# Comparison of Different Fixed-bed Methanation Reactors for Renewable Natural Gas Production

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Technical University of Munich

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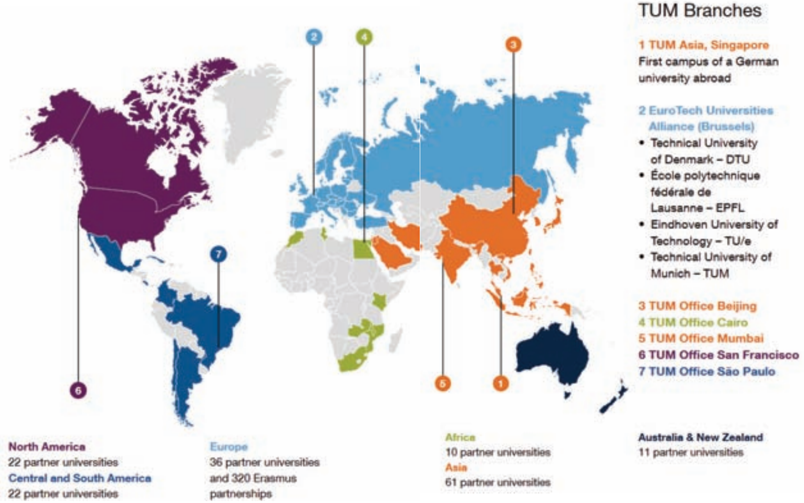
# Technical University of Munich (TUM)

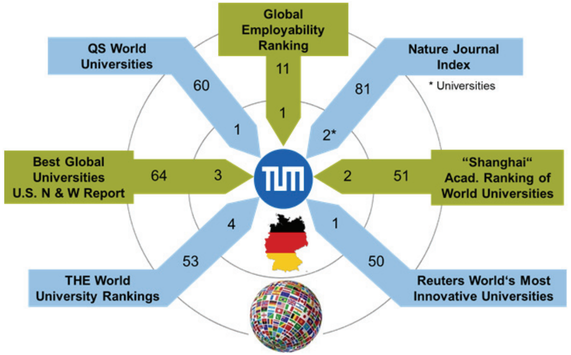
## Overview

**TUM in numbers:**

- 14 departments
- 3 Integrative Research Centers
- 7 Corporate Research Centers
- 10,103 staff members
- 40,124 students
- 545 professors
- 172 degree programs
- 411 buildings
- 17 Nobel prizes
- 800 Start-ups
- Budget: 1,329 Mio. € /
  285 Mio. € third-party

→ https://www.tum.de/



# Chair of Energy Systems, TUM

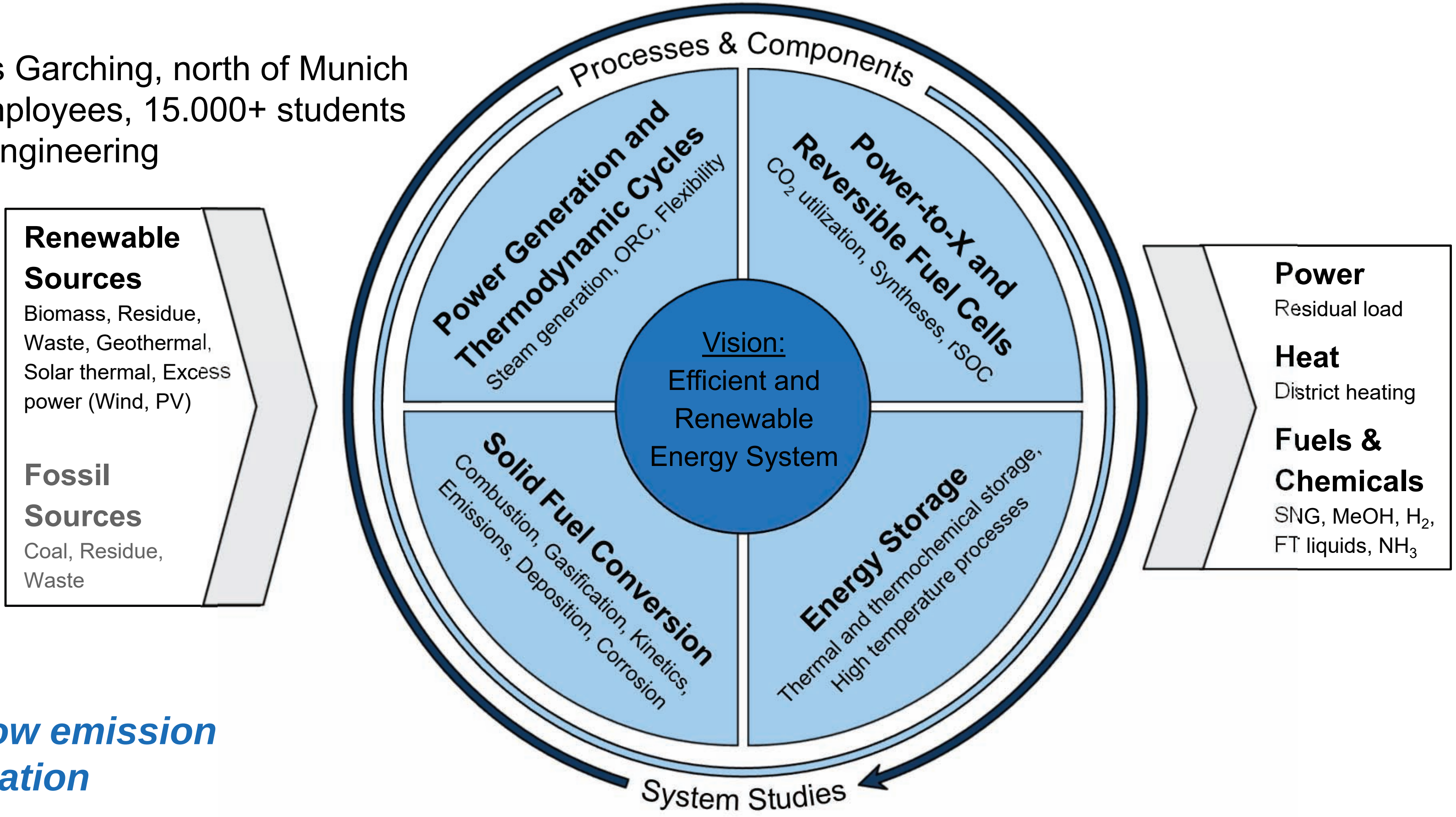
**Head of Chair: Prof. Dr.-Ing. Hartmut Spliethoff**

- Located at the TUM Campus Garching, north of Munich
- Campus Garching: 6.000 employees, 15.000+ students
- Department of Mechanical Engineering

- CES staff:
  45 Employees
  30 PhD students
  5 Postdocs

- Group leaders:
  Sebastian Fendt
  Annelies Vandersickel
  Christoph Wieland
  Stephan Gleis
  Stefan DeYoung

