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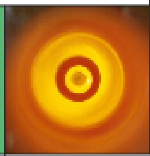
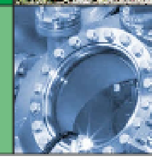


LEM – Laboratory for Energy and Materials Cycles



Optically accessible channel reactor for kinetic investigations of autothermal reforming

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Introduction

- Autothermal reforming of gasoline provides an efficient method to produce hydrogen for fuel cell applications.
- To control a fuel processor a simple yet accurate kinetic model is necessary.
- A new experimental reactor concept was developed, which allows surface temperature measurements through a quartz window and gas sampling through a capillary to measure the concentration profile in the reactor.
- The measurements will be used to develop a kinetic model which takes into account the complex reaction network of autothermal gasoline reforming in a monolithic reactor.

Complex reaction network

TOX: $C_nH_m + \left(n+\frac{m}{4}\right)O_2 \rightarrow n\,CO_2 + \frac{m}{2}H_2O$

SR: $C_nH_m + n\,H_2O \leftrightarrow n\,CO + \left(n+\frac{m}{2}\right)H_2$

POX: $C_nH_m + \left(\frac{n}{2}\right)O_2 \rightarrow n\,CO + \frac{m}{2}H_2$

WGS: $CO + H_2O \leftrightarrow CO_2 + H_2$

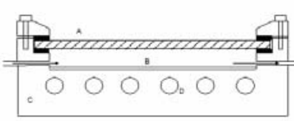
MET: $CO + 3H_2 \leftrightarrow CH_4 + H_2O$

Experimental

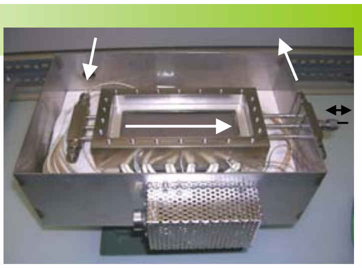
Reactor

Idea: Design an optically accessible reactor with a flow field similar to a single monolith channel

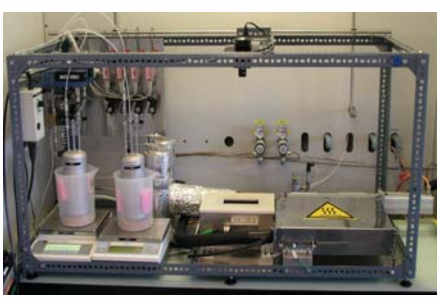
Schematic side view of the channel reactor



A: quartz window, B: catalytic plate, C: reactor housing, D: heating cartridge.



Channel reactor with catalytic plate (insulation removed). The white arrows visualize the flow through the reactor. On the right side of the reactor the septum port with the movable sampling capillary (black arrow) is visible



Lab setup of channel reactor with the feed system (left side of picture), channel reactor (in the middle of the picture), heating box for sampling capillary (right side of picture) and the infrared camera (top section of the picture)

• *Infrared Camera*
- Spectral range: 900 - 1700 nm
- Detector: Indium Gallium Arsenide
- Array format: 320 x 256
- Pixel: 30 x 30 microns
- Frame rate: 30 Hz
- Temperature range: 20 - 1200°C

• *Setup Control*
LabVIEW™ program to control the step motor, the mass flow meters and the controllers of the different heaters

Experimental Setup

• *Feed System*
- Water and Gasoline are fed using pressurised containers and LiquiFLOW™ controllers
- Oxygen and nitrogen are fed through flow controllers
- 2 Separate evaporators for water and gasoline
- Superheater up to 750°C

• *Reactor*
- Stainless steel, channel height = 4 mm
- Maximum heating temperature = 750°C

• *Gas Sampling*
- Movable stainless steel sampling capillary, internal diameter = 0.5 mm, external diameter = 0.8 mm
- Capillary enters reactor through high temperature septum port
- Capillary is placed directly above catalyst plate
- Electrical step motor is coupled to a linear positioning system (accuracy < 0.1 mm)
- Heated transfer line to GC and MS

