Supported by:
Federal Ministry for Economic Affairs and Energy
on the basis of a decision by the German Bundestag

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

International Conference on Polygerneration Strategies 2019

Trickle-Bed Reactor for Biological Methanation

Tobias Weidlich, M.Sc.
Chair of Energy Process Engineering
Friedrich-Alexander-Universität Erlangen-Nürnberg

Lab-scale trickle-bed reactor in Nürnberg

Trickle-bed reactor in Regensburg (project ORBIT) ©Michael Heberl

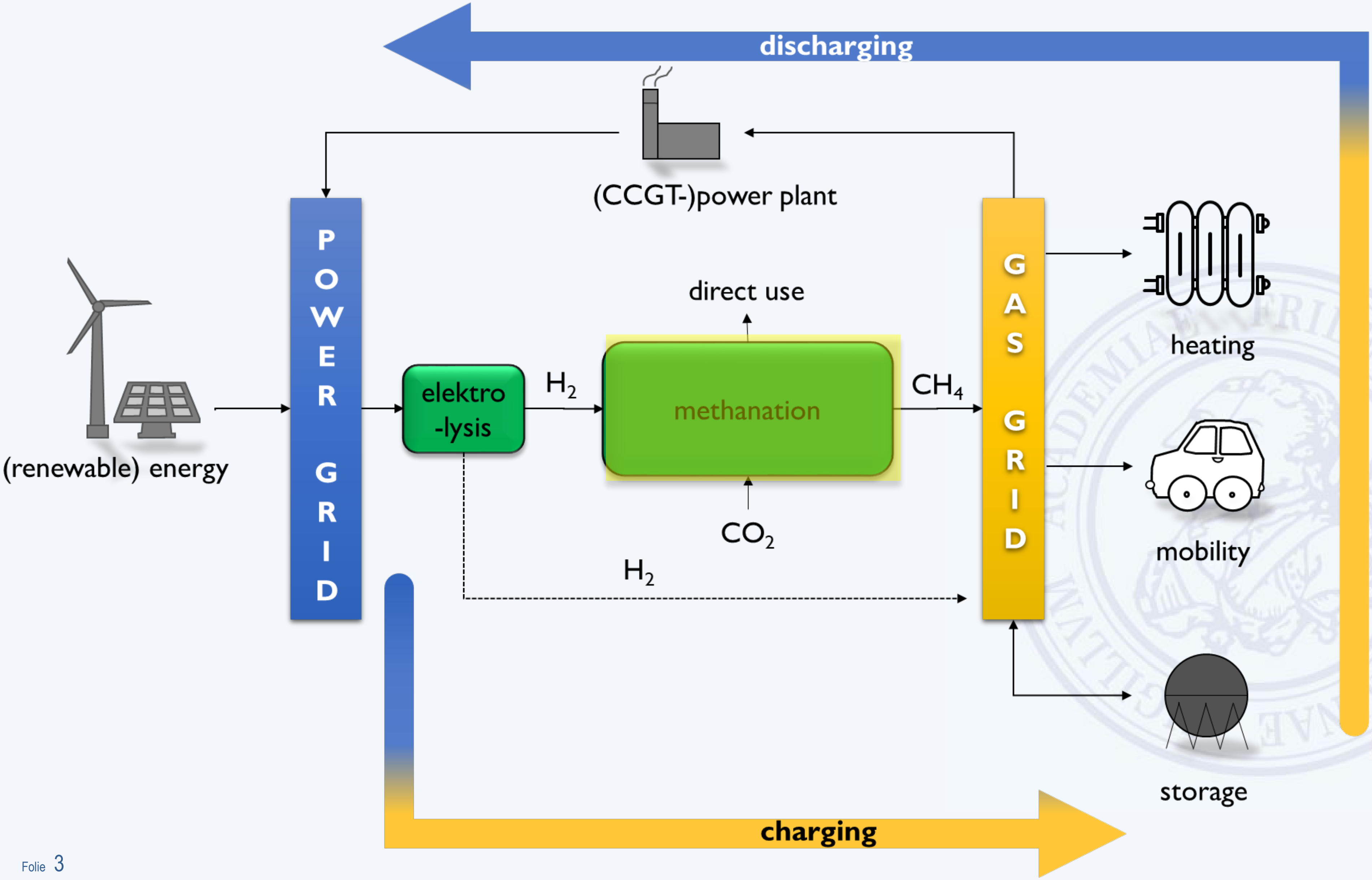
FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Outline

- Power-to-Gas
- TBRs for biological methanation
- Results pure culture
- Results mixed culture
- Standardization
- Conclusion and Outlook

FRIEDRICH-ALEXANDER
UNIVERSITÄT
ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Power-to-Gas



FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

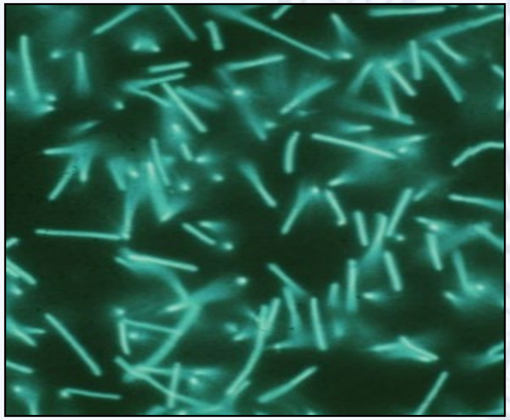
Biological methanation

- Biological methanation is the generation of methane by microorganisms from gaseous feedstocks (most commonly CO_2 or CO and H_2)
- The main challenge is the solution of H_2 in the liquid phase
- The methane production rate (MPR_R) shows the performance of the biological methanation process

$$CO_2 + 4\,H_2 \Rightarrow CH_4 + 2\,H_20$$

$$CO + 3\,H_2 \Rightarrow CH_4 + H_20$$

$$MPR_R = \frac{\dot{V}_{CH_4,out} - \dot{V}_{CH_4,in}}{V_R}\left[\frac{m^3}{h \cdot m^3} = h^{-1}\right]$$



Source: Institute of Microbiology and Archaea Centre, University of Regensburg

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Biological Methanation @FAU

1) <u>Lab-scale trickle-bed reactor TBR (V = 5.0 l)</u>

- Operation mode: batch and continuous
- Variation of parameters and optimization of the stable running process
- Running with a pure culture and a mixed culture
- Up to 2.5 bar_a

Lab-scale trickle-bed reactor in Nürnberg

2) <u>Lab-scale stirred reactor CSTR (V = 6.8 l)</u>

- Operation mode: batch and continuous
- Variation of parameters and optimization of the stable running process
- Running with a pure culture and a mixed culture
- Up to 2.5 bar_a

Lab-scale CSTR in Nürnberg

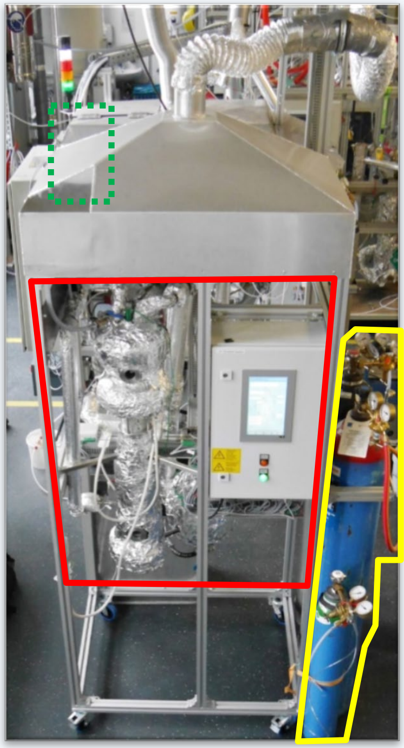
3) <u>Prototype trickle-bed reactor (V = 50.0 l)</u>

ORBIT

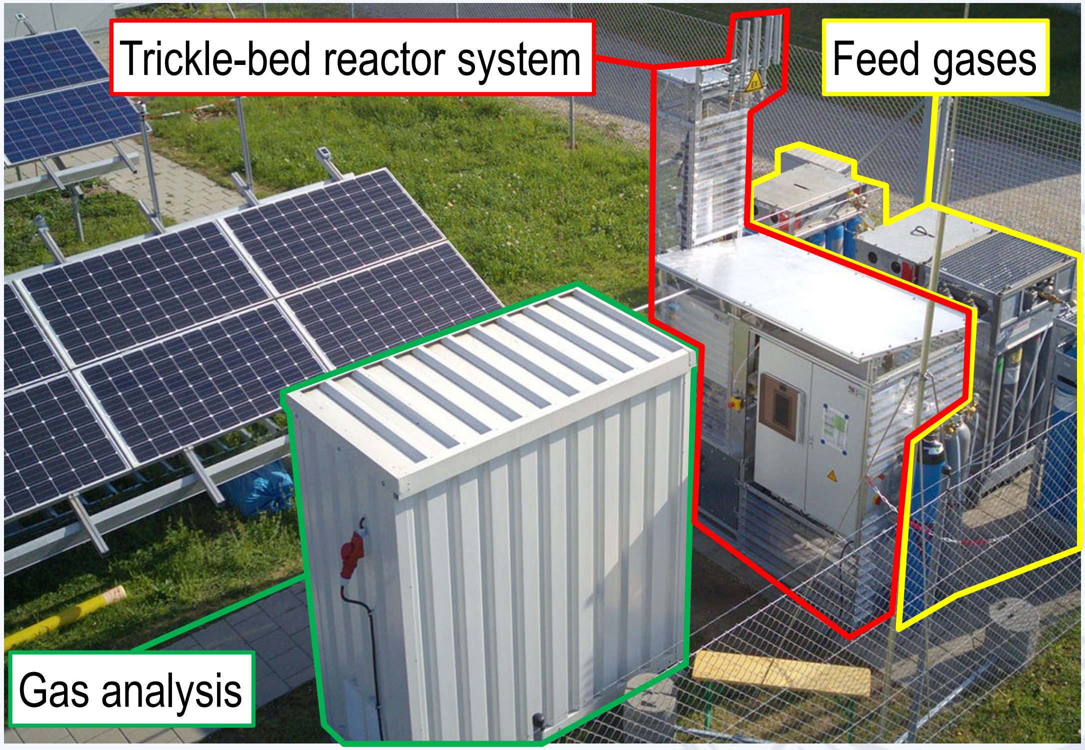
- Part of the research project “ORBIT”
- Designed for 16 bar_a
- First experiments running with a pure culture