***Mission: Efficient and low emission renewable power generation***

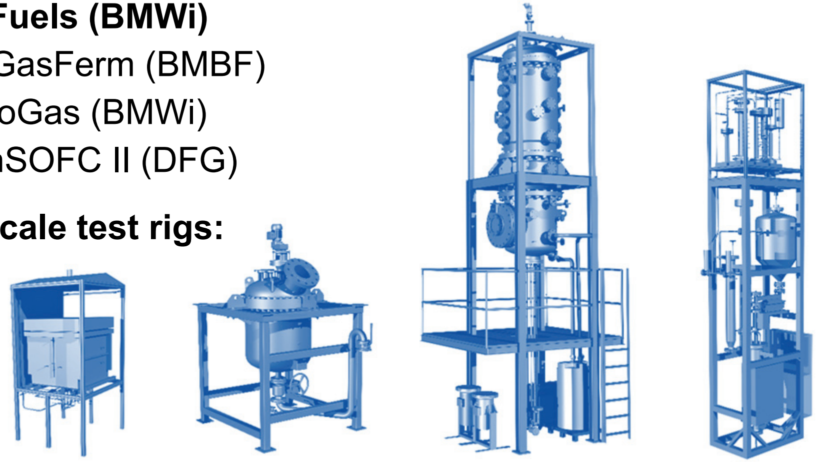


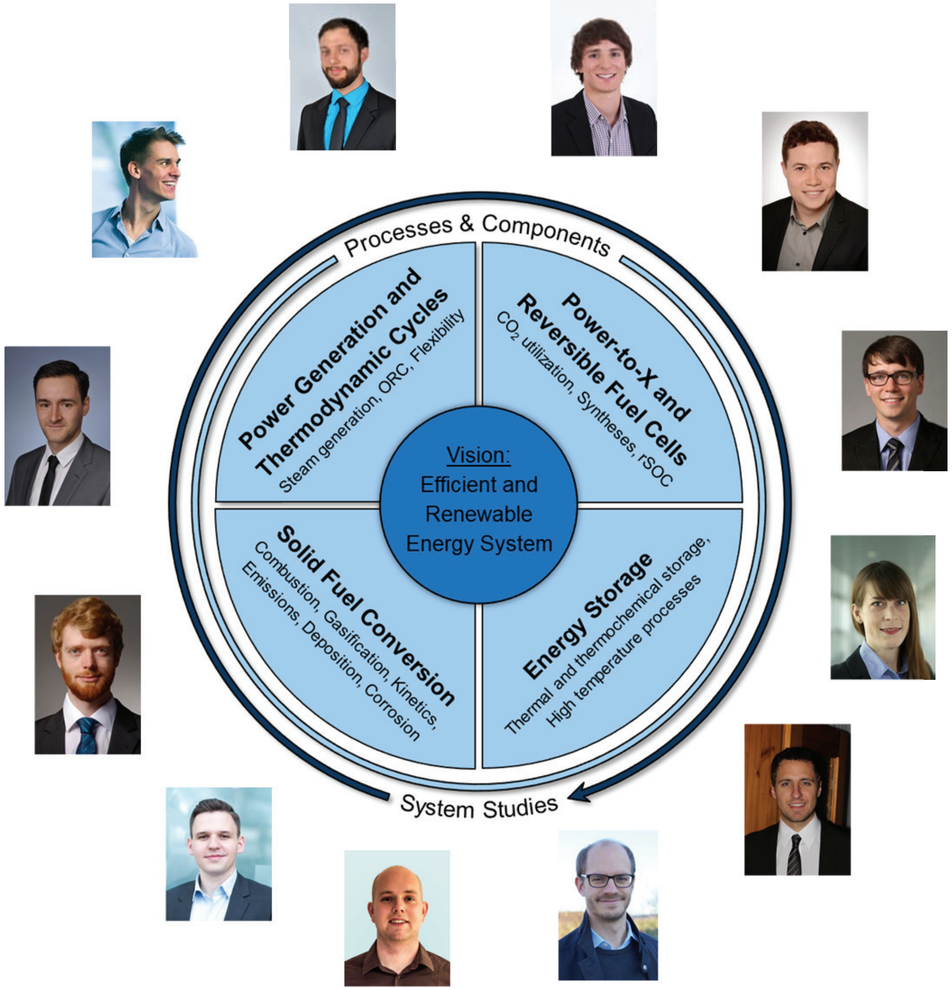
# Research group:
# Biomass and Renewable Fuels

**Research topics:**
- Biomass pre-treatment
- Biomass gasification and combustion
- Power, heat and fuel production from biomass

**Projects:**
- Biofficiency (EU/H2020)
- **BioCORE (BMBF)**
- **E2Fuels (BMWi)**
- ReGasFerm (BMBF)
- PyroGas (BMWi)
- SynSOFC II (DFG)

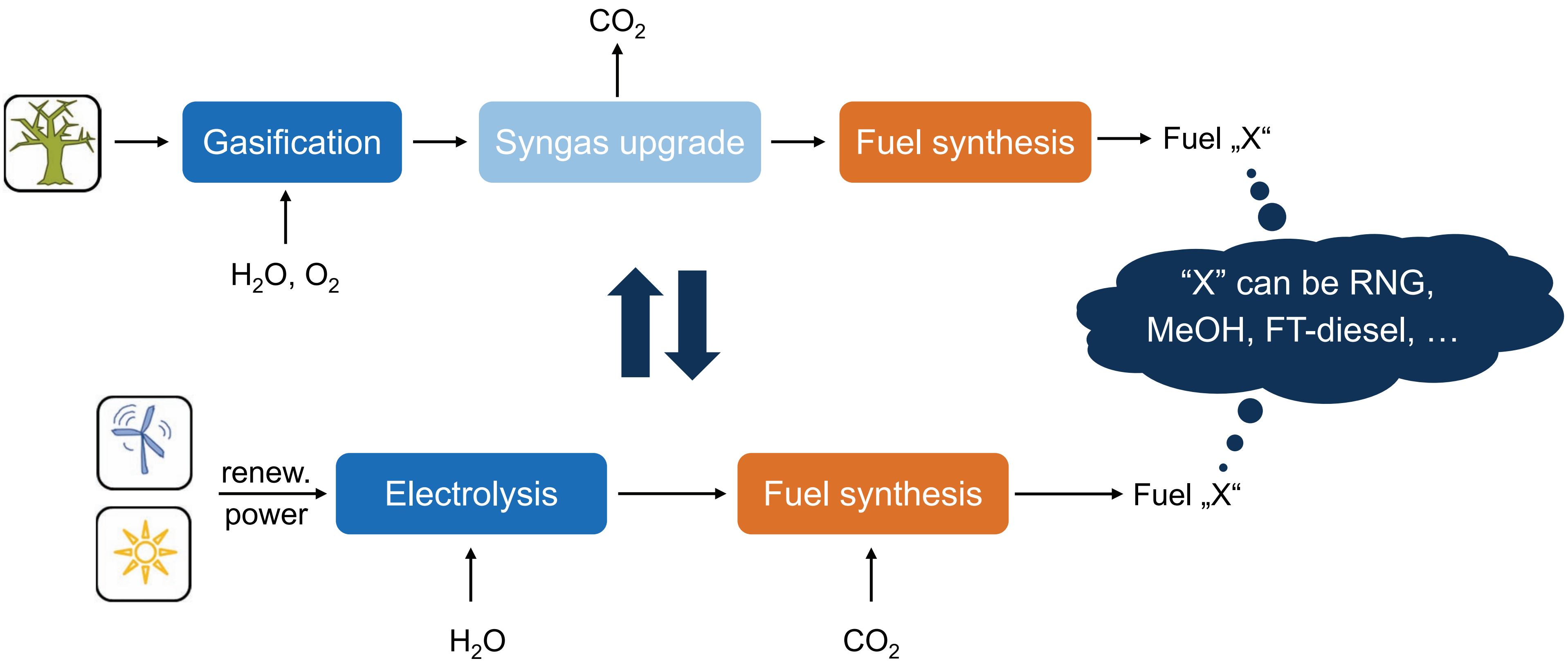
**Pilot-scale test rigs:**





# Motivation and wording

**Biomass-to-Fuel (BtG, BtL, BtF → BtX) and Power-to-Fuel (PtG, PtL, PtF → PtX)**



# Wind and solar, the drivers of PtX

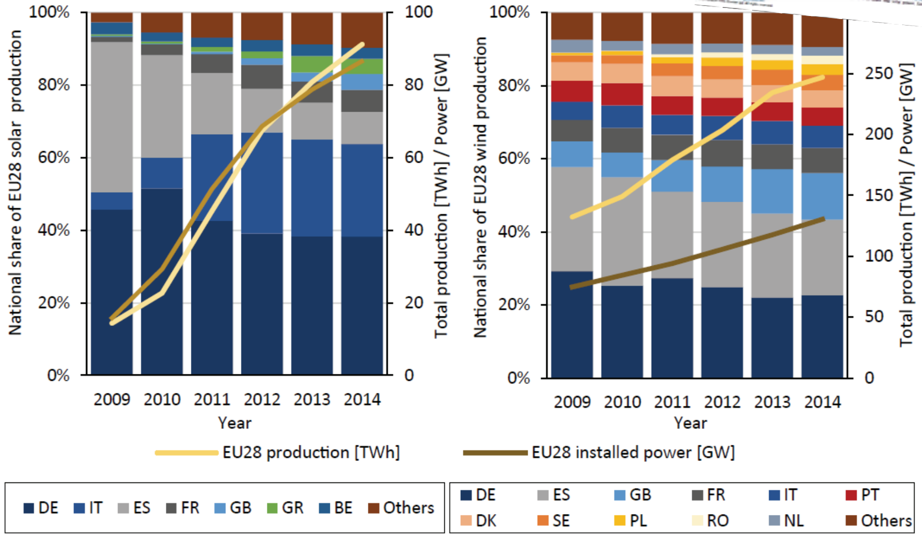
## System studies and review studies by CES

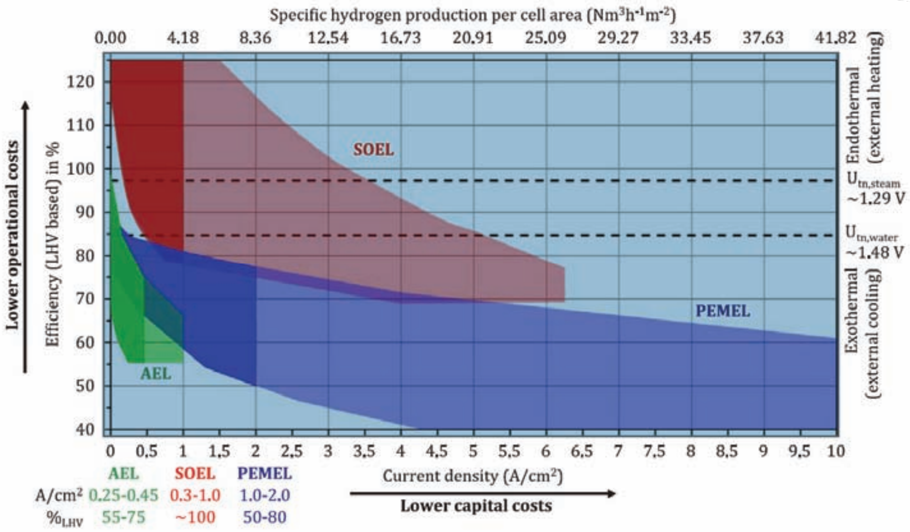
**Variability of wind and solar power**
**An assessment of the current situation in the European Union based on the year 2014**

A. Buttler et al., Energy 106 (2016) 147-161

**Current status of water electrolysis for energy storage, grid balancing and sector coupling via power-to-gas and power-to-liquids: A review**

