Co-funded by the European Union's Horizon 2020 research and innovation programme
under Grant Agreement no. 691797

STORE&GO

Innovative large-scale energy storage technologies and power-to-gas concepts after optimisation

D5.9

Final report on evaluation of technologies and processes

Due Date	29 February 2020 (M48)
Deliverable Number	D5.9
WP Number	WP 5
Responsible	DVGW
Author(s)	Friedemann Mörs (DVGW), Schlautmann, Ruth (EBI), Jachin Gorre (HSR), Robin Leonhard (HSR),
Reviewer	Frank Graf (DVGW)
Status	Started / Draft / Consolidated / Review / Approved / Submitted / **Accepted by the EC** / Rework

Dissemination level

✘	**PU**	Public
	PP	Restricted to other programme participants (including the Commission Services)
	RE	Restricted to a group specified by the consortium (including the Commission Services)
	CO	Confidential, only for members of the consortium (including the Commission Services)

Document history

Version	Date	Author	Description
0.1	2019-11-18	Mörs/Schlautmann	First draft to discuss table of content
0.2	2020-03-06	Mörs/Schlautmann	First draft to discuss
0.8	2020-03-27	Mörs/Schlautmann	Feedback from partners integrated
1.0	2020-04-09	Mörs/Schlautmann	Feedback from reviewer integrated
2.0	2020-05-15	Mörs/Schlautmann	Working in feedback from EU officer

Table of Contents

Executive Summary

Within the STORE&GO project, three innovative PtG plants had been built, tested and operated in three different countries. In this report, the three demo sites are evaluated regarding technical and economic aspects. Furthermore, optimization potential of the demo sites is pointed out and future development potential is shown.

At the demo site in Falkenhagen, Germany, an innovative catalytic methanation process based on metallic honeycomb structures was tested. In order to improve the heat transfer, the main reactor contains multi-tube channels, honeycombs that are coated with a catalyst. Hydrogen and carbon dioxide which had not reacted to methane in the honeycomb reactor was converted to methane in the subsequent polishing reactor. Thus, the requirements for injection into the gas grid were reached. In Solothurn, Switzerland, a biological stirred bubble column methanation reactor was built at an already existing 'Hybridwerk'. Here, the feed gas was converted to SNG via a biocatalyst (Archaea). The required hydrogen was produced via an on-site proton exchange membrane (PEM) electrolyser. A nearby waste water treatment plant served as CO_2 source. At both plants, Falkenhagen and Solothurn, the produced SNG was injected to the gas grid, whereas at the third demo site in Troia, Italy, the produced methane was liquefied to LNG. At the Troia demo site, the interaction of the liquefaction unit with another two innovative technologies was tested: The required CO_2 was captured from air using direct air capture from Climeworks, and for methanation an innovative milli-structured reactor was used. Due to the liquefaction of the methane and the direct air capture, the process is independent of the location, since no gas grid or CO_2 source nearby is required.

The technical and economic evaluation of the demo sites is based on different methods and definitions described in chapter 3. In a first step, the system boundaries needed to be carefully chosen for the technical evaluation. Since all demo sites include different PtG concepts, careful consideration must be given to how the technologies are evaluated, as for example all demo sites contain different CO_2 sources. For the energetic evaluation, the demo sites were assessed regarding defined performance indicators (PIs), as the gas hourly space velocity (GHSV), the conversion, and different efficiencies. In addition, the flexibility and the dynamics of the demo site should be investigated. Therefore, the load change rate was calculated. Beside the evaluation of the measurement data, ASPEN simulations of the demo sites had also been produced and compared to the measured PIs. In a second step, the demo sites were evaluated according to economic aspects. For this, the capital expenditure (CAPEX) was determined using the add-on factor method. Afterwards, the production costs were calculated via the annuity method.

One of the most important performance indicators is the efficiency of the overall power-to-gas process chain (see example in Figure 1-1). The overall PtG efficiency takes into account the heat usage and the energy demands for the following process steps: CO_2 conditioning, H_2 production, methanation unit, and injection into a high pressure gas grid or liquefaction. One of the main goals of the STORE&GO project was to demonstrate an overall PtG efficiency of higher than 75 %. On the one hand the efficiency of the individual process steps must be sufficient. On the other hand, the efficiency is mainly dependent on the possibility of heat usage, which requires a suitable location for the plant. A further goal of the project was to reach a high methane content y_{CH4} > 90 % in the product gas. Two more goals were to prove a load flexibility in the range of 20 – 100 % load and a load change rate of at least 5%/min.

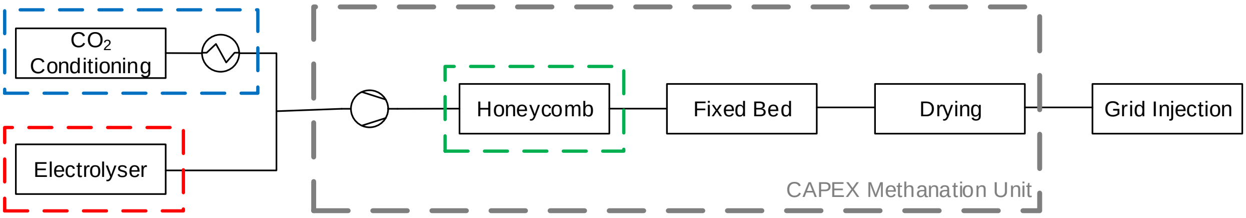

Figure 1-1: Block Flow chart of a power-to-gas process chain (Falkenhagen).

Table 1-1 gives an overview of the technical evaluation of the demo sites. For the Falkenhagen plant, an overall PtG efficiency of 53 % is reached based on the measurement results. The methanation unit reaches an overall methanation efficiency of 85 % (including heat usage and electricity demand). The relatively low overall PtG efficiency arises from the poor efficiency of the existing alkaline electrolyser (AEL). Due to this fact, the biggest optimization potential in Falkenhagen is to use a state-of-the-art (SoA) electrolyser (optimized PtG efficiency of 69 %). The core technology in Falkenhagen (methanation unit) was capable of producing high quality SNG (volumetric methane content y_{CH4} > 99 vol.-%) for a wide variation of the load (40 – 100 %). Also during load changes, the SNG quality always fulfilled the limits for injection of the gas.

Table 1-1: Overview of methane fraction after the methanation, overall PtG efficiency, and optimized efficiency if all optimization potentials are considered

	Project goals	Falkenhagen	Solothurn	Troia
Methane content of the product gas	y_{CH4} > 90 vol.-%	> 99 vol.-%	> 99 vol.-%	96 vol.-%[1]
Overall PtG efficiency based on measurements	$\eta_{PtG,HHV,ov}$ > 75 %	53 %	76 %	29 %
Optimized overall PtG efficiency		69 %	89 %	46 %

In Solothurn, the methanation unit also reached a product gas quality of more than 99 vol.-% of methane. During the operation of the plant, the biocatalyst was slowly adapted to higher loads: At the end of the project, the plant was capable of operation at nearly 100 % load. The overall methanation efficiency (without heat usage) in Solothurn is 73 %. The overall PtG efficiency is 76 %, which includes the usage of the low temperature (T_{use} < 60 °C) waste heat from the electrolysis. In Solothurn the nearby 'Hybridwerk' was able to use this waste heat at relatively low temperature, since the heat was boosted via a heat pump for district heating. The energy demand for the CO_2 source was neglected, since the CO_2 stream to the plant was a waste product. Further optimization potentials is the integration of the waste heat from the methanation reactor in the 'Hybridwerk', which was planned in the project but not tested. Another potential is the reduction of the electrical energy demand of the methanation unit. If both potentials would be realised, an overall PtG efficiency of 89 % could be reached. This shows that the efficiency of the overall a PtG process chain is very dependent on the local conditions and requirements.

Due to the innovative character of the overall process chain in Troia and the relatively small capacity of 0.1 MW SNG output, a huge potential for energetic improvement exists. During the project, an overall PtG efficiency of 29 % was reached. It has to be considered that the DAC and the liquefaction of the SNG had a comparably high energy demand. Due to the recycle of lean gas to the front of the

[1] This is the methane fraction in front of the liquefaction. The gas quality is reached by gas separation and recycling the lean gas.

methanation unit, the overall conversion of CO_2 and H_2 is in the range of 99 %. A methane content of y_{CH4} = 96 % in front of the liquefaction was reached. It was also shown that the process could be operated dynamically from 20 – 80 % of load with a load change rate of 5 %/min. By heat integration and energetic optimization of the process units, an overall PtG efficiency of 46 % could be reached. Due to the DAC in Troia, the potential for internal heat usage is very high. In combination with the liquefaction, the process chain of Troia can be operated more or less stand-alone and does not require a connection to the gas grid or a nearby heat sink.

Beside a technical evaluation, also an economic evaluation of the demo sites was performed. The economic evaluation of the three demo sites includes (i) the calculations of the capital expenditure (CAPEX) for the methanation unit, (ii) future expectations of the methanation units' $CAPEX_{Meth}$ development until 2050, and (iii) the calculations of the production costs.

Regarding (i): Table 1-2 summarizes the results of the economic evaluation. In a first step, the $CAPEX_{Meth}$ of the demo sites' methanation units was determined using the Add-on factor method. Afterwards, the plant design was optimized, and the $CAPEX_{Meth}$ was calculated for the plants scaled to an SNG/LNG output of 5 MW, 10 MW and 50 MW. Thereby, the optimized specific $CAPEX_{Meth}$ for the three sites is in the same range, between 720 €/kW and 1090 €/kW for a plant scaled to 5 MW SNG/LNG output. The investment costs of the Solothurn methanation unit with an SNG output of 5 MW are 870 €/kW. The relatively low costs can be explained, since only one simple reactor was required to reach the gas quality. Also, the biocatalyst reproduces itself, so that no additional costs are incurred. The optimized specific $CAPEX_{Meth}$ of Falkenhagen methanation unit is 720 €/kW for a 5 MW plant. It must be considered that two reactor stages are necessary, due to the higher restrictions for injection in Germany. Furthermore, there is a high potential to reduce the costs of the reactor by optimizing the reactor design. Troia has the highest specific $CAPEX_{Meth}$ for the methanation unit with 1090 €/kW for 5 MW LNG output. Partly, this technology of a milli-structured methanation reactor is relatively new and in part investigated in pilot scale for the first time. A huge potential is available in Troia to optimize the plant and thus reduce the $CAPEX_{Meth}$. The aim of the STORE&GO project was to achieve a cost reduction for industrial scale methanation plants by 15 % compared to state-of-the-art technologies. This aim has been fulfilled for the Falkenhagen and Solothurn sites, where cost reduction was about 31 %.

Table 1-2: Optimized $CAPEX_{Meth}$ and $CAPEX_{PtG}$ for the methanation units and optimized production costs for the three demo sites based on an SNG/LNG output of 5 MW

	Falkenhagen	**Solothurn**	**Troia**
$CAPEX_{Meth}$ of an optimized plant with 5 MW SNG output	720 €/kW	870 €/kW	1090 €/kW
$CAPEX_{PtG}$ of an optimized plant with 5 MW SNG output	3100 €/kW	3130 €/kW	5120 €/kW
Methane production costs for an optimized plant with 5 MW SNG output (8000 h/a, German electricity price)	0.123 €/kWh	0.098 €/kWh	0.135 €/kWh

Regarding (ii): To estimate the potential of future cost reductions for methanation units, learning curves were implemented on the determined $CAPEX_{Meth}$. The learning curves were calculated within the project by the project partner Energieinstitut Linz, see Deliverables D7.5 and D7.7. Due to scaling effects and high technical development potential, the $CAPEX_{PtG}$ of power-to-gas technologies will strongly decrease. Based on the CAPEX evaluations, the costs will be reduced by 60 % in 2050, for the methanation unit of Falkenhagen for an SNG output of 5 MW (based on HHV). The CAPEX of the Solothurn methanation plant (5 MW SNG output) will be reduced by 61 % until 2050. The CAPEX of the methanation plant in Troia for an SNG output of 5 MW is reduced by about 52 %. Furthermore, a higher H_2 content in the gas grid is currently discussed (current limit y_{H2} = 2 vol.-% in Germany). If a higher injection of H_2 would be introduced for the gas grid, the $CAPEX_{MEth}$ of Falkenhagen and Solothurn could be further reduced. For example, in Falkenhagen only one reactor stage could be capable to reach sufficient methane content. In Solothurn the reactor volume could be reduced.

Regarding (iii): Based on the optimized $CAPEX_{Meth}$ calculations, the production costs for the entire process chain were calculated. In contrast to the calculations of the $CAPEX_{Meth}$, the electrolyser, the CO_2 conditioning/capture, the methanation and the injection/liquefaction are taken into account for the production costs' calculations. The CAPEX data for the remaining process units are based on literature data. For the calculations of the production costs, several assumptions must be made, due to the fact that a lot of operational experiences still need to be gathered. The production costs were determined for 8000 h/a, 4000 h/a and 1500 h/a operational hours. Since the network charges and taxes vary in the different countries, only day-ahead market electricity prices were taken into account for the calculations. In order to better compare the production costs of the plant with each other, firstly, the productions costs were calculated based on the German electricity price.

Thus, based on the day-ahead market electricity prices in Germany, the SNG production costs are 0.123 €/kWh for the Falkenhagen plant with an SNG output of 5 MW and 8000 h/a. For comparison, the production costs for electricity from onshore wind and open space photovoltaic are 0.037 – 0.068 €/kWh and 0.04 – 0.082 €/kWh, respectively [1]. The import costs for fossil gas in Germany are in the range 0.02 €/kWh (2018) [2].

The calculated costs for the Falkenhagen plant include, on the one hand, the optimization of the plant design. The optimizations are taken into account in the CAPEX calculations and, in addition, further optimization potential with regard to operation was integrated. For Falkenhagen, the heat usage was improved and integrated into the calculations. In future, a high cost reduction of the honeycomb reactor is expected. For the Solothurn plant, the production costs are 0.098 €/kWh, assuming an SNG output of 5 MW, 8000 h/a operational hours and the German electricity price. Due to the infrastructure of the 'Hybridwerk', in Solothurn heat usage of a low temperature level could be considered. This leads to high efficiencies and relatively low production costs. In addition, a strong drop of the nutrient cost is assumed, if the nutrients are commercially available. For the same parameters (8000 h/a, 5 MW SNG output, German electricity price), the production costs of SNG amount to 0.135 €/kWh for the process chain in Troia. Compared to the other demo sites, the production costs are slightly higher. This can be explained by the high effort to capture CO_2 from air, and by the liquefaction unit, which is complex compared to the injection technology at the other sites. A huge potential is available in Troia to optimize the plant and thus reduce the CAPEX.

1 Introduction

Aiming at the reduction of global warming according to the Paris agreement, Europe's energy supply must be decarbonized, and the shares of renewable energies in the European electricity grid must grow. However, this implies technological challenges, for instance as electricity supply is becoming increasingly volatile due to the dependence of renewable energy sources on weather conditions. Bridging this gap of energy supply and demand, power-to-gas (PtG) applications represent a promising technology in the European energy system by providing both long-term and large-scale energy storage. By converting hydrogen and carbon dioxide to synthetic natural gas (SNG) via electrolysis and a subsequent methanation, large amounts of energy can be stored through SNG with high energy density. Within the STORE&GO project, three innovative PtG plants have been built, tested and operated in three different countries.

In Falkenhagen, Germany, a newly developed honeycomb methanation reactor was constructed nearby to an existing alkaline electrolyser (AEL). In the project proposal it was planned to extract the CO_2 from a nearby biogas plant via absorption. This was not realized in the project and the CO_2 was delivered from a bioethanol plant. In Solothurn, Switzerland, a biological stirred bubble column methanation reactor was built. A proton exchange membrane (PEM) electrolyser is used as H_2 source. The CO_2 comes from a nearby waste-water treatment plant. At both demo sites, the produced SNG is injected into the gas grid. In Troia, Italy, an innovative milli-structured methanation reactor as well as a direct air capture (DAC) for the carbon dioxide (CO_2) supply were erected. In contrast to the other two demo sites, in Troia the produced SNG is liquefied (LNG). Since the CO_2 is captured from air and the LNG can efficiently be transported without an existing gas grid, this process configuration is less dependent on the location of the plant.

Within this report, the technical as well as the economic evaluation of the three different demo sites is performed. Therefore, the system boundaries are carefully chosen and performance indicators are defined within the project. To test the long term stability and the load flexibility of the demo sites, different test profiles were defined and applied to the demo sites. Generally, all three demo sites reached the defined goal of a methane content y_{CH4} higher than 90 vol.-% at different loads. It was shown that an overall PtG efficiency of higher than 75 % can be achieved. Furthermore, energetic optimizations are discussed within this Deliverable.

For the economic evaluation, optimization potential of the plant design regarding energetic and economic aspects was detected and considered in the calculations. Subsequently, the capital expenditure of the methanation unit ($CAPEX_{Meth}$) at every demo site was calculated using a default factor-method. The capital expenditures for the overall PtG process ($CAPEX_{PtG}$) are calculated based on project results and literature data. Finally, a scale-up of the plants was carried out and the SNG/LNG production cost were determined.

The following report is structured in four parts. First, the design of each demo site is briefly described and the operation history of the demo sites during the project is shown. Then, the applied methods and definitions are explained. In chapter 4 and 5 the results of the technical and economic evaluation are presented, respectively.

2 Description of the Demo Sites

This chapter gives a brief overview of the three power-to-gas demo sites. This chapter also includes the operation hours and a short operation history.

2.1 Falkenhagen

The new methanation plant at the Falkenhagen power-to-gas (PtG) site (see Figure 2-1) was used to produce SNG. The hydrogen was mixed with CO_2 and converted into methane by the Sabatier reaction in a novel reactor concept on suitable catalysts. The generated SNG was fed to the natural gas transport pipeline system of ONTRAS via the existing compression and feed-in infrastructure.

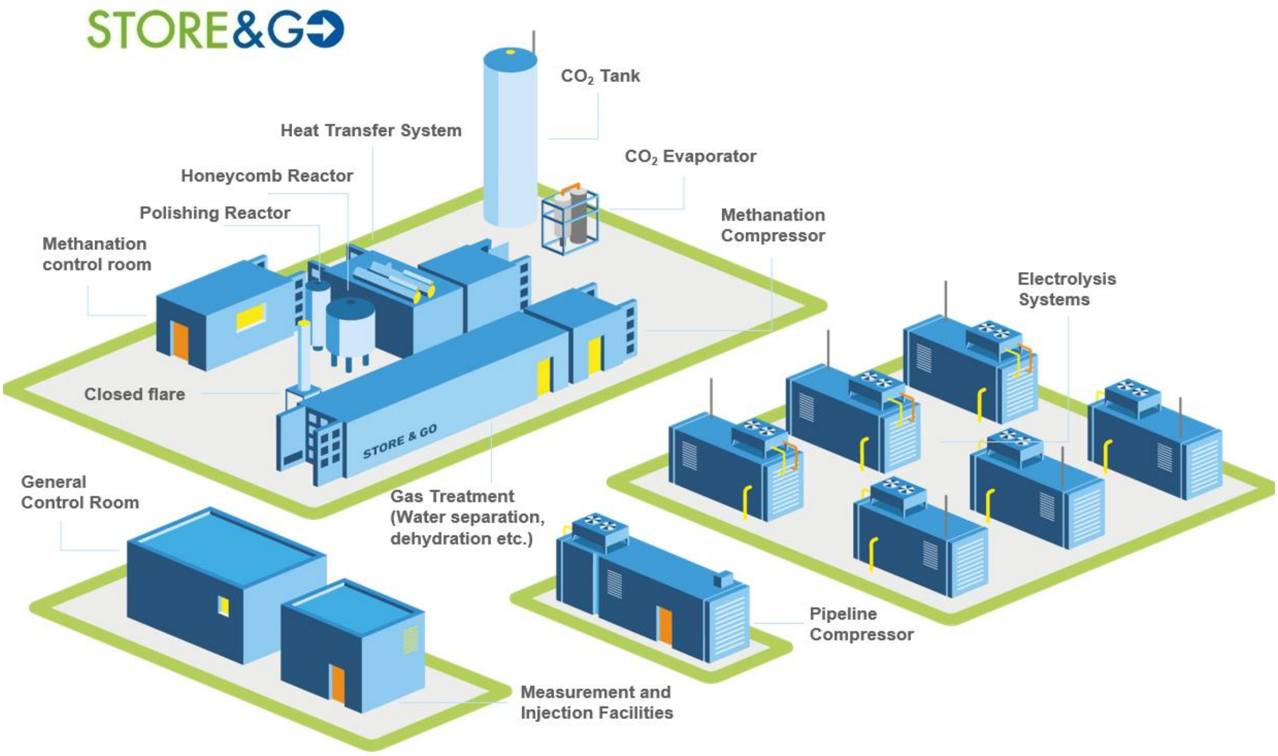

Figure 2-1: General setup of the power-to-gas plant in Falkenhagen.

The hydrogen was provided by an electrolysis plant (6 alkaline electrolysers), already existing in Falkenhagen (more details see in D2.4). A maximum volumetric flow of 210 m³/h (STP) of hydrogen was used for the methanation. This quantity corresponds to an electrical load of about 1 MW. Originally it was planned to separate the CO_2 from a biogas plant nearby. However, it was not realised during the project. Therefore, the CO_2 was delivered in liquid form from a bioethanol plant. It was fed stoichiometrically into the methanation (maximum CO_2 volumetric flow $\dot{V}_{CO_2,STP} = 52.5\ \mathrm{m^3/h}$). The methanation reaction was carried out in two stages. The first reaction stage converted more than 80 % of the CO_2 into methane. A second polishing reactor completed the methanation reaction to achieve the SNG product quality required for the injection into the natural gas grid (more than 99 % conversion).

In front of the first methanation stage, the educt gas was compressed up to 14 bar (operation pressure). In the first reaction stage of the process, the gas mixture entered the honeycomb methanation reactor. This reactor is designed as a multi-tube reactor, in which metallic catalyst carriers made of stainless steel were placed in parallel tubes (see Deliverable D2.4). The inlet gas flow entered the catalytically coated channels, the reaction started. The reaction heat was dissipated through the cooling medium (thermal oil) located on the shell side. The reaction heat was transferred to the cooling medium via the honeycomb structure and the tube walls. The outlet temperatures of the reactors were regulated by the throughput of the cooling medium, which allowed an effective cooling of the system.

