Theoretical Insights in the Heat and Mass Transfer Limitations in Single Pellet String Reactors of different Lengths

Christian Bauer[1,2], Tabea Gros[1,2], Olaf Hinrichsen[1,2]

1 Technical University of Munich, School of Natural Sciences, Department of Chemistry, Lichtenbergstr. 4, 85748 Garching, Germany

2 Technical University of Munich, Catalysis Research Center, Ernst-Otto-Fischer-Str. 1,

85748 Garching, Germany

**ch.bauer@tum.de*

Highlights

- Evaluation of internal and external mass transfer in Single Pellet String Reactors
- Identification of the hotspot formation and its evaluation with changing flow rate
- Dependency of the reactor performance on the pellet string length

1. Introduction

The single pellet string reactor (SPSR) has shown potential for testing different catalyst shapes as a scaled-down version of a fixed-bed reactor [1]. The reactor tube has a diameter that closely matches the diameter of the catalyst pellets. The main goal of this study is to examine the scalability of different single pellet string lengths with respect to external mass transfer limitations. The exothermic CO_2 methanation over a Ni-based catalyst serves as the model reaction. Reactive CFD modelling is used to get insights into the SPSR system with the highest level of detail. The reliability of the model is shown by comparing the simulative studies with experimental results.

2. Methods

Reactive CFD simulations are conducted by a fully resolved multi-region model. In the fluid phase, conservation of total mass, momentum, species and energy is ensured. The porous catalyst pellets are fully resolved in terms of heat and mass transfer by Fick's law or Fourier's law. The model is implemented in OpenFOAM v2006 as steady-state solver. The kinetic model by Xu and Froment [2], which serves as the source term for heat and species in the catalytic reaction, has been refitted to powder measurements of crushed pellets. Our spherical catalyst pellets were prepared by impregnating Al_2O_3 pellets with a nickel-nitrate solution, resulting in a nickel loading of 20 wt.-%. For all cases, a stoichiometric molar feed gas ratio of $H_2/CO_2 = 4/1$ was used for the reaction. All feed gas flow rates result in a particle Reynolds number below 50 which allows the assumption of laminar flow conditions. Internal mass transfer resistance can be evaluated by efficiency factors. The efficiency factor of a species, which is evaluated for each pellet, is defined by the ratio of its volume averaged formation rate and its surface averaged formation rate. The temperature uptake in the pellets due to the exothermic reaction and the heat transfer resistance is postprocessed as well. Moreover, the deviation from plug-flow conversion is evaluated by section wise 1D modelling of the SPSRs. In this attempt the maximum temperature of a pellet and its efficiency factor for methane production is passed to the corresponding section in the 1D model. In this way the effects of internal mass transfer resistance and of heat transfer are assumed to be compensated. The deviation between 1D and CFD modelling are then assumed to result from non-ideal transport phenomena in the gas phase.

3. Results and Discussion

In general, conversion results of SPSRs with different lengths show that under laminar flow conditions a 12 pellet and a 18 pellet SPSR result in nearly the same performance at the same GHSV while a 6 pellet long SPSR shows less carbon dioxide conversion and methane production. This can also be indicated in figure 1 where the deviation from plug-flow conversion is evaluated. By comparing the methane production, it can be seen that a 6 pellet long SPSR deviates the most. In any case the higher the methane production rate, which is related to a higher GHSV at constant temperature, the more a SPSR deviates from plug-flow behavior. This can be ascribed to the external mass transfer resistance which becomes dominant with increasing flow rate. Figure 2 supports this finding as it shows the Damköhler number for methane production in a 6 pellet SPSR. Damköhler numbers above unity indicate that mass transport is dominant. In this context, the first pellets in a SPSR are more influenced by external mass transport than the last ones. However, with decreasing flow rate more pellets are operating in the kinetic regime as the Damköhler numbers dropping below unity. This indicates that for spheres the SPSR is a suitable reactor to reach plug-flow behavior at lower flow rates.

