

Power-to-Gas: CO₂ Methanation Concepts for SNG Production at the Engler-Bunte-Institut

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Power-to-Gas approaches comprise different activities to store electric power in form of gaseous energy carriers like hydrogen or methane. The synthesis of SNG (substitute natural gas) and its injection into the natural gas grid allows the utilization of the well-established infrastructure for natural gas storage, distribution and utilization without the need for further changes to the energy system. At the Engler-Bunte-Institut research focuses on catalytic methanation in catalytically coated metallic honeycomb reactors and in slurry bubble column reactors with the aim of evaluating alternative reactor concepts offering optimized heat transfer characteristics as well as maximizing the possibility of dynamic operation. Both concepts are attractive for small to medium scale power-to-gas applications. Hence, a scale-up was performed for both reactor concepts with the aim of implementation on commercial scale.

Keywords: CO₂-consuming processes, Methanation, Power-to-Gas, Reactor concepts, Renewable energy

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1 Power-to-Gas

The increasing share of fluctuating renewable electric energy in future energy systems causes an increasing dynamic behavior of the energy system depending on the availability of wind and sunlight. In this context, storage of electric energy in form of chemical energy carriers is a viable solution to match production with demand. Besides hydrogen, methane qualifies as a gaseous energy carrier because of its versatility and established process technologies. CH₄ is an energy carrier of growing importance in industry, energy and transportation sectors. Furthermore, the existing natural gas infrastructure provides huge storage capacities and a well-established network for distribution and utilization.

There are many possible routes to store electric energy in form of gaseous energy carriers using Power-to-Gas (PtG) processes [1, 2]. The basic approach is illustrated in Fig. 1: mobility sector, industry and power plants routinely access both the power network and the gas grid to cover their energy demand. However, the gas grid can also be used for storing energy in times of surplus. PtG approaches establish a link between the electric energy network with limited storage capacities and the natural gas network with the aim of overcoming the lack of mid- to long-term storage capacities of the first.

Methane production from electric energy usually needs two steps. In a first step the reaction educts are generated. Typically, hydrogen is produced via water electrolysis using electric energy. Carbon dioxide can be absorbed from either unavoidable exhaust gases or ambient air. The gas mixture

of H₂ and CO₂ is subsequently transformed into methane in the methanation reactor. Thema et al. [3] recently published an overview of currently operating PtG plants (for H₂ as well as for SNG production) showing a high research interest and exponential global trend to increase installed PtG power as a key technology for future energy systems.

1.1 Catalytic CO₂ Methanation – State of the Art

The catalytic methanation of CO₂ (Eq. (1)) is a highly exothermic reaction firstly described by Sabatier [4] in 1902, which is recently gaining attention with regards to CO₂-consuming processes producing energy carriers. An alternative reaction route not discussed in this paper is the use of CO as carbon source for methanation [5, 6], which is relevant especially for BtG (Biomass-to-Gas) routes based on biomass gasification processes with or without PtG options.

Due to the reaction equilibrium, high methane yields are obtained for CO₂ methanation at low temperature and high pressure [1].

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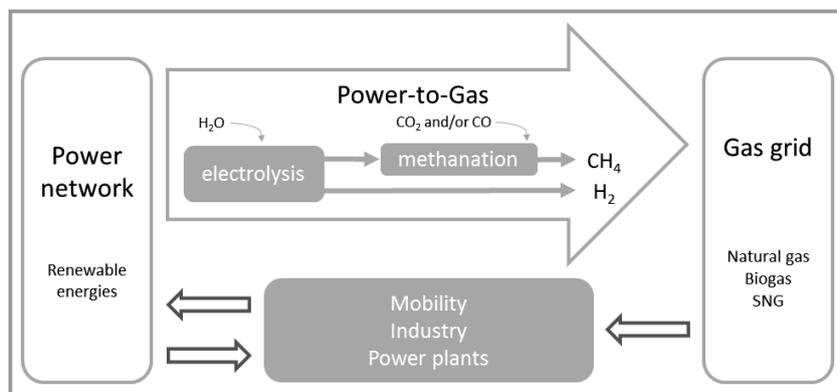


Figure 1. Role of PtG processes in the energy system.