The product gas leaving the first reactor stage was first cooled in the heat exchanger against the feed gas. Subsequently, the gas stream was cooled in an air cooler ($T_{Gas,out}$ = 60 °C) and in a water cooler ($T_{Gas,out}$ = 10 °C). The major portion of the reaction water formed was condensed and separated from the product gas in the following liquid separator. This gas was fed to the second methanation stage (Polishing Reactor). This reactor is designed as a cooled tube reactor with a catalyst pellets filling in the tube bundle tubes and oil cooling medium at the shell side, which allowed an isothermal operation. In this reactor the required conversion rate was achieved to supply the product gas to the natural gas grid.

After exiting the Polishing Reactor, the gas stream was again cooled in two heat exchangers to 10 °C. The formed water condensed in a liquid separator and was separated from the product gas. To feed the produced methane gas into the natural gas grid, the gas had to be treated further according to the DVGW rules [3]. Two parallel drying vessels were used to adjust the residual moisture content required for feeding into the natural gas grid. Subsequently, the SNG was sent to the existing compression unit for final supply to the natural gas grid.

2.1.1 Operation Hours

Table 2-1 summarizes the operation status of the demo site in Falkenhagen for the entire operation period. The operation of the demo site started in January 2019 and lasted until February 2020. In the total project period, the demo site was operated 1186 h, which corresponds to 27 613 m³ of injected SNG.

Table 2-1: Status of operation hours of the demo site in Falkenhagen at the end of the project, 14 Feb. 2020.

	Operational time	Volume of gas	Mass of gases
H_2 supply	1322 h	122 666 m³	
CO_2 supply[2]	1186 h	27 613 m³	51 034 kg
Flare (in use)	511 h		
SNG-Injection (ONTRAS)	668 h	17 328 m³	11 367 kg
Thermal oil heated	2759 h		
Compressor run	2108 h		

In Figure 2-2 the distribution of the load hours during the operation of the Falkenhagen methanation plant are shown. Nearly 80 % of the time, the plant was operated between the 40 and 65 % of load.

[2] Count of hours with CO_2 supply defines the operating hours in the project

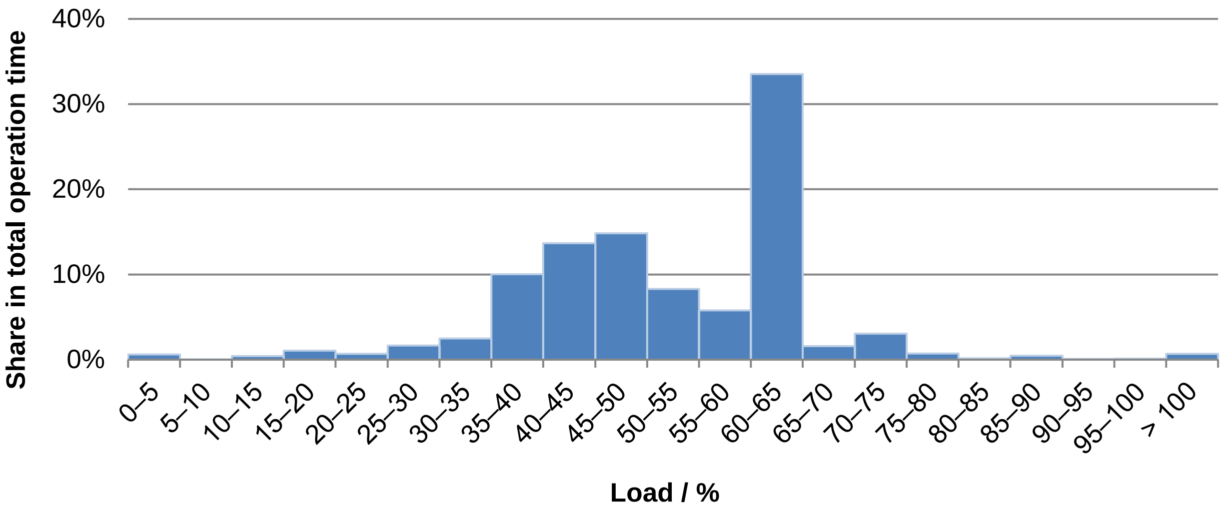

Figure 2-2: Distribution of load hours during operation of the Falkenhagen plant

2.2 Solothurn

In Solothurn the PtG plant (see Figure 2-2) consisted of a new methanation plant receiving H_2 and CO_2 from existing facilities. The hydrogen production facility of Solothurn consisted of two PEM-C30 (PEM: Proton exchange membrane) electrolysers from ProtonOnSite with a rated electrical power of 175 kW each. Each electrolyser can produce 30 m^3/h (STP) hydrogen at 30 bar and 15 m^3/h (STP) oxygen at atmospheric pressure. For the hydrogen production 27 l/h ultrapure water (American Society for Testing and Materials (ASTM) Type 1) was required. The water was prepared by a reversed osmosis system. The electrolysers (AC/DC converters) were connected to the low voltage main distribution panel (3x 400 V, 50 Hz).

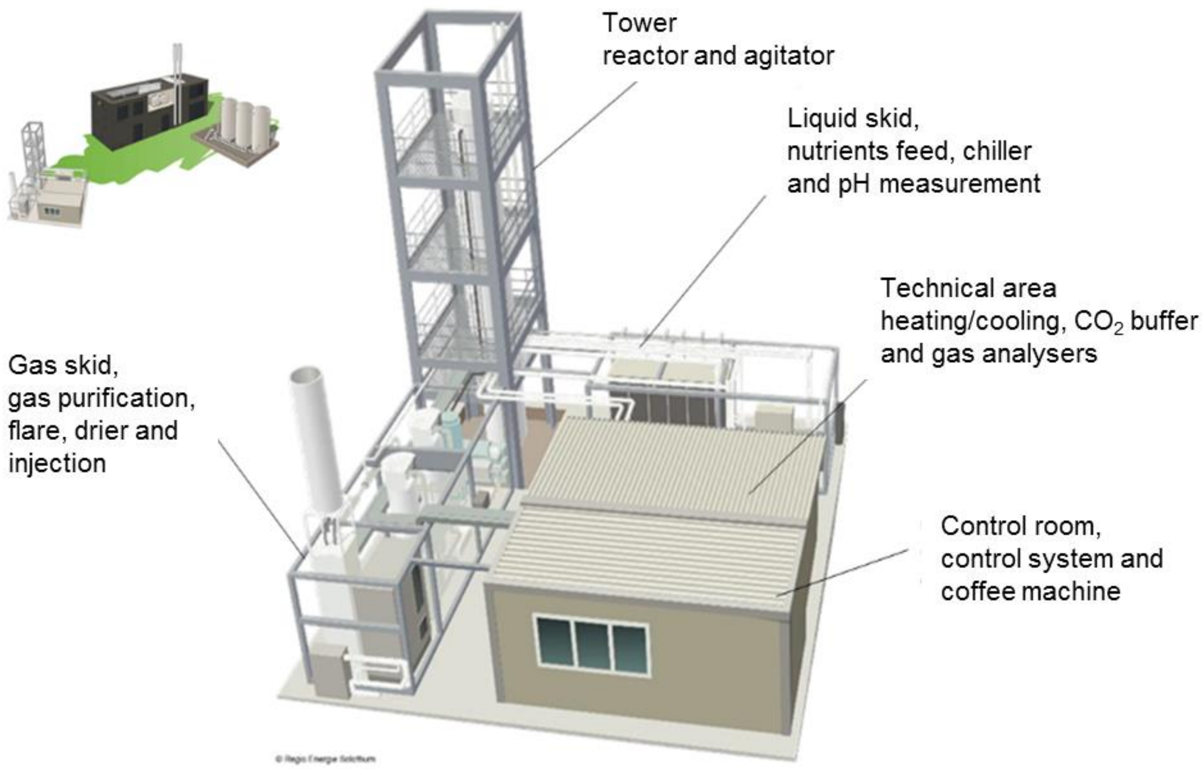

Figure 2-3: General setup of the power-to-gas plant in Solothurn. © Regio Energie Solothurn

The CO_2 for the methanation came from a wastewater treatment plant, which is 2.5 km away from the ‘Hybridwerk’. The raw gas of the wastewater treatment consisted of approx. 50 vol.-% CO_2 and 50 vol.-% CH_4. The gas mixture was separated at the wastewater plant via an existing membrane system. The CH_4 was injected into the natural gas grid and the CO_2 was supplied as a waste product to the methanation plant. The CO_2 which was not used, was fed into a waste incinerator to burn residual concentrations of CH_4. When the PtG plant indicated a need for CO_2, a fan was activated to transport the CO_2 from the wastewater treatment plant to the ‘Hybridwerk’. In the basement of the ‘Hybridwerk’ was a compressor that compressed the CO_2 from 1 bar to approx. 13.5 bar. A tank of 2 m^3 volumetric storage capacity buffered the consumption of the methanation and the delivery quantity of the compressor.

The H_2 produced by the electrolysis and the CO_2 from the wastewater treatment plant were provided at the system boundary with a gauge pressure of about 12 bar. The methanation was based on a biological process with Archaea. The Archaea metabolise H_2 and CO_2 and convert these substances into methane and water in an exothermic process at 11 bar and 61.5 °C.

Archaea are unicellular microorganisms and one of the oldest organisms on earth. They have been identified as a new domain of life about 30 years ago by pioneers like Prof. Karl-Otto Stetter, Germany, and Prof Carl Woese, USA. The Archaea can be cocci, rods, sarcinae-, spirillum- or thread-shaped. They typically live in extreme environments, e.g. hot springs, high salt or low pH pools. Archaea can be found in many environments depending on their metabolic capabilities. One of the subgroups, the so-called methanogenic Archaea, is specialized in methanogenesis, and they use H_2 and CO_2 as their energy and carbon source for the generation of CH_4. The methanogenic Archaea are anaerobic organisms, and various species are known. Methanation is their energy-generating metabolism and therefore a key feature of these organisms.

The range of process conditions for methanogenic Archaea and the conditions in the demonstration plant are listed in the table below.

Table 2-2: Range of process conditions for methanogenic Archaea, and the process conditions at the demo site for a biological methanation with Archaea.

Process parameter	Range	Demo Site
Gauge pressure	1 – 150 bar	10 bar
Temperature	15 – 98 °C	62 °C
pH value	5 – 9.5	7.5 – 8.5
Salt content	0.1 – 15 % NaCl	3 % NaCl

The methanogenic Archaea which were used for the demonstration plant in Solothurn were an optimised strain of *Methanothermobacter thermoautotrophicus,* called ECH0100. The strain is unique in its robustness, tolerance towards contaminants, and it is highly flexible and ideal for following different loads due to changing availability of renewable energy sources.

2.2.1 Operation Hours

Table 2-3 and Table 8-2 sum up the operation results accumulated since May 2019 and show the improvements made by further adapting operation and start-up procedures. In Figure 2-4 the distribution of the load hours is shown. Nearly 60 % of the time, the plant was operated at loads of 50 %. One explanation is the needed adaption time of the biological system to the higher loads. Another reason is the limitation of the full load hours by the hydrogen supply.

Table 2-3: Status of the operation hours of the demo site Solothurn at the end of the project (last update: 24 February)

Parameter	Value	
Operation time (BES10BU034)	1299	h
Hours of flared SNG (BES10BU039)	242	h
Injection hours SNG (BES10BU040)	1057	h
Duration of injected SNG with y_{CH_4} > 90 vol.-% (BES10BU047)	549	h
Duration of injecting SNG with y_{CH_4} > 96 vol.-% (BES10BU048)	214	h
Equivalent full load hours (BES10BU035)	612	h
Flared gas (BES10BU039)	2357	kg
Mass of injected Gas (BES10BU040)	11 165	kg
Energy content of injected gas related to higher heating value (CH_4 + H_2)	172 720	kWh
Energy content of injected SNG related to higher heating value (only CH_4)	166 240	kWh

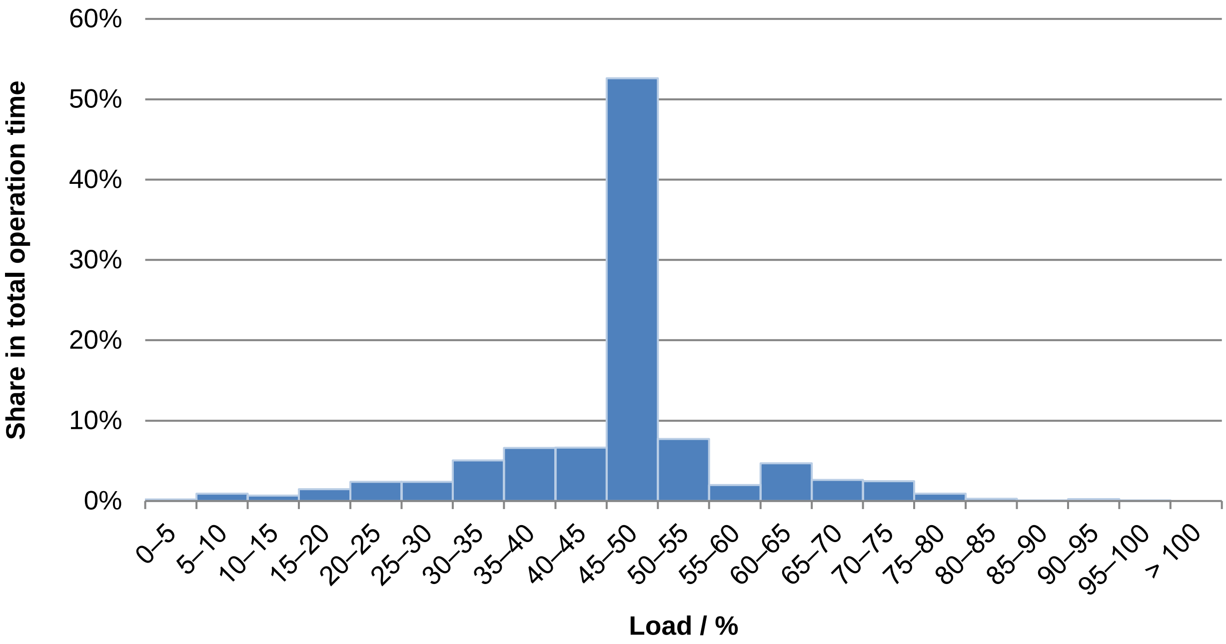

Figure 2-4: Distribution of load hours during operation of the Solothurn plant

The total operation hours consist of the injection and flaring hours, see Figure 2-5. The injection hours (indicator "Time injection") includes "Time injection y_{CH_4} > 90 vol.-%", which includes "time injection y_{CH_4} > 96 vol.-%". The blue part indicating injection including SNG with a CH_4 content of y_{CH_4} < 90 vol.-%. The following relationships apply:

$$\text{I:} \quad \text{Total operation hours} = \text{Time injection} + \text{Time flare} \tag{2.1}$$

$$\text{II:} \quad \text{Time injection} \ni \text{Time injection } y_{CH_4} > 90 \text{ vol.} - \% \tag{2.2}$$

$$\text{III:} \quad \text{Time injection } y_{CH_4} > 90 \text{ vol.} - \% \ni \text{Time injection } y_{CH_4} > 96 \text{ vol.} - \% \tag{2.3}$$

As can be taken from Figure 2-5, the quality of the gas could constantly be improved towards the end of the project. The reason for this initially low CH_4 content was presumably the incorrect calibration of the hydrogen mass flow controller, which led to a higher than expected H_2/CO_2 ratio, and as a consequence hydrogen was available in excess. Measurement results show that the measured ratio changed from M42 on, indicating a drift in the mass flow measurements at the inlet of the plant. This problem was fixed by adjustment of the mass flow controller in M47.

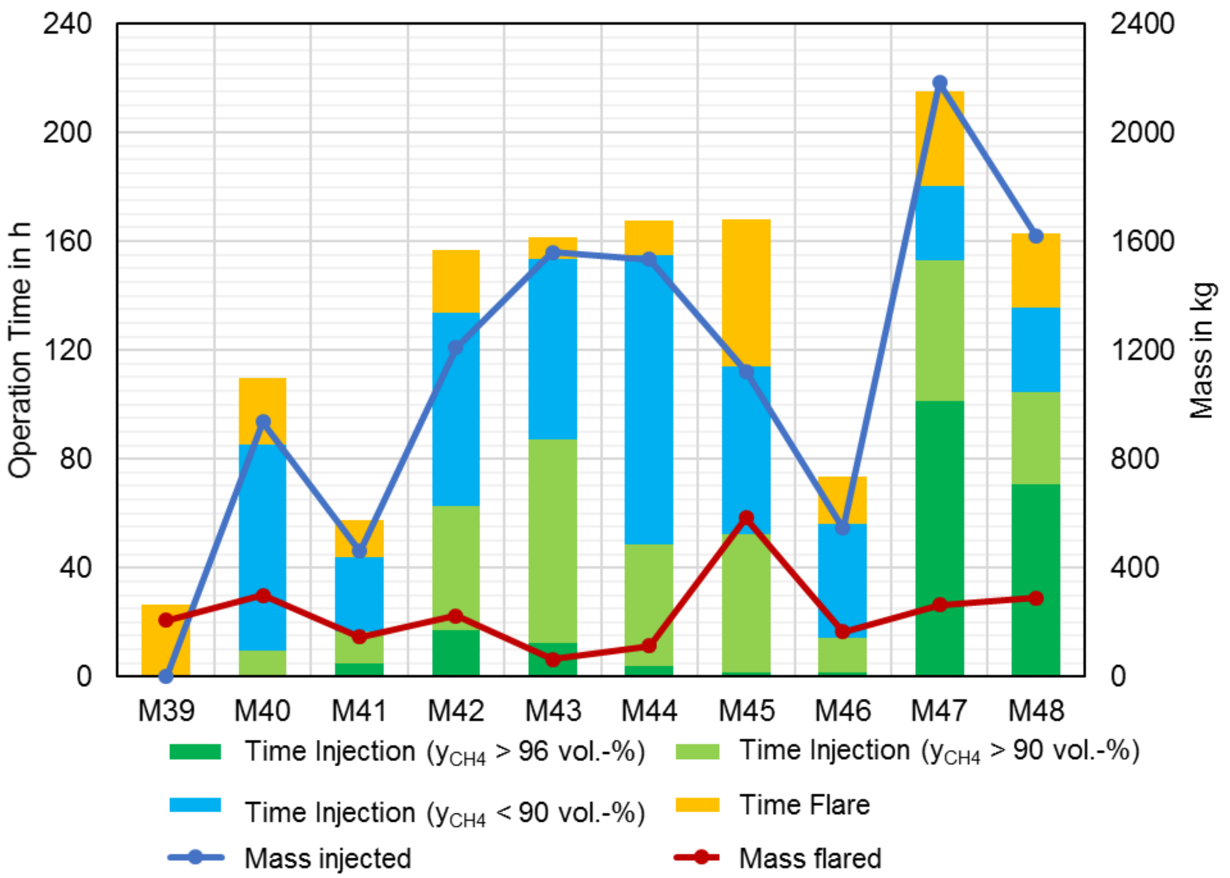

Figure 2-5: Monthly overview of the operating status with operation time. For each month the mass of flared gas and the mass of injected Gas are calculated.

When comparing the result of plant operation in October 2019 (M44) with January 2020 (M47), it becomes obvious that the adjustment of stoichiometry resulted in much better conversion of CO_2 and H_2. After this issue was fixed, the conversions and methane fraction in the SNG were nearly 100 vol.-% at a constant level. The gas composition of the first operation phase did not meet the requirements of the SVGW injection guidelines G13 and G18 for unlimited injection. On the one hand, the methane content was too low and the Wobbe Index was not reached. Due to the possibility of limited injection, which takes special care of the mixture after injection, it was still possible to inject the gas. This was because the flow rate in the grid was high enough to ensure that the weak methanation gas and the gas in the grid maintained a sufficiently high quality in the mixture. In January 2020 the limits for unlimited injection were met.

2.3 Troia

In Troia, Italy, a 200 kW (electrical input equivalent for an electrolyser) plant had been built to implement and to demonstrate the PtG concept. The concept of the Troia plant was based on the production of synthetic Liquid Natural Gas (LNG) using hydrogen generated from the excess of renewable electricity and carbon dioxide captured from atmosphere, as illustrated in Figure 2-4. The implemented concept is a promising solution in converting the excess of renewable electricity into a different energy carrier. An already existent water electrolysis process was used to convert electricity coming from renewable energy sources (RES) into hydrogen. The produced H_2 was then mixed with the CO_2 capture directly from the air on-site in order to produce Synthetic Natural Gas (SNG) in a methanation unit. Because an injection into the gas grid was not feasible at the site location, the SNG was liquefied through a cryogenic process and ready for transportation by tankers or trucks.

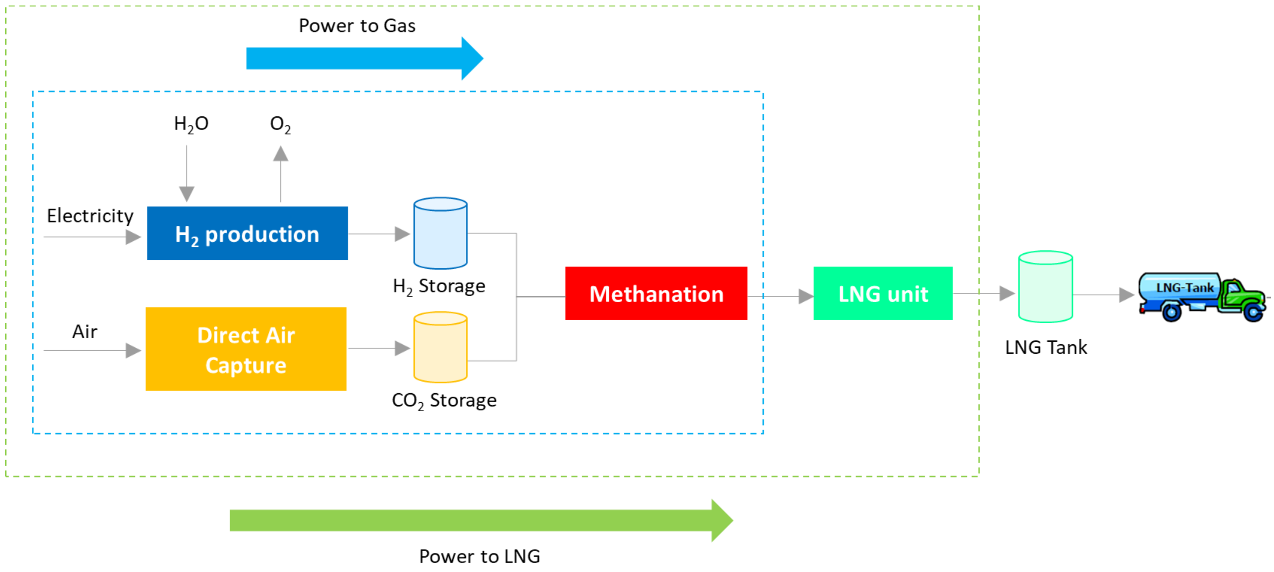

Figure 2-6: Power-to-gas/LNG concept at the demo site in Troia.

