

Coupling of spatially and temporally resolved reaction kinetic data in a channel type reactor with a novel kinetic model describing transient reactor behavior

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Introduction

The development of new reactor concepts based on suitable kinetic models in order to achieve the highest possible reactor performance under fluctuating inlet conditions is becoming increasingly important. Previous work has shown that the concentration of reaction intermediates on the catalyst surface play a key role in reaction dynamics [1]. Therefore, investigation of these surface species under reaction conditions is crucial for an improved understanding of the reaction mechanism.

Methods

The CO₂ methanation reaction was investigated using an in-house developed reactor with multiple taps along the reaction coordinate, allowing for spatially and temporally resolved gas phase sampling in between dedicated reaction zones [2]. The reactor further allows for optical access, which was used to gather data on adsorbed species on the catalyst's surface via DRIFTS (**Figure 1**). This data was subsequently used for parametrization of a novel kinetic model.

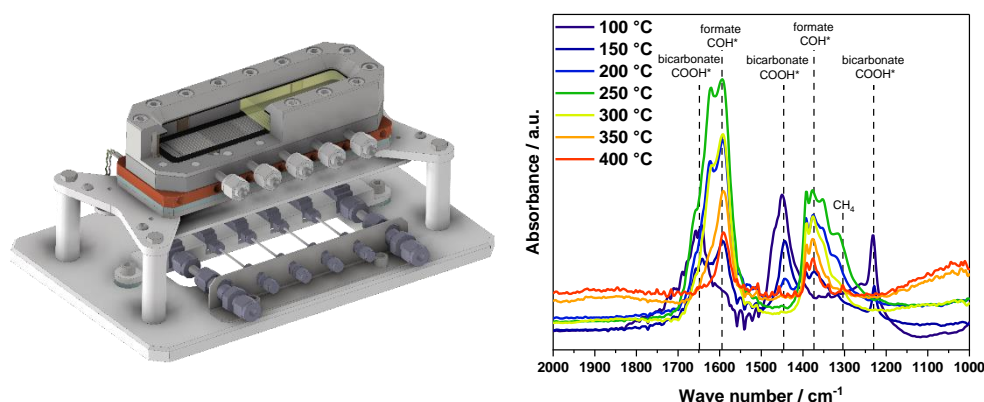


Figure 1. (a) CAD drawing of the reactor used to gather temporally and spatially resolved in operando data. (b) DRIFTS spectra of catalyst surface at different TR ($p_R = 5$ bar, $SV = 4.6$ ml_N min⁻¹ mg_{cat}⁻¹, CO₂:H₂ = 1:4 with 50 vol.-% N₂).

A CFD model of the reactor was implemented in ANSYS Fluent® to investigate the influence of sampling and flow phenomena on the experimental data. The results of the CFD simulations were compared with 1- and 2-dimensional pseudo-homogenous and heterogeneous reactor models. A novel dynamic kinetic model approach (Rate affecting step, RAS model) was implemented. This model is capable of describing the surface coverage of reaction intermediates on the catalyst surface and thus can be coupled with the gathered data from the DRIFTS measurement.

Simulations were performed for different operation conditions. The temperature was varied from 200 to 450 °C and the pressure from 1 to 10 bar. A volume flow range from 100 to 500 Nml min⁻¹ and varying feed compositions were simulated.

Results and discussion

The simulations show a good agreement with experimental data (see **Figure 2**). The different models were evaluated regarding computational effort and accuracy in describing the reaction progress.

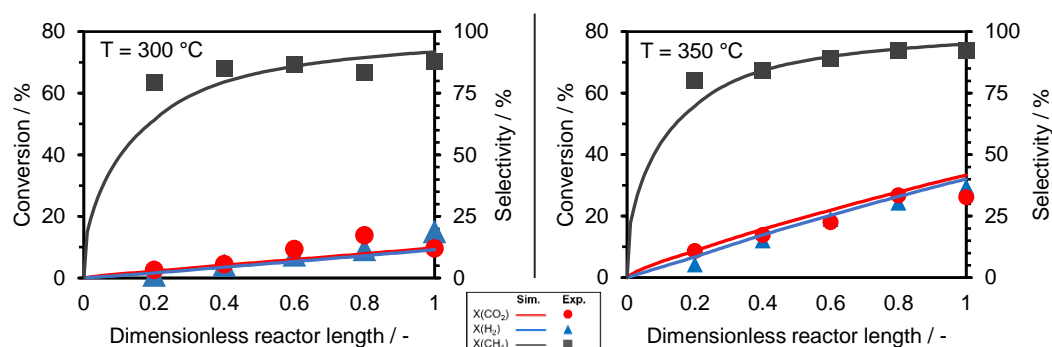


Figure 2. Comparison of experimental data versus simulation results of a PFR model at different temperatures. At Temperature $T = 300$ and 350 °C as indicated, $p_R = 5$ bar and $SV = 4$ mL_N min⁻¹ mg_{cat}⁻¹, $m_{cat} = 121.7$ mg with stoichiometric feed ($CO_2/H_2 = 1:4$), 50 vol.-% diluted with N_2 .

Using the experimental data, the RAS kinetic model approach describing the surface reactions according to Langer et al. [1] was reparametrized. As a result, the model shows good agreement with the data from the experimental reactor and is suitable for describing transient behavior.

References

- [1] M. Langer, D. Kellermann, H. Freund, Chem. Eng. J. 467, 143217, 2023
- [2] T. Engl, M. Langer, H. Freund, M. Rubin, R. Dittmeyer, Chem. Ing. Tech. 95(5), 658–667, 2023