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Applied kinetics for modeling of reactive hot gas filters

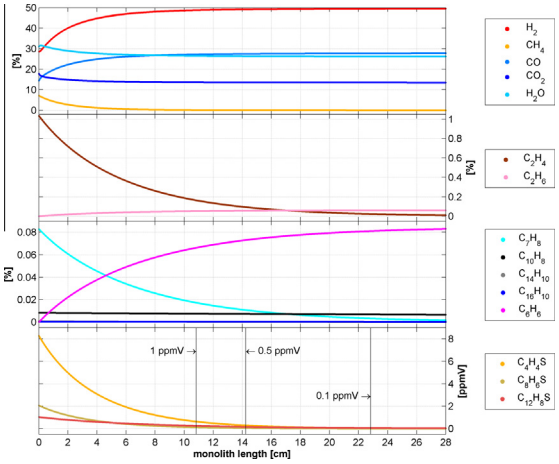
Urs Rhyner, Philip Edinger, Tilman J. Schildhauer *, Serge M.A. Biollaz

Paul Scherrer Institut (PSI), 5232 Villigen PSI, Switzerland

HIGHLIGHTS

- Applied first order kinetics could be developed for a noble metal catalyst.
- Activation energies were determined for steam reforming of tars and sulfur tars.
- Catalytic tar conversion of producer gas from biomass gasification was simulated.
- Possibilities regarding catalyst integration into hot gas filter units were evaluated.

GRAPHICAL ABSTRACT



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ABSTRACT

First order kinetics were developed based on experimental results of a noble metal catalyst. Activation energies and pre-exponential factors were determined by parameter estimation for steam reforming of tars, sulfur tars and ethene. The formation of ethane and benzene was assumed to be at constant rate depending on the decomposition of ethene and toluene respectively. Further, for steam reforming of methane and water gas shift reaction, the kinetic parameters of Langmuir–Hinshelwood–Hougen–Watson (LHHW) type rate laws including equilibrium term and adsorption of sulfur could be determined.

With the applied kinetics, catalytic tar conversion of producer gas from biomass gasification was simulated. Simulation results at operating temperatures of 850 °C showed significantly higher conversions rates for sulfur free tars, ethene and methane than at 600 °C or 750 °C while the conversion of sulfur tars was less temperature dependent and high at all temperatures.

The simulation results were used to evaluate different possibilities regarding the integration of catalytic material into hot gas filter units with vertical and horizontal filter design. The option of catalytic active filter elements, additional catalytic foam type packing at the inside of the filter element and a monolith at the exit of the filter vessel are feasible assuming the same catalyst material as applied in the reforming catalyst used in this study. These three options can be applied independently of the horizontal or vertical filter design. Placing a catalytic monolith or foam structure at the filter candle exit of a horizontal filter design was found to be unrealistic because the monolith or foam structure would be too long to reach sulfur tar concentrations below 1 ppm V.

* Corresponding author. Tel.: +41 (0)56 310 27 06.
E-mail address: tilman.schildhauer@psi.ch (T.J. Schildhauer).

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Nomenclature

AbbreviationsB-IGFC
Biomass-Integrated Gasification Fuel Cell System
Bio-SNG Biomass Synthetic Natural Gas
Ca Carberry number
CHP combined heat and power
CPO catalytic partial oxidation
cpsi channels per square inch
Da_{II} Dahmköhler number type II
DEN denominator
GC/SCD gas chromatograph sulfur chemiluminescence detector
GC/FID gas chromatography with flame ionization detector
GHSV gas hourly space velocity
HGC hot gas cleaning
HGF hot gas filtration
HPD highest probability density
LHHW Langmuir–Hinshelwood–Hougen–Watson
LHV lower heating value
LOD limit of detection
μGC micro gas chromatograph
PPI pores per inch
RDS rate determining step
Re Reynolds number
Sh Sherwood number
SOFC solid oxide fuel cells
SRM steam reforming of methane
WGS water gas shift

Latin letters
A area
D_m molecular diffusion
D_k Knudsen diffusion
D diffusion
E_a activation energy
F Molar flow
ΔH heat of adsorption
ΔH_R^0 standard reaction enthalpy
K adsorption constant
L washcoat thickness (characteristic length)
P pressure
ΔP pressure drop
Q volume flow
R gas constant
T temperature
X conversion
a' specific surface area
c concentration
d_h hydraulic diameter
d_o outer diameter
d_i inner diameter
k rate constant
K_0 pre-exponential factor
k_0 pre-exponential factor
k_f mass transfer coefficient
l length
p partial pressure
r reaction rate

Greek letters
ε void fraction
η efficiency
Φ Wheeler–Weisz modulus
φ Thiele modulus
ρ density
τ tortuosity factor

Superscripts
in inflow
n exponent
out outflow

Subscripts
b bulk
ch channel
eq equilibrium
eff effective
i species i
n norm conditions (T_n = 273.15 K, P_n = 100,000 Pa)
obs observed value
ref reference
v per volume

1. Introduction

Renewable energies are supposed to cover a substantial part of the future energy supply. Efficient processes have to be developed regarding environmental impact and costs. Small scale combined heat and power plants (CHPs) can be attractive solutions as part of a decentralized energy supply. A promising approach to reach high electrical efficiencies is the combination of biomass gasification with high temperature fuel cells, such as solid oxide fuel cells (SOFCs). The combination with fuel cells is referred to as "Biomass-Integrated Gasification Fuel Cell System" (B-IGFC). Biomass Synthetic Natural Gas (Bio-SNG) is the name of the gas produced by the combination of biomass gasification and methanation. The main technical challenge is the adjustment of the three main system components gasification, gas processing and fuel cell or methanation.

Hot gas cleaning (HGC) of producer gas derived from biomass gasification is assumed to have the potential of an efficient and economical attractive process. Hot gas filtration (HGF) technology prevents exposing heat exchangers to particle loaded producer gas because the hot gas can be filtered at exit temperatures of gasifiers. HGF temperatures stay above the condensation temperature of tars and water. This has several advantages. Processing tars above their dew points prevents fouling of equipment due to tar condensations. Because condensation in quenching columns can be avoided by HGF, depending on the gasification process, no steam needs to be added to the producer gas again. Steam content is needed downstream of the filter unit for steam reforming or to prevent soot formation in catalytic process units (e.g. fuel cell). In addition, contaminated liquid of quenching columns can be avoided which improves energy efficiency and reduces environmental impact and costs.

Exploiting high temperatures of producer gases downstream of a gasifier unit avoids heat losses because cooling and reheating of the gas is not needed. Catalytic partial oxidation (CPO) can be applied to heat the producer gas but will reduce the heating value. High temperatures are needed e.g. to catalytically convert sulfur containing hydrocarbons (sulfur tars) and sulfur free tars to lower molecular hydrocarbons and H_2S. Hydrogen sulfide can be adsorbed by a fixed bed of metal oxides completing the desulfurization of the producer gas. Sulfur concentration in the producer gas below 1 ppm V will reduce losses in the performance of nickel