So, the demonstration site in Troia, developed inside WP4 activities, is paving the way for an integration of PtG storage into flexible energy supply and distribution systems with a high share of renewable energy. The complete process chain was composed of the following units developed by the indicated key partners:

1. <u>H_2 Production Unit</u>: hydrogen production from water electrolysis by using the legacy electrolyser developed inside the INGRID's project and customised for STORE&GO (TROIA);

2. <u>Direct Air Capture Unit</u>: CO_2 capture from air, processing and storage (Climeworks (CW));

3. <u>CH_4 Production Unit</u>: The milli-structured catalytic reactor was designed and optimised for heat recovery, stream recycle and gas quality for high yield/recovery towards the LNG liquefaction unit (KHIMOD (formerly ATM) and CEA);

4. <u>LNG Purification and Liquefaction Unit:</u> SNG purification, liquefaction and LNG production (Hysytech (HST)):

In detail, H_2 was produced by the alkaline electrolyser powered with renewable electricity from sources in the surroundings, especially from solar, but also from wind. It could be fed with up to 200 kW of electricity. The electrolyser was scaled down from the original 1 MW to the PtG requirement of 200 kW. The hydrogen was then mixed with CO_2 captured from the air: a constant H_2-to-CO_2

ratio was maintained. The educt stream was fed into the milli-structured reactor where the methanation took place. After the methanation step, the stream was mainly composed of methane, water, hydrogen, and carbon dioxide. Humidity and carbon dioxide had to be below a certain concentration (CO_2 concentration < 50 ppm; moisture < 1 ppm) to avoid freezing of CO_2 and water traces during the liquefaction process. The first unit after the methanation reactor was dedicated to the gas conditioning. The gas stream was cooled down to ambient temperature to remove most of the water content. Subsequently, carbon dioxide was separated by using a membrane gas separation system, and the permeate (rich in CO_2 and H_2) was recycled upstream to the methanation unit in order to optimize the plant performances. In order to reach the required specification for the liquefaction unit, a Temperature Swing Adsorption (TSA) unit was used for the final abatement of the remaining CO_2 and H_2O. The last step was the liquefaction of the SNG to obtain LNG, which was then stored in a special cryogenic tank at controlled temperature, while a boil-off stream was then recycled to the process.

2.3.1 Operational Hours

Table 2-4 summarizes the operation status of the demo site in Troia divided into the different process units for the entire operation period. In the total project period, the methanation unit was operated for 824 h, which corresponds to 4669 m^3 of produced SNG. Out of these hours, the methanation unit operated coupled with the liquefaction unit for 260 h. The DAC run more than 2300 h in the total project period.

Table 2-4: Summarized operation hours of the demonstration plant Troia at the end of the project

Parameter	**Value**	**Unit**
Operational time DAC	> 2300	h
Worked time	1620	h
	115	d
Operational time	1142	h
	1362 (incl. stand-by)	h
Methanation time	824	h
	260 (coupled with LNG)	h
Liquefaction time[3]	191	h
	305 (incl. cool down)	h
SNG production	4669	m^3 (STP)
	1663 flared (35.6 vol.-%)	m^3 (STP)
	3006 to LNG (64.4 vol.-%)	m^3 (STP)
LNG production	441	kg
CO_2 injection	4129	m^3 (STP)
H_2 injection	17195	m^3 (STP)
Equivalent full load hours (32 m^3/h)	537	h

In Figure 2-7 the distribution of load hours during operation of the Troia PtG plant is shown. Nearly 30 % of the time, the plant was operated at about 80 % of load.

[3] LNG amount discharged into the LNG Tank. However, during the LNG unit operation, all the methane produced by the methanation unit was liquefied.

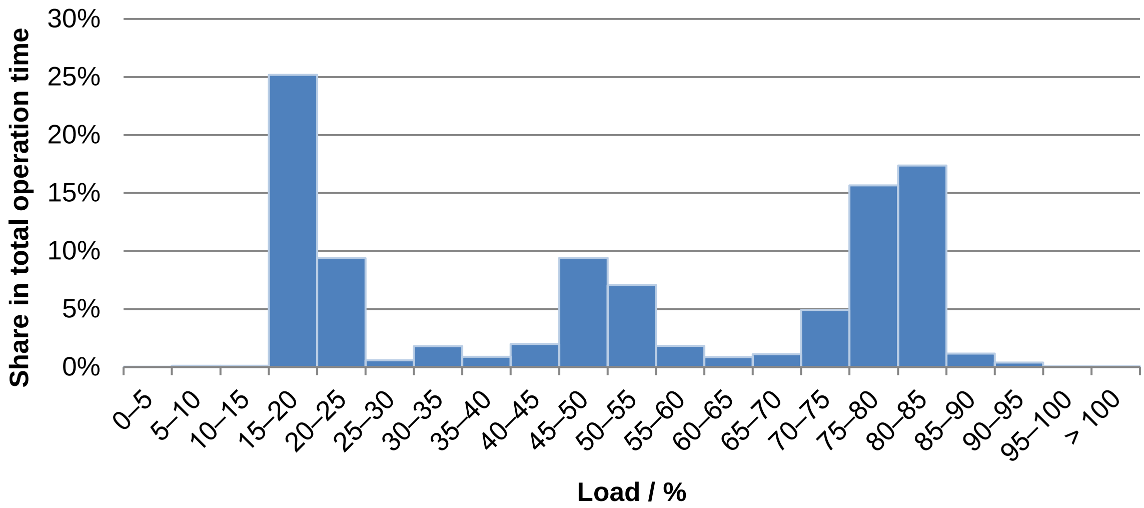

Figure 2-7: Distribution of the load hours during the operation of the Troia plant

3 Methods and Definitions

This chapter gives a brief overview of the applied methods for the technical, energetic and the economical assessment of the technologies realized at the three demo sites. A detailed description of the methods and definitions can also be found in the Deliverables D5.2, D5.5 and D5.6 of WP5.

3.1 Definition of the system boundaries

In order to compare the three different demo sites, the system boundaries for the energetic evaluation and the definition of the performance indicators (PI) are chosen carefully. In Figure 3-1, the systems boundaries are shown in a simplified block flow diagram. The challenge arise from the technical differences of the three demo sites. The three major differences are the CO_2 sources, the methanation concepts and the products (e.g. gas quality). The differences are discussed in detail in the confidential Deliverable D5.5 which is briefly summarised below:

Different H_2 sources which already existed before the project start:

- Alkaline electrolyser in Falkenhagen and Troia
- Proton exchange membrane electrolyser in Solothurn

Different CO_2 sources with different CO_2 fraction in the source $y_{CO2,Source}$:

  - Liquid CO_2 separated from bioethanol plant. Originally it was planned in the project proposal to separate the CO_2 from biogas via an absorption process ($y_{CO2,Source} \approx 40 – 60$ vol.-%). Therefore, CO_2 separation from biogas was considered as the CO_2 source for the Falkenhagen process chain evaluation.
  - CO_2 separation at a waste water treatment plant via membranes ($y_{CO2,Source} \approx 40 – 60$ vol.-%)
  - Direct air capture (DAC) ($y_{CO2,Source} \approx 400$ ppm)

Different products at each demo site (gas grid injection or LNG):

- In **Falkenhagen** SNG with a maximum hydrogen fraction of $y_{H2} < 2$ vol.-% and $y_{CO2} < 2$ vol.-% was injected in a transport grid ($p_{gas\ grid} > 45$ bar) [3].
- In **Solothurn** the produced gas is injected into a local gas distribution grid (max. pressure 5 bar). The gas in the grid after adding the SNG must fulfil the following limits: H_2 fraction y_{H2} has to be lower than 2 vol.-%, for oxygen $y_{O2} < 3$ vol.-% is the limit.
- In **Troia** the product gas, after the methanation and a methane enrichment via membrane separation, is fed in to a liquefier to produce the final product LNG.

Also the **heat usage options** and the available **heat temperature level** were different.

In order to exclude the different H_2 and CO_2 sources and product gas processing steps after the methanation (gas injection, gas liquefaction), the methanation was analysed based on the gas inlet (H_2, CO_2) and outlet flow. As outlet flow, the flow in front of the last step (see red dashed line in Figure 3-1) was taken. The different pressure levels of the product gas flows after the respective methanation (Falkenhagen: 14 bar; Solothurn: 10 bar; Troia: 4 bar) were neglected in this attempt.

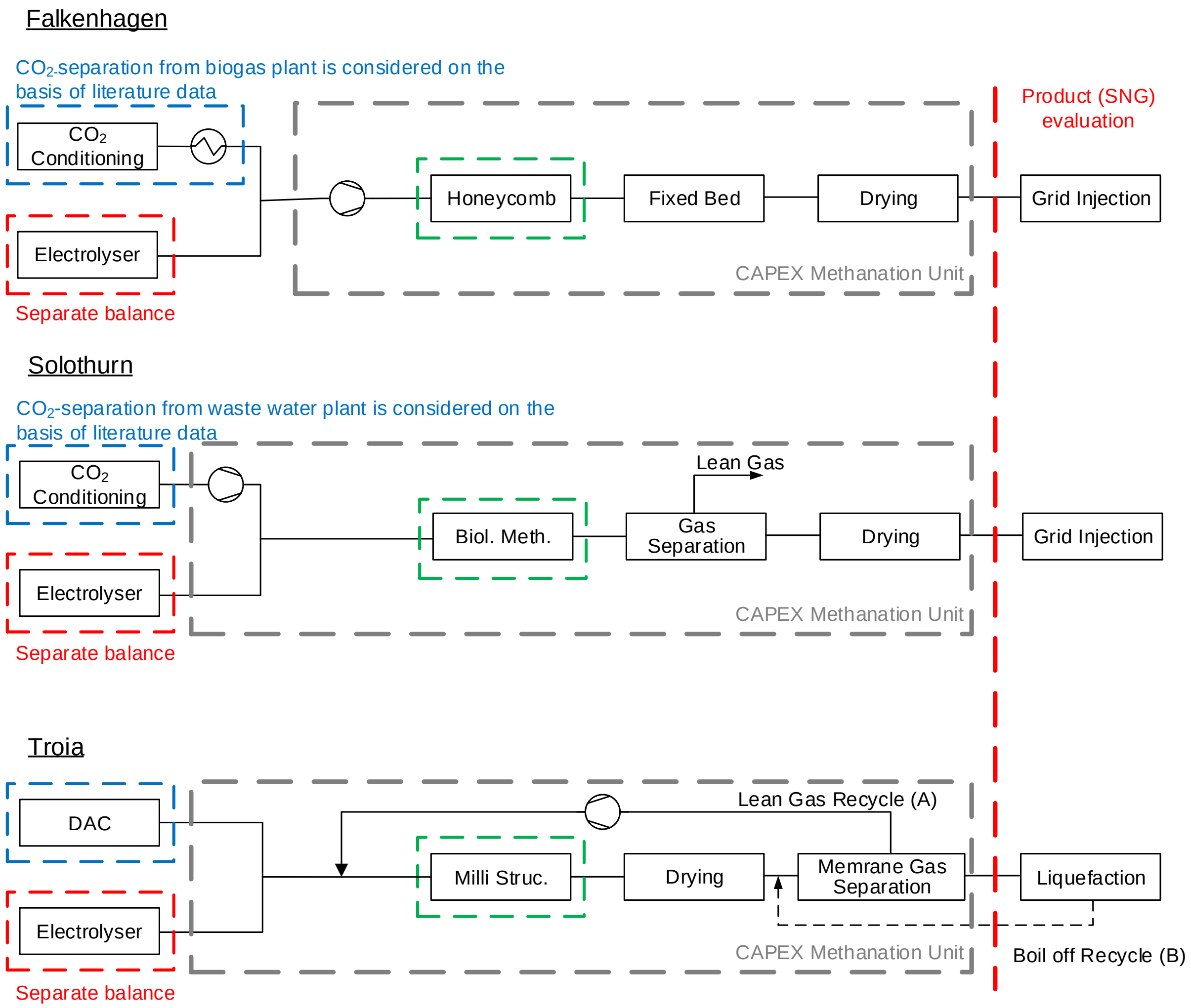

Figure 3-1: Simplified block flow diagrams of the three demo sites. Red dashed line: product gas flow for the evaluation of the methanation unit (grey box); Blue box: system for the CO_2 conditioning, Red box: electrolyser system, Green dashed line: System boundaries for the reactor evaluation.

3.2 Evaluation of the Methanation and PtG Efficiency

To evaluate the demo sites, a common set of performance indicators (PIs) were used (e.g. GHSV, product gas quality, efficiency). Additional specific PIs were defined for the three demo sites (e.g. conversion rate in the honeycomb reactor). Constant load profiles should have been run at regular intervals to evaluate the performance of the system over time and under various external conditions. A technical characteristic program (TC) was used to get data for the calculation of the (key) performance indicators. The aim of the long term TC was to run the system over a longer period and check if the quality of the output and the efficiency remains constant. In section 3.3, the different testing profiles are explained. Defined load profiles were used to show and to quantify the flexibility and dynamic of the power-to-gas system. The sub-systems electrolysis and methanation were analysed with stress tests (ST) to determine the limits of each sub-system. The duration of the stress test profiles were configured for one workday. To ensure standardized parameter set, all volumetric flows are given at standard temperature and pressure (STP, T_{STP} = 0 °C, p_{STP} = 1.01325 bar).

The Gas Hourly Space Velocity (GHSV) gives the feed of the reactant per volume of the reaction zone (V_{cat}) and is defined as follows:

$$GHSV = \frac{\dot{V}_{\mathrm{in,STP}}}{V_{\mathrm{cat}}} \tag{3.1}$$

The reaction volume of the honeycomb is the volume of the monolith structure. In the stirred bubble column reactor, the reaction volume is the bubbly flow. The reaction volume for the milli-structured reactor is the overall channel volume. A detailed description of the reaction volume is given in D5.5.

The conversion X_i for all the reactors and for the overall system is defined as follows:

$$X_{\mathrm{i}} = \frac{\dot{N}_{\mathrm{i,in}} - \dot{N}_{\mathrm{i,out}}}{\dot{N}_{\mathrm{i,in}}} \tag{3.2}$$

In order to evaluate different aspects of the three demo sites, different efficiencies were defined. The simplest definition is the methanation efficiency (see equation (8.1)). As aligned within the WP 5 the energy flow of the product gas $\dot{E}_{ch,SNG}$ includes only the methane fraction. The hydrogen fraction is not considered. Equation (8.2) and (8.3) also take the heat usage and electricity demand into account, respectively. The energy flow is always calculated based on the higher heating value (HHV) of the gases. Since the usable heat from the methanation in Troia was integrated within the process (heat supply for the DAC), the methanation and the CO_2-separation could not be evaluated separately. Hence, the comparison of the three demo sites based on the overall methanation efficiency (see equation (8.3)) including the heat use and the electrical energy demand is unfair. To overcome this issue, the energy demand for the CO_2-separation was theoretically added for the Falkenhagen and Solothurn demo site. In order to consider the additional energy demand within the overall methanation efficiency $\eta_{\mathrm{SNG,HHV,ov}}$, the definition was extended compared to the Deliverables D2.4 and D5.2 to the overall methanation efficiency with CO_2 separation:

$$\eta_{\mathrm{SNG,HHV,ov,CO_2-Con}} = \frac{\dot{E}_{\mathrm{ch,SNG}} + \dot{E}_{\mathrm{th,use,SNG}}}{\dot{E}_{\mathrm{ch,H_2,in}} + P_{\mathrm{el,Meth}} + \dot{E}_{\mathrm{th,CO_2-Con}} + P_{\mathrm{el,CO_2-Con}}} \tag{3.3}$$

As stated before, the energy flow of SNG $\dot{E}_{\mathrm{ch,SNG}}$ consists only of the methane fraction. By considering the efficiency of the electrolyser η_{Ely} and potential heat usage from the electrolyser $\dot{E}_{\mathrm{th,use,Ely}}$, the PtG efficiency can be calculated:

$$\eta_{\mathrm{PTG,HHV}} = \frac{\dot{E}_{\mathrm{ch,SNG}} + \dot{E}_{\mathrm{th,use,SNG}} + \dot{E}_{\mathrm{th,use,Ely}}}{\dot{E}_{\mathrm{ch,H_2,in}} / \eta_{\mathrm{Ely}} + P_{\mathrm{el,Meth}} + \dot{E}_{\mathrm{th,CO_2-Con}} + P_{\mathrm{el,CO_2-Con}}} \tag{3.4}$$

The electrolyser efficiency η_{Ely} could be calculated from the measurement data, given from the project partners (e.g. UST), or the state of the art was applied.

In order to evaluate the efficiency of the overall process chain from electricity to the product (injected SNG or produced LNG), the energy demand for the product upgrading ($\dot{E}_{\mathrm{el,Inj/Liq}}$, gas grid injection (Inj) or liquefaction) needs to be included in the PtG efficicency (see equation (3.4)). This results in the definition of the overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$:

$$\eta_{\mathrm{PtG,HHV,ov}} = \frac{\dot{E}_{\mathrm{ch,SNG}} + \dot{E}_{\mathrm{th,use,SNG}} + \dot{E}_{\mathrm{th,use,Ely}}}{\dot{E}_{\mathrm{ch,H_2,in}} + P_{\mathrm{el,Meth}} + \dot{E}_{\mathrm{th,CO_2-Con}} + P_{\mathrm{el,CO_2-Con}} + P_{\mathrm{el,Inj/Liq}}} \tag{3.1}$$

The specific energy demand for the product upgrading is either based on the mass of the liquefied SNG (kWh/kg) or on the nominal volume of the injected SNG (kWh/m³).

3.3 Evaluation of the Flexibility and Dynamics

To evaluate the flexibility and dynamics of the methanation process, the minimum load $L_{\mathrm{Meth,min}}$ and the load change rate (LCR) were calculated.

$$L_{\mathrm{Meth,min}} = \frac{\dot{V}_{\mathrm{part\ load,min}}}{\dot{V}_{\mathrm{nominal\ load}}} \tag{3.5}$$

$$LCR = \frac{\frac{\partial \dot{V}_{\mathrm{H_2}}}{\partial \mathrm{t}}}{\dot{V}_{\mathrm{H_2,full\ load}}} \tag{3.6}$$

During start-up, the deployment time of the start-up process from the cold standby to the state of hot standby (operational readiness) was measured. Likewise, the duration and the total amount of poor/lean gas produced (m³ (STP)) per ramp up could also be measured or calculated when the plant starts from hot standby and then reaches the state when the gas quality meets the requirements.

$$\int_{\mathrm{start\ ramp\ on}}^{\mathrm{operation}} \dot{V}_{\mathrm{feed\ gas}}\,\mathrm{dt} \tag{3.7}$$

In order to examine the operational durability and flexibility of the different methanation technologies as well as the corresponding electrolysis systems, several testing profiles had been defined and were partially carried out by the plant operators. The results are presented in the sections 4.1.1, 4.2.1 and 4.3.1.

Different test procedures were foreseen (see section 8.2):

- The PtG system was analysed with stress tests (ST) to determine the limit of the system and to get data for the monthly calculation of the (key) performance indicator for full load. The duration of the stress test profiles was configured for one workday. If possible, the ST should be done individually for the sub-systems electrolysis and methanation.

- The technical characteristic program (TC) was used to get data over different loads (TC001) and long process operation (TC002) to check if the quality of the output and the efficiency remain constant. The aim of the 72 h test (TC002) was to run the system over a longer period with maximum possible (ideally 100 %) load.

3.4 Data Reconciliation

To classify the measurement results and the calculated PI, it is useful to validate the data. But the measurements inevitably contain inconsistencies because of, for example:

- Intrinsic sensor inaccuracies (*random* errors),
- Improper sensor calibration (*systematic* or *gross* errors),
- Issues in the data transmission process (systematic or gross errors).

Data reconciliation and validation (DRV) techniques use information redundancy and mathematical methods to validate raw sensor data and allow the calculation of reliable plant indicators. Data reconciliation aims at correcting measurement errors due to intrinsic sensor inaccuracies (random errors), while data validation targets calibration issues (systematic errors) before and after. These techniques build on the following formulation of the measurement variables:

A measurement $\hat{y}$ can be expressed as the sum between its true value $\underline{y}$ and two error terms ε_b and ε_r, respectively the systematic and random errors.

$$\hat{y} = \underline{y} + \varepsilon_s + \varepsilon_r \tag{3.8}$$

The statistic over $\hat{y}$ is said to be an unbiased estimate of parameter $\underline{y}$ only if the systematic error (bias) is absent (or equal to zero). This assumption lets us rewrite the above relation as:

$$\hat{y} = \underline{y} + \varepsilon_r \tag{3.9}$$

Considering ε_r as an independent random variable that is normally distributed with mean 0 and variance σ^2 ($\varepsilon_r \sim N(0, \sigma^2)$), the above relation tends towards a normal distribution with mean $\underline{y}$ and variance σ^2 ($\hat{y} \sim N(\underline{y}, \sigma^2)$).

