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Catalytic Oxidation of CO over Rh/Al_2O_3 and Ni/Al_2O_3 Catalysts in a Stagnation-Flow Reactor

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Introduction

Catalytic oxidation of CO is one of the side reactions that occur in partial oxidation and steam reforming of methane, which are two major efficient syngas (H2/CO) production technologies [1-3].
Noble metals such as Ru, Rh, Pd, Pt are the most effective catalysts for the syngas production [3]. Although Ni suffers coking more than noble metals, its high activity and low cost make it a good candidate for reforming reactions [4].
In order to understand the reforming behavior of Ni and Rh catalysts, measurements are carried out in a stagnation-flow reactor.

Stagnation-Flow Reactor

Stagnation-flow reactor has been designed to specifically facilitate numerical modeling based on multi component diffusive transport and elementary chemical kinetics.

With the simplicity of the stagnation-flow reactor configuration:

- Problem can be reduced to a boundary value problem of a one dimensional ordinary differential equation system.
- Temperature and concentration profiles of reactive gas-phase species with the boundary layer are measured as a function of distance from the catalytic surface.
- These profiles are independent of the radial position.

The derived reaction mechanisms can then be incorporated into models for more practical reactor configurations, e. g. high surface area porous foams or honeycomb monolith structures, to provide quantitative design insights [5].

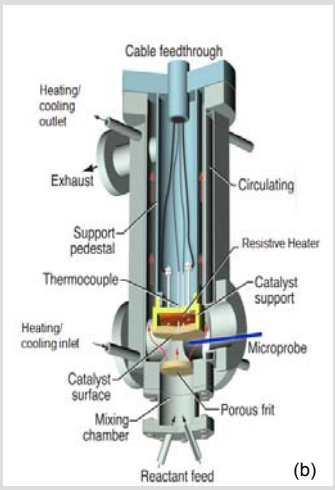

Figure 1a,b: Stagnation-flow reactor

References
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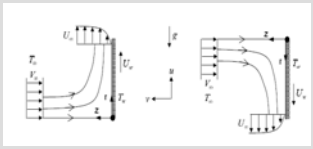
Modelling Approach

Basic Assumptions:

- There is a viscous boundary layer
  T, Ci, ρi = f(z)
- Potential flow:
- No vorticity
- No curl
- Pressure gradient is constant
- Steady state



Computational simulations are performed using the software SPIN, which is a part of CHEMKIN package [6].

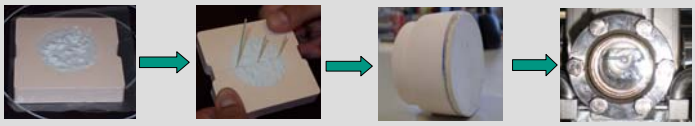
Overall mass balance: $\frac{d(\rho u)}{dz} + 2\rho V = 0$

Species: $\rho u \frac{dY_k}{dz} = -\frac{d}{dz}(\rho Y_k V_k) + \dot{m}_{s,k} \quad (k = 1, K_g)$

Momentum balance: $\rho u \frac{dV}{dz} + \rho V^2 = -\Lambda_0 + \frac{d}{dz}\left(\mu \frac{dV}{dz}\right) \quad \Lambda_0 = \frac{1}{r}\frac{dp}{dr}$

Energy balance: $\rho u c_p \frac{dT}{dz} = \frac{d}{dz}\left(\lambda \frac{dT}{dz}\right) - \sum_{k=1}^{K_g} \rho Y_k V_k c_{p,k} \frac{dT}{dz} - \sum_{k=1}^{K_g} h_{s,k} \dot{m}_{s,k}$

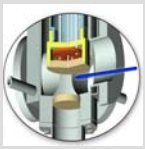
Experimental



Stagnation surface is made of Alumina.

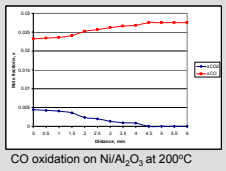
5% Rh/Al_2O_3 and 15% Ni/Al_2O_3 catalysts are prepared by spin-spray coating method to ensure a homogeneously distributed active phase on the stagnation surface.

- 95% Ar diluted CO:O2 mixture (1:1.2) is fed to the reactor at room temperature.
- Pressure is kept constant at 500 mbar. A quartz microprobe (with a 50 μ opening) is used to sample the gas composition.
- Boundary layer distribution of CO, CO_2 species are analyzed by a mass spectrometer (V&F).

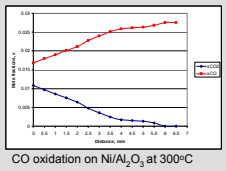


Microprobe sampling

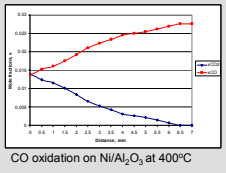
Results



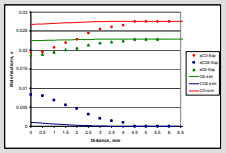
CO oxidation on Ni/Al_2O_3 at 200°C



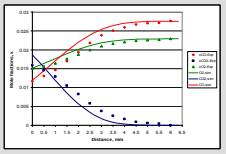
CO oxidation on Ni/Al_2O_3 at 300°C



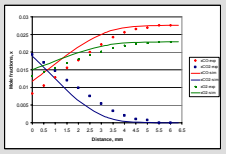
CO oxidation on Ni/Al_2O_3 at 400°C



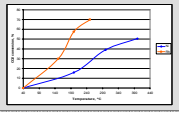
CO oxidation on Rh/Al_2O_3 at 150°C: Experiments vs Simulations



CO oxidation on Rh/Al_2O_3 at 200°C: Experiments vs Simulations



CO oxidation on Rh/Al_2O_3 at 250°C: Experiments vs Simulations



Comparison of CO_2 conversions on Rh and Ni loaded catalysts

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