Results

Reactor Modeling

Assumptions:
- Flow velocity: 0.14 m/s
- Pure Nitrogen
- Incompressible flow (Mach number << 1)
- Laminar flow (Re < 30)

Navier-Stokes (incompressible flow):

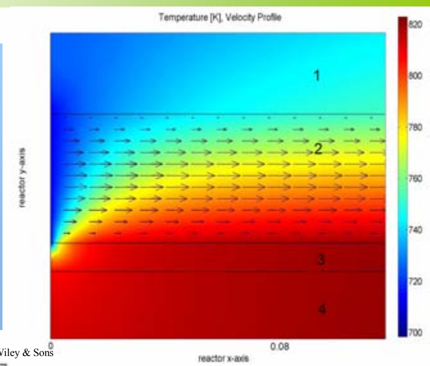
$$\begin{cases} \rho\frac{\partial u}{\partial t} + \rho(u\cdot\nabla)u = -\nabla p + \eta\nabla^2 u + F \\ \nabla\cdot u = 0 \end{cases}$$

$$F_y = g\rho\beta(T - T_\infty)$$

Convection and conduction:

$$\rho c_p\frac{\partial T}{\partial t} + \nabla\cdot\left(-k\nabla T + \rho c_p T u\right) = Q$$

$Q = \alpha(T - T_{amb.})$ (Convective heat flux)

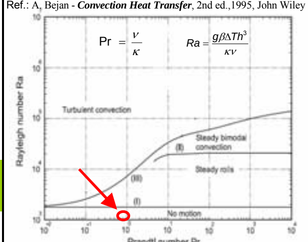


Reactor inlet section (1 quartz window, 2 channel, 3 catalytic plate, 4 heating block) with parabolic velocity profile. The arrows show the velocity distribution in the gas channel. Temperature distribution (side view)

→ Buoyancy force has no significant influence on flow behavior in reactor channel for pure nitrogen

→ The parabolic velocity profile is maintained along the reactor

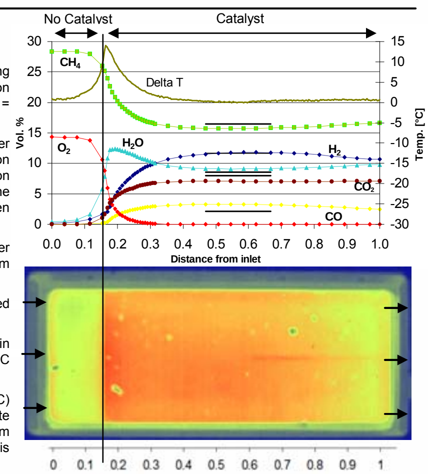
Ref.: A, Bejan - *Convection Heat Transfer*, 2nd ed.,1995, John Wiley & Sons



→ Rayleigh-Bénard convection does not occur in reactor channel: Ra is far into the no-motion area

First Experiments

- Measured concentration profiles (MS) along the catalyst plate during dry partial oxidation of methane at 500°C (O/C = 1, GHSV = 100'000 h^{-1}, p = 2 bar).
- The oxygen is consumed completely after 32 % of reactor length by total oxidation and partial oxidation, producing water, carbon monoxide, carbon dioxide and hydrogen. The measured hot spot correlates with the oxygen concentration.
- After 18% of reactor length the water concentration decreases due to the steam reforming and water gas shift reactions.
- The thermodynamic equilibrium is reached at 50% of the reactor length (black lines).
- After 80% of the reactor length changes in concentration profiles are due to the 15°C cooler reactor outlet section.
- Measured (yellow = 500°C, red = 514°C) temperature distribution on the catalyst plate during the experiment. The gas flow is from left to right (arrows). The sampling capillary is visible in the centre of the reactor (top view).



Conclusions

- The reactor modeling showed that the parabolic flow in the channel is maintained.
- First experiments showed that the gas sampling and the surface temperature measurements can be carried out with a high local resolution.
- The measured concentration profiles indicate that CO_2, CO, H_2, and H_2O are primary products in the early stage of reaction.
- The measured concentration profiles and surface temperatures will allow to parameterise a kinetic model based on a large number of experimental data.

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