In technical applications temperatures above 200 °C are used with pressures ranging from 5 to 100 bar, depending on the specific application. There are various catalysts available to accelerate the methanation reaction; in commercial applications nickel-based systems are dominant because of their high catalytic activity and low price.

The vast majority of reactor concepts discussed for catalytic CO₂ methanation are fixed-bed reactors, structured reactors, fluidized-bed reactors and slurry bubble column reactors [1,6]. Fixed-bed reactors present the state of the art for large-scale methanation applications. In adiabatic operation mode multiple fixed-bed reactors are used and reaction heat is removed by intercooling between the reactors and/or staged addition of feed gas. This operation mode causes high heat stress on catalyst and reactor material, typical reactor temperatures range between 250 and 700 °C.

Using cooled fixed-bed reactors usually leads to significantly lower temperatures in the reactor. Hence, the overall process is simplified and less reactor steps are needed. However, one of the main drawbacks of cooled fixed-bed reactors is the formation of temperature hotspots due to the restricted heat transfer through the catalyst bed and reactor walls to the cooling medium. In tube bundle reactors limited heat transfer restricts the individual tube diameter and consequently more reactor tubes are needed.

Efficient heat management is vital for all reactor concepts to reach the usual requirements on process efficiency ($\eta > 75\%$), load flexibility and methane concentrations ($y_{\text{CH}_4} > 95\%$) in catalytic methanation. There are several reactor concepts available that show improved heat transfer to fulfill these requirements.

In fluidized-bed reactors gas flow introduced into the reactor fluidizes the catalyst particles and causes a high degree of mixing. This effect and the high heat capacity of the catalyst particles allows for nearly isothermal operation and the avoidance of temperature hotspots. Regarding process design, a single reactor is most often sufficient to

reach the desired conversion for injection into the natural gas grid. However, attrition processes reduce catalyst particle size and particles are involuntarily discharged from the reactor.

In structured reactors often metallic structures are part of the reactor interior or are used as catalyst carrier significantly enhancing the heat transfer from the catalyst to the cooling medium on the outer shell of the reactor tube. As these structures show significantly higher radial heat conductivity compared to fixed-bed reactors, lower hotspot temperatures are achieved. Structured reactors are characterized by high GHSV,

main drawback is the complex procedure to immobilize the catalyst on the structures.

Slurry bubble column reactors include a liquid phase in the reactor, directly present on the catalyst surface where the heat of reaction is produced. Due to its high heat capacity and heat conductivity this liquid facilitates heat management. The reactor design allows for highly dynamic operation modes due to its thermal indolence dampening hotspots or coldspots caused by times of high or low load. However, slurry bubble column reactors show low values for GHSV due to additional mass transfer resistances in the liquid phase that are not present in most other methanation reactors.

2 Reactor Concepts Developed at the Engler-Bunte-Institut

Catalytic methanation has been extensively studied in the context of PtG applications [1,2]. Generally, the reactor types used can be divided into two-phase and three-phase systems. In commercial applications mostly adiabatic and polytropic fixed-bed reactors are applied, which therefore form the state of the art for catalytic methanation. Other two-phase reactor concepts are structured reactors such as microchannel or honeycomb reactors. Fluidized-bed reactors can be operated with two or three phases, whereas slurry reactors are operated with three phases exclusively. Structured reactors, fluidized-bed reactors and slurry reactors are subject to ongoing research [1,7,8].

At the Engler-Bunte-Institut (EBI) two innovative methanation reactor concepts were developed and are subject to ongoing research activities with focus on dynamic operation and optimized heat transfer characteristics for PtG applications: a two-phase honeycomb reactor (HCR) and a three-phase slurry bubble column reactor (SBCR).