A. Buttler et al., International journal of hydrogen energy 40 (2015) 38-50



# Carbon source for fuel synthesis: EFG of biomass and biogenic waste

## PiTER

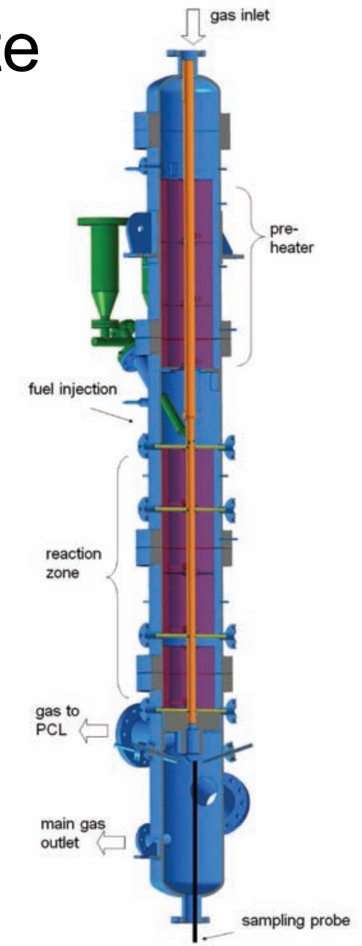
→ Investigated fuels:
- Biomass: HTC coal, torrefied wood, beech wood
- Coal mixtures/blends

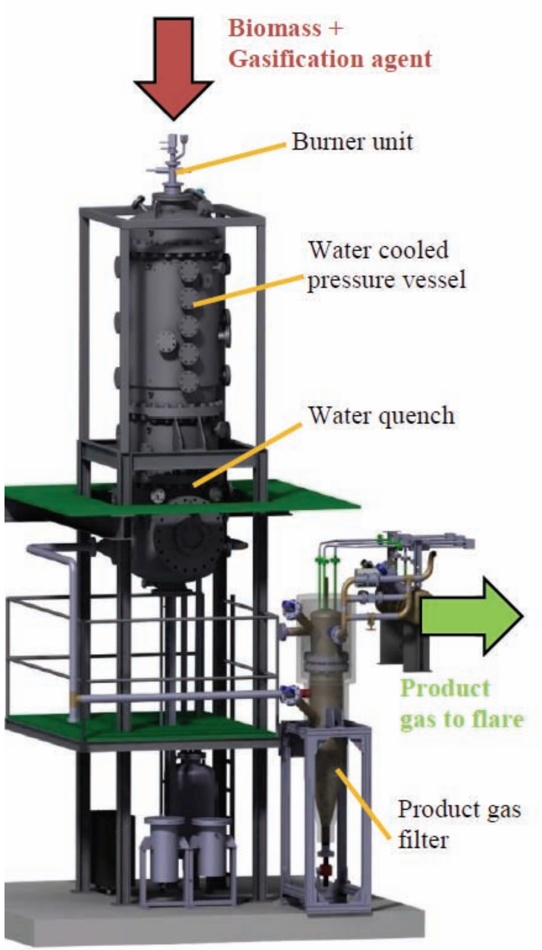
→ *Operating conditions:*
$T$ = 1200-1600°C, $p$ = 1-40 bar, O/C = 1, $O_2/H_2O/CO_2/N_2$

## BOOSTER (autothermal gasification)

→ Investigated fuels:
- HTC biomass (green waste, beech wood and compost)
- Torrefied wood (deciduous and coniferous wood)
- Beech wood
- Corn cob
- Grass waste

→ *Operating conditions:*
$P_{th}$ = 70-150 $kW_{th}$, $p$ = 1 bar, $\lambda$ = 0.3-0.6, $O_2/H_2O$/Air



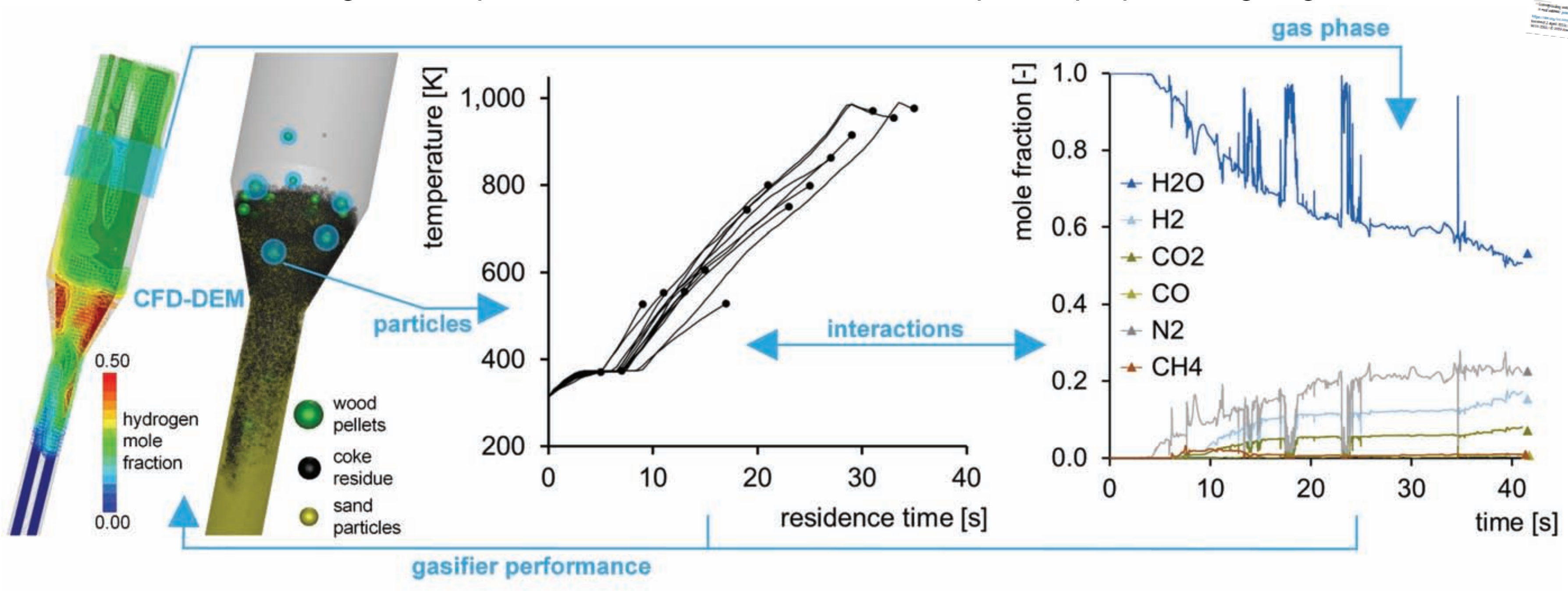



# CFD-DEM simulation of biomass gasification in a fluidized bed reactor

P. Ostermeier et al., Fuel 255 (2019) 115790

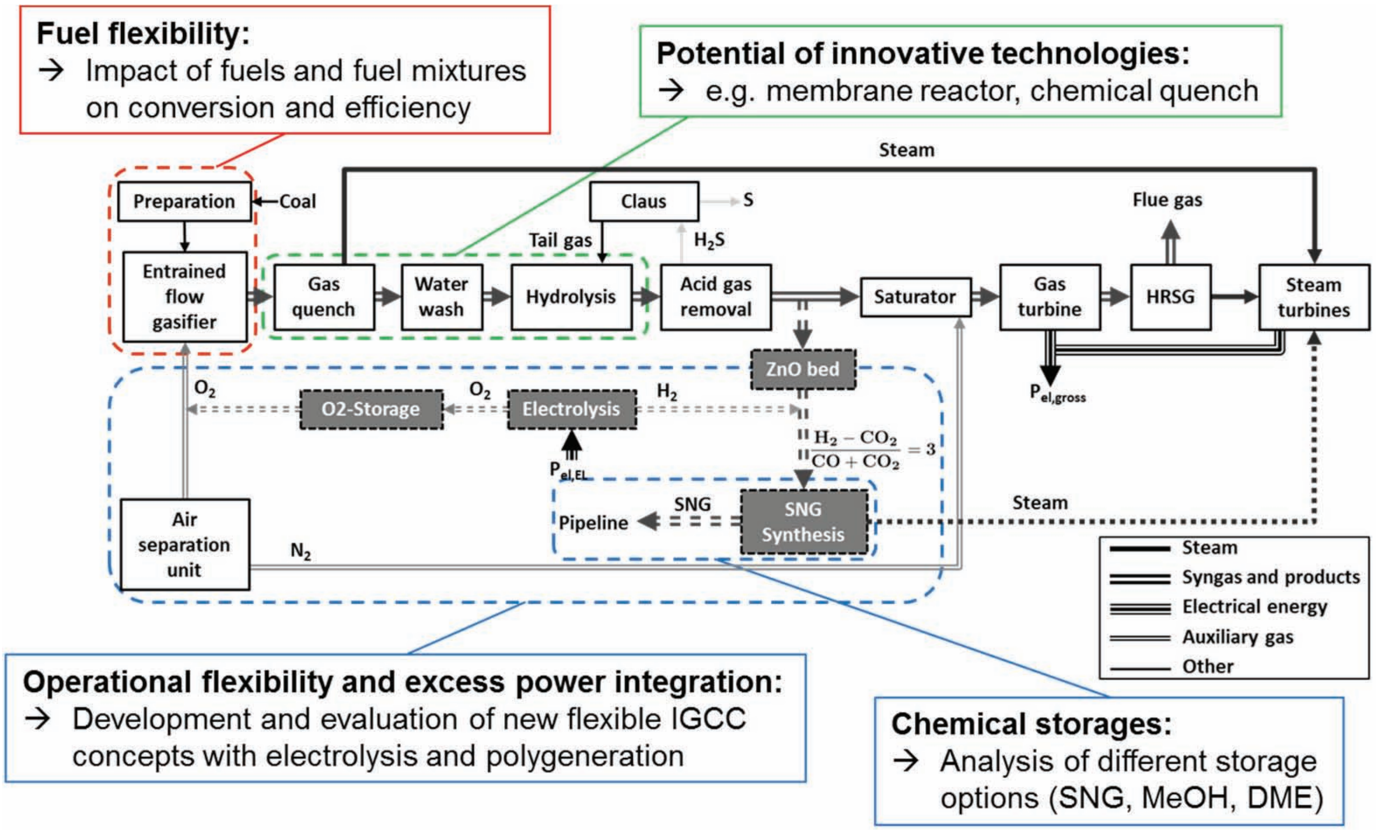
**Motivation:**

- Gas-solid fluidized beds play an important role in many industrial operations
- But: lack of knowledge of the processes inside the bed which impedes proper designing