Trickle-bed-reactor in Regensburg (project ORBIT)

TBRs for biological methanation



Lab-scale reactor at
FAU Erlangen-Nürnberg



© Michael Heberl

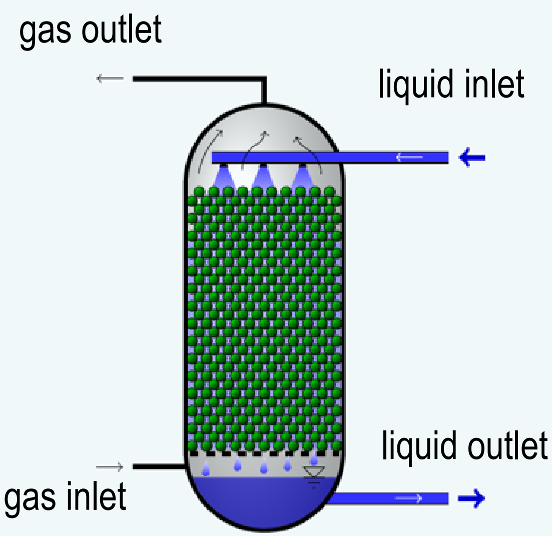
ORBIT reactor at OTH Regensburg

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Two reactors – one concept

- Trickle-bed reactors (TBR) are used for enhanced solution of gases in a liquid
- High surface between liquid and gas phase
- Most common: gas and liquid in countercurrent


Lab-scale trickle-bed reactor in Nürnberg



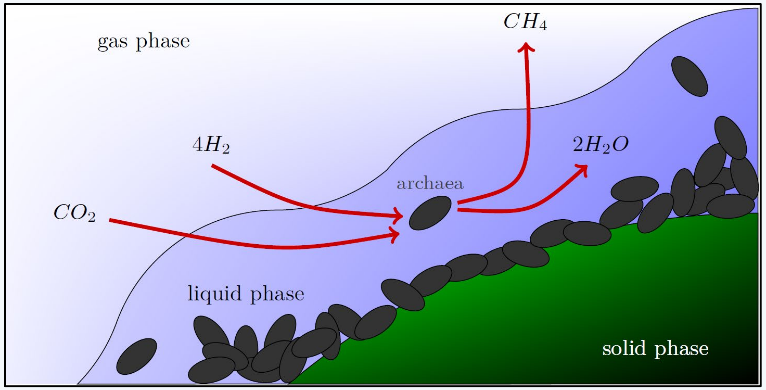

Trickle-bed reactor in Regensburg (project ORBIT)

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Two reactors – one concept

- Trickle-bed reactors (TBR) are used for enhanced solution of gases in a liquid
- High surface between liquid and gas phase
- Most common: gas and liquid in countercurrent
- Archaea live in the liquid, but can also create a biofilm on the random packing