The probability density function (pdf) of $\hat{y}$ (also known as the likelihood of $\hat{y}$) is therefore given by:

$$P(\hat{y}\,) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{\left(-\frac{(\hat{y}-E[\hat{y}])^2}{2\sigma^2}\right)} \tag{3.10}$$

For the sake of ease, this density function can be simplified into the so-called logarithmic likelihood function $l(\hat{y}\,)$.

$$l(\hat{y}\,) = \log\big(P(\hat{y}\,)\big) = \frac{1}{\sqrt{2\pi\sigma^2}}\left(-\frac{(\hat{y}-E[\hat{y}])^2}{2\sigma^2}\right) \tag{3.11}$$

Data reconciliation aims at maximizing the likelihood of $\hat{y}$ (or logarithmic likelihood of $\hat{y}$) through the minimisation of the term $\frac{(\hat{y}-E[\hat{y}])^2}{\sigma^2}$. In any DR application, the true value (or expectation $[\hat{y}] = \underline{y}$) of a measurement $\hat{y}$ is always unknown while only the measured value of the sensor (which is affected by an error) is object of knowledge. The goal of DR is therefore to approximate the true value of $\hat{y}$, also called validated value, in such a way that the likelihood of $\hat{y}$ is maximized while satisfying all the system constraints. A typical DR problem is therefore an optimization problem with an objective function that corresponds to the sum of the weighted measurements' square errors, and the resolution of which allows to maximize the overall likelihood of the whole set of measurements.

A steady-state DR problem with normally distributed measurements can be formulated as follows. Suppose $\hat{y}_{i,t}$ to be the measured value of sensor *i* at time *t*, and $y_{i,t}$ its corresponding reconciled value. By considering the unmeasured variables x_t, the data reconciliation problem can be mathematically expressed as:

$$\sum_{i}^{t} \frac{\left(y_{i,t} - \hat{y}_{i,t}\right)^2}{\sigma_i^2} \quad s.t. \quad \{f_t(y_t, x_t) = 0\,, g_t(y_t, x_t) \geq 0\;, \tag{3.12}$$

Where σ_i is the standard deviation of sensor *i*, and where f_t and g_t are respectively the set of process equality and inequality constraints at time *t*. The term $\left(\frac{y_{i,t} - \hat{y}_{i,t}}{\sigma_i}\right)^2$ is called the penalty of measurement *i* and it quantifies the contribution of the correction of measurement *i* to the overall objective function. The DR problem is stated as a Non-Linear Programming (NLP) optimization problem with the goal of minimizing the overall severity of correction (measured as sum of weighted least square errors) that is needed in order to satisfy the system constraints.

These errors (denoted ε_b and ε_r in the mathematical formulation of DRV) lead to apparent violations of the mass and energy conservation laws. For example, the following ones were observed when screening the measurement data:

- Differences of mass flowrates (Figure 3-2) between the inlet and outlet of a component, or over an entire process (violation of the *mass conservation principle*)
- Apparent increases of temperature after cooling of a synthetic gas stream with negligible heat gains/losses, or increases of pressure over a pipe channel (violation of the *energy conservation principle*)

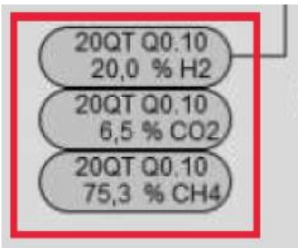

Figure 3-2: Example of violation of the mass conservation principle for measurements of the Falkenhagen plant – the sum of the volume fractions for the three main gas components exceeds 100 vol.-%.

These errors need to be addressed when analyzing the process measurements to ensure appropriate plant monitoring. The **data reconciliation** technique consists of correcting the measurement errors from the retrieved data (random errors). It is based on the actual measurements & sensor accuracy information, and respects the mass and energy conservation laws, and possibly other constraints.

The data reconciliation technique generally requires redundancy of the process measurements. For example, for a vapour-liquid separator (Figure 2), the flow rate at point (a) can be deduced from the mass balance if the flow rates at points (b) and (c) are measured. The measurements are therefore *not* redundant.

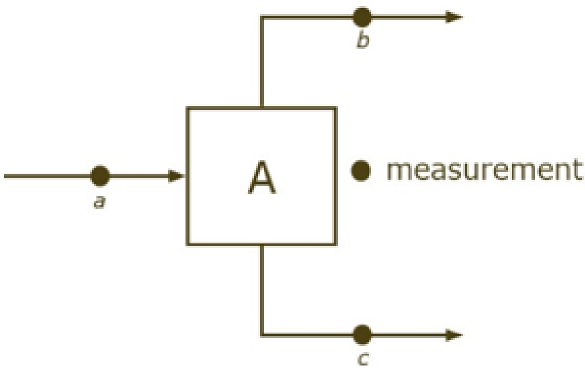

Figure 3-3: Schematic illustration of a vapour-liquid separator and of the redundancy required for data reconciliation.

On the contrary, performing measurements of the mass flow rates at points (a), (b) and (c) is redundant, as the mass flow rate at (a) can be deduced from the two others. This redundancy is necessary for performing data reconciliation, as no measurement is exempt of random errors. This illustrates that the more sensors are placed the more reliable the data reconciliation process is.

In the cases of the Solothurn and Falkenhagen plants, several process units (such as the electrolyser and CO_2-supply systems) did not have enough sensors, and thus not enough measurements, to perform data reconciliation techniques. Data from the catalogues and design data were therefore used to complete the models when relevant. On the contrary, the methanation systems presented redundant measurements because of multiple sensors placed after the electrolysis and storage systems (Solothurn and Falkenhagen), in the cooling loops (Solothurn) and after the methanation reactor(s) (Solothurn and Falkenhagen).

The detailed results of the data reconciliation are available in the confidential Deliverables of the demo sites (D2.5, D3.5 and D4.10).

3.5 Process Modeling based on ASPEN

In order to be able to determine the ideal process within the design specifications, Aspen models were developed for the methanation units at all three demo sites by DVGW (Part of WP5). The calculated performance indicators (PIs) based on the ASPEN model were compared to the ones based on measurement data. The models are based on flowsheets provided by HSR, and present the ideal process within the respective process design. As example, the flowsheet for the Falkenhagen demo site generated in ASPEN is shown in Figure 8-4. In general, all three models are based on the assumptions of an ideal stoichiometric ratio of H_2 to CO_2 (y_{H2}/y_{CO2} = 4.0), ideal heat transfer and neglectable heat losses to the air. In addition, the CO_2 as well as the H_2 extraction were neglected in the ASPEN model. The modelling of the methanation reactors are based on the measured methane yield for respective demo site. The electrical power demand and the Balcene of Plant (BoP) are not included in the ASPEN model. The electrical power demand of the compressors and the pumps had to be calculated with help of measurement data or engineering data. The assumptions for each demo site are briefly summarised in the appendix 8.4.

Furthermore, the results of ASPEN simulations were used for the scale-up of the methanation processes, and subsequently for the calculation of the $CAPEX_{Meth}$ (see section 3.6) for different SNG output (1 – 50 MW).

3.6 Investment Cost Calculation

The costs for the demo sites were calculated by a cost estimation tool developed by DVGW. Within this tool, the capital expenditure ($CAPEX_{Meth}$) was calculated based on the ratio factor method. Therefore, the cost for the major equipment (e.g. reactors, compressors, heat exchanger) were calculated based on cost correlations [4]. The parameters required for the calculations (e.g. volumetric flow or heat flux) are based on the Aspen simulations. The currency had been transferred to Euro using the average exchange rate of 2017. Furthermore, costs were adjusted for inflation. By means of ADD-on factors, the costs for plant equipment (e.g. piping, instruments, etc.) were determined (see Table 3-1). The costs for engineering were considered by a second group of ADD-on ratio factors, which also includes a size factor.

To estimate the effect of the scale-up on the $CAPEX_{Meth}$, the costs were calculated for 1, 5, 10 and 50 MW SNG output. The scale-up was done based on the ASPEN model. The used ADD-on ratio factors during calculation of the demo sites costs and the scale-up procedure to different plant sizes are given in Table 3-1.

Within the Task 5.2 and this Deliverable, the absolute costs for a specific plant size (based on SNG output in HHV) of the methanation units $K_{\mathrm{Meth,5MW(SNG)}}$ were given in Euro (€). In order to simplify the comparison of the different plants and to show the effect of the scale-up on the costs, the costs are also given as specific costs. The specific costs are based on the produced SNG $k_{\mathrm{Meth,5MW(SNG)}}$ (see equation (2.13)). As for the energetic evaluation of the process, the energy flow of the produced SNG $\dot{E}_{\mathrm{ch,SNG}}$ includes the energy content of methane only. The hydrogen fraction is neglected.

$$k_{\mathrm{Meth,5MW(SNG)}} = {K_{\mathrm{Meth,5MW(SNG)}}}\Big/{\dot{E}_{\mathrm{ch,SNG}}} \tag{2.13}$$

Table 3-1: Ratio factors for the estimation of the CAPEX of the demo sites based on literature [5]

Ratio Factors	Plant Size					Description
	%	Demo Site	1 MW	5 MW	10/50 MW	
Installation equipment	15	15	15	15	15	
Process piping	7 – 60	45	45	45	45	Gaseous mediums
Instrumentation	2 – 15	13	13	13	13	It is based on a far-reached instrumentation.
Building and site development	5 – 100	50	50	50	40	For larger sites, plant will increasingly be realized as open-air construction.
Auxiliary services	0 – 100	0	0	0	0	Assumed that the methanation will be integrated into an existing plant
Outside lines	0 – 25	0	0	0	0	Heat transfer pipes and educt gas pipes are not included in the methanation unit.
Engineering and construction	20 – 50	35	35	35	30	Average engineering factor, since experience increases with increasing plant numbers, resulting in a decrease of the factor.
Contingencies	0	0	0	0	0	Since the cost estimate is made in comparison with an existing plant, the factor is 0.
Size factor	0 – 35	30	25	10	5	

3.7 Estimation of the SNG/LNG Production Costs

Additionally to the plant's investment costs, the production costs of the injected or rather the liquefied gas were calculated for the different demo sites. In contrast to the above described calculations of the $CAPEX_{Meth}$, the production costs include the costs for the entire process chain. Thus, beside the costs for the methanation unit, the costs for the electrolyser, CO_2 conditioning/capture and injection or liquefaction must also be determined and taken into account in the calculations. The production costs consist of the initial $CAPEX_{PtG}$ as well as of the plant's operational expenditure (OPEX). Thereby, the OPEX can be divided into fixed and variable costs. The fixed OPEX includes for example maintenance costs, insurance and personnel costs, while energy, material costs and revenues from the excess heat were considered in the variable OPEX. In order to obtain the production cost, the annuity method was used, see equation 2.13. Using equation 2.13, the entire arising expenses I_{A} and C_{t}, the by-products' revenues R_{t} and the total energy output E_{t} in the determined period were discounted by a defined interest rate [6–8].

$$\text{production costs} = \frac{\mathrm{I_A} + \sum_{\mathrm{t}=1}^{\mathrm{n}} \frac{\mathrm{C_t} - \mathrm{R_t}}{(1+\mathrm{r})^{\mathrm{t}}}}{\sum_{\mathrm{t}=1}^{\mathrm{n}} \frac{\mathrm{E_t}}{(1+\mathrm{r})^{\mathrm{t}}}} \tag{2.14}$$

I_{A} : Initial investment in €

C_{t} : Costs in period t in €/a

R_{t} : Revenues of by-products in period t in €/a

E_{t} : Energy output in period t in MWh/a

r : Interest rate in %

n : Calculatory operating life of the plant in years

Thereby, the used interest rate is 6.86 % consisting of the regulated equity interest rate (5.64 %) and the corporate tax (1.23 %) [9]. The estimated amortization period *n* of the plant was set to 20 years. The initial investment corresponds to the summarized $CAPEX_{PtG}$ of each process unit. The total costs of a period t involve the fixed and variable OPEX as well the potential interim investment costs for the period.

Since the variable OPEX, in particular the electricity costs, depends on the operating hours per year of the plant, three different scenarios were determined for the calculations of the production costs. These are as follows: 1500 h/a, 4000 h/a and 8000 h/a. According to the operating hours, the electricity prices of the day-ahead market vary. In Table 3-2 the day-ahead market electricity prices of the reference year 2017 are shown for different countries. Due to no standardized network charges and taxes within Europe, only day-ahead market prices were considered into the calculations.

Table 3-2: Electricity prices at the day-ahead-market for individual hours divided into different bidding zones, based on the reference year 2017

	DE	FR	CH	IT-SUD	DK1	Unit
Averaged electricity prices for the 1500 cheapest hours	10.6	22.8	23.7	34.4	14.7	€/MWh
Averaged electricity prices for the 4000 cheapest hours	22.2	29.9	30.9	40.4	22.9	€/MWh
Averaged electricity prices	34.2	45.0	46.0	49.8	30.1	€/MWh

4 Technical Evaluation

Within in this chapter the results of the technical evaluation of the three demo sites are presented. The focus is the evaluation of the methanation units. However, the efficiency of the overall process chain (include e. g. electrolyser, CO_2-Source) is also assessed. The evaluation of the sites is based on performance indicators (PI) and efficiencies, which were defined in chapter 3. All volumetric flows are given at standard temperature and pressure (STP, T_{STP} = 0 °C, p_{STP} = 1.01325 bar). The PIs were calculated on the basis of the measurement data during constant operation. To ensure constant operation the standard deviation (SD) of the data was checked. If the SD was too high, the data were checked more carefully to ensure constant conditions. In order to find optimization potential, the data were compared with the results of an ASPEN model. The section 3.5 gives a brief description of the approach.

One of the major goals of the STORE&GO project was to prove the capability of dynamic operation at the demo sites. For this purpose, different test scenarios were defined (see section 8.2). The results can be found in section 4.1.1, 4.2.1 and 4.3.1.

Based on the evaluation of the demo sites and the comparison with an ASPEN simulation, technical optimization potentials were found. The focus of the optimization is to increase the efficiency of the overall PtG process chain. Furthermore, some technical improvements have an effect on the investment costs of the methanation plant. The investment costs are discussed more in detail in chapter 5.

4.1 Demo Site Falkenhagen

In the following, the calculated PIs of the demo site Falkenhagen (see block flow diagram in Figure 4-1) based on measurements are discussed (see Deliverables D2.4 and D2.5). Additionally, the results are compared with an ASPEN simulation. In order to compare the PIs, the same operation points were selected. For both cases, an inlet molar flow of H_2 of 9.0045 kmol/h ($\dot{V}_{H_2,STP} = 200$ m³/h) were chosen, which corresponds to a load of 95 % of the methanation unit. Table 4-1 shows the selected PIs for the defined operation point of 1 hour. This period was chosen, since this period had the highest load at constant operation, (see Deliverable D2.5). It was chosen since at higher load the highest efficiency can be reached.

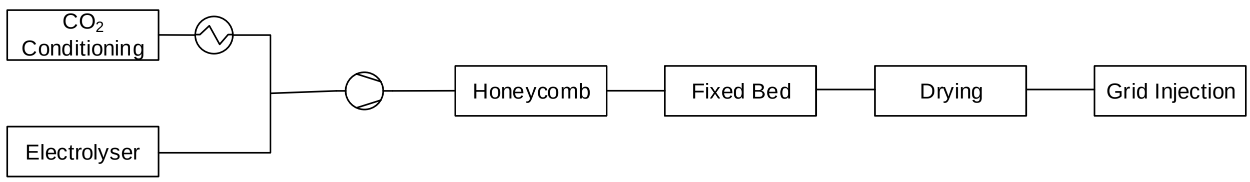

Figure 4-1: Simplified block flow chart of the demo site Falkenhagen

At first, the high stoichiometry ratio of the feed gas stands out. In this case, it is most probably a measurement error. Since the methane fraction in the product gas is y_{CH4} > 99 vol.-%, the feed gas stoichiometric ratio had to be in the range of 4. For the ASPEN model an ideal stoichiometric ratio of four is assumed. Hence, the inlet flow of CO_2 is 50 m³/h, which is higher than the measurement results. As a consequence, the GHSV in the ASPEN model is also higher. The same effect can be seen for the SNG output, which is also higher for the ASPEN results.

Due to the high conversion of 99.8 %, the maximum thermodynamically reachable methanation efficiency of 78 % is almost achieved in the ASPEN model. However, the methanation efficiency of 77.5 % based on measurement data is very close to the ideal efficiency of 78 %. The effect of neglecting the H_2 content in the product gas is lower than 0.2 %, since y_{CH_4} > 99 vol.-%.

Also, the dissipated heat by the oil circuit as well as the heat loss in the air cooler are slightly optimized in the ASPEN model towards the measurement data. Due to the assumption of an ideal oil circuit (e.g. neglecting heat losses), the heat usage increases by nearly 20 % to 103 kW. According to the improved heat integration, the heat loss in the air cooler and the demand for electrical heating decreases. The improved heat transport/usage also leads to an 2.5 % increase in overall methanation efficiencies (ASPEN: $\eta_{\mathrm{SNG,HHV,ov}} = 87.3$ %).

The comparison of the PIs for the methanation plant shows that the calculated performance indicators are close to the ideal values being modelled by ASPEN. It can be assumed that the methanation unit is operated close to its design limits. There is only slight potential for increasing the overall methanation efficiency without changing the plant design.

Based on the calculated methanation efficiency, the efficiency of the overall process chain was evaluated. In the project proposal, the CO_2 separation form biogas was planned for the demo site in Falkenhagen. Realized was the supply by liquid CO_2 from a bioethanol plant. Nevertheless, the energy demand for CO_2 separation from biogas was considered in the evaluation, which is in the same range as the electrical energy demand for the methanation unit (see Table 4-2). This additional energy demand reduces the overall methanation efficiency by nearly 5 %. Due to the low efficiency of the electrolyser in Falkenhagen, the overall PtG efficiency is $\eta_{\mathrm{PtG,HHV,ov}} = 52.7$ %. To reach the project goal of 75 % overall PtG efficiency, optimization has to be performed, which are discussed in section 4.1.2 .The energy demand for the injection of the product gas into the grid is comparably low ($P_{\mathrm{el,Inj}} = 0.1\ \mathrm{kWh/m^3}$). Thus, the effect on $\eta_{\mathrm{PtG,HHV,ov}}$ is minor. In Deliverable D2.5 more constant operation were analysed. At a load of 66 % ($\dot{V}_{\mathrm{H_2,STP}} = 140$ m³/h), the overall PtG efficiency drops by nearly 2 % due the higher specific energy demand of the methanation unit.

Table 4-1: Comparison of the PIs of Falkenhagen demo site. The PIs based on the ASPEN model are compared with PIs which are calculated based on measurement data (04/17/2019, 1:30 pm to 2:30 pm (local time)). The definition of the PIs are given in section 3.2 and 8.1.

Performance Indicator	Measurement data $\dot{V}_{H_2,STP} = 200\ m^3/h$	ASPEN model $\dot{V}_{H_2,STP} = 200\ m^3/h$	Unit
PtG plant H_2 input	708	715	kW
Feed gas stoichiometry	4.4	4.0	-
PtG plant SNG output (only CH_4)	549	557	kW
PtG plant SNG output (CH_4 + H_2)	550	558	kW
Electricity demand methanation plant	41	41	kW
GHSV Honeycomb (as- built)	732	751	1/h
GSHV Honeycomb (as designed)	1465	1502	1/h
Honeycomb conversion rate	92	92	%
GHSV overall (as-built)	500	512	1/h
GHSV overall (as designed)	821	842	1/h
Overall conversion rate of H_2	99	100	%
Methane fraction after methanation	99	99	vol.-%
Heat usage	86	103	kW
Temperature level of heat usage	184	-	°C
Methanation efficiency $\eta_{\mathrm{SNG,HHV}}$	77.5	78.0	%
Methanation efficiency with heat usage $\eta_{\mathrm{SNG,HHV,T_{use}}}$	89.8	92.4	%
Overall Methanation efficiency $\eta_{\mathrm{SNG,HHV,ov}}$	84.8	87.3	%
Overall methanation efficiency with energy demand for CO_2 conditioning $\eta_{\mathrm{SNG,HHV,ov,CO_2-Con}}$	79.7	83.3	%
PtG efficiency $\eta_{\mathrm{PtG,HHV}}$ ($\eta_{Ely} = 63.7\ \%$)	52.9	54.3	%
Overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$ ($P_{\mathrm{el,Inj}} = 0.1\ \mathrm{kWh/m^3}$)	52.7	53.8	%

4.1.1 Dynamic Operation / Test Program / Long Term Operation

As described in section 3.3, the dynamics and flexibility of the catalytic honeycomb reactor with subsequent polishing reactor were tested in the course of several measurements.

In Figure 4-2, the red and green lines in the upper diagram show the load cycles of the hydrogen and carbon dioxide flow. The three lower diagrams show the composition measured in the product gas after the honeycomb reactor and in front of the injection (after the polishing reactor).

When starting up from hot standby until injectable gas quality was reached, the following steps were performed. The aim during start-up was to burn as little product gas as possible in the flare that does not fulfil the injection conditions. During the start-up, the hydrogen was recycled in a closed loop to heat up the system. Starting conditions for start-up were that the reactor had been adjusted in the range of operating temperature and operating pressure under hydrogen atmosphere. Then carbon dioxide was added and the hydrogen and carbon dioxide reacted to methane and water. The reaction itself and the condensation of the water vapour led to a reduction in volume. This caused the pressure to drop in the in the closed loop. The addition of fresh hydrogen up to a stoichiometric ratio to carbon dioxide and the reduction or interruption of the recirculation led to an increase in pressure. As soon as the operating pressure was reached, the gas was sent to the flare for combustion. When the injection conditions were met, the gas was directed to the injection point and the flare was switched off.

The duration of the transition from hot standby until the injection criteria are met depends on the desired load and on the reactor and piping volume. The case shown in Figure 4-2 produced approx. ~33 m^3 (STP)[4] of gas which had to be flared before the SNG reached the required injection quality. In the specific case of Falkenhagen, a large dead space volume was chosen for the reactor to facilitate access to the reactor and maintenance of the honeycombs. For the construction of the same methanation on an industrial scale, a different design would be preferred in order to shorten the transition period during start-up and to be able to measure changes in controlled variables in the product gas composition.

In the range of time on stream TOS $\approx$ 10 – 17 h, the SNG flow drops to zero. Probably during this period, the gas was sent to the flare instead of injecting the SNG into the gas grid. Since the gas flow was measured after the injection compressor, the gas flow could not be measured. This position of the measurement site also explains the relatively high signal noise, which is related to pressure fluctuations due to compression of the gas. Also the methane fraction of the gas is dropping to less than 97 vol.-%. From the data, it can be seen that during this time the pressure in the reactors increases. The conversion in the honeycomb reactor is not affected by this. One possible explanation is that the gas analyser is affected by the changing conditions.