2.1 Reactors

The honeycomb reactor discussed in this paper is a two-phase structured fixed-bed reactor that contains coated catalyst carriers made of stainless steel. It is designed as a multitube reactor, in which the metallic catalyst carriers are placed in parallel tubes. A schematic drawing of the honeycomb-like bodies is shown in Fig. 2a. They are made of a combination of corrugated and plane metal sheets, which are jointly coiled up. The layers are form-fit pressed in a cladding tube. Typically, honeycomb structures are characterized by the number of parallel channels per square inch (CPSI). For the discussed application, honeycomb struc-

tures of 100–600 CPSI are used corresponding to channel diameters of 0.1–2.8 mm. This fits the typical dimensions of microreactor channels, but the honeycombs are per definition not microreactors [9, 10]. The feed gas flow enters the catalytically coated channels and if the reactor temperature is high enough (above 200 °C) the catalytic methanation reaction starts. CO₂ and H₂ are converted to CH₄ in the porous catalyst layer and reaction heat is released mostly at the channel inlets. As a result, characteristic temperature profiles with a defined peak at the inlet and an outlet temperature close to the cooling medium temperature ($\Delta T < 10$ K) are obtained [11, 12].

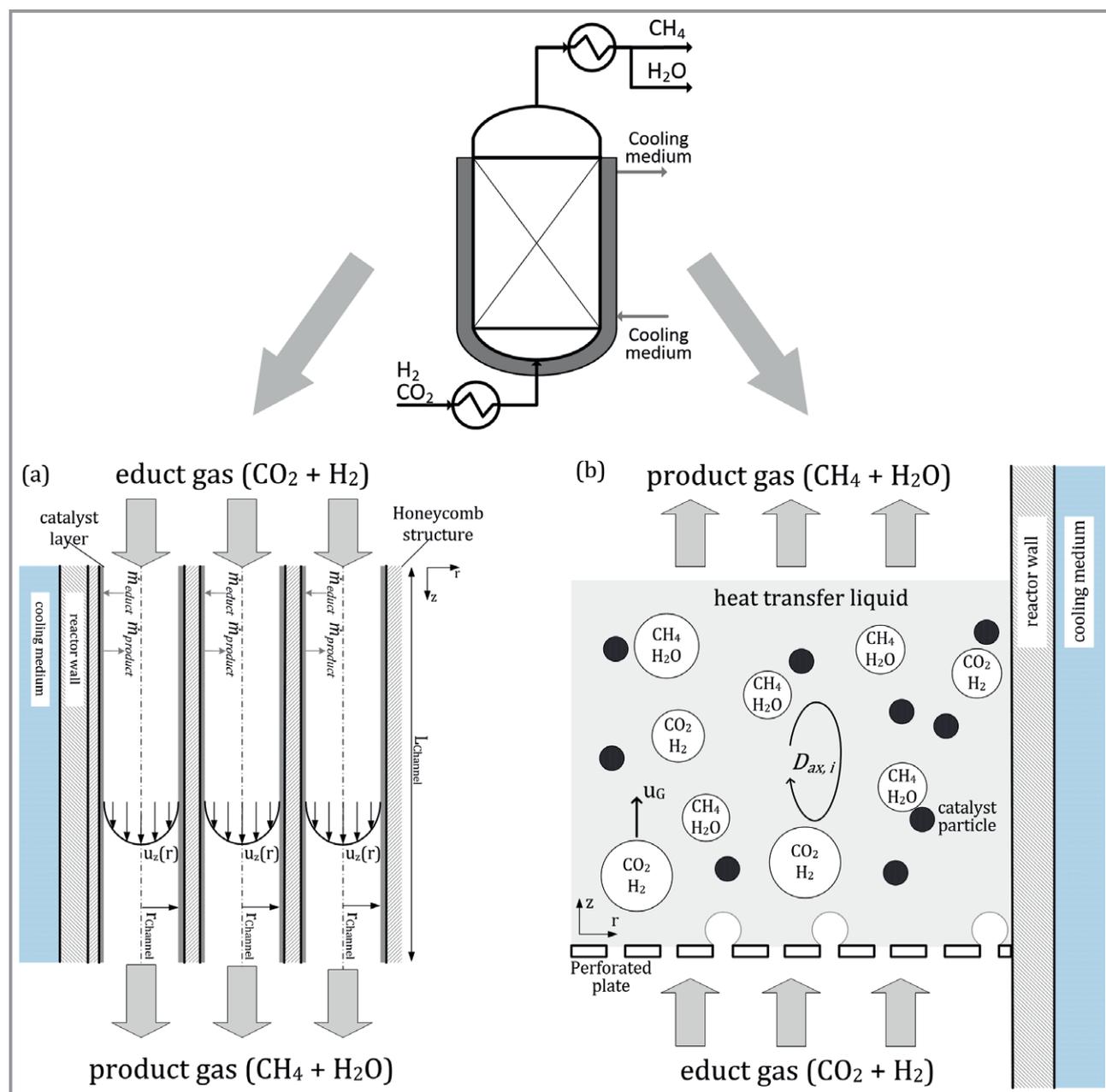


Figure 2. Schematic drawing of the honeycomb reactor (a) and the three-phase slurry bubble column reactor (b).

For typical conditions of the HCR the Reynolds number in the channels is in the range of 10–500. Based on a channel diameter of 1 mm and low Reynolds numbers the channels can be described as ideal plug flow reactor ($Bo > 100$) with nearly complete cross mixing in radial direction [10]. This illustrates the intensification of transfer phenomena in the channels.

Reaction conditions are set to allow for a maximum hotspot temperature of 550 °C in the reactor. To achieve this, the temperature inside the reactor is measured using temperature sensors and the temperature of the cooling medium is adjusted accordingly. For the described reactor system, the cooling medium temperature is set to 220 °C. Pressures between 6 and 21 bar (absolute) are chosen since higher pressures influence the thermodynamic equilibrium only to a small extent. The catalyst coating on the metallic honeycombs is a commercially available product.