# Process modeling of innovative IGCC and polygeneration concepts

**Detailed process analysis (techno-economic analysis)**





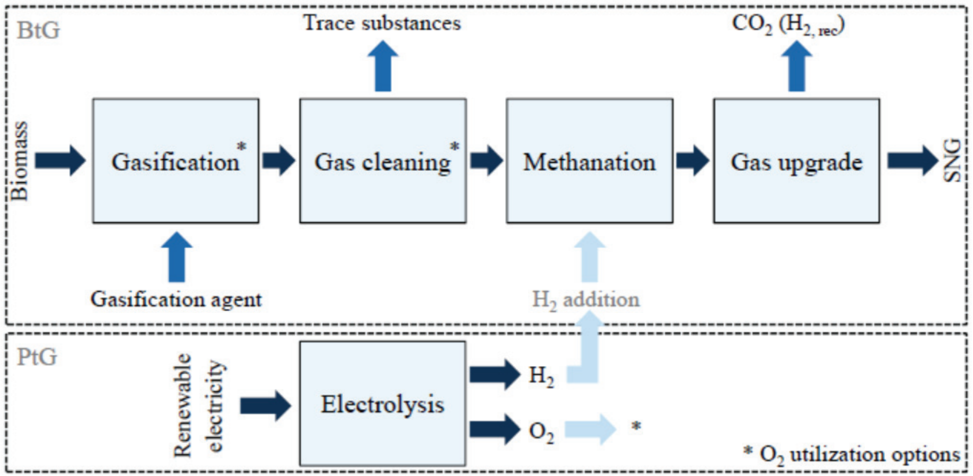
Dissertation of
A. Buttler 2018
(German)

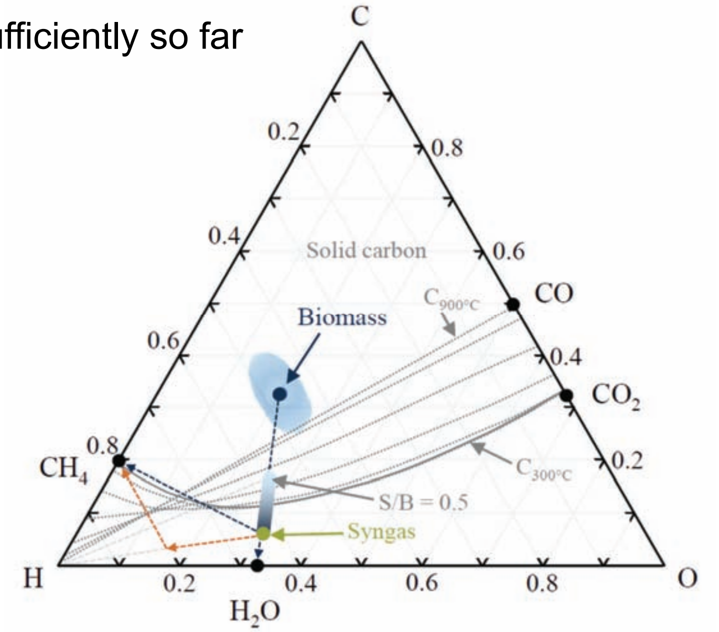
# RNG production from lignocellulosic biomass

## Experimental investigation of RNG production under realistic conditions

<u>Motivation:</u>

- Inherent lack of $H_2$ for RNG production from woody biomass
- At the same time: PtG concepts needs a renewable $CO_2$ source.
- The two processes fit well together for a sustainable solution
- But: Main technological obstacles have not been addressed sufficiently so far



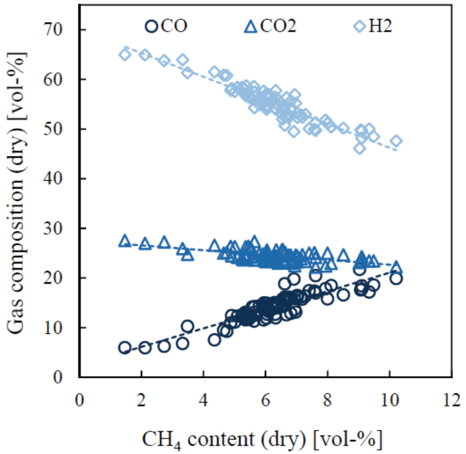



# RNG production from lignocellulosic biomass (BFB)

## Experimental investigation of RNG production under realistic conditions

**Gasification**

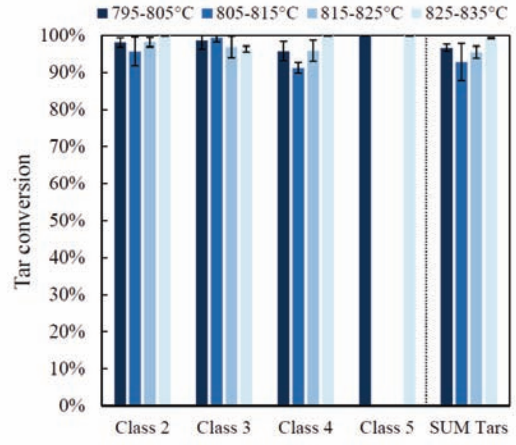
Adaption of gas quality for later methanation




Influence of parameter variation on syngas composition

**Gas cleaning**

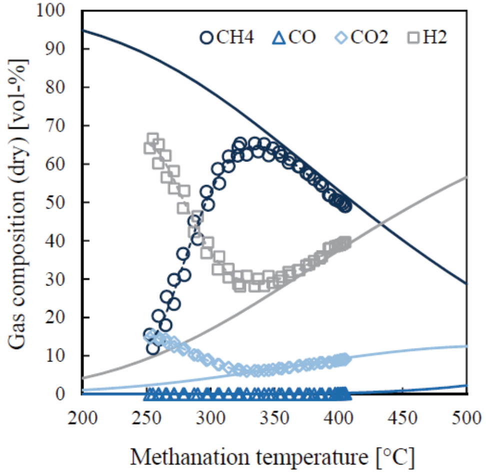
Tar conversion (long-term testing)




