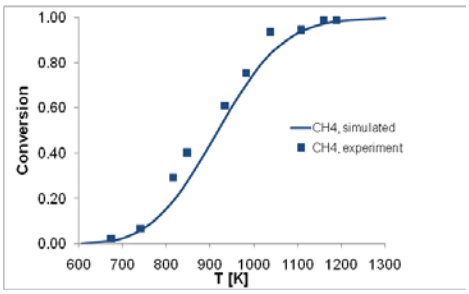
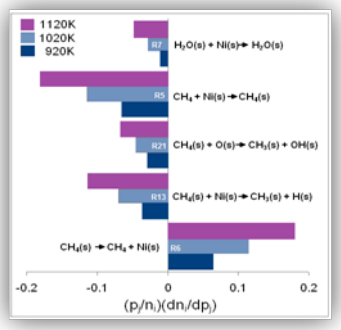
Conversion of light hydrocarbons of over nickel

Background: During the last years, there has been an increased interest in catalytic combustion and conversion of hydrocarbons to more valuable products. These more valuable products are produced by either direct routes such as oxidative coupling reactions, or by indirect routes utilizing synthesis gas as an intermediate. Synthesis gas (H_2/CO in various compositions) plays a key role as a feedstock in many catalytic processes such as synthesis of methanol, oxo-synthesis, and Fischer–Tropsch synthesis; the synthesis gas production via the reforming of methane with either steam or steam/CO_2mixtures is the most widely employed route from natural gas to commodity chemicals. Due to the increasing concern of global warming and oil depletion the catalytic (dry) reforming of methane with carbon dioxide offers a potential opportunity to convert greenhouse gases into H_2/CO. Industrial practice relies on Ni catalysts due to fast turnover rates, good availability and low costs, although it is more sensitive to coke formation and growth of carbon filaments than noble metals.



Methane conversion is simulated as function of temperature over metal monolith washcoated with nickel catalyst, S/C=3; experimental data taken from literature.



Sensitivity coefficients for methane mole fraction as function of temperature for methane steam reforming.

Project: The objective of our work is the development of a surface reaction mechanism over nickel catalyst applicable for catalytic conversion of light hydrocarbons under oxidative, reforming, and pyrolysis conditions at a wide range of temperature, pressure, and residence time. Our research focus is to model carbon formation and deactivation of nickel catalyst during the reforming of methane, which gives us a better understanding of the mechanism at a molecular level. The development of the heterogeneous reaction mechanism over nickel is based on theoretical studies and experimental results from literature and our own laboratories.

Sensitivity analyses are carried out to evaluate crucial parameters and determinate *most important reactions* in the mechanism for each species at different inlet conditions.

The applicability of the mechanisms is evaluated by simulation of new experimental data as well as data taken from experimental studies in literature for different reactor types (plug flow, fixed bed, batch and *stagnation-flow reactor)*.

Numerical simulation is performed using the software DETCHEM^{TM}; a program specifically designed for numerical simulation of the flow fields coupled with detailed gas phase and surface kinetics in chemical reactors at laboratory and technical scale.

Co-workers: Karla Herrera, Lubow Maier, Steffen Tischer

Collaboration:

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Selected publications: L. Maier, B. Schädel, K. Herrera Delgado, S. Tischer, O. Deutschmann, Top. Catal 54 (2011) 845-858; J.-H. Ryu, K.Y. Lee, J. Power Sources 171(2007) 499; V. M. Janardhanan, O. Deutschmann. J. Power Sources. 162 (2006) 1192-1202; D. Chen, R. LØdeng. Chem. Eng. Sci. 56 (2001) 1371-1379; L.M.Aparicio. J.Catalysis. 165 (1997) 262-274; M. C. J. Bradford, M.A. Vannice, Cat. Rev. - Sci. Eng. 41 (1999) 1-42; J. Xu, G. F. Froment, A.I.Ch.E. Journal. 35 (1989) 88-96.