[4] This flow sensor had a measuring error of approx. 10–20% because it was not calibrated correctly (temperature, pressure and compositions deviated). The product gas was recirculated in the system until a methane content of ~80% was reached, and was flared from there until it had >= 96 vol.-% CH_4 and <= 2 vol.-% H_2.

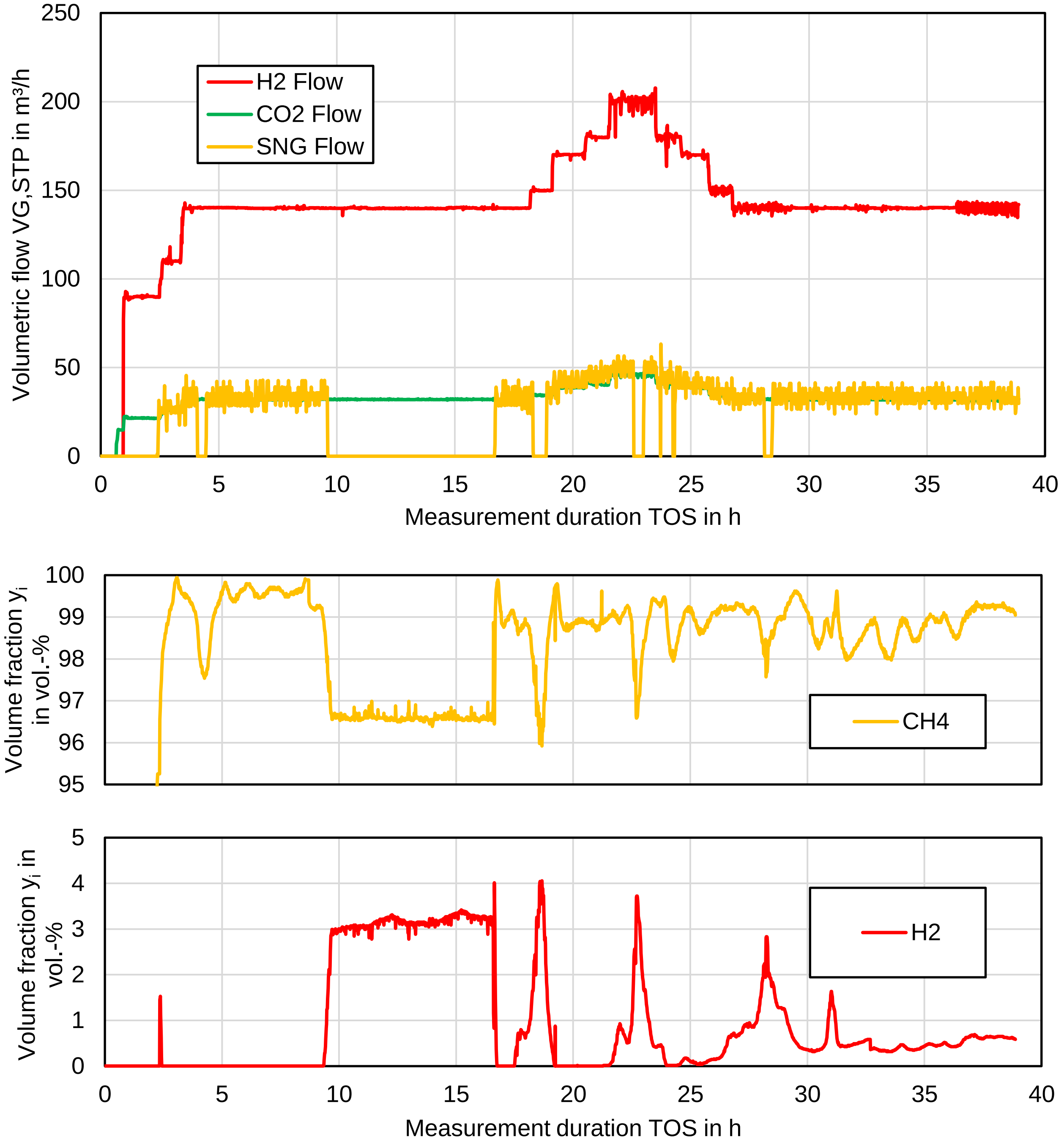

Figure 4-2: Test program Falkenhagen on the basis of TC001 and TC002 (04/16/2019 12 am – 04/18/2019 3 am). The gas fractions are measured in the dry product gas in front of the injection (CO_2 fraction was not exported correct).

The fact that the methanation technology can handle load changes had been proven in various tests (see Figure 4-2). The load profiles for the tests are shown in the appendix 8.2. Load changes from 67, 71, 81, 86 up to 95 % and vice-versa were executed. The load change rate was analysed in this report (see definition in equation (3.6)). In Figure 4-3, two different load changes are shown based on the hydrogen flow. The black line shows the mean load change rate during the load change. The mean values for these two load changes are 3.2 %/min and 3.1 %/min. In the left load change, the slope in the beginning is high and decreases when it is getting near the new set point. In the right load change, the controlling of the H_2 flow could be optimized. From this behavior it could be stated that also higher mean load change rates are possible with this plant configuration.

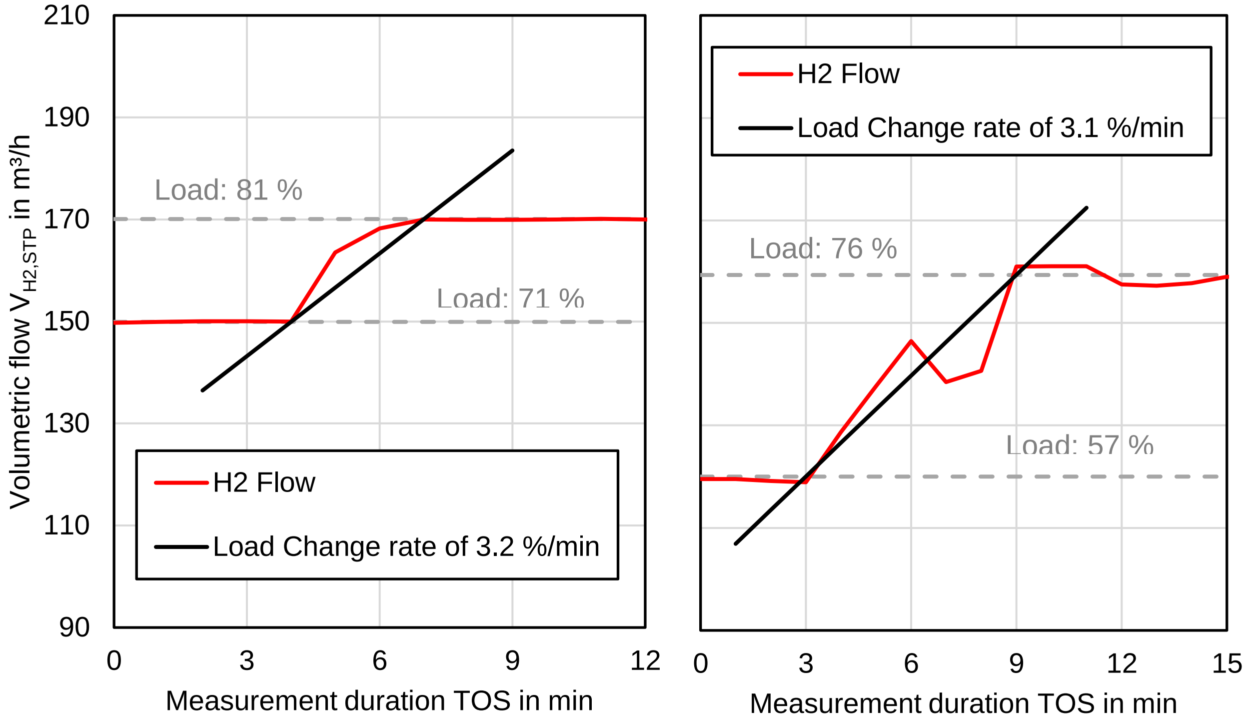

Figure 4-3: Analyses of the load change rate based on measurement on 01/28/2020. Left: Load change from 71 to 81 %; Right: Load change from 57 to 76 %. The black line indicates the mean load change rate.

After TOS = 18 h, the load was changed stepwise until 95 % of load were reached. During the load changes, the product gas quality hardly changed. But is has to be considered that the effect of the load changes on the product gas quality delayed time-wise due to residence time of the product gas in the reactor and in the pipes. Due to the construction of the honeycomb reactor, the dead volume below the honeycomb structures is relatively high. As a consequence, the residence time is also high. But from the date in Figure 4-2 it can be seen that the product gas quality is in not changing noticeably during and after the load changes. This test shows that the methanation plant in Falkenhagen is capable of producing SNG (y_{CH4} > 98 vol.-%) during dynamic operation.

In general, the methanation technology in Falkenhagen was able to compensate variable loads (40 – 100 %) with the built plant design, and no quality losses due to the methanation technology could be detected. Due to the grid injection limitation of y_{H2} < 2 vol.-% in the SNG, the injection into the grid was occasionally interrupted. Especially towards the end of the project, however, the process was optimized to reduce the number of interruptions. From a process engineering point of view, it would be better to adapt the injection guideline and apply the limit of y_{H2} < 2 vol.-% to the mixture in the network after the feed-in. In this way, gases richer in hydrogen can also be fed into the grid.

4.1.2 Energetic Optimization

In this section, the energetic optimization potential for the Falkenhagen plant is discussed. The considered potentials are listed below (see also Table 4-2):

- Use a State-of-the-Art (SoA) Electrolyser. This reduces the energy demand for the hydrogen production from 22.4 kWh/m³ (based on SNG) to 18.7 kWh/m³ (assumption SoA incl. BoP: $\eta_{\mathrm{Ely,H_2}} = 76\,\%$).

- Decrease heat losses in the oil cycle. This could increase the heat usage at high temperatures (T_{use} = 180 °C) by 20 %.
- Use heat of the product gas cooling after the honeycomb reactor at T_{use} = 60 °C (condensation of water: $y_{H20,Honneycomb,out}$ = 58 vol.-%).
- Decrease the energy demand of the methanation unit by changing the compressor configuration (0.7 kWh/m³). A CO_2 compression is needed only if the H_2 is produced at reaction pressure. In Deliverable D5.6 a further decrease of the BoP to 0.3 kWh/m³ (based on SNG output) is indicated.

In D2.4 and D2.6 these potentials are discussed in detail.

Table 4-2: Energy demand for the overall PtG process chain (see block flow diagram in Figure 3-1) for the Falkenhagen plant based on the measurement (04/17/2019, 1:30 pm to 2:30 pm (local time)). The energy data are based on the SNG-Output.

	CO_2-Source (Absorption process)	**H_2 Source (Electrolyser)**	**Methanation**	**Injection**	**Unit**
Initial situation					
Energy demand	1.0	22.4	0.8	0.1	kWh/m³
Heat usage potential (without optimization)	-	-	1.7	-	kWh/m³
Optimization					
Energy demand	1.0	18.7	0.7	0.1	kWh/m³
Heat usage hot water ($T_{\mathrm{use}} = 180\,°C$)	-	-	2.1	-	kWh/m³
Heat usage product gas cooling ($T_{\mathrm{use}} = 60\,°C$)	-	-	0.9	-	kWh/m³

In order to show the effect of these optimization, the calculated overall PtG efficiencies are given in Table 4-3 if these potentials are considered. Since the electrolyser is the biggest energy consumer in the overall process chain (see Table 4-2), the increased efficiency of the electrolyser has a major impact on the overall efficiency. The effect of the improved heat usage at high temperature and the effect of the slight decrease of the energy demand of the methanation unit have only a minor effect. The heat usage a lower temperature could increase the overall PtG efficiency by 4 %. However, this requires a heat user at 60 °C at the location of the plant. If there is an additional heat user for low temperature heat, the waste heat from the electrolyser could also potentially be used. This would have a major effect on the overall PtG efficiency and would help to reach an overall efficiency higher than 75 %.

Table 4-3: Effect of the optimization for the Falkenhagen plant on the overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$. These optimizations are given based on measurements (04/17/2019, 1:30 pm to 2:30 pm (local time)).

Case	Overall PtG efficiency $\boldsymbol{\eta}_{\mathbf{PtG,HHV,ov}}$
Initial situation from Table 4-1	53 %
+ Increase the heat usage by decreasing heat losses in oil cycle ($T_{\mathrm{use}} = 180°°C$)	54 %
+ Optimized electrical energy consumption methanation	54 %
+ Including heat usage Methanation ($T_{\mathrm{use}} = 50°°C$)	58 %
+ Optimized electrolyser (SoA, $\eta_{\mathrm{Ely,H_2}} = 76$ %)	69 %

4.2 Demo Site Solothurn

The technical evaluation of the demo site in Solothurn is based on a constant operation of 1 hour. This constant condition at a relatively high load of 75 % was reached at the end of the project. At this load, it is possible to achieve a high methane fraction y_{CH4} > 99 vol.-% in the product gas, which fulfils the project goal of a methane content of higher than 90 %. Longer periods of lower loads were analyzed in detail in WP4 of the STORE&GO project. This period of operation was used for the evolution of the process since it was the constant measurement at the highest load. This corresponds most likely to the nominal 100 % load of the plant. As for the Falkenhagen plant, the efficiency will increase at higher loads

In order to achieve this performance of the reactor, the Archaea need to be adapted to the high flow rate of CO_2 and H_2. During the commissioning of the plant, high fractions of methane (y_{CH4} > 90 vol.-%) were only reached at low loads of less than 50 %. If the load was increased to nearly 100 %, the methane fraction dropped to 40 vol.-% (see Figure 8-5). After decreasing the load, higher methane fractions were reached again. The biocatalyst as any other living being is subject to environmental adaptation. This can be considered as an evolutionary process, where an organism becomes able to live better in a specific habitat. As mentioned above, the biocatalyst was adapted to live in an environment which provided a certain amount H_2 and CO_2. As soon as the availability of substrate (H_2 and CO_2) was increased, the biocatalyst could not catch up with it and conversion went down. Proteins, enzymes, cofactors etc. need to be build inside the Archaea to make it able to access more process gasses. This adaptation process for a simple organism as an Archaea is relatively quick compared for instance to mammalians or other evolved species. For the Archaea, this process can take hours to days depending on the organism fitness. This process is characterized by a gradual increase of the process gasses made by steps, followed by an adaptation period before the next step up. After the adaption of the Archaea and optimization of the nutrients dosing system, it could be seen that the biocatalyst was able to convert higher volumetric flows with constant gas quality of y_{CH4} > 90 vol.-% methane in January 2020 (see Figure 4-4). The strong fluctuations of the SNG flow were caused by control issues of the pressure regulation in the reactor and the injection unit.

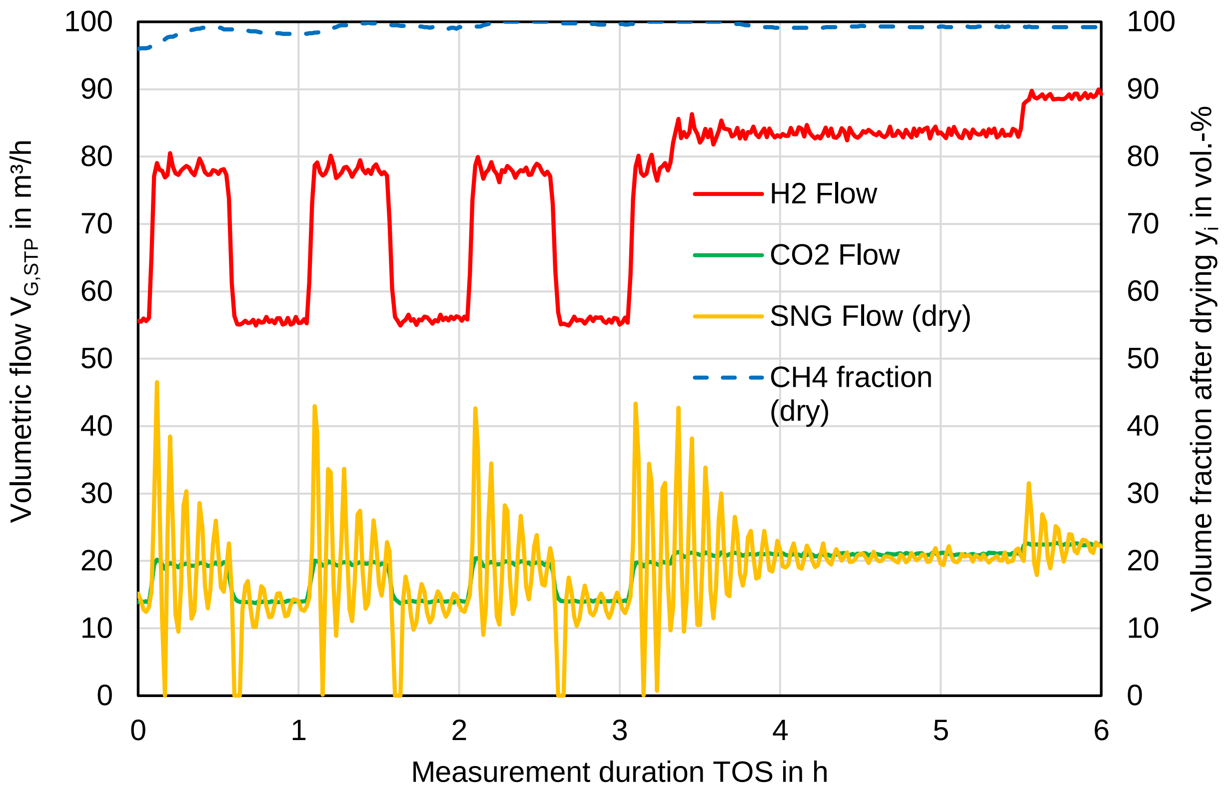

Figure 4-4: Measurement results from the demo site Solothurn (01/21/2020 9:00 pm – 01/22/2020 3:00 am).

After the adaption of the process, the reactor performance was evaluated. The results are shown in Table 4-4 for an operation at 75 % of load. This refers to H_2 input of $\dot{E}_{\mathrm{ch,H_2,in}} = 296\ \mathrm{kW}$. Also the PIs based on the ASPEN model were determined. By means of the Aspen simulation, a comparison with the ideal process can be made.

As defined in chapter 3, only the energy content of the methane is considered for the SNG output. Due to the high conversion of 99.8 %, the additional consideration of the energy content of the hydrogen would increase the SNG output by less than 1 %. Compared to the results from ASPEN, the measured SNG output is 2 % higher. This can be attributed to minor measurement errors. The electrical energy demand for the methanation is $P_{\mathrm{el,Meth}}$ = 18 kW, which correspond to 0.8 kWh/m³ based on the SNG output (based on measurement). The main consumers are the agitator (operated at 70 % of nominal power), the CO_2 compressor and possible trace heating (T_{amb} < 5 °C).

Despite the high conversion, the methanation efficiency is low compared to the theoretical efficiency ($\eta_{\mathrm{Meth}}\left(X_{\mathrm{CO_2/H_2}} = 99.8\%\right) > 78\ \%$). The Aspen results show the same behavior. This could be explained by the solubility of the gases in water at higher pressure (p_{Reactor} = 10 bar) and low temperature ($T_{\mathrm{Condensation}}$ = 7°C) in water. Also H_2 and CO_2 is consumed by the biocatalyst. Since the biocatalyst is reproducing itself, there is a demand for energy and material. This energy and material is gained by the consumption of nutrition, carbon and hydrogen. Due to the high calculated conversion of the biological methanation, the gas consumption of the biomass must be in the range of lower than 1 vol.-%. The byproduct water is discharged from the reactor and the knock out vessel after the reactor. Within this water, CO_2 and CH_4 is dissolved and also discharged from the process. The methanation efficiency is further decreased to $\eta_{\mathrm{SNG,HHV,ov}}$ = 73 % by considering the energy demand of the methanation plant.

Table 4-4: Comparison of PIs of Solothurn demo site (01.23.2020, 2:30 am to 3:30 am (local time)) with Aspen modelling. The definition of the PIs is given in D5.2.

Performance Indicator	Measurement data (75 % load)	ASPEN model	Unit
Feed gas stoichiometry	4.0	4	-
PtG plant H_2 input	315	315	kW
PtG plant SNG output	244	237	kW
PtG plant SNG output (only CH_4)	244	236	kW
Electrical power demand methanation plant (incl. trace heating)	23	23	kW
GHSV (according to liquid volume)	31	31	1/h
CO_2 conversion (methanation)	100.0	99.8	%
H_2 conversion (methanation)	100.0	99.8	%
Methane fraction (in front of injection)	100.0	98.9	vol.-%
Heat usage electrolyser ($\eta_{\text{Ely,therm}} = 32\ \%$)	168	168	kW
Heat usage Methanation	0	0	kW
Temperature level of heat usage	48	61	kW
Methanation efficiency $\eta_{\text{SNG,HHV}}$	77	75	%
Methanation efficiency with heat usage $\eta_{\text{SNG,HHV,T}_{\text{use}}}$[5]	77	75	%
Overall methanation efficiency $\eta_{\text{SNG,HHV,ov}}$	73	71	%
Overall methanation efficiency with energy demand for CO_2-Conditioning $\eta_{\text{SNG,HHV,ov,CO}_2\text{-Con}}$	73	71	%
PtG efficiency $\eta_{\text{PtG,HHV}}$ ($\eta_{\text{Ely,H2}} = 60\ \%$) [6]	76	74	%
Overall PtG efficiency $\eta_{\text{PtG,HHV,ov}}$ ($P_{\text{el,Inj}} = 0\ \text{kWh/m}^3$)[7]	76	74	%

In order to calculate the overall PtG efficiency the H_2 production (PEM electrolyser), the CO_2 conditioning and the injection to the gas grid must be considered. In the case of the Solothurn, the waste

[5] Heat usage potential for the methanation is not considered
[6] Heat usage potential for the electrolyser is not considered
[7] The energy demand can be neglected since the grid pressure is below the reactor pressure

heat (168 kW at 75 % load) from the electrolyser can be used for district heating in the 'Hybridwerk' [10]. The additional energy demand for the heat pump is not considered in this Deliverable. Since in Solothurn the CO_2 is a waste product from the waste water treatments plant, the energy demand for CO_2 separation can be neglected. Furthermore, the energy demand for the injection can also be neglected in Solothurn, since grid pressure is lower than the reactor pressure which supersede the compression. This results in an overall PtG efficiency of 76 % based in the measurement result in Table 4-4. The low temperature (T_{use} = 48 °C) heat from the biological methanation (3.0 kW/m³ based on SNG output) could be utilized in the 'Hybridwerk' as well. But the usage of the methanation waste heat is not considered since it was not realized within the project. The effect on the overall PtG efficiency is discussed in section 4.2.2.