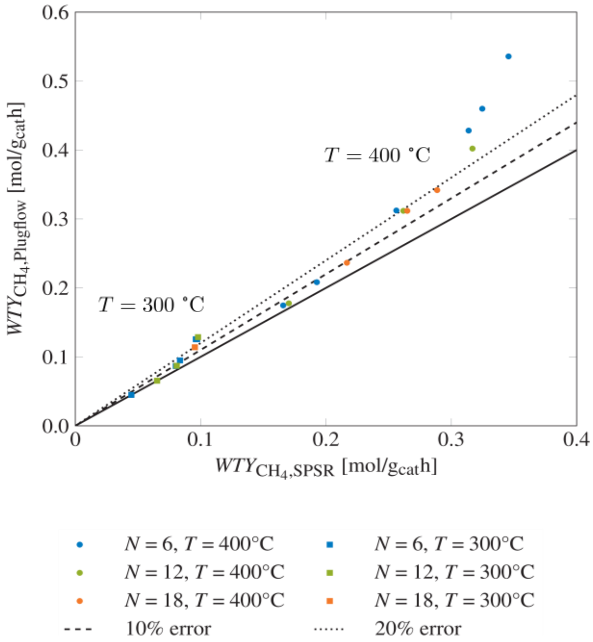

Figure 1. Comparison of methane weight time yield between section wise 1D plug-flow modelling and reactive CFD modelling of SPSRs with different lengths.

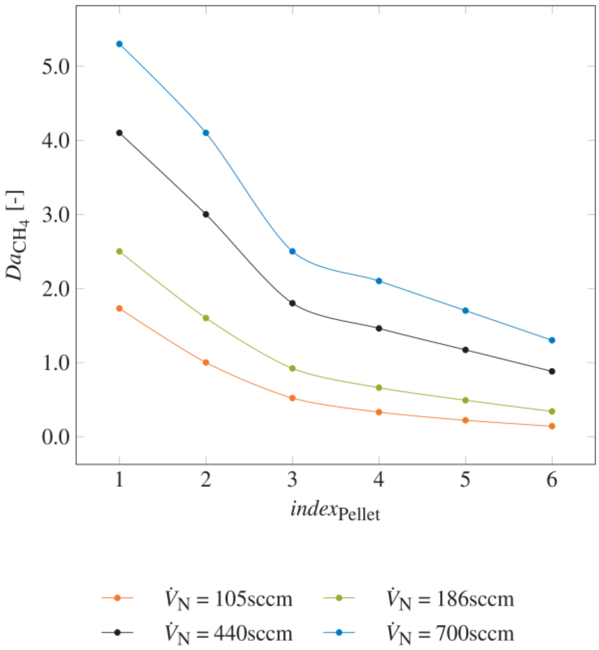

Figure 2. Damköhler numbers for methane production in a 6 pellet SPSR at different flow rates. The pellets are counted in ascending order in flow direction.

4. Conclusions

For the investigated system a 12 pellet SPSR is sufficient to yield results which are independent of the pellet number at constant GHSV in the laminar flow regime. The internal mass transfer resistance is evaluated by the effectiveness factor which relates the surface reaction rate with the mean reaction rate of each sphere. Relatively high surface temperatures due to the exothermicity of the methanation reaction and the limited diffusion rate due to the small pores of our pellets result in quite low effectiveness factors for methane production. By comparing the results of section wise 1D modelling of the SPSRs and reactive CFD modelling we address the deviations to non-ideal mass transport in the gas phase. With increasing flow rate the deviations also increase which means external mass transfer becomes relevant. This can also be found by the evaluation of the Damköhler number for each component. More pellets operate in the mass diffusion regime at higher flow rate while most pellets in the SPSR operate in the kinetic regime at lower flow rates.

References

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[2] J. Xu. and G. F. Froment, AIChE Journal 35 (1989) 88–96.

Keywords
Single Pellet String Reactor; Heat and Mass Transfer Limitations; CO_2 Methanation; Reactive CFD Modelling