The slurry bubble column reactor (SBCR) presented in this paper has three distinctive phases: the commercially available solid catalyst (particle size of 50–100 μm) is suspended in a heat transfer liquid and is fluidized by the educt gases. A schematic drawing of all involved components is shown in Fig. 2b.

Heat management in the SBCR is implemented by the heat transfer fluid, which shows high heat capacity and, thus, enables efficient heat transfer from the catalyst particles to the cooling medium in the cooling jacket. The educt gases entering the bubble column through a perforated plate at the bottom enable back mixing resulting in isothermal operation [13]. Requirements on the heat transfer fluid are high educt and product gas solubility, high heat capacity and high thermal stability.

Reaction conditions are set to a temperature of approx. 320 °C and pressure of 20 bar (absolute). Kinetically a high temperature is favorable, but its maximum is limited by the thermal stability of the liquid phase present in the reactor and the thermodynamic equilibrium presetting the maximum of conversion and corresponding methane yield.

2.2 Reaction Kinetics

For both reactor systems research at the EBI focuses on reaction kinetics as well as heat and mass transfer phenomena. Furthermore, hydrodynamic behavior in the SBCR is investigated [14]. Götz [15] identified dibenzyl toluene (DBT) as suitable fluid, which is recently gaining attention due to its application as liquid organic hydrogen carrier [16]. Lefebvre [17,18] determined reaction kinetics in the three-phase reactor. Regarding hydrodynamics Götz [19] developed a novel gas holdup correlation for the SBCR that can be used to describe the homogeneous flow regime.

Schollenberger [11,12] determined reaction kinetics for the HCR. Recent research focusses on heat transfer phenomena and experimental determination of the effective axial and radial heat conductivity in commercial honeycombs.

2.3 Mass Transfer

An overview of mass transfer phenomena is presented in Fig. 3. For both systems reaction takes place in the catalyst particle or in the catalyst layer, so transfer of gaseous educts and products to and from the catalyst has to be considered.

2.3.1 Honeycomb Reactor

In the honeycomb reactor, educts are transferred from the bulk phase to the catalyst layer (thickness of about 100 μm) where the reaction takes place resulting in a concentration gradient in the gas phase. This process can be described by mass transfer coefficients β_i (1). From the outer surface of the catalyst layer, the gas molecules diffuse to the active sites in the catalyst. This process can be described using molecular diffusion coefficients D_i (2).

