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Oxidation and reforming of light hydrocarbons over Rh/Al_2O_3 catalyst by using a stagnation-flow reactor

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Introduction

The complexity of reaction mechanisms for the simultaneous description of catalytic partial and total oxidation as well as reforming and pyrolysis of light hydrocarbons such as methane (CH_4) and propane (C_3H_8) requires linking of well-defined experimental and numerical studies to gain a better understanding on a molecular level [1]. The stagnation-flow reactor is a valuable laboratory-scale reactor concept to study catalytic chemistry, because a zero-dimensional catalytic surface can be realized, i.e. well-defined gas-phase concentrations and temperatures can be used to explore the intrinsic kinetics of the system [2]. In this study; under varying conditions (fuel/oxygen ratio, temperature) total oxidation of H_2, CO,as well as the water gas shift reaction (WGS), reverse WGS, steam reforming and partial oxidation of CH_4, C_3H_8 species have been investigated at varying conditions to improve our understanding of the reaction mechanism

Stagnation-Flow Reactor

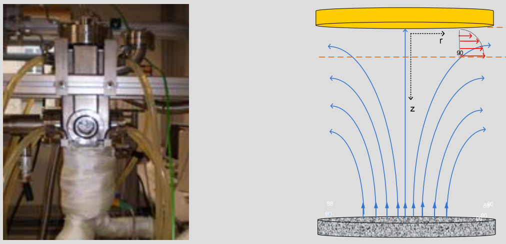

Figure 1: Stagnation-flow reactor

- Zero-dimensional on the catalyst surface
- Temperature and species profiles do not vary in radial directions
  - T, Ci, pi = f(z)
- Potential flow:
  - No vorticity, no curl,
  - Pressure gardient is constant
- Problem can be reduced to 1D steady-state problem.

Experimental

- 5% Rh/Al_2O_3 catalyst is prepared by spin-spray coating.
- Pressure is kept constant at 500 mbar. A quartz microprobe (with a 50µm opening) is used to sample the gas composition.
- Profiles of the species resolved in the gas-phase as function of distance of the catalytic surface using mass spectrometer and FT-IR analytics.
- Computational simulations are performed using the software SPIN, which is a part of CHEMKIN package [3].

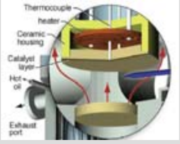

Figure 2: Microprobe sampling [2]

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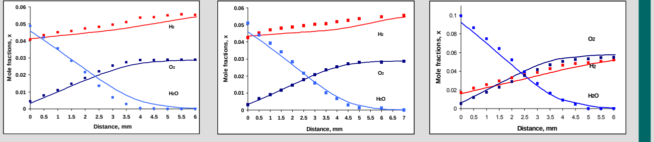
Results

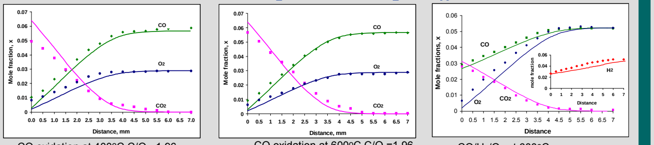
H_2 Oxidation : $H_2 + 1/2\ O_2 \rightarrow H_2O$ $\Delta H_R = -286$ kJ/mol



H_2 oxidation at 400ºC H/O =2.0 | H_2 oxidation at 600ºC H/O =2.0 | H_2 oxidation at 400ºC H/O =1.0

Simulations are performed based on the reaction mechanism of Karakaya et al. [4].

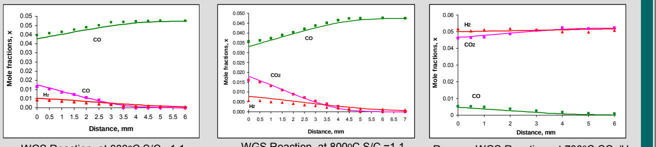
CO Oxidation : $CO + 1/2 O_2 \rightarrow CO_2$ $\Delta H_R = -393$ kJ/mol



CO oxidation at 400ºC C/O =1.96 | CO oxidation at 600ºC C/O =1.96 | CO/H_2/O_2 at 600ºC CO/H2/O2=1/1/1

Simulations are performed based on the reaction mechanism of Karakaya et al. [5].

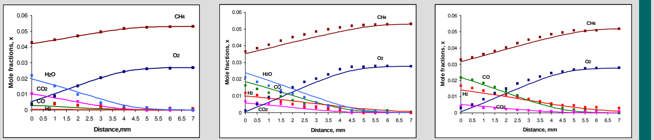
Water Gas Shift (WGS) and Reverse WGS Reactions



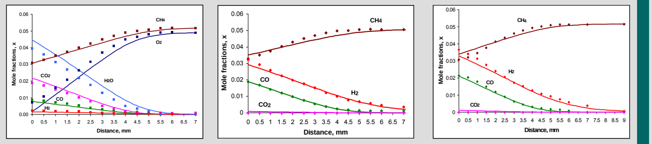
WGS Reaction at 600ºC S/C =1.1 | WGS Reaction at 800ºC S/C =1.1 | Reverse WGS Reaction at 700ºC CO_2/H_2 =1.0

Simulations are performed based on the reaction mechanism of Karakaya et al. [5]

Reforming of CH_4: Partial oxidation and Steam Reforming



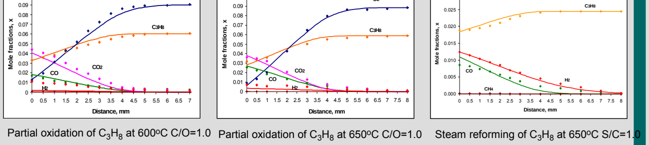
CH_4 partial oxidation at 600ºC C/O =1.0 | CH_4 partial oxidation at 700ºC C/O =1.0 | CH_4 partial oxidation at 750ºC C/O =1.0



CH_4 partial oxidation at 700ºC C/O =0.53 | Steam reforming of CH_4 at 700ºC S/C =1.1 | Steam Reforming of CH_4 at 735ºC S/C =1.1

Simulations are performed based on the reaction mechanism of Deutschmann et al.[6]

Reforming of C_3H_8 : Partial oxidation and Steam Reforming



Partial oxidation of C_3H_8 at 600ºC C/O=1.0 | Partial oxidation of C_3H_8 at 650ºC C/O=1.0 | Steam reforming of C_3H_8 at 650ºC S/C=1.0

Simulations are performed based on the reaction mechanism of Karakaya et.al.[7].

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