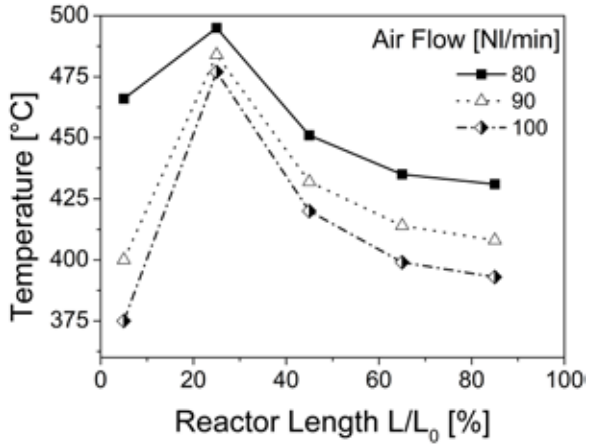

Figure 2. : Experimental temperature profiles along the reactor bed during methanation of 1.4 Nm^3/h of syngas as a function of the air flux (entering the reactor at 150°C) for cooling purposes.

The literature kinetics has been modified in its pre-exponential factor to fit the experimental but further investigations needs to be performed in order to adjust the overall pressure dependence.

Conclusions

In summary, the manufactured reactor was designed and tested successfully carrying out methanation of CO/CO_2 mixtures with throughputs of up to 1.4 Nm^3/h and air, steam and water as coolants. The used device was reproduced on the numerical level regarding most critical parts e.g. cooling side or packed bed. The reactor concept has been approved and will be scaled to > 100 kW methane chemical energy output in the near future.

Acknowledgments

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References

[1] Xu, J. and Froment G.F, Methane steam reforming, methanation and water-gas shift: I. Intrinsic kinetics, .AIChE Journal, 35(2004)