Lab-scale trickle-bed reactor in Nürnberg

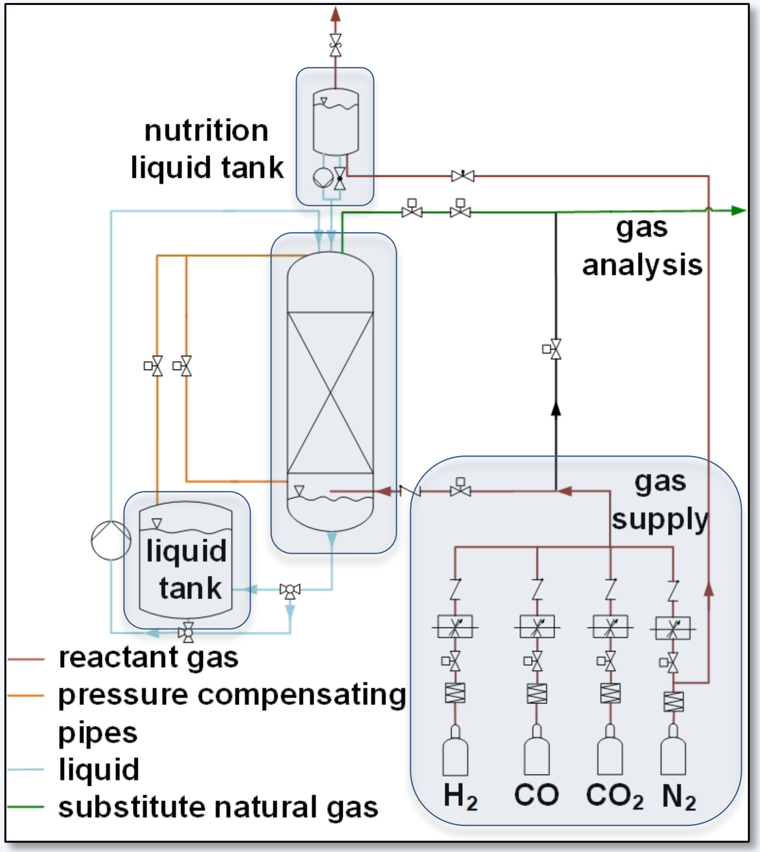


Trickle-bed reactor in Regensburg (project ORBIT)

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Two reactors – one concept

Lab-scale trickle-bed reactor in Nürnberg



Scheme of the trickle-bed reactor

Trickle-bed reactor in Regensburg (project ORBIT)

Two reactors – one concept

Parameter	Lab-scale reactor (experimental evaluation)	Prototype reactor (Orbit)
Reactor volume	5 L	50 L
Pressure of the reactor Pressure of the gas grid	1 – 2.5 bar(a) -	14-15 bar(a) 12.5 bar(a)
Temperature	ca. 65°C	ca. 65°C
MPR_R	$1.4\ \frac{Nm^3{}_{CH4}}{m^3{}_R \cdot h}$	$2\ \frac{Nm^3{}_{CH4}}{m^3{}_R \cdot h}$ *
Educts feed gas	0.02 $Nm^3/h\ H_2$ 0.005 $Nm^3/h\ CO_2$	0.26 $Nm^3/h\ H_2$ 0.063 $Nm^3/h\ CO_2$
Maximum methane concentration	98 vol.-%	> 95 vol.-% *
Power (methane)	70 W	1000 W *
Recirculation rate	0.3 – 3.5 L min^{-1}	1 - 10 L min^{-1}
Random packing	Bioflow 9 (RVT)	variable
Microorganisms	pure culture & mixed culture	pure culture
Operation mode	continuous	continuous

* state of design

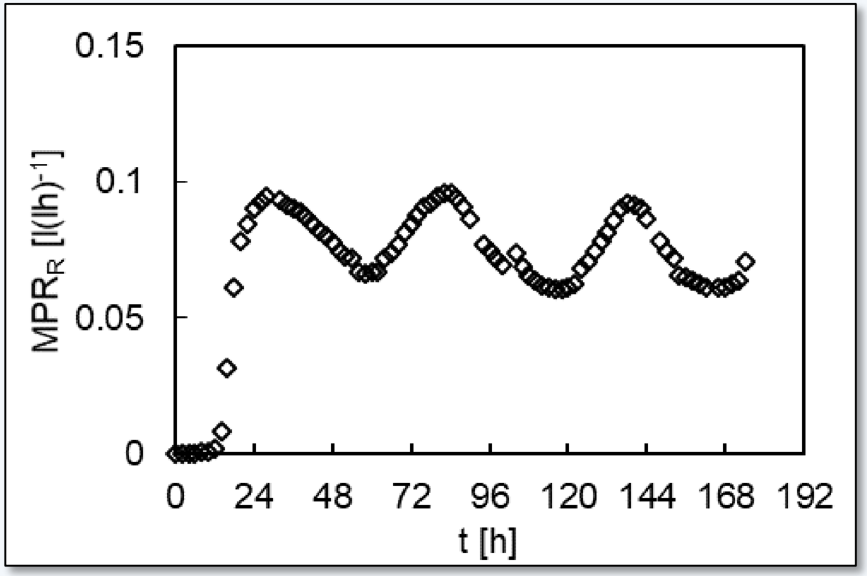
Trickle-bed reactor in Regensburg (ORBIT)

Lab-scale trickle-bed reactor in Nürnberg

Methanation with a pure culture

- Experiments with special archaea culture
- Fluctuating methane production observed without apparent cause
- Pure culture tends to self inhibition