Tar conversion under realistic conditions

**Methanation**

Catalyst performance



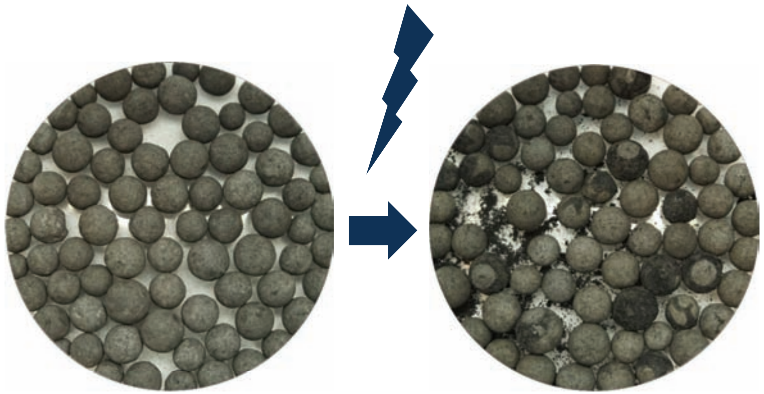

Lab-scale tests on catalyst activity

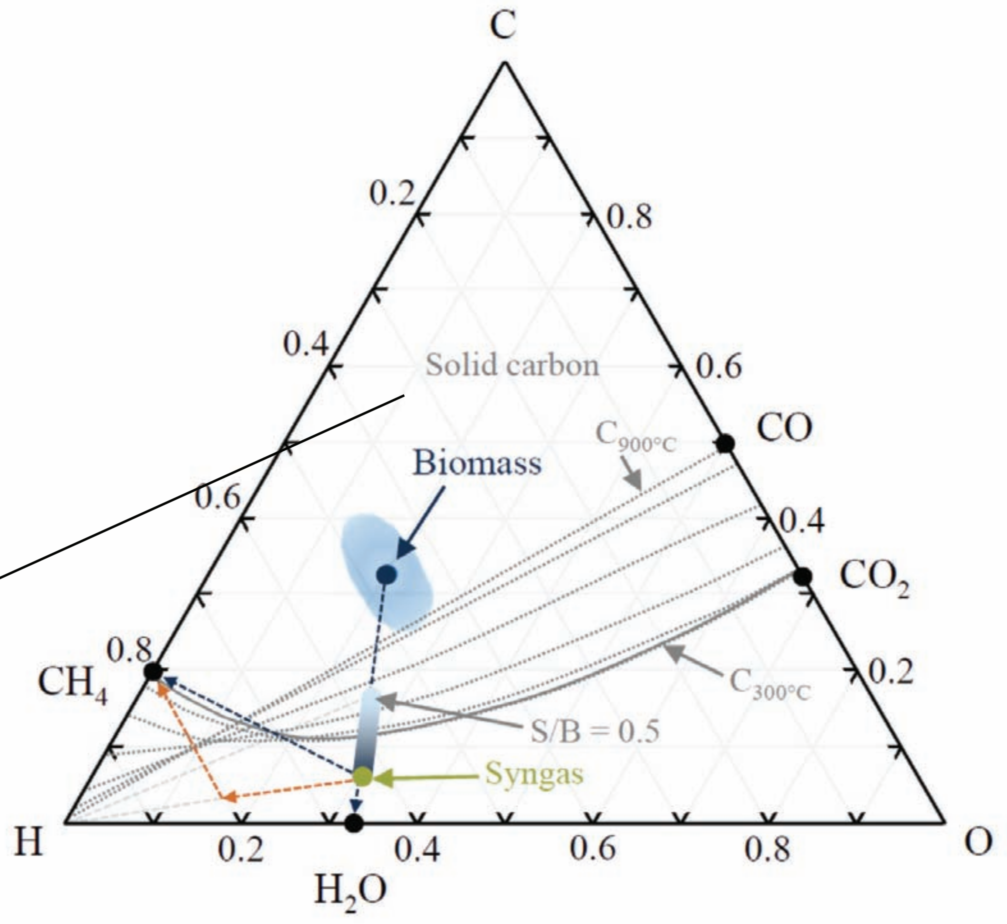
# Bio-RNG production from syngas – Fixed-bed methanation

Aims:

- Simple, optimized fixed-bed reactors for decentralized bio-based methanation
- Hot-spot control and reduction (→ catalyst life-time)
- Avoidance of sintering and carbon formation

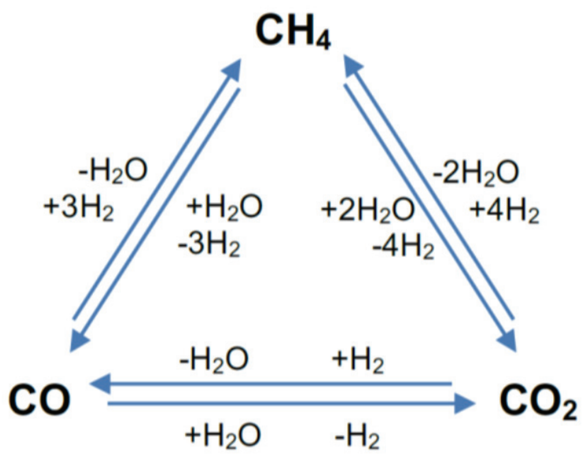
- Model-based optimization of future designs → CFD

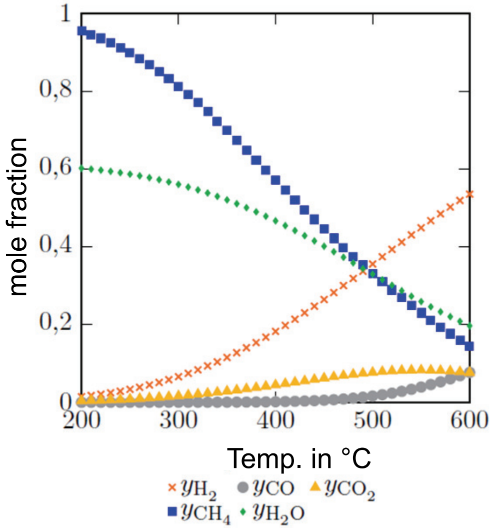


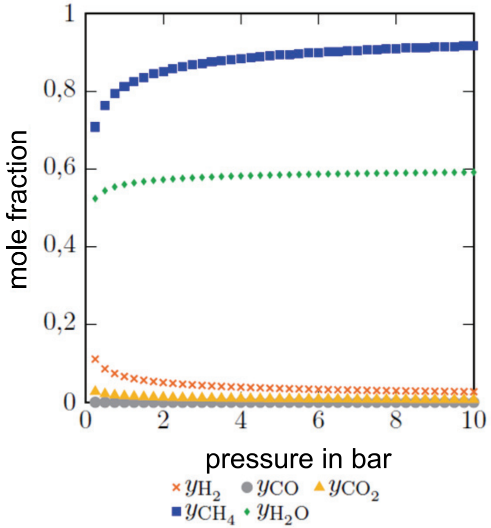


# Methanation – Fundamentals

## Influencing factors on $CH_4$-yield



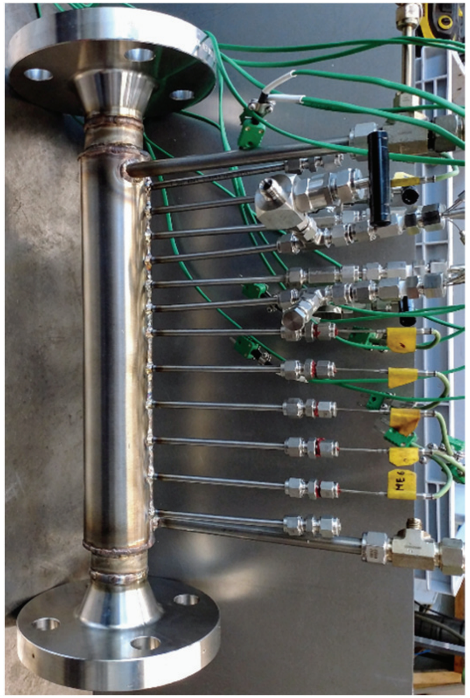



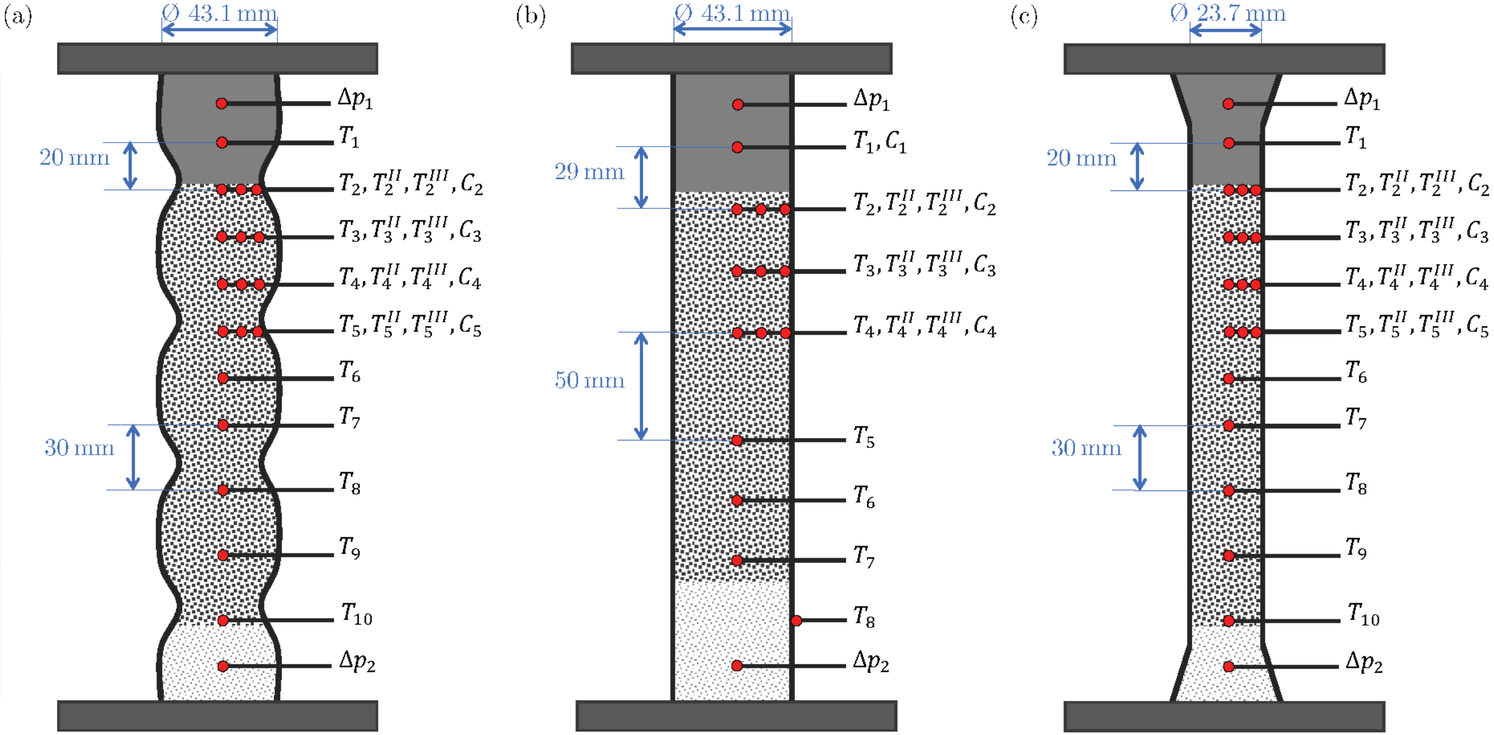

- Temperature (highly exothermal, reaction kinetics)
- Pressure
- GHSV / CSV
- Inlet concentration (e.g. as $H_2/CO$)
- Catalyst (usually Ni-based)