To use the heat at a low temperature level (T_{use} < 60 °C) in a nearby heat sink is a key advantage of the location in Solothurn. Also the low grid pressure and the advantageous CO_2 source have an effect on the efficiency. Table 4-5 shows the effect of these advantageous conditions at the location in Solothurn. If the waste heat from the electrolysis (7.8 kW/m³ based on SNG output) could not be used the overall PtG efficiency would drop to 45 %. If additional an energy demand for the CO_2 conditioning and the injection had to be considered, the overall PtG efficiency would drop to 43 %. This shows, the strong dependency of the overall PtG efficiency on the location and the energetic integration of the PtG plant.

Table 4-5: Change of the overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$ if the waste heat form the electrolyser could not be used and the energy demand for the CO_2 conditioning and the injection had to be considered.

Case	Overall PtG efficiency $\boldsymbol{\eta_{\mathrm{PtG,HHV,ov}}}$
Initial situation from **Table 4-4**	76 %
- Without heat potential electrolyser available ($\eta_{\mathrm{Ely,therm}} = 32\ \%$)	45 %
- With energy demand for CO_2 conditioning	43 %
- With energy demand for SNG Injection	43 %

4.2.1 Dynamic Operation / Test Program / Long Term Operation

In the first six months of operation, the stoichiometric flow rate, which was measured in the control system, contained a systematic error, due to improper sensor calibration. After this was adapted by iteration and the ratio was set correct (H_2/CO_2 = 4:1) in January, the produced SNG contained up to 100 vol.-% of methane at constant loads.

As described in section 3.3, the methanation was tested for its flexibility as well. Figure 4-5 shows the change in feed gas quantity over time and the corresponding gas composition measured in the product gas. It becomes obvious that the methanation is easily able to deal with load changes up to 75 % of the nominal load. At a hydrogen flow of $\dot{V}_{H2,STP}$ = 90 m^3/h, the quality drops slightly, which indicates that the culture still needed to be adapted for higher flows as described above. After adaption of the Archaea, the plant was operated at almost full load (93 % of load, $\dot{V}_{H2,STP}$ = 111 m^3/h) for two hours on February 13th. It was possible to reach a high methane fraction of y_{CH4} > 93 vol.-% (see Table 4-6).

The process for starting up biological methanation in Solothurn is different from that for starting up chemical methanation. The aim during start-up is the same, to burn as little product gas as possible in the flare that does not fulfil the injection conditions. The starting conditions for biological methanation are also that the reactor was set in the range of operating temperature and operating pressure. A hydrogen atmosphere is not necessary. The gas from the last production phase can remain in the head of the reactor. The catalyst or the material will not be damaged. Thus it can be started directly with the quality of the previous run. Thanks to that, no gas has to be flared during start-up, if the correct feed gas ratio is used from the outset. In Solothurn, the methane quality often collapsed shortly during start-up, because the reactor pressure was readjusted with pure hydrogen, when the plant was shut down.

Carbon dioxide and hydrogen are added together in stoichiometric ratio and react to methane and water. The condensation of the produced water in the reactor leads to an increase of level in the reactor. The water is continuously discharged from the reactor, and the continued production of methane causes the pressure in the reactor to rise. A pressure control valve keeps the pressure in the reactor constant to 10 bar. This has been designed relatively large and switches slow in the lower control ranges. Another pressure control valve downstream is used to control the inlet pressure before the injection and the flare. This two-stage control can lead to a fluctuating volume flow measurement in the injection pipe due to overlapping of two control units (see SNG flow in Figure 4-5). This can also cause the interruption of the SNG flow. The SNG flow is interrupted when the SNG in Figure 4-5 is zero and the produced gas is sent to the flare. Depending on the pressure in the reactor and in front of the injection, the length of interruption varies.

When the injection conditions are met, the gas is directed to the injection point, and the flare is in operation with a pilot flame. The duration of the transition from hot standby until the injection criteria are met depends on the quality of the product gas before the last interruption. The case shown in Figure 4-5 started with a methane concentration of 98 vol.-% of the last period, and reached therefore the required injection quality immediately. For methanation on an industrial scale, the same procedure and design would be used in order to avoid flaring product gas.

The fact that the methanation technology can handle load changes has been proven in various tests. The load profiles for the tests are shown in Appendix 8.2.

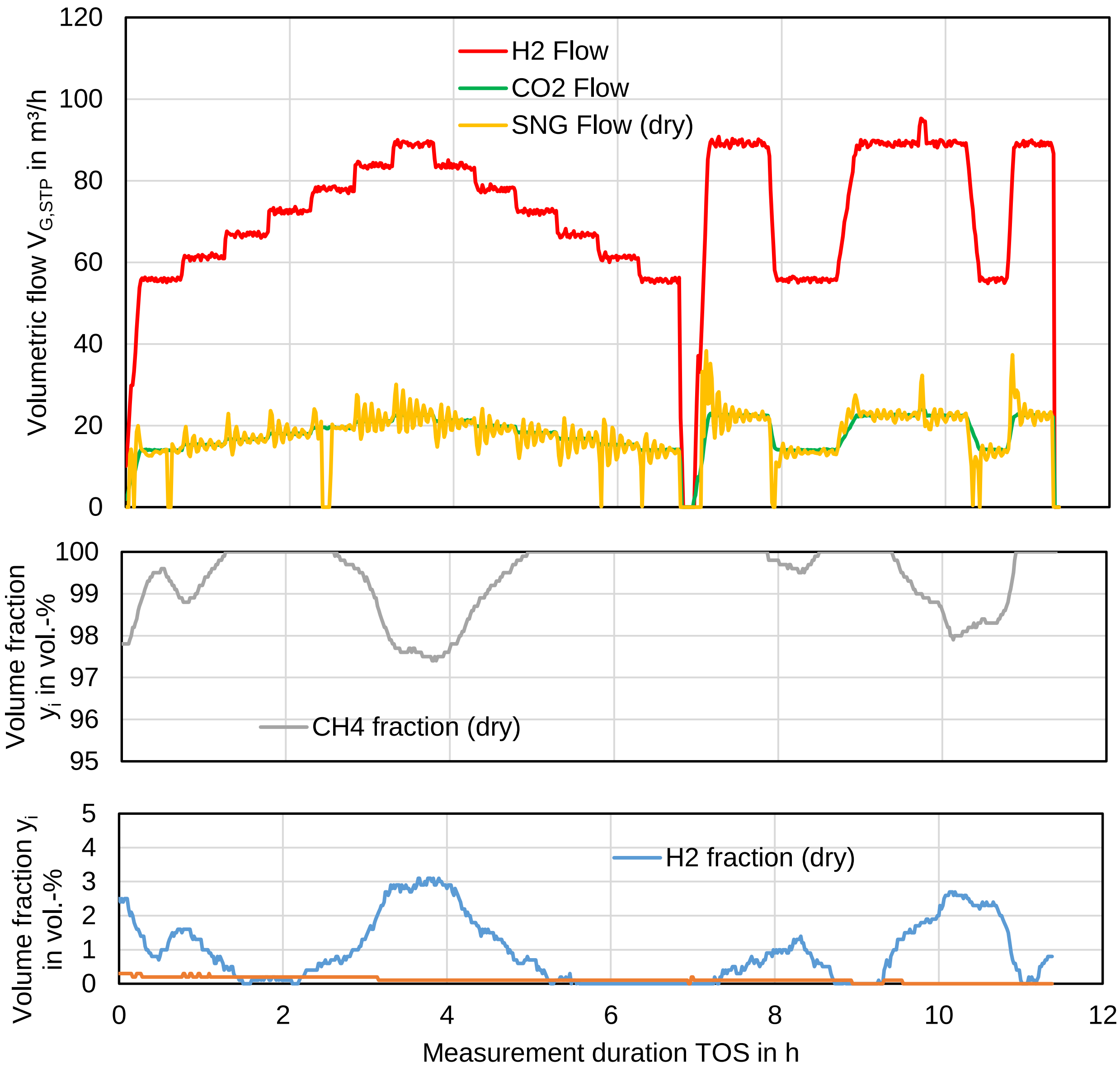

Figure 4-5: Test for technical characteristics (TC001) and Stress test (ST008) of the Solothurn plant

The aim of the load profiles shown in Figure 4-4 was on the one hand to determine the partial load efficiencies and to check how dynamic the load changes can be. It was successfully shown, that the product gas quality hardly changes for all loads and load change rates. The load changes between 45 and 75 % with a step size of 5 %-points every 30 minutes. Even after immediate shutdown, a 15 minute break and a start-up from 0 to 75 % within 15 minutes took place without quality loss. Also with load changes between 75 and 45 % and vice-versa with a step size of 30 %-points per 5 minutes and 15 minutes vice-versa, the injection quality was maintained throughout. Short-term load changes have hardly any influence on the gas quality.

The test program further showed that the plant can be operated over a long period without major complications. Individual process engineering flaws could be evaluated and would have to be designed differently when the plant was rebuilt (for example: flare, dosing skid). The detailed optimization potentials were analysed in the confidential Deliverable D3.6.

Other important parameters regarding the operational flexibility (deployment time, transient tests) or long-term durability (stationary tests) of the methanation unit can be looked up in Deliverable D3.5 (confidential). Summarized, the biological methanation copes with load changes from 40 to 95 %with instant load change rates between 1.8 and 4.2 %/min without loss of quality. In another experiment, an LCR of 5.5 %/min. was applied directly from hot standby to 60% load. The high load change rate is possible because the GHSV of methanation is very low and the liquid phase is a buffer for the changes to the feed.

4.2.2 Energetic Optimization

In this section the energetic optimization potential for the Solothurn plant is discussed in detail. Table 4-6 shows the energy demand for the overall PtG-process chain based on the SNG output. The biggest consumer of electrical energy is by far the electrolyser ($\eta_{\mathrm{Ely,H2}} = 60\ \%$). Compared to the SoA (SoA, $\eta_{\mathrm{Ely,H_2}} = 76\ \%$), the efficiency is low, which results in a high potential of heat usage from the electrolyser. The energy demand of the methanation is $P_{\mathrm{el,Meth}} = 1.1\frac{\mathrm{kWh}}{\mathrm{m^3}}$, which corresponds to approx. 10 % of the energy content SNG.

Further, the Table 4-6 indicates some optimization potential. The efficiency of the electrolyser can be increased (assumption SoA incl. BoP: $\eta_{\mathrm{Ely,H_2}} = 76\ \%$). Thus, the energy demand for the production of H_2 sinks. Assuming that the overall efficiency of the electrolyser ($\eta_{\mathrm{Ely,H_2+therm}} = 92\ \%$) stays constant, the potential heat usage decreases by more than 50 %. Additionally, there is some potential to optimize the energy demand for the methanation. On the one hand the energy demand of the CO_2 compressor can be improved. The size of the compressor is too big, which leads to a less-than-ideal control strategy. Based on the ASPEN results, the energy demand can be reduced up to 70 %. Furthermore, the operation strategy and insolation of the plant can be improved to reduce or avoid trace heating. This would decrease the electrical energy demand by 0.25 kWh/m³ based on the SNG output.

Table 4-6: Energy demand for the overall PtG process chain (see block flow diagram in Figure 3-1) for the Solothurn plant based on the measurement (01.23.2020, 2:30 am to 3:30 am (local time)). The energy data are based on the SNG-Output

	CO_2-Source (Membrane Separation)	**H_2 Source (Electro-lyser)**	**Methanation**	**Injection**	**Unit**
Initial situation					
Energy demand	1.3	23.8	1.0	0.1	kWh/m³
Heat usage potential		7.6			kWh/m³
Optimization					
Energy demand	0.0	18.8	0.5	0.0	kWh/m³
Heat usage		2.9	3.0		kWh/m³

The effect of the optimization potential on the overall PtG efficiency is shown in Table 4-7. By decreasing the energy demand for the methanation and using the heat from the methanation reactor, the overall efficiency can be increased to 89 %. Since the biological methanation is near to the 'Hybridwerk', it would be possible to use this heat in the case of Solothurn. This is an example of a nearly ideal integration of a PtG process in an overall energy system. By optimization of the electrolyser, the efficiency is almost the same. But the share of produced SNG is increased, and the waste heat production is decreased. This can be explained by the assumptions which were made. For the optimization of the electrolyser, a higher efficiency based on the H_2 output was taken (SoA: $\eta_{\mathrm{Ely,H_2}} = 76\,\%$). But the overall efficiency of the electrolyser including H_2 and heat output stays constant. It is worth to mention that the overall energy demand for the production of SNG can be decreased by 5 kWh/m³ based in SNG output by increasing the H_2 production efficiency.

Table 4-7: Effect of optimization on the overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$

Case	**Overall PtG efficiency** $\boldsymbol{\eta_{\mathrm{PtG,HHV,ov}}}$
Initial situation from Table 4-4	76 %
+ Optimized energy consumption methanation	77 %
+ Including heat usage Methanation ($T_{\mathrm{use}} = 50\,°\mathrm{C}$)	89 %
Optimized electrolyser	
+ without heat usage (SoA, $\eta_{\mathrm{Ely,H_2}} = 76\,\%$)	73 %
+ with heat usage ($\eta_{\mathrm{Ely,therm}} = 17\,\%$)	88 %

4.3 Demo Site Troia

After the plant in Troia initially had to overcome a few difficulties, the plant could be operated constantly from October 2019 onwards. As mentioned above, the Troia site was comprised of four demonstration units, and this involved facing many challenges in getting the four interconnected plants to work together continuously. In Figure 4-6 a block flow diagram of the overall process configuration is shown. The CO_2 was provided by a direct air capture (DAC), and the H_2 was produced in an AEL. The feed gas was mixed and sent to the methanation unit. The methanation unit was a single stage methanation. After the reactor, the gas was separated by means of a membrane system in front of the liquefaction. The product gas (high methane fraction) was processed in the liquefaction. The lean gas (lower methane fraction) was compressed and fed to the inlet of the methanation reactor (recycle A). The boil-off gas from the liquefaction was also recycled and fed to the methanation (recycle B).

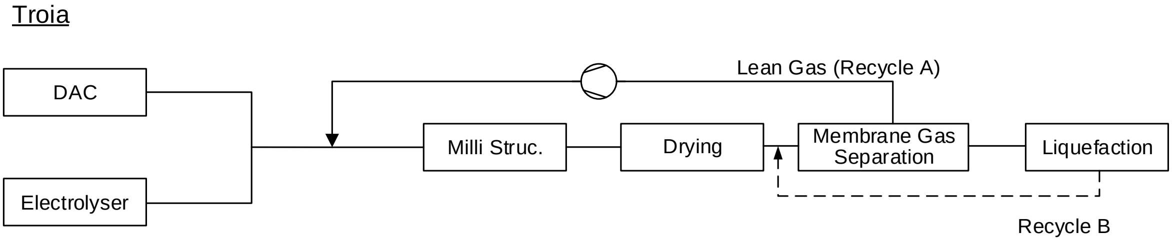

Figure 4-6: Simplified Block flow diagram of the PtG / PtLNG plant in Troia

CO_2 Conditioning via Direct Air Capture

Due to the challenges during the commissioning of the overall plant, the DAC was also operated stand-alone. Figure 4-7 shows the mass of harvested CO_2 per cycle normalized to the average. Due to the fluctuation of the weather conditions (temperature and humidity), the CO_2 mass flow fluctuates up to 15 %. In Troia, the DAC was operated in broad weather conditions. The ambient temperature ranges from approx. 5 °C (relative humidity up to 90 %) to 30 – 35 °C at low relative humidity of 20 – 40 % (see Figure 8-7). Since the DAC is a circular process and the harvested mass per cycle varies, the data are integrated over time. In average, the DAC provides 12 kg/h of CO_2 to the methanation. The CO_2 is delivered with a high purity of y_{CO2} > 99.9 vol.-%. Afterwards the CO_2 is compressed to 10 bar and is stored in a buffer vessel.

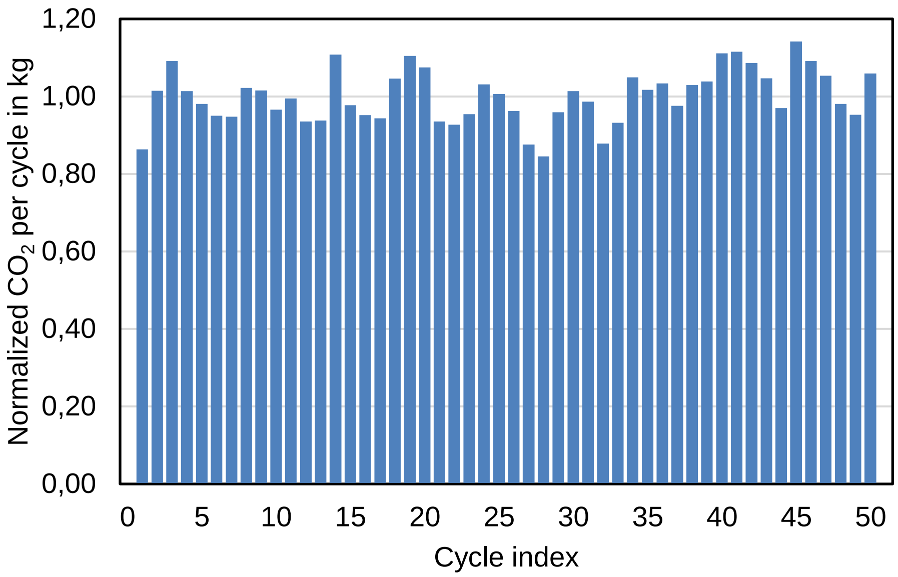

Figure 4-7: Measurement data of the direct air capture in Troia during a period in April 2019. The data are normalized to the average separated CO_2 mass per cycle

Methanation Unit

After the H_2 and CO_2 storage vessels, the gases were mixed and fed into the methanation unit. During the first 200 hours of operation, the product gas quality was in the range of $y_{CH4} \approx 80$ vol.-%. After the first 200 h, the methane fraction dropped noticeably to approx. 50 vol.-% of CH_4. A hydrogen leakage had been formed, and consequently a carbon deposit was suspected. Hence, the reactor was removed from the methanation unit and the leak was welded. After the welding works adjustments in the shutdown process were performed, in which the reactor was flushed sufficiently with pure hydrogen. Due to the hydrogen flushing, the catalyst was regenerated and a high methane fraction was achieved again.

In Table 4-8 the important performance indicators for the methanation are shown for a constant operation of 20 h at 80 % of load. The feed gas (CO_2, H_2) stoichiometry is 4. When the plant is operated without recycle (see Figure 4-6), the volumetric methane fraction after the reactor is in the range of $y_{CH4} = 80 - 90$ vol.-% (load 25 – 50 %). At constant reaction temperature, the quality depends on the load. If the load is increased to 80 % ($\dot{V}_{H2,STP} = 32$ m³/h), the methane fraction drops to 80 vol.-%. This indicates that the thermodynamic equilibrium is not reached in the reactor. Due to the milli-structured reactor concept, the conversion could be optimized by adjusting the reaction temperature. The gas hourly space velocity at 80 % load is GHSV = 9100 1/h[8]. At 100 % load ($\dot{V}_{H2,STP} = 40$ m³/), this would increase to a comparably high GHSV of 11 400 1/h.

[8] Based on the fed gas from the H_2 and CO_2 storage vessel. The additional recycle flow is not considered.

Table 4-8: Comparison of PIs of the Troia demo site. The PIs based on the ASPEN model are compared to PIs calculated from measurement data (02/12/2020 – 02/13/2020, 2:00 pm – 10:00 am). The definition of the PIs are given in section 3.2 and 9.1.

Performance Indicator	**Measurement data** $\dot{V}_{\mathbf{H_2,STP}} = \mathbf{32\ m^3/h}$	**ASPEN model** $\dot{V}_{\mathbf{H_2,STP}} = \mathbf{32\ m^3/h}$	**Unit**
PtG plant H_2 input	113	113	kW
Educt stoichiometry (inlet)[9]	4.00	4.0	-
SNG output (only CH_4)	87	88	kW
Electricity power demand methanation plant	55	26	kW
Volumetric flow of CO_2 from DAC	8	8	m³/h
Heat demand DAC	36	36	kW
Electricity power demand DAC	20	20	kW
Electricity power demand lique-faction	18	18	kW
GHSV (methanation) ($V_{\mathrm{cat}} = 4.4 \cdot 10^{-3}\ \mathrm{m}^3$)	9090	9089	1/h
CO_2 conversion (methanation re-actor)	99	100	%
H_2 conversion (methanation reac-tor)	91	96	%
Methane fraction (after methana-tion reactor)	73	78	vol.-%
Methane fraction in front of mem-brane	67	88	vol.-%
Methane fraction in front of lique-faction	96	100	vol.-%
Methane fraction in recycle A	42	51	vol.-%
Hydrogen fraction in recycle A	57	48	vol.-%
Methane fraction recycle B	92		vol.-%
Heat usage	0	17	kW
Temperature level of heat usage	290	290	°C
Methanation efficiency $\eta_{\mathrm{SNG,HHV}}$	77	77	%

[9] This educt stoichiometry refers to the inlet of H_2 and CO_2 in front of the mixing of the recycle. The H_2/CO_2 ratio in front of the reactor is higher due to the mixing in the recycle.

Methanation efficiency with heat usage $\eta_{\mathrm{SNG,HHV,T_{use}}}$	77	92	%
Overall methanation efficiency $\eta_{\mathrm{SNG,HHV,ov}}$	52	75	%
Overall methanation efficiency with energy demand for CO_2-conditioning $\eta_{\mathrm{SNG,HHV,ov,CO_2-con}}$	39	53	%
PtG efficiency $\eta_{\mathrm{PtG,HHV}}$ ($\eta_{\mathrm{Ely}} = 65.6\ \%$)	31	41	%
Overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$ ($P_{\mathrm{el,Liq}} = 3.4\ \mathrm{kWh/kg}$)	29	38	%

When the plant was operated with recycle, the product gas was treated in a gas separation unit. The lean gas from the separation unit and the feed gas were mixed in front of the reactor. Due to membrane separation, the methane fraction in the retentate (product gas in front of the liquefaction) increases to y_{CH4} = 96 vol.-%. This proves the capability to produce SNG with a methane content of higher than 90 %, which fulfils the goal of the project. The CO_2 fraction in the retentate is y_{CO2} < 0.5 vol.-%. The H_2 rich permeate (see Figure 4-6, recycle A (lean gas): y_{H2} = 57 vol.-%) is compressed and mixed with the feed gas in front of the reactor. Due to the recycle, the GHSV in the reactor is increased. Hence, the methane fraction after the reactor drops to $y_{CH4,dry}$ = 73 vol.-% (gas analyser in methanation unit, see Figure 4-8 (average)). The measurement in front of the membrane (gas analyser in liquefaction unit) indicates a methane fraction of y_{CH4} < 70 vol.-%. The methane fraction after the reactor in the ASPEN results is higher ($y_{CH4,dry}$ = 78 vol.-%), since thermodynamic equilibrium is assumed after the reactor.