→ Application of a mixed culture

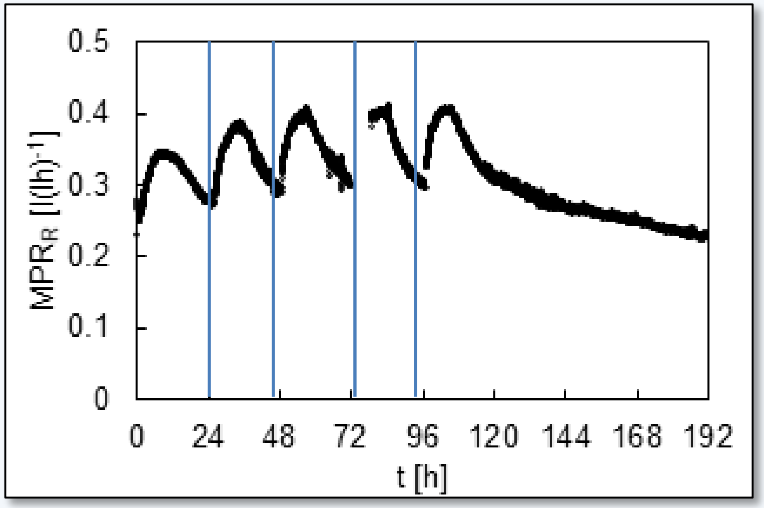


MPR fluctuation of the pure culture

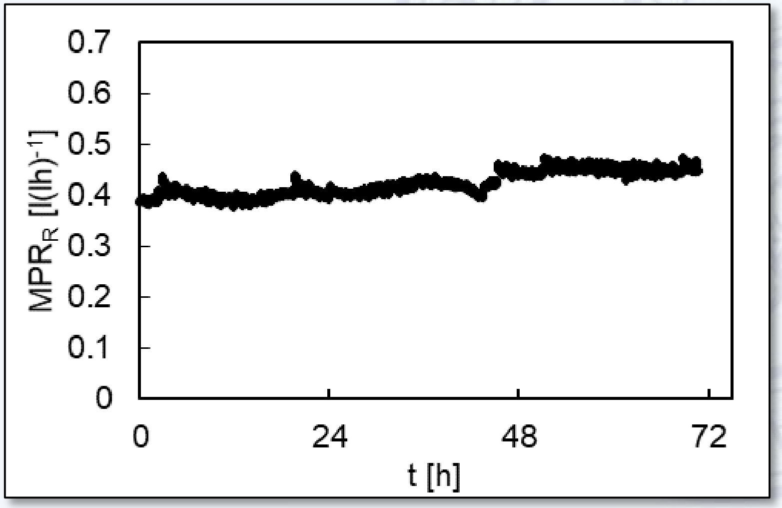
TBR at Nuremberg and the trickle zone

Mixed Culture

- Biogas digestate includes nutrients and several mircroorganisms
- Biogas digestate was added to the pure archaea culture
- The combination stables the system (most recent 10 months without changing the culture)
- Resulting in a higher performance level and less fluctuations
- Fluctuations caused by nutrient media feed (left figure)
- After adding the dosing pump the nutrient media was added continuous (right figure)



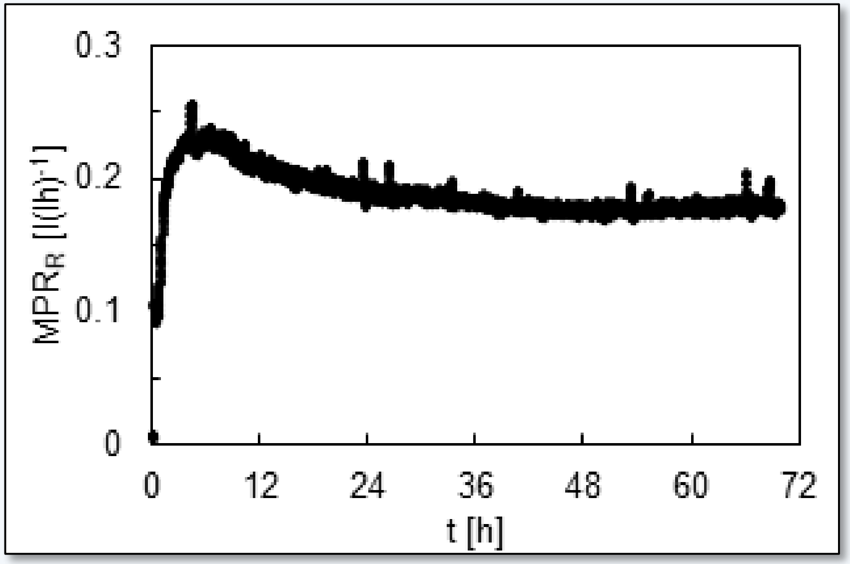
Mixed culture with daily adding of nutrient media