**Carbon Deposition Reactions**

$$2CO \leftrightharpoons C + CO_2$$
$$CO + H_2 \leftrightharpoons C + H_2O$$
$$CH_4 \leftrightharpoons C + 2H_2$$
$$C_nH_{2n} \leftrightharpoons nC + nH_2$$

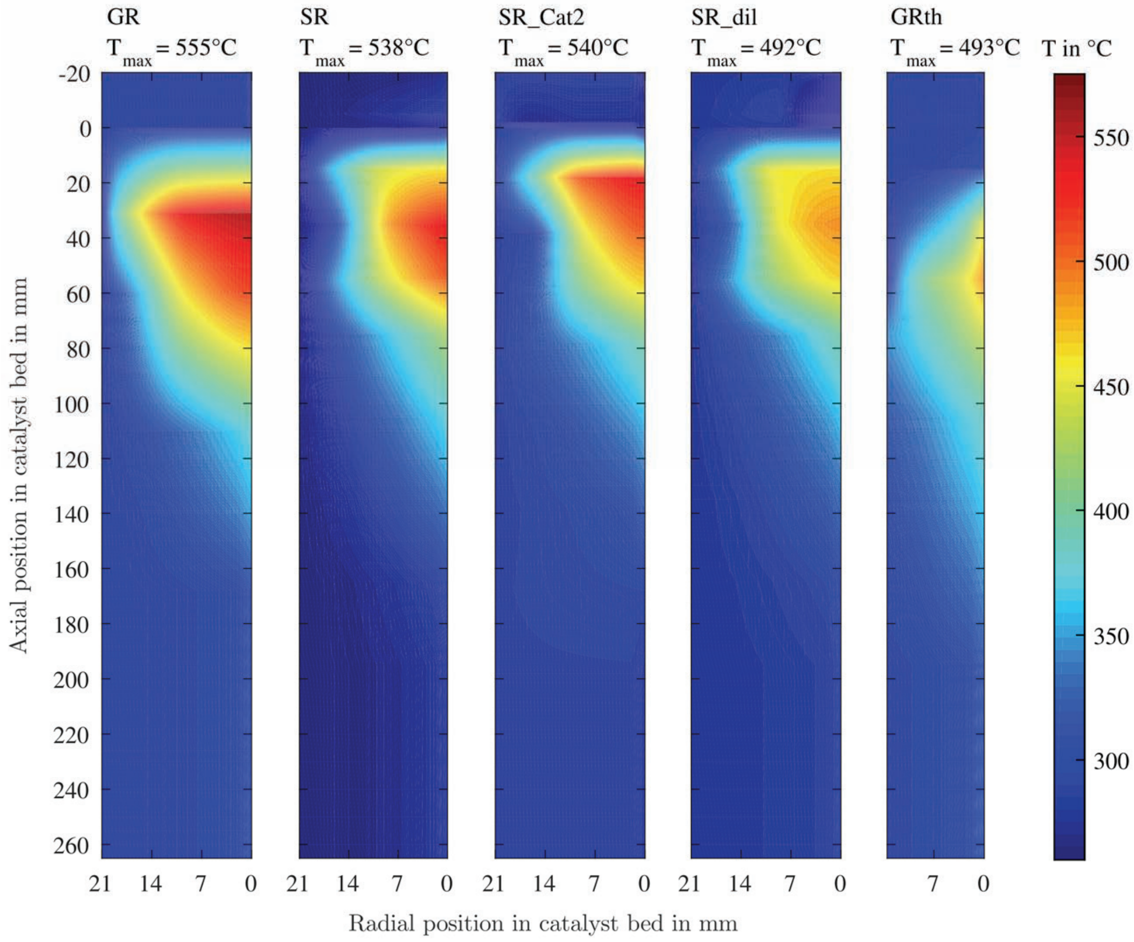
# Investigation of different oil-cooled fixed bed reactors

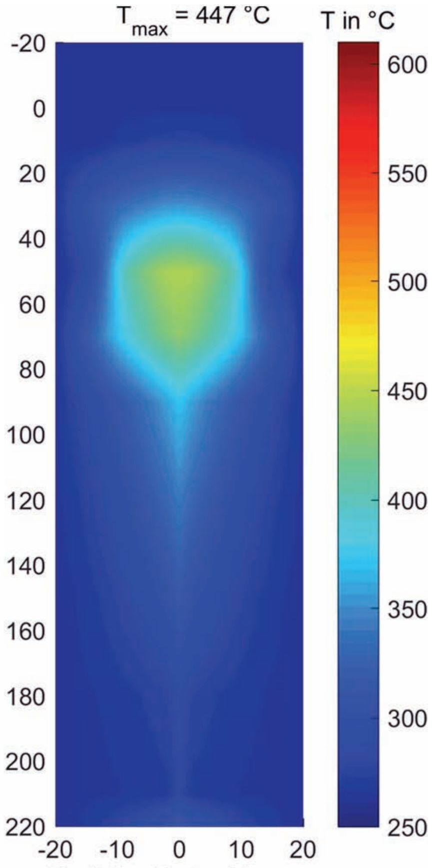





- 3 geometries: ............................... a) Dented pipe (SR) b) Standard pipe DN40 (GR) c) Thinner pipe DN25 (GRth)
- 2 catalysts: Meth134: 25% $Ni/Al_2O_3$, spherical
  Cat2: 40% $Ni/Al_2O_3$, cylinders

# Reactor geometry influences hot spot → load changes

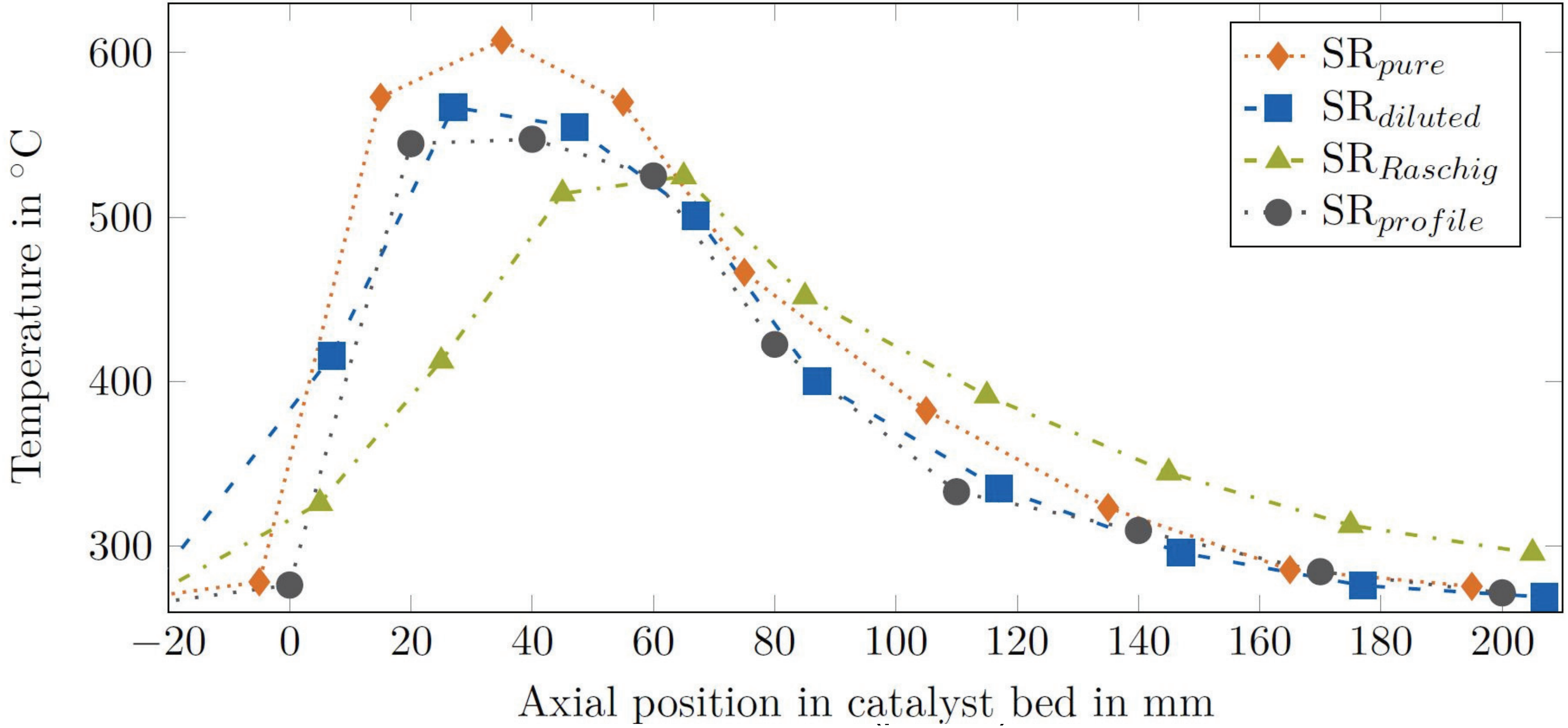


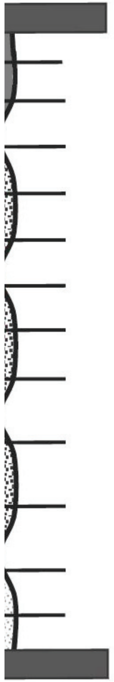



# Hot spot reduction measures

**Raschig rings, catalyst dilution using inert materials**

Raschig rings

sta
(pure

rings
n")