Within the reactor, the CO_2 conversion is $X_{CO2} \approx 99$ %. Since the recycle A (lean gas) contains almost 60 vol.-% of H_2, the stoichiometric ratio in front of the reactor is shifted to higher than 4. Hence, the H_2 conversion in the reactor is only $X_{H2} \approx 92$ %.

To avoid clogging of the liquefaction process, the product gas (retentate from gas separation) was treated in an adsorption unit. Only a small amount of CO_2 is left in the gas after the product gas after the membrane separation unit (y_{CO2} < 0.5 vol.-%). This CO_2 was separated via adsorption and is discharged (CO_2 and H_2O separated were sent to the flare) from the process. Due to the low losses of CO_2 (in the range of 1 vol.-% of the inlet CO_2) in the adsorption in front of the liquefaction, almost the complete CO_2 is converted to CH_4. The CH_4 output is $\dot{V}_{CH4,STP}$ = 7.9 m^3/h. By means of the recycle, also hydrogen is almost completely converted in methanation process.

Liquefaction Unit

After the membrane separation and the adsorption unit, the fine cleaned methane stream is liquefied in the liquefaction process. The recycle B contains the boil-off gas from the liquefaction process. The measurement results indicates a lower methane concentration in the recycle B (boil-off) than the inlet gas to the liquefaction. This is reasonable, since the boil-off gas contains a slightly higher amount of hydrogen than the inlet gas to the liquefaction process. The recycle B is compressed and mixed with the product gas from the reactor in front of the separation unit.

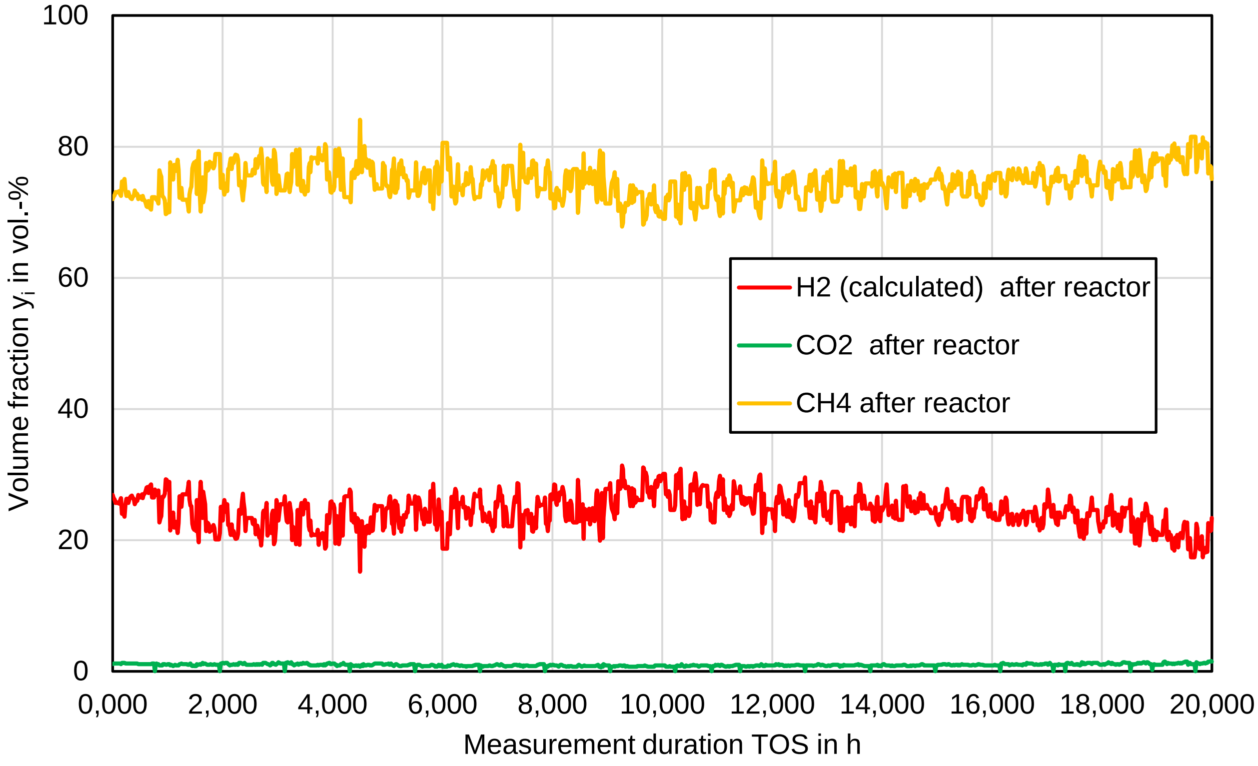

Figure 4-8: Measurement results from the demo site Troia: CH_4, H_2 and CO_2 fraction (dry) after the reactor (02/12/2020 2:00 pm – 02/13/2020 10:00 am). Operation conditions: Load: 75 %, $T_{oil,out,top}$ = 290 °C, $T_{oil,out,bottom}$ = 310 °C, Recycle A and B in operation.

Energetic Analyses

In the following, the energy demand of the four main units are analysed, as well as the efficiency of the methanation process and the overall PtG efficiency (resp. power-to-LNG efficiency). The energy demand of the units is presented in Table 4-8. Additionally, in Table 4-9 the energy demand normalized to the SNG output in front of the liquefaction unit is given.

The electrolyser is the biggest consumer of energy. The energy demand is 5.4 kWh/m³ based on the H_2 output. This corresponds to an efficiency of 66 %. Due to the low CO_2 concentration in the air, the DAC is the second biggest consumer. In Figure 4-9, the energy demand per cycle is shown. As for the CO_2 mass harvested per cycle, the energy demand per cycle varies, due to weather conditions and the amount of adsorbed CO_2. As for the mass flow, the energy demands are integrated over time. The average thermal and electrical energy demand are 2.3 kWh/kg and 1.3 kWh/kg based on CO_2, respectively. It is important to note that significant parts of the DAC plant's central process unit were oversized, leading to increased losses compared to what had been demonstrated in previous installations.

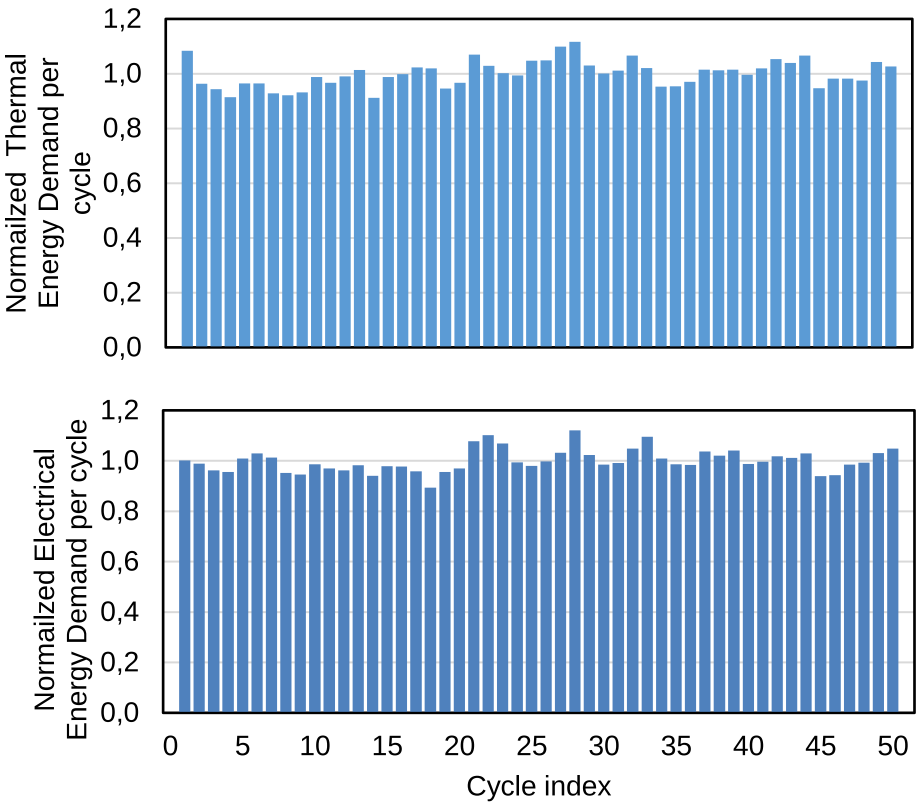

Figure 4-9: Thermal and electrical energy demand of the direct air capture in Troia during a period in April 2019. The data are normalized to the average thermal and electrical energy demand, respectively.

To calculate the efficiencies of the process, all the energy consumers are normalized to the produced SNG (resp. LNG). This corresponds to 87 kW. The calculated efficiencies are given in Table 4-8.

From the H_2 input and the CH_4 output, the methanation efficiency gives 77.4 %, which is close to the ASPEN results and the thermodynamic maximum. This high efficiency of the methanation with only one reactor is only possible due to the overall conversion of almost 100 % due to the recycle of the lean gas. Due to the high electrical energy consumption of the pilot methanation unit (3.3 kWh/m³ based on SNG output), the methanation efficiency drops to 52 % (compare D4.11). The energy demand of the methanation is very high compared to the state-of-the-art. On the one hand, this can be explained by the high heat losses of the reactor. The heat losses emerge from a non-ideal insulation. On the other hand the electrical energy demand includes the compressors of the two recycles. In general, the realized methanation is very small, which increases the specific BoP and heat losses. The given value for the ASPEN simulation (26 kW) represents the reachable energy consumption (see section 4.3.2).

The integration of reaction heat from the exothermic methanation reaction in the DAC unit – an essential advantage of the plant's concept with CO_2 capture directly on site – was demonstrated in the last week of the plant operations at Troia. However, it was not tested sufficiently to provide accurate quantitative data to be considered in the present analysis. Including the energy demand for the DAC and the H_2 production, a PtG efficiency of 31 % could be reached. Compared to the injection of the produced SNG into a gas grid, the energy consumption for the liquefaction is higher. In the end, an overall PtG efficiency of 29 % is reached.

Due to the innovative character of the overall process chain and the relatively small capacity of 0.1 MW SNG/LNG output, a huge potential for energetic improvement exists. From the ASPEN results it can be concluded, that the heat integration of DAC and methanation alone could increase the efficiency to 38 %. All the potentials to increase the efficiency are discussed in detail in section 4.3.2.

4.3.1 Dynamic Operation / Test Program / Long Term Operation

As described in section 3.3, the dynamics and flexibility of the catalytic milli-structured reactor with subsequent membrane system were tested in the course of several measurements.

First the start-up procedure is described. When starting up from hot standby until the maximum gas quality after the reactor is reached, the following steps are performed. The aim during start-up is to burn as little product gas as possible in the flare that does not fulfil the required quality. Starting conditions for start-up are that the reactor has been adjusted in the range of operating temperature and operating pressure under hydrogen atmosphere. Then carbon dioxide and hydrogen are added and react to methane and water. Due to a relatively high GHSV and good cooling of the reactor, the time of the start-up is quiet fast. Since methanation start-up has never been tested at the same time as recirculation, the amount of rejected lean gas cannot be determined. The start-up strategy could be adapted and the same procedure could be chosen as in Falkenhagen. However, for this purpose, the composition of the recirculate would have to be known in order to maintain the stoichiometric ratio before the gases enter the reactor.

In Figure 4-10 the results of a constant operation at 50 % load are shown without recycle. At TOS = 2 h, the load was decreased from 80 % to 50 %. Until TOS = 18 h, this load was held constant. In the lower figure, the gas composition after the reactor is shown. Since the gas after the reactor was flared, the volumetric flow after the reactor is zero. It was observed that the product gas quality dropped from 85 vol.-% to 82 vol.-% (CH_4 fraction) within 12 hours. The cause could not be determined from the measured data. The flow rates, the temperature of the cooling circuits and the operating pressure remained unchanged. The outside temperature did not change simultaneously with the gas quality. A reversible carbon deposition is possible, but unlikely, since the quality increases again after a few hours without any apparent changes. Laboratory tests have shown a time-dependent deactivation of the catalyst. When product gas is recirculated, the product gas quality decreased. That is because the feed gas flows of H_2 and CO_2 were not adjusted to keep the stoichiometry of approx. 4:1 constant. The methane concentration drops due to hydrogen surplus. This process must be better regulated in future.

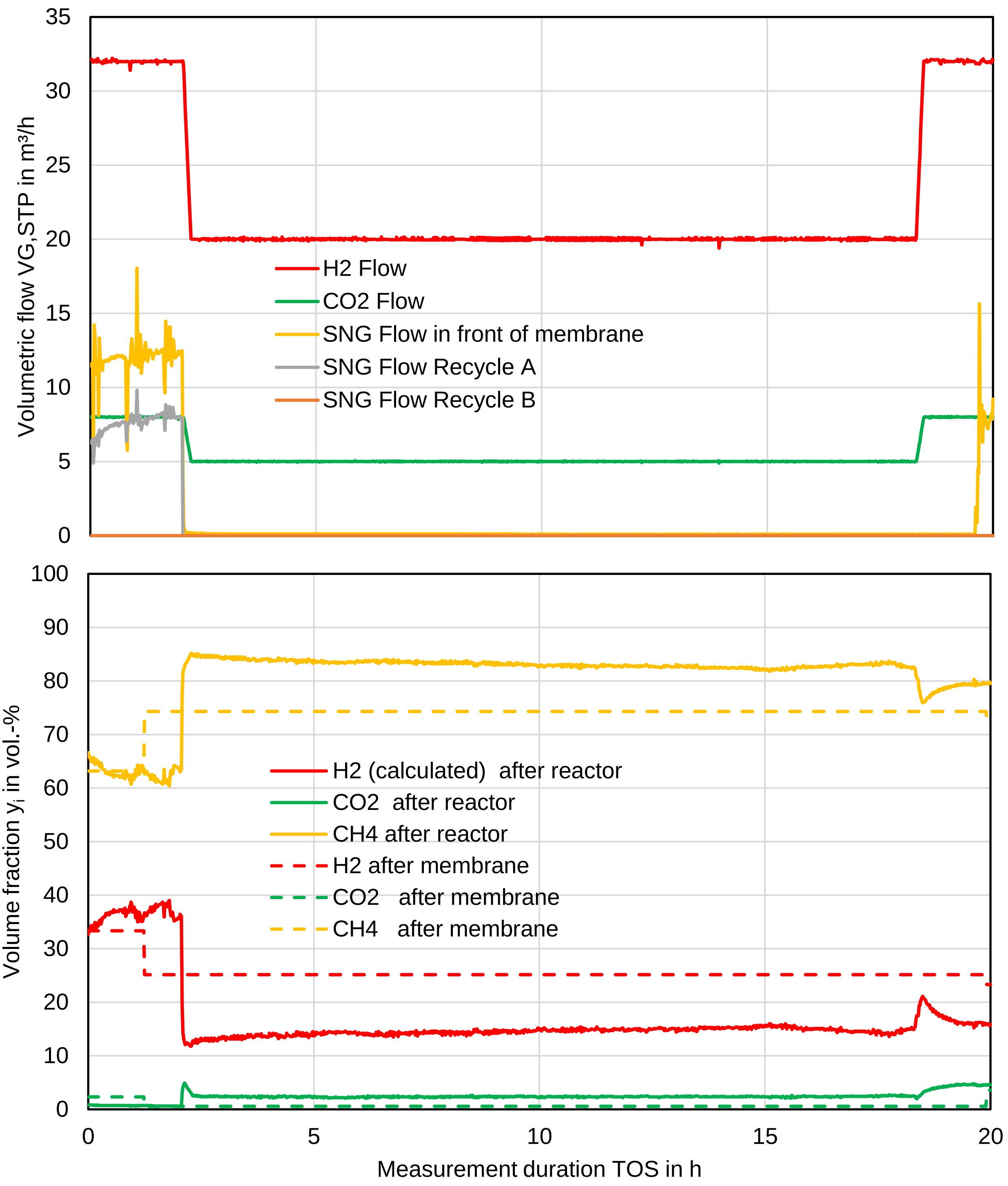

Figure 4-10: Test program Troia; Constant operation and gas recycle (01/27/2020 3:00 pm – 01/28/2020 11:00 am).

The fact that the methanation technology can handle load changes has been proven in various tests (see Figure 4-12). The load profiles for the tests are shown in Appendix 8.2. In Figure 4-12, the red and green line in the upper diagram show the load cycles of the hydrogen and carbon dioxide flow and in the lower two diagrams the corresponding gas composition measured in the product gas after the milli-structured reactor (continuous lines). The gas quality after the membrane system (dotted line) is only measured when the recirculation (green or purple line in the upper diagram) are active. The load change rate analysis is shown for the load change from 20 to 80 % of load (see Figure

4-12, TOS = 4 h). It was proven that a load change rate of 5 %/min is possible. During this load change, the methane fraction only shortly drops to 76 vol.-%. After less than 5 min the methane fraction is stabilized at $y_{CH4} \approx 80$ vol.-%. The slight drop of the methane fraction should not be a problem, since the membrane separation unit can increase the methane fraction to more than 95 vol.-% of methane. The data in Table 4-8 indicate that the membrane unit can increase the methane fraction from 69 to 96 vol.-% at 80 % of load.

In general, the load changes are smooth, and the quality of the product gas after the milli-structured reactor follows the load changes. This in turn indicates that the thermodynamic equilibrium is not reached within the reactor. A lower load, which corresponds to a longer residence time, increases the methane concentration in the product gas and vice versa. This means that the subsequent membrane must be designed for a wide range of gas compositions in order to achieve the required quality.

The goal that the methanation can be operated between 20 and 80 % of load was proven within the STORE&GO project. It was also proven that the load change range is in the range of 5 %/min, which meets the challenges at the start of the project.

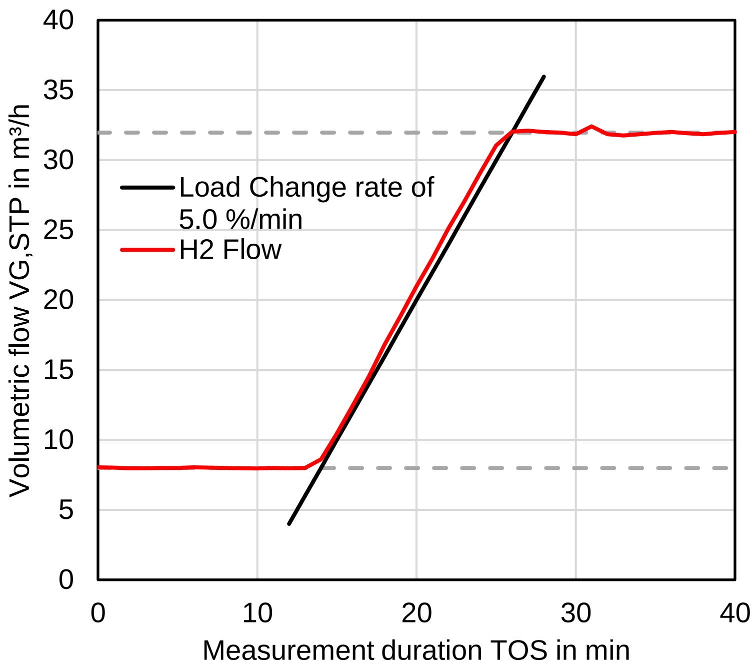

Figure 4-11: Analyses of the load change rate based on measurement on the 01/20/2020 (see Figure 4-12). Load changes from 20 to 80 %. The black line indicates the mean load change rate.

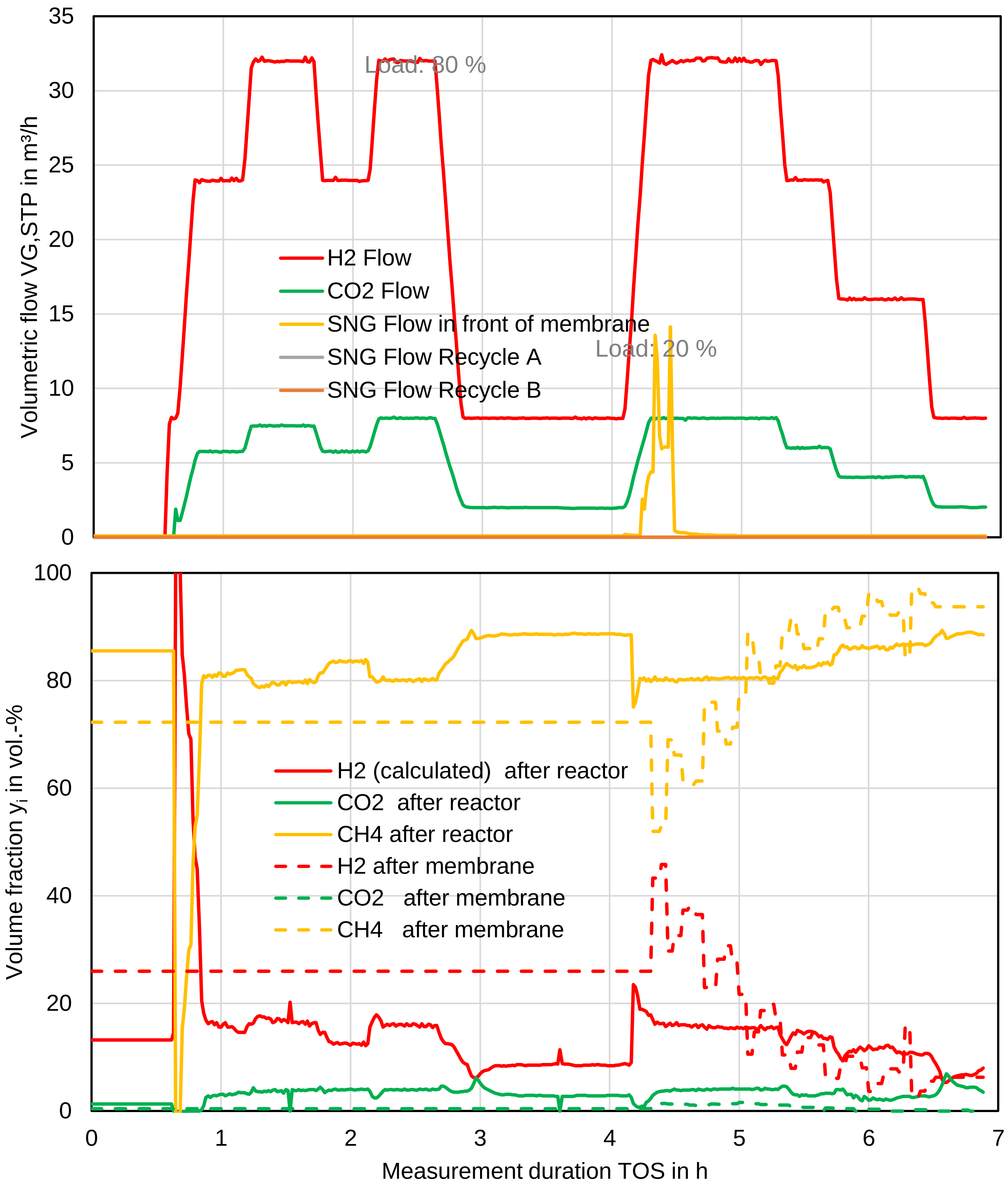

Figure 4-12: Test program Troia on the basis of ST008 (01/20/2020 11:00 pm – 01/20/2020 5:53 pm).