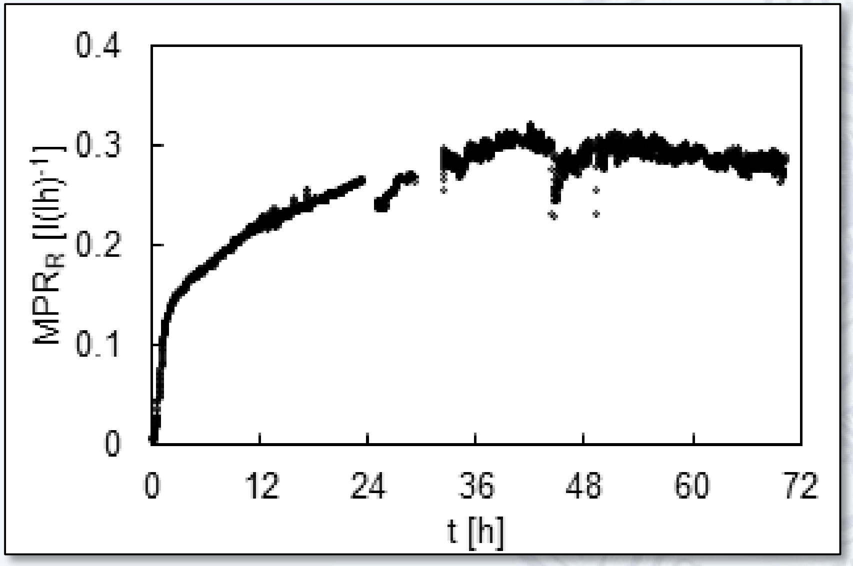
Mixed culture with continous adding of nutrient media

Mixed Culture

- Cold standby: no nutrient media, no heating, no pumping of liquid
- Shut down: empty reactor, archaea stored in the refrigerator
- For energy storage system the starting time after cold standby is important
- After cold stand by it took less then 5 hours (left)
- After shut down it was slower (right)



startup after cold standby



startup after shut down

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Mixed culture parameter studies

- The same culture stayed in the reactor for months
- Experiment started (t = 0) with the change of one parameter
- Before the experiment the reactor performed in a baseload to keep the microorganisms (MOs) alive and active (same initial situation)
- There are two options of experimental process:

Changing the parameters ongoing

	Pressure (bar_{abs})	Time (h)
Baseload	2	2
Parameter step 1	1.5	1
Parameter step 2	1.6	1
Parameter step 3	1.7	1
Parameter step 4	1.8	1
Parameter step 5	1.9	1
...	...	...
Drain of liquid		

+ quick
+ adaption of MOs to pressure
− not exactly the same initial situation

Going back to the baseload after each change

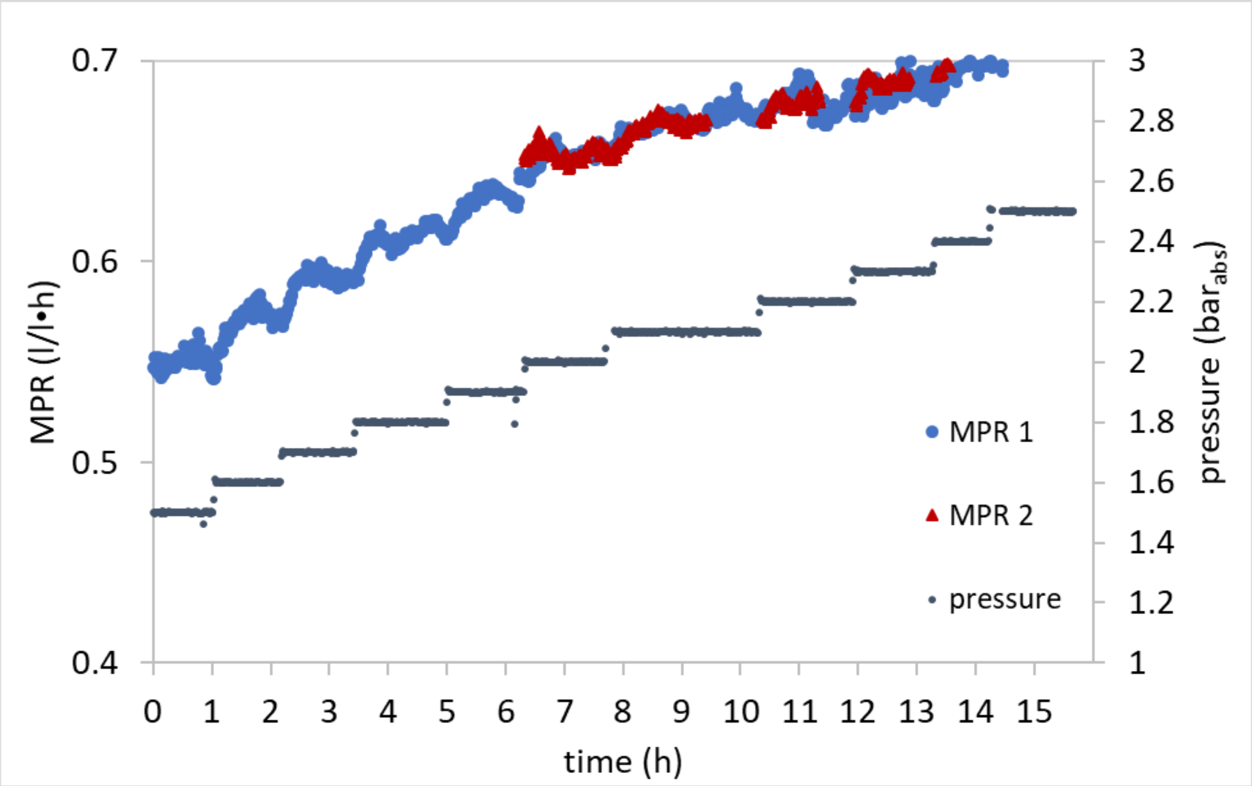
	Pressure (bar_{abs})	Time (h)
Baseload	2	2
Parameter step 1	1.5	5
Baseload	2	5
Drain of liquid		
Baseload	2	2
Parameter step 2	2.5	5
Baseload	2	5
Drain of liquid		