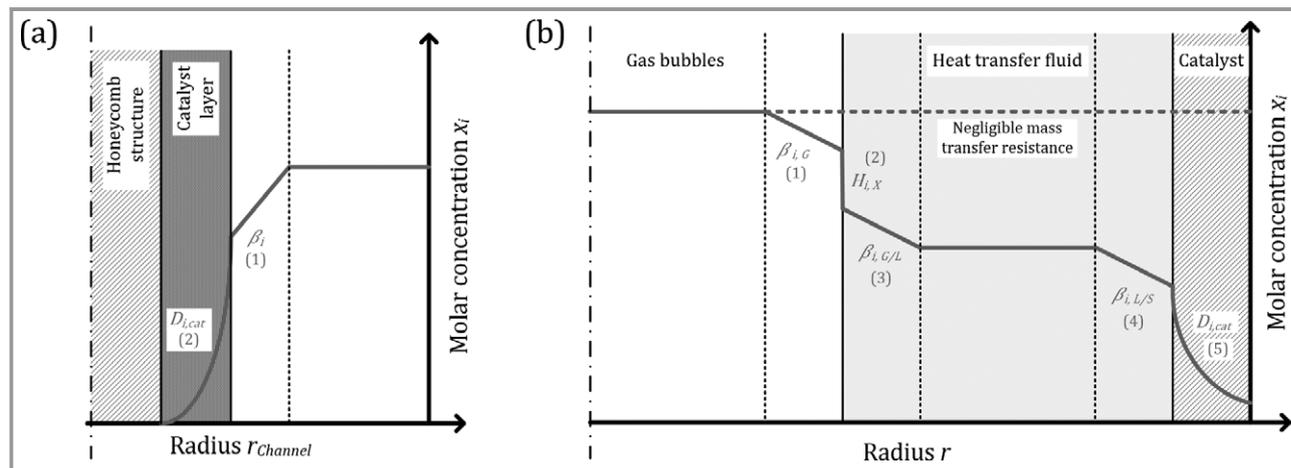


Figure 3. Mass transfer phenomena in the honeycomb reactor (a) and the three-phase reaction system (b) based on theoretical considerations.

2.3.2 Slurry Bubble Column Reactor

Introducing a heat transfer liquid in the reactor offers benefits regarding heat management and dynamic operation, but it also introduces additional mass transfer resistances. A schematic concentration profile is presented in Fig. 3b.

Transfer of the educt gas components from the bulk phase to the gas-liquid layer is described by the mass transfer coefficients $\beta_{i,G}$ (1). The Henry coefficients $H_{i,X}$ of the components describe solubility in DBT (2); this is a rate-limiting step especially for hydrogen as it shows low solubility in most liquids. Solved in the liquid phase, mass transfer from the gas-liquid layer (3) is considered using the mass transfer coefficients $\beta_{i,G/L}$. The bulk liquid phase can be assumed as ideally mixed.

The mass transfer resistance from the liquid phase to the solid catalyst particles is described using the mass transfer coefficients $\beta_{i,L/S}$ (4). At the catalyst particle gas molecules diffuse from the surface to the active catalytic sites in the pores of the particle (5). This process can be described using the diffusion coefficients $D_{i,cat}$.

2.3.3 Comparison

Mass transfer in the SBCR plays a crucial role determining the effective reaction rate. In the HCR, mass transfer resistances are minimized. Since the channel diameter is in the range of 1 mm, radial diffusion plays a significant role and convective transfer is not dominant. The intensification of transfer phenomena leads to high conversion rates and high specific methane yields compared to established fixed-bed reactors.

In the SBCR, mass transfer resistances are limiting the effective reaction rate leading to low GHSV values compared to two-phase reactor systems. To reach high conversion, larger reactor volumes are needed, which is a slight drawback compared to established fixed-bed reactor systems.

2.4 Heat Transfer

Both reactor concepts introduced show advanced heat management characteristics compared to conventional methanation reactor concepts. An overview of temperature profiles in the HCR and the SBCR is shown in Fig. 4.