# Pseudo-homogenous reactor modeling

**... enables time-efficient simulation of complex reactor geometries**

**Approach:**

## Modeling of the kinetics

Micro kinetic $r_{int}$

Mass transport limitation via Thiele modulus

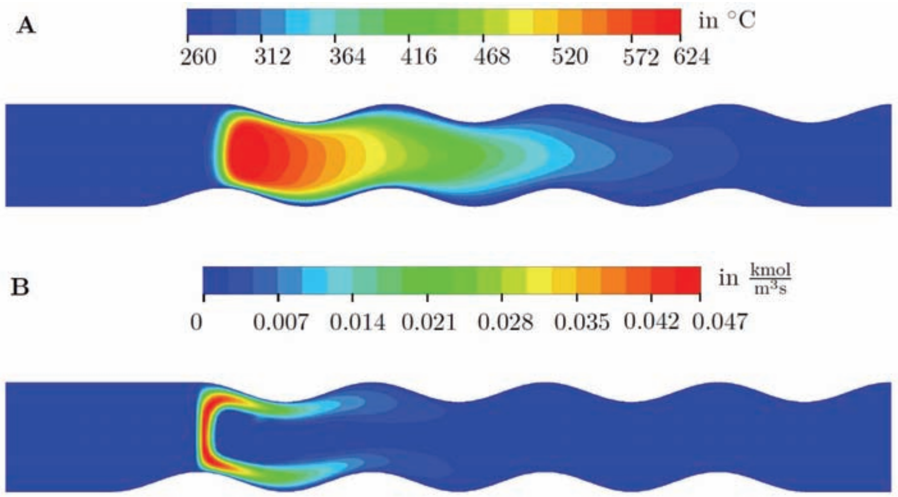
→ Effectiveness factor: $\eta$

Macro kinetic $r_{eff} = r_{int} \times \eta$

(Rönsch, Koschany, Kopyscinski, …)