+ exactly the same initial situation at every parameter step
− slow

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Mixed Culture – pressure (1)

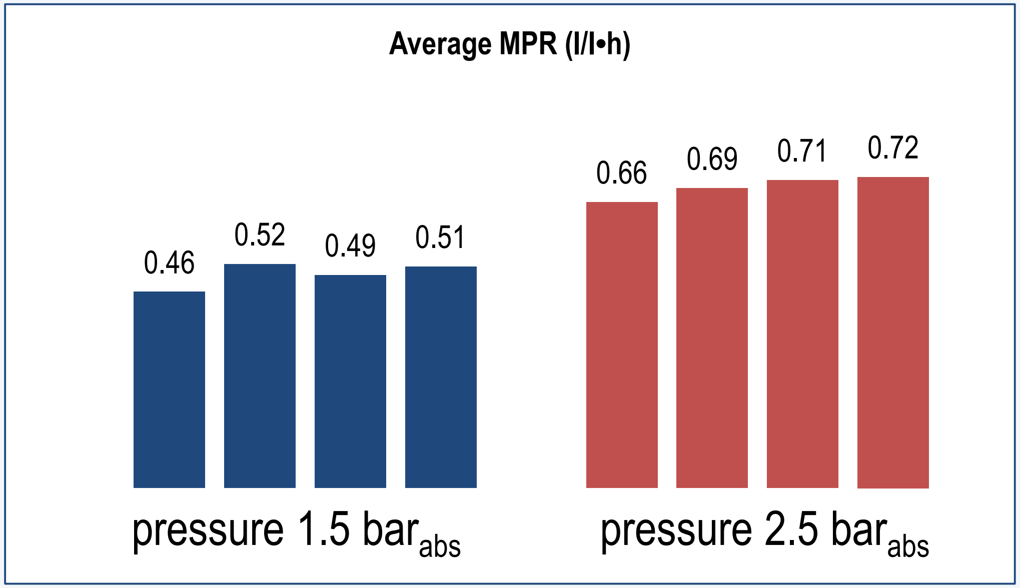
Pressure analyzing by increasing the pressure about every hour



- Increasing the pressure led to a higher MPR (better mass transfer)
- A validation one month later (red) showed quite similar results

Mixed Culture – pressure (2)