2.4.1 Honeycomb Reactor

In general, the approaches to describe heat transfer in the honeycomb channels are analogous to mass transfer mechanisms. A characteristic radial temperature profile is shown in Fig. 4a. Reaction heat is released in the catalyst layer and is transferred to the cooling medium by conduction through the catalyst carrier. Depending on the feed volume flow and the outer heat transfer coefficient a more or less axially and radially pronounced temperature gradient is obtained. Consequently, the HCR is operated in polytropic mode. Radial conduction through the catalyst carrier dominates heat transfer, whereas convective heat transfer in the channels is almost negligible. Due to radial heat transfer resistances in the catalyst carrier higher temperatures are obtained with increasing throughput as more reaction heat is produced. Temperature restrictions of the catalyst (hotspot temperature $T_{HS} < 550^\circ\text{C}$) are limiting the throughput of the honeycomb reactor.

To describe heat transfer in metallic honeycombs established approaches in the literature are based on monoliths not considering additional resistances between the coiled metal layers [11,20]. For the metallic honeycombs heat transfer is described by a model developed at the EBI for cubic cells based on the interconnection of the heat transfer resistances.

2.4.2 Slurry Bubble Column Reactor

In the SBCR reaction heat is removed using a heat transfer liquid. A temperature profile of the reaction zone is pre-

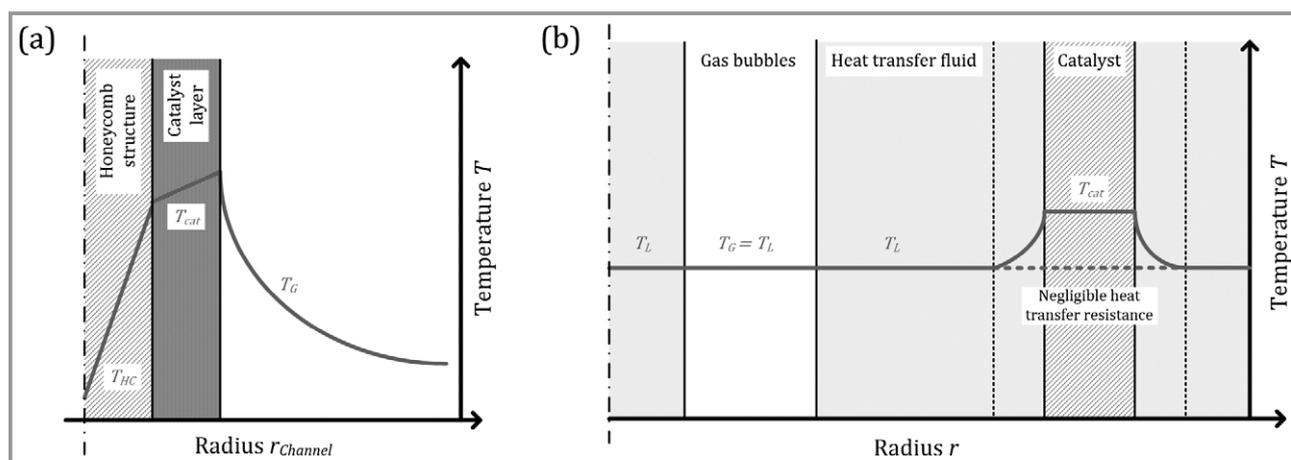


Figure 4. Heat transfer phenomena in the honeycomb reactor (a) and the three-phase reaction system (b) based on theoretical considerations.

sented in Fig. 4b. The mixing effect in the bulk liquid phase causes a constant temperature in the fluid phases. At the catalyst particle, temperature is higher because of the reaction heat released. However, the temperature increase is not critical since convection is high. The combination of high heat capacity of the liquid phase and high degree of mixing enables isothermal reactor operation.

The liquid phase also enables dynamic operation of the reactor, because of its heat management capabilities. Due to inevitable fluctuations in renewable energy supply, educt gas load of the methanation reactor can be fluctuating. The high heat capacity of the system prevents the reactor from cooling down or heating up (too) fast, so periods of high or low gas load can be buffered and the necessary temperatures of operation are assured. The transient operation of the SBCR has been investigated by Lefebvre et al. [21].