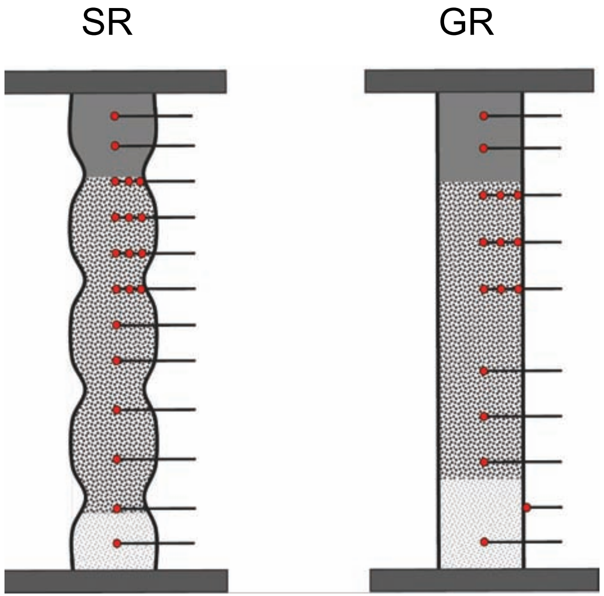
## Modeling of the fixed bed

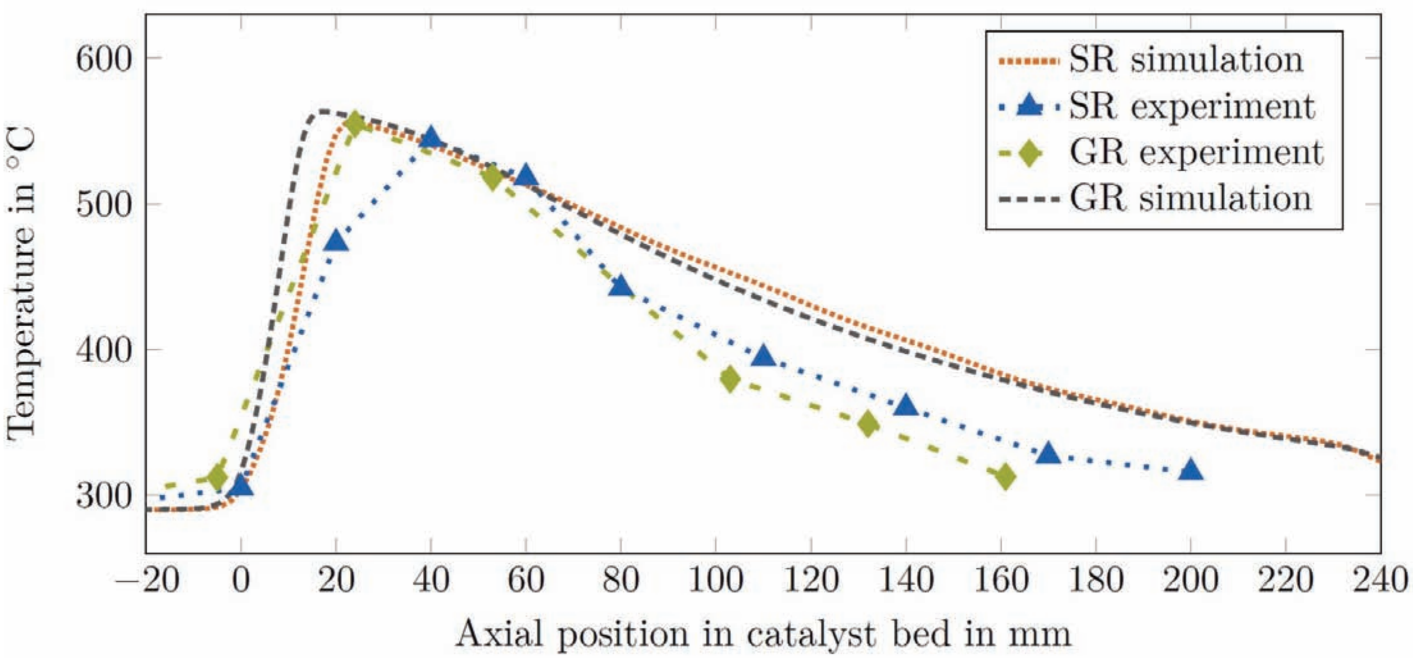
Fixed bed




Publication submitted

# CFD model is capable of predicting temperatures for a broad variety of geometries and conditions



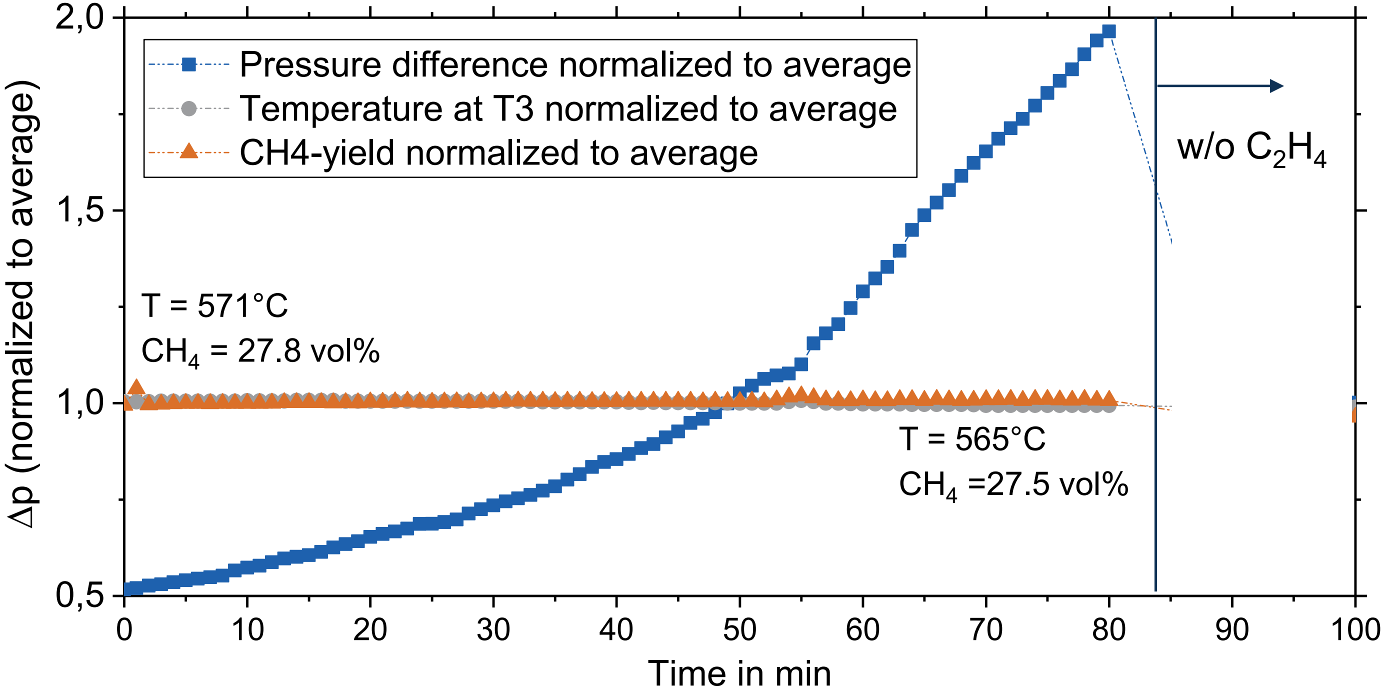



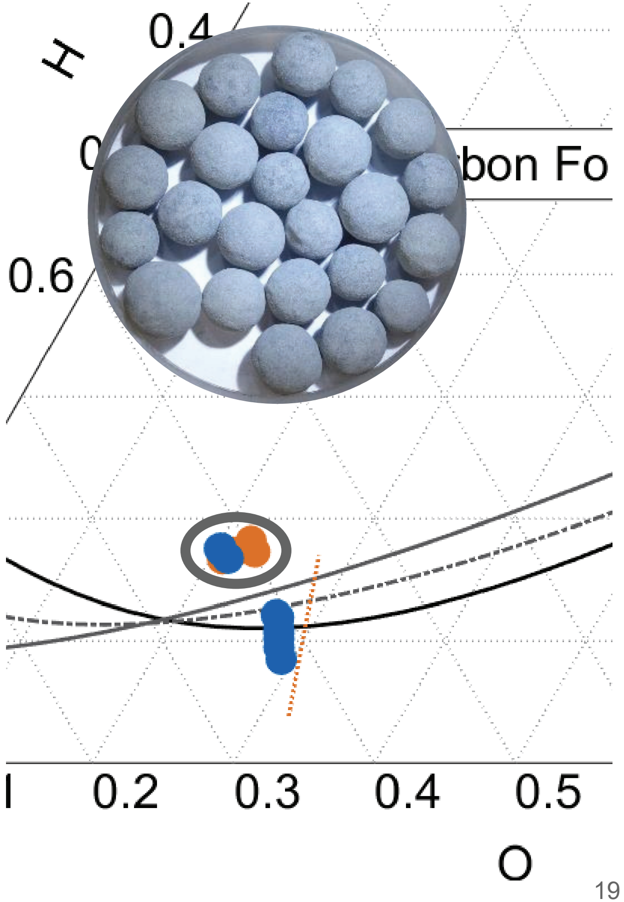

→ Simulations are in good agreement with experimental results (not only regarding temperature profiles)

# Excursus: Carbon formation running on syngas

**Fixed-bed methanation with UC Davis bio-syngas**

- Obvious carbon formation immediately detected if $C_2H_4$ is present
- Same conditions without $C_2H_4$: no further increase in diff. pressure

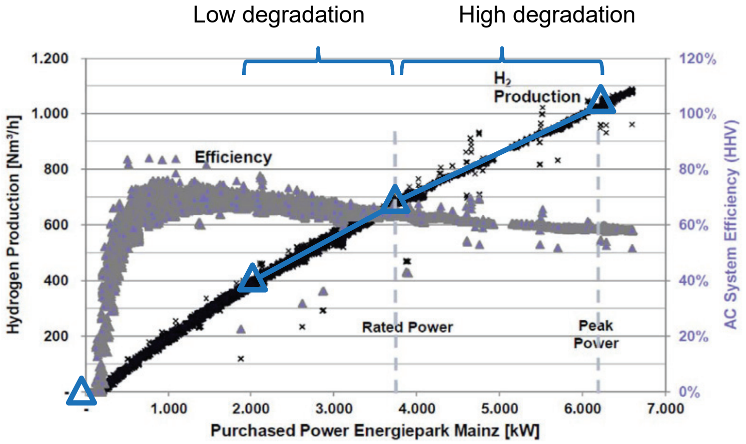


# Evaluation of dynamic operation (PtG plant)

## Dimensioning and integration of electrolysis and methanation in the model

**Electrolysis:**



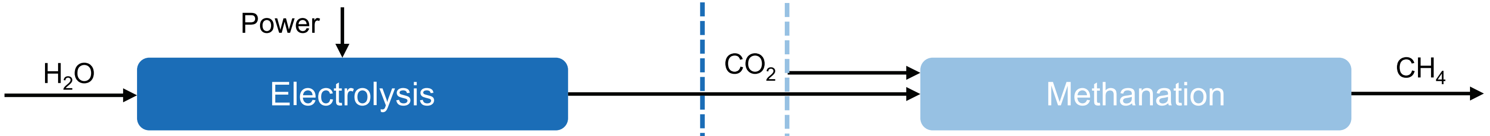

Piece by piece interpolation of the characteristic curves for load-dependent $H_2$-production and efficiency

**Methanation:**

- Dimensioned to peak load of electrolysis
- Constant product gas composition
- $\eta_{HHV} = 78.2\ \%$

Dynamic behavior:

- Operation only if reactor warm
- Cool-down via half-life time $(12\ \mathrm{h})$
- Different start types:
  - After long downtime: $26\,\frac{€}{\mathrm{MW}}$
  - After e.g. 12 h of downtime: $14.2\,\frac{€}{\mathrm{MW}}$



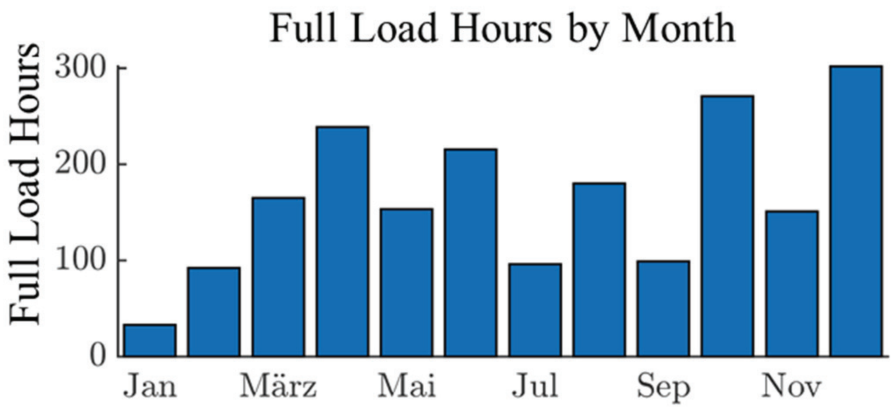

A. Buttler et al., http://dx.doi.org/10.1016/j.rser.2017.09.003

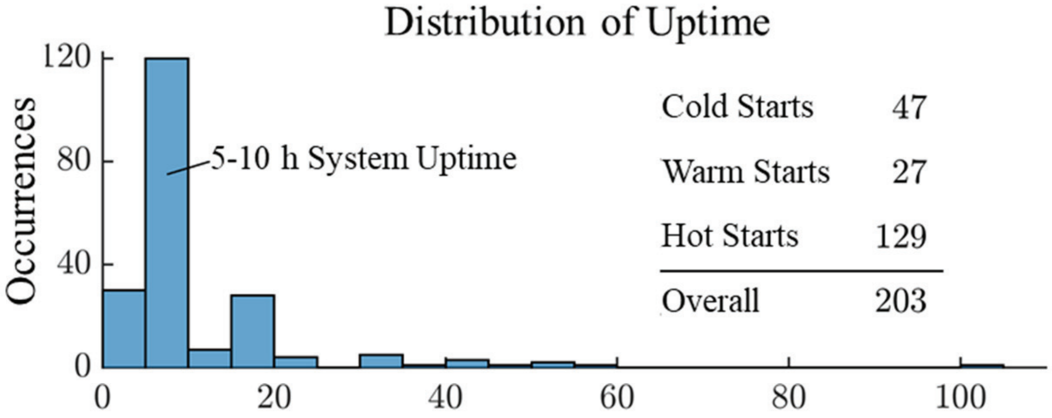
# Resource scheduling for a PtG plant

## Exemplary results for April 2017

e2 Fuels
Erneuerbare Emissionsarme Kraftstoffe

Federal Ministry for Economic Affairs and Energy




Tumbling resource scheduling based on mixed integer linear optimization

# Conclusion and Outlook

- Understanding methanation is crucial for the design of any RNG process (BtG and PtG)
- A holistic research approach from lab-scale kinetics to reactor design via modelling is extensive but needed → cooperation
- Different reactor technologies and process layouts might be suitable for different applications and scales
  → e.g. fluidized bed vs. fixed bed, multi-zone design, etc.
- Transferability of lab-scale results to full-scale reactor systems (integration of transport limitations in form of effectiveness factor to kinetic data sets) is crucial and needs still more work
- The findings might be transferrable to other highly exothermic reactions
- Heat management crucial for any methanation research

<u>Outlook:</u>

- Catalyst development and optimization ongoing → tests with „new“ catalysts
- Enhanced analysis of used catalysts regarding loss of activity, specific surface area, etc.
- Testing of different reactor geometries and catalyst systems in order to further optimize fixed bed methanation
- Develop new ideas and concepts, which might not be economic today, but show an increased potential efficiency and flexibility for the future
- Investigation of biological alternatives

# Thanks…

**… to my group and colleagues at CES**
**… to the German Federal Ministry for Economic Affairs and Energy**
**… to the Federal Ministry of Education and Research**
**… to BaCaTeC**

Federal Ministry of Education and Research

Federal Ministry for Economic Affairs and Energy

BaCaTeC

## … and thank you for the attention!

More information: http://www.es.mw.tum.de/