Pressure analyzing by going back to the baseload after each change



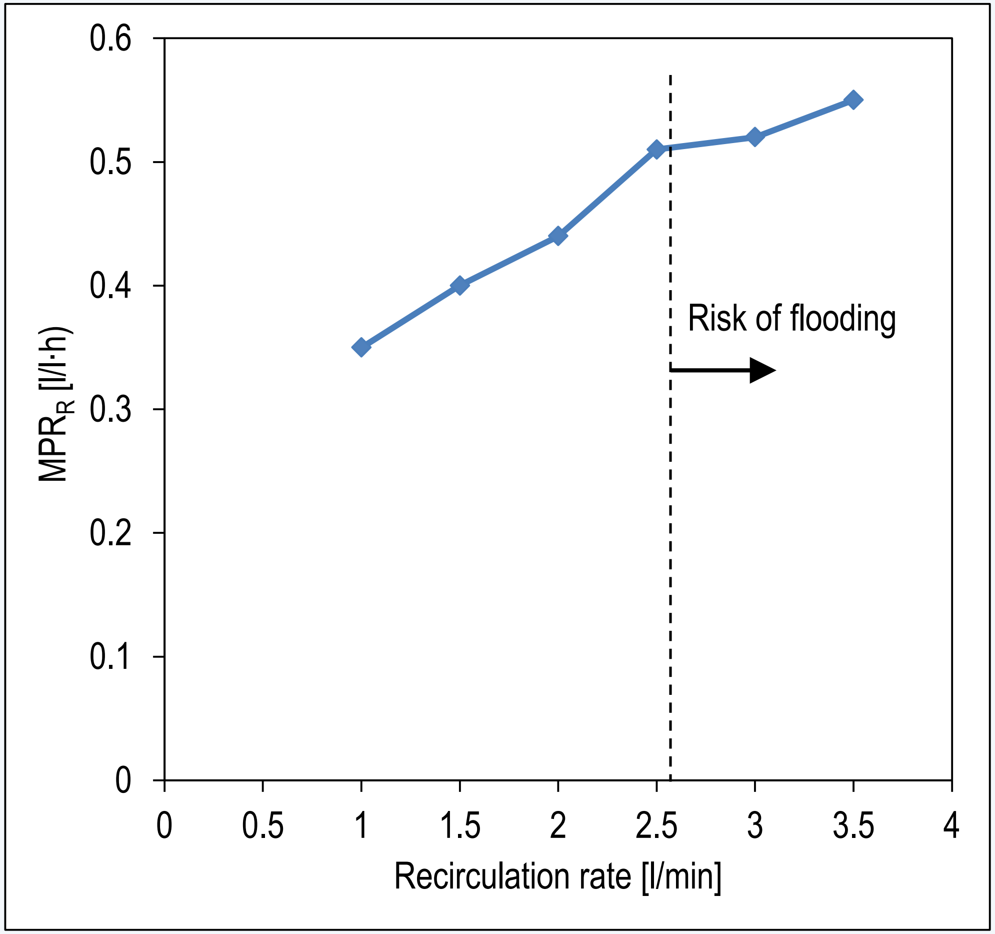
4 times:

	Pressure (bar_{abs})	Time (h)
Baseload	2	2
Parameter step 1	1.5	5
Baseload	2	5
Drain of nutrition		
Baseload	2	2
Parameter step 2	2.5	5
Baseload	2	5
Drain of nutrition		

- Similar results as before
- Increasing of the pressure resulted in a higher MPR (better mass transfer)
- Still a variation “form of the day”

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Mixed culture – recirculation rate



trickle zone of the lab scale reactor

- A higher recirculation rate leads to a higher hold-up and more turbulence
- Better mass transfer with higher recirculation rate ⇨ higher MPR
- Above 2.5 l/min: unstable system because of flooding

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Standardization

- Increasing number of scientific articles desribe the technology
- Difficult to compare because of no coherent nomenclature
- Approach to standardization for a better comparibility of the different systems was made [1]
- Work on VDI Guideline 4635 PtX in progress

Normalized to liquid volume

$$MPR_L = \frac{\dot{V}_{CH_4,out}}{V_L} \left[\frac{m^3}{day \cdot m^3}\right] = 120 \frac{m^3}{day \cdot m^3} = 5 \frac{m^3}{h \cdot m^3}$$

Normalized on the trickle bed zone

$$MPR_{TB} = \frac{\dot{V}_{CH_4,out}}{V_{TB}} \left[\frac{m^3}{day \cdot m^3}\right] = 240 \frac{m^3}{day \cdot m^3} = 10 \frac{m^3}{h \cdot m^3}$$

Suggested definition

$$MPR_R = \frac{\dot{V}_{CH_4,out} - \dot{V}_{CH_4,in}}{V_R} \left[\frac{m^3}{h \cdot m^3}\right] = 1.4 \frac{m^3}{h \cdot m^3}$$

[1] Thema, M.; Weidlich, T.; Hörl, M.; Bellack, A.; Mörs, F.; Hackl, F.; Kohlmayer, M.; Gleich, J.; Stabenau, C.; Trabold, T.; Neubert, M.; Ortloff, F.; Brotsack, R.; Schmack, D.; Huber, H.; Hafenbradl, D.; Karl, J.; Sterner, M. Biological CO_2-Methanation: An Approach to Standardization. Energies 2019, 12, 1670.

FRIEDRICH-ALEXANDER UNIVERSITÄT ERLANGEN-NÜRNBERG
TECHNISCHE FAKULTÄT

Summary

1. Biological Methanation
- One option for energy storage by production of renewable methane (SNG)
- TBR is a promising reactor system for the biological methanation

2. Lab-scale TBR
- Mixed culture shows better results
- High pressure is advantageous for biological methanation
- Recirculation rate and nutried media feed have direct influence on the performance
- Lab-scale reactor systems work well

3. ORBIT-reactor
- Upscale of the lab-scale TBR
- Experiments just started with pure culture at OTH and University Regensburg

4. Outlook
- Optimizing the amount of nutrition for the lab-scale reactor
- Experimental analysis of the ORBIT-reactor
- Feed the gas grid with continuously produced SNG from the ORBIT-reactor