2.4.3 Comparison

In the honeycomb reactor the throughput is limited by heat transfer. However, all relevant steps have been identified and can be quantified for reactor scale-up. The slurry bubble column reactor is characterized by isothermal operation and heat transfer is technically not an issue. This presents a major advantage compared to two-phase reactors, in which characteristic temperature hotspots are unavoidable. Hence, no limiting heat transfer resistances are present enabling dynamic operation of the reactor system.

3 Scale-Up

The two reactor systems are experimentally investigated at various scales with the aim of obtaining reliable data for scale-up to commercial applications. Lab-scale plants at the EBI provide first insights into the systems and are used in ongoing research projects for academic research accompanying the experiments at pilot- and demo-scale.

Fig. 5 shows an overview of the scale-up activities performed for the two presented reactor concepts. For methanation in the HCR a lab-scale plant was built in 2011 for basic research topics. Within the scope of the KIC DemoSNG project [22] a mobile demo plant was designed in 2014 with an installed methanation output of 60 kW. In the course of the project the demo plant was transported to Sweden to the project partner CORTUS SE and operated with a syngas from a biomass gasification.

During the operation of the plant a total carbon conversion (CO and CO₂) of more than 99 % was achieved.

In 2017 the up to now biggest plant using HCR technology was built in Falkenhagen. This PtG plant with an installed electric power of 1 MW and the accompanying research activities are part of the EU-funded project STORE&GO (see Fig. 6a). Commissioning of the plant was completed in 2018 and the plant is up to now more than 1200 h on stream. The produced SNG is injected in a natural gas transportation grid, which means that all relevant German requirements ($y_{H_2} < 2 \text{ vol } \%$ and $y_{CH_4} > 95 \text{ vol } \%$) fixed in [23, 24] are met. The pilot plant demonstrates the fully integrated operation of a PtG plant with high efficiency [25]. Ongoing activities at the EBI focus on refining the technology in cooperation with industrial partners for a plant size of approx. 20 MW.

For methanation in the SBCR a first lab-scale plant was built in 2009 after preceding bench-scale tests. The plant is equipped with both a SBCR and an autoclave reactor to investigate different aspects of the reaction system.

In 2019, a unique SBCR plant was commissioned as part of the KIT Energy Lab 2.0 (see Fig. 6b) with a CH₄ output equivalent to 100 kW [26]. The reactor has a diameter of about 250 mm and a total height of roughly 3 m. The dimensions allow a realistic evaluation of the reaction system for technical application as influences of the reactor diameter on gas holdup can be neglected for diameters above 150 mm [27]. Basic research activities will be performed to give reliable information on long-term operation under technical conditions, e.g., with synthesis gas from the adjacent bioliq[®] gasification plant or with highly dynamic

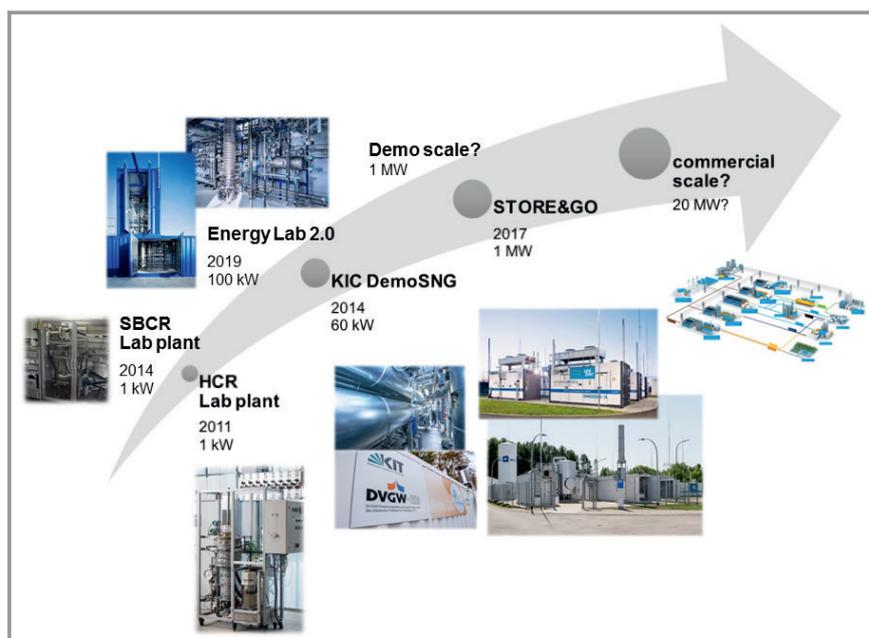


Figure 5. Scale-up strategy for methanation reactor concepts at the Engler-Bunte-Institut.



