Oxidation and reforming of light hydrocarbons over Rh/Al$_2$O$_3$ catalyst by using a stagnation-flow reactor

Canan Karakaya$^1$, Lubow Maier$^2$, Olaf Deutschmann$^{1,2}$

Nicholas E. McGuire$^3$, Neal P. Sullivan$^3$, Robert J. Kee$^3$

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$_3$H$_8$) 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$_3$H$_8$ species have been investigated at varying conditions to improve our understanding of the reaction mechanism.

Stagnation-Flow Reactor

![Stagnation-Flow Reactor Diagram](Image)

Figure 1: Stagnation-flow reactor

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

Experimental

- 5% Rh/Al$_2$O$_3$ 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].

![Microprobe Sampling Diagram](Image)

Figure 2: Microprobe sampling [2]

Results

- H$_2$ Oxidation: $H_2 + \frac{1}{2} O_2 \rightarrow H_2O + \Delta H = -286 \text{kJ/mol}$
- CO Oxidation: $CO + \frac{1}{2} O_2 \rightarrow CO_2 + \Delta H = -393 \text{kJ/mol}$
- Water Gas Shift (WGS) and Reverse WGS Reactions
- Reforming of CH$_4$: Partial oxidation and Steam Reforming
- Reforming of C$_3$H$_8$: Partial oxidation and Steam Reforming

References


[1] Institute for Chemical Technology and Polymer Chemistry, Germany
[2] Institute for Catalysis Research and Technologies, Germany
[3] Engineering Division, Colorado School of Mines, Golden, USA

KIT – University of the State of Baden-Wuerttemberg and National Research Center of the Helmholtz Association

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