Figure 6. HCR plant with 1 MW installed electric power in Falkenhagen (a) and SBCR plant with 100 kW CH₄ output in the Energy Lab 2.0 (b).

load changes that can be expected in a technical PtG process.

Commissioning of the pilot plant and first tests on dynamic methanation were completed successfully in June 2019. In August 2019, further tests have been performed on stationary and dynamic operation. Different educt gas ratios and gas loads were set to investigate influences on CO₂ conversion with the prospect of using the reactor in combination with a downstream liquefaction unit.

Further scale-up of HCR and SBCR designs will use different approaches. The honeycomb reactor consists of multiple pipes containing metallic honeycomb structures; scale-up is performed up to now by numbering-up. Recent research activities aim at alternative reactor designs like radial flow reactors as numbering-up is no longer beneficial for methanation reactors significantly above the 10-MW scale.

For the slurry bubble column reactor, hydrodynamics and its impact on reactor performance has to be observed very closely during the scale-up procedure. At pilot scale, hydrodynamic data should be acquired under process conditions as close as possible to those of the full-scale unit to avoid false estimation of important design parameters such as the gas holdup or gas velocities of regime transition.

4 Comparison and Outlook

Methane is a highly versatile chemical energy carrier, which offers huge storage capacities and a well-established network for distribution and utilization. Catalytic methanation requires high temperatures to obtain the reaction rates necessary for high product yields per pass. However, temperature is limited by thermodynamic equilibrium of the highly exothermic reaction. Suitable reactor concepts should address this issue and still allow for high throughput and methane yield.

Future methanation applications need plants of different size and with various abilities regarding (load) flexibility. Large-scale plants of more than 10 MW SNG output could be used to continuously convert a minimal fluctuating base load of electric energy and/or could be combined with, e.g.,

(biomass) gasification giving constant base load with PtG option in times of surplus electricity. Plants with a SNG output of less than 10 MW could help to stabilize decentralized PtG systems by offering a highly dynamic synthesis as storage technology [28].

The honeycomb methanation system shows good load flexibility and high specific CH₄ output, which is especially favorable in PtG plants of high base load. In comparison, the SBCR shows lower specific methane

production due to mass transfer limitations. However, the possibility to operate the SBCR highly dynamic presents a main advantage for its aimed application in PtG plants with low or even no base load.

For both reactor concepts scale-up from laboratory to pilot scale was successfully performed. Ongoing projects focus on system optimization and the transfer to commercial scale. A promising approach is the implementation of an HCR as a second reactor step after the SBCR to achieve high specific CH₄ output combined with highly dynamic operation.

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Symbols used

Bo	[-]	Bodenstein number
D_{ax}	[m ² s ⁻¹]	axial dispersion coefficient
D_i	[m ² s ⁻¹]	diffusion coefficient
$\Delta_R H$	[kJ mol ⁻¹]	heat of reaction
H_i	[bar]	Henry coefficients
L	[m]	length
\dot{m}	[kg s ⁻¹]	mass flow
T	[K]	temperature
ΔT	[K]	temperature difference
t_D	[s]	time constant of diffusion
u	[m s ⁻¹]	velocity
y	[-]	molar concentration

Greek letters

β_i	[-]	mass transfer coefficient
η	[-]	process efficiency
τ	[-]	time constant of convection/ residence time

Sub- and Superscripts

cat	catalyst
G	gaseous
L	liquid
S	solid

Abbreviations

BtG	Biomass-to-Gas
CPSI	channels per square inch
DBT	dibenzyl toluene
GHSV	gas hourly space velocity
HCR	honeycomb reactor
PtG	Power-to-Gas
SBCR	slurry bubble column reactor
SNG	substitute natural gas

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