4.3.2 Energetic Optimization

As discussed earlier, there is a huge potential to decrease the energy demand for the process units, and thus to increase the overall PtG efficiency. In Table 4-9, the energy demand for the four main units are given. For comparison, the given values are normalized to the SNG output which is fed to the liquefaction.

In Deliverable D4.11, the optimization potentials for the process chain are discussed in detail. To reduce costs within the project, the DAC the design of all the instruments (pumps etc.) is based on a DAC plant with 6 adsorption units. Therefore it is oversized for the 3 implemented adsorption units, and the energy demand is relatively high compared to what had been demonstrated in previous installations. It is stated in D4.11 that the overall energy demand can be reduced by 46 %. If a SoA electrolyser is installed, the energy demand for the H_2 production could also be decreased. A further potential is the heat usage of the waste heat from the electrolyser, e.g. in the DAC unit, which is not considered in this Deliverable.

In the methanation unit, the energy consumption could be decreased by reducing the energy losses to the environment. Since the capacity of the methanation unit is very small compared to e.g. Falkenhagen, the BoP could be further decreased for large scale plants. Like for the DAC, the liquefaction is oversized and only operates at 12 – 20 %. If the liquefaction is operated at nominal load, the energy demand could be decreased by almost 75 %.

When heat integration is considered, the thermal energy demand for the DAC can be reduced by the potential heat, which can be extracted from the methanation reactor.

Table 4-9: Energy demand for the overall PtG process chain (see block flow diagram in Figure 3-1) for the Troia plant based on the measurement data (01/12/2020 - 01/13/2020, 2:00 pm – 10:00 am). The energy data are based on the SNG-Output

	CO_2-Source (DAC)	H_2 Source (Electrolyser)	Methanation	Liquefaction	Unit
Initial situation					
Energy demand	7.2	21.9	3.3	2.2	kWh/m³
Heat usage potential (without optimization)			2.1		kWh/m³
Optimization					
Energy demand	3.9	19.6	1.9	0.6	kWh/m³
Energy demand with heat integration	1.9	19.6	1.9	0.6	kWh/m³

The effect of the optimization of the four main units can be seen in Table 4-10. If all the optimization potentials are realized, the overall PtG efficiency could be increased to 46 %. It needs to be said that this is not the maximum efficiency which can be reached with this overall process chain. There is further potential for heat usage:

- Cooling of the product gas after the reactor

- Waste heat from the electrolyser in the DAC (requires heat at 95 – 100 °C). Theoretically, the entire heat demand of the DAC can be provided by excess heat from electrolysis and methanation.

Also the BoP will be lower if the capacity of the overall process chain is increased. But it must be kept in mind that this process chain differs strongly from the Solothurn and Falkenhagen plant. The CO_2 source has to deal with much lower CO_2 concentration (400 ppm, i.e. 0.04 vol.-% instead of higher than 40 vol.-%). And the produced CH_4 is liquefied. Both process steps need a high energy input, which reduces the efficiency, but they make the plant a stand-alone solution even for remote locations.

Table 4-10: Effect of the optimization on the overall PtG efficiency $\eta_{\mathrm{PtG,HHV,ov}}$

Case	**Overall PtG efficiency** $\boldsymbol{\eta}_{\mathbf{PtG,HHV,ov}}$
Initial situation from Table 4-9	29 %
+ Optimization of the electrolyser	31 %
+ Optimization of the DAC	33 %
+ Optimization of the Methanation	40 %
+ Optimized of the Liquefaction	42 %
+ Including heat integration (reaction heat to DAC)	46 %

5 Economic Evaluation

Besides the technical evaluation, the demo sites were also economically assessed within the STORE&GO project. One goal of this project is to reduce the $CAPEX_{Meth}$ by 15 % compared to the state-of-the-art technologies. In a first step, calculations of the current investment costs of the methanation units ($CAPEX_{Meth}$) were performed, followed by a future perspective of costs development until 2050 using learning curves from EIL, see Deliverables D7.5 and D7.7. In the section 5.3 the calculations of the production costs for methane or rather LNG are shown. These calculations consider the entire process chain – from the CO_2/H_2 source to the injected/liquefied methane and are based on optimized cost estimations. .

5.1 Investment Costs for the different Methanation Units

In the first step of the economic evaluation the current capital expenditure (CAPEX) of the different methanation units of the demo sites were determined by using the ADD-on factor method, as described in section 3.6. In the $CAPEX_{Meth}$ calculations only the methanation units were considered. Figure 3-1 shows a simplified block flow diagram of the demo sites. The process units framed with the grey dashed line are included in the $CAPEX_{Meth}$ calculations. At first, the specific $CAPEX_{Meth}$ of each demo site was calculated. Afterwards, the $CAPEX_{Meth}$ was scaled to a methanation unit with an SNG/LNG output of 1 MW and 5 MW. As an example, the specific $CAPEX_{Meth}$ calculations of the demo site in Falkenhagen are shown in Figure 5-1.

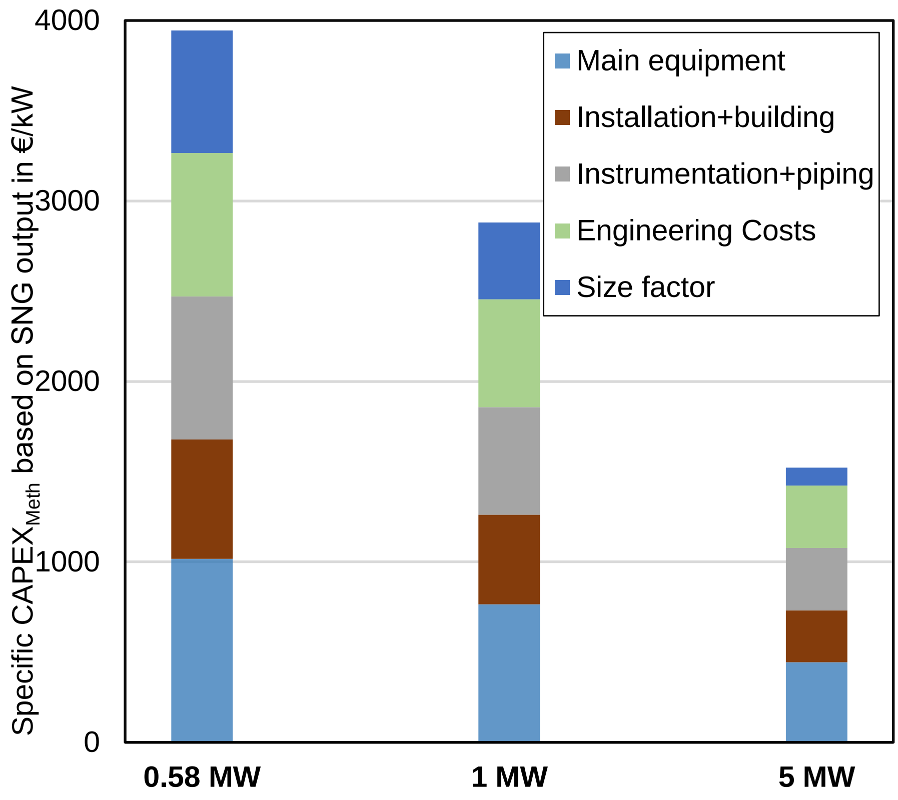

Figure 5-1: Specific CAPEX of the Falkenhagen methanation unit and the specific $CAPEX_{Meth}$ scaled to 1 MW and 5 MW based on the SNG output.

For the Falkenhagen demo site, the total specific $CAPEX_{Meth}$ is 3740 €/kW. By increasing the plant size or rather the SNG output, the specific $CAPEX_{Meth}$ is reduced. As an example, the specific $CAPEX_{Meth}$ of the demo site scaled to an SNG output of 5 MW is decreased to 1430 €/kW. Also, as shown in Figure 5-1, the total costs can be split into different factors. The bar chart shows that the costs of the size factor decreases with increasing SNG output, whereas the shares of the main equipment costs remain in the same range.

The $CAPEX_{Meth}$ calculations for the Solothurn and Troia methanation units are described in detail in Deliverable D5.5. For Solothurn, the $CAPEX_{Meth}$ is 4320 €/kW for the demo site and 940 €/kW for a plant with an SNG output of 5 MW. For Troia, the $CAPEX_{Meth}$ is 5400 €/kW for the demo site and 1090 €/kW for the methanation unit scaled to 5 MW based on SNG output. A closer look on the results is shown in Figure 8-8 in the appendix.

In a second step, the plant design optimization discussed in chapter 4 were included in the $CAPEX_{Meth}$ calculations. As already described in section 4.1.2, the methanation unit of the Falkenhagen plant could be optimized regarding the compression of the gas inlet flow and the required catalyst amount. Afterwards, the optimized $CAPEX_{Meth}$ had been calculated for a plant scaled to 10 MW and 50 MW SNG output. The diagram in Figure 5-2 gives an overview of the various optimization steps and their impact on the specific $CAPEX_{Meth}$.

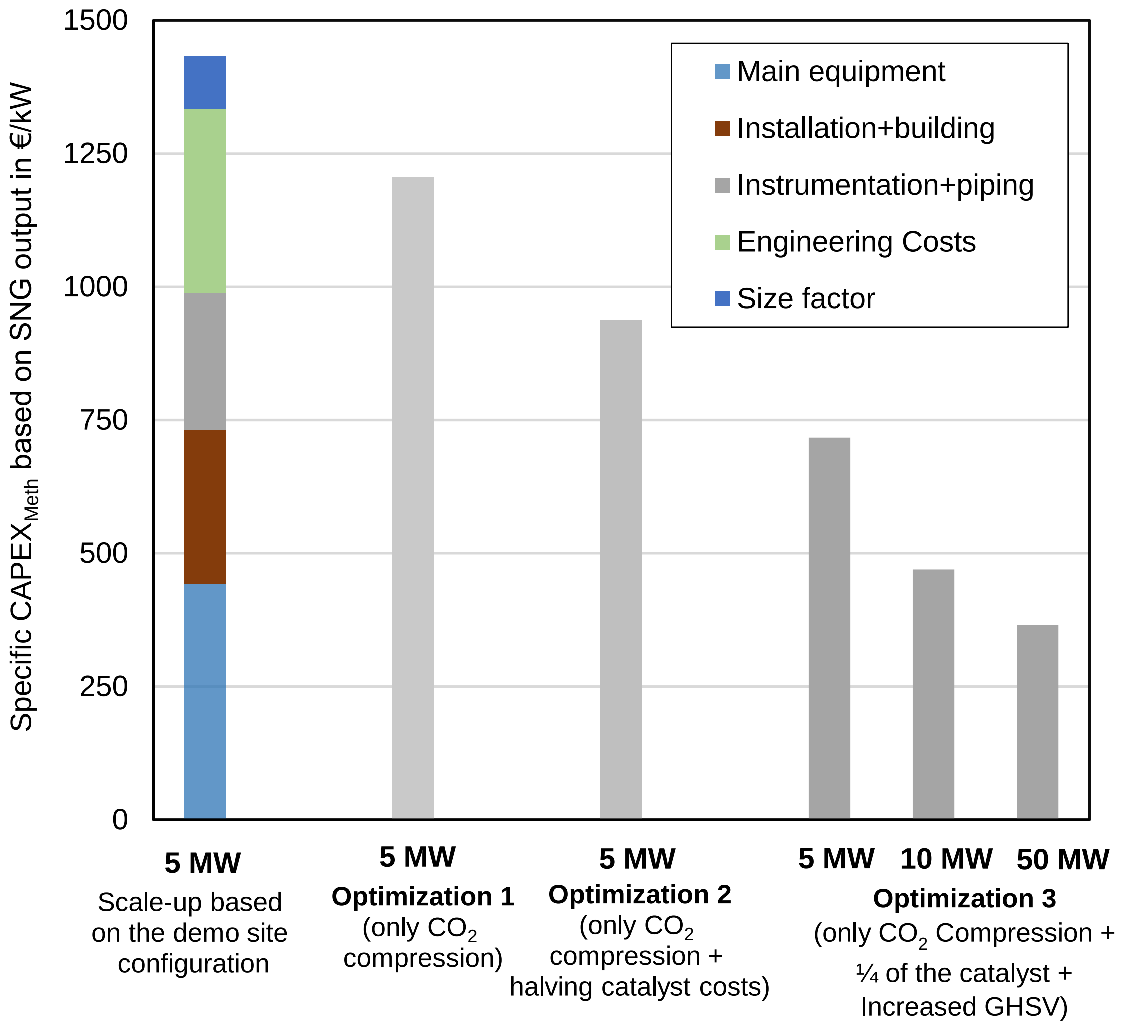

Figure 5-2: Optimized $CAPEX_{Meth}$ calculations for a scaled Falkenhagen methanation unit

As seen in the diagram in Figure 5-2, the first optimization step includes the improvement of the compression of the volumetric flow into the reactor. In Figure 4-1, a simplified block flow diagram of the plant in Falkenhagen is shown. It can be seen that in the current plant design the H_2 and the CO_2 were first mixed and then compressed to the desired reaction pressure of 14 bar. Building a new plant inclusive of the electrolyser, the outlet pressure of the H_2 stream from the electrolyser would be adjusted to the pressure level of the methanation. This leads to a reduction of the volumetric flow which needs to be compressed, resulting in a smaller compressor. As shown in the diagram (Figure 5-2), the specific $CAPEX_{Meth}$ of a 5 MW methanation plant can be reduced to 1200 €/kW, if the compression of the inlet volumetric flow into the reactor is improved. This means a cost reduction of 16 %. In a next step, the catalyst were reduced, since only 2 instead of 4 monolith structures per tube are sufficient. Originally, the reactor was designed with 2 monolith structures per tube. However, in order to have a catalyst reserve in case of a catalyst's deactivation, 4 monolith structures per tube were implemented (as built). Considering these optimizations, the specific $CAPEX_{Meth}$ decreases to 940 €/kW for 5 MW SNG output. The third optimization step includes further reduction of catalysts and a reduction of the reactor volume by increasing the GHSV. Lab testings have shown that a quarter of the initially used catalyst is sufficient to reach a conversion rate of 90 %. Hence, a further reduction of the catalyst was assumed, so that only one monolith per tube is installed. The reduction of the catalyst volume leads to an increased GHSV. In addition, the GHSV can be increased by increasing the heat transfer from the honeycombs to cooling agent. By reducing the gap between the honeycombs and the tubes, the radial heat transfer is improved, leading to a higher GHSV. Considering both facts, the GHSV can be increased to 7000 1/h, see the confidential Deliverable D2.6. Applied to the reactor in Falkenhagen, this implies a reduction of the reactor volume by a factor of 10 compared to the as-built configuration. Including these optimizations in the $CAPEX_{Meth}$ calculations, the specific $CAPEX_{Meth}$ of a 5 MW plant is reduced to 720 €/kW.

Further, the costs for the optimized Falkenhagen plant were calculated by scaling to an SNG output of 10 MW and 50 MW. Scaling the methanation plant from 5 MW to an SNG output of 10 MW, the specific CAPEX is reduced by about 34 %. For an optimized plant configuration, the specific CAPEX of a 50 MW SNG output is further decreased to 360 €/kW, which corresponds to a reduction of only 21 % compared to the CAPEX of a 10 MW plant. The honeycomb reactor has the largest share of the investment costs for the main equipment, considering a 5 MW plant the share is determined to 41 %. In order to scale the investment costs of the honeycomb reactor, the number of catalyst layers in the reactor is first increased. If the maximum catalyst volume is reached, the reactor is numbered up. Due to the chosen scaling method, the reactor is scaled with a degression coefficient of 0.7 for a scale-up to 5 MW. For a scale-up from demo site to a 10 MW plant, the degression coefficient is already increased to 0.74. Considering both factors, the scaling method of numbering-up and the high reactor's share of the main equipment costs, the specific costs are not strongly reduced for higher SNG output. In Deliverable D2.6, a different reactor design is submitted for the honeycombs. By optimizing the reactor design of the honeycomb reactor, the investment costs can be reduced in future.

Using the same procedure, the $CAPEX_{Meth}$ for the demo sites in Solothurn and in Troia were calculated and then scaled to larger plant sizes regarding SNG output. The optimization potential of the plant configuration in Solothurn is described in section 4.2.2. Due to an increased methane formation rate (MFR), the reactor volume can be reduced. For a plant with an SNG output of 5 MW, the specific $CAPEX_{Meth}$ is reduced by about 8 % from 980 €/kW to 870 €/kW, see Figure 5-3.

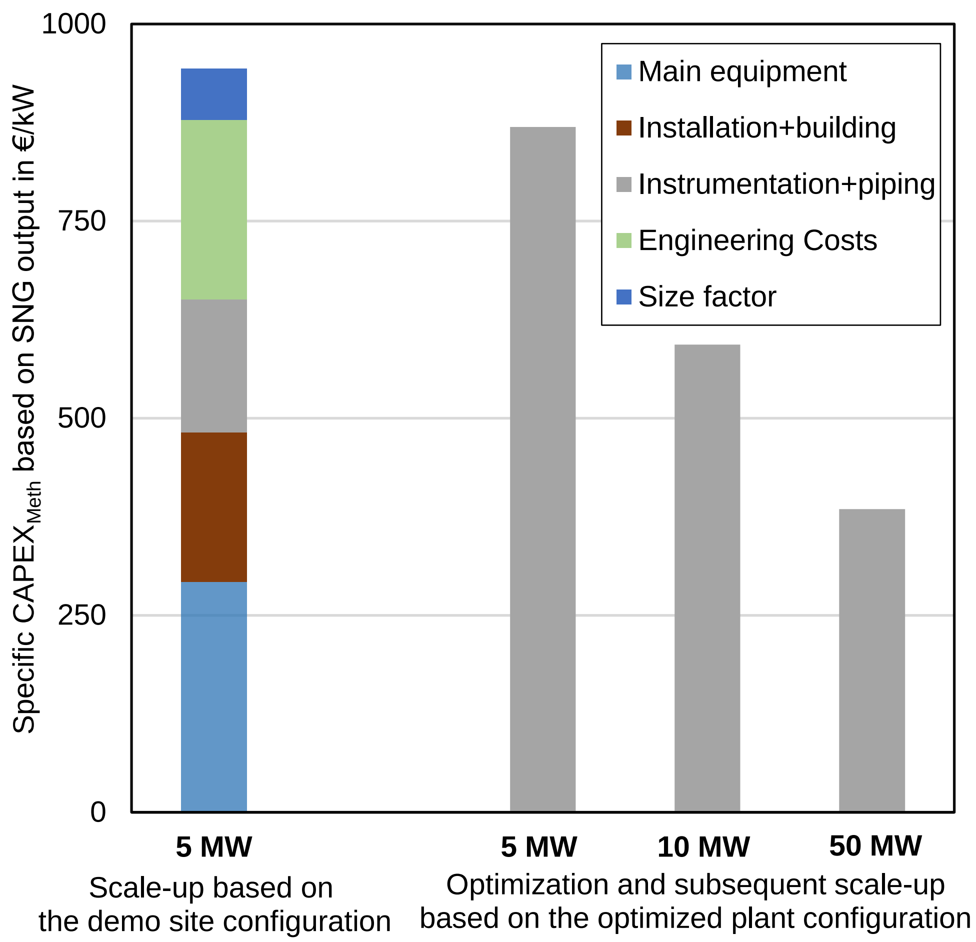

Figure 5-3: Specific $CAPEX_{Meth}$ for an optimized Solothurn methanation unit

By scaling the plant to an SNG output of 10 MW and 50 MW, the specific $CAPEX_{Meth}$ can be reduced to 590 €/kW and 380 €/kW, respectively. This implies that the specific $CAPEX_{Meth}$ of the methanation plant with a 5 MW SNG output can be decreased by 57 %, if a scale-up to 50 MW based on SNG output is performed. Compared to the demo site Falkenhagen, there is a strong drop in the specific $CAPEX_{Meth}$ regarding the scale-up from 10 MW to 50 MW. Although the reactor costs are numbered up, the specific $CAPEX_{Meth}$ further decreases for larger plants. Assuming the improved MFR, only one reactor is sufficient for a 10 MW plant. According to this, five reactors must be implemented for a 50 MW plant. However, the reactor costs account for only 30 % of the main equipment costs. Hence, the total $CAPEX_{Meth}$ can be further reduced by scaling the demo site to 50 MW, since the costs of equipment such as heat exchangers and compressors continue to decrease.

In Figure 5-4, the specific $CAPEX_{Meth}$ is shown for the demo site Troia as well as for a scale-up of the plant. By scaling the methanation unit of the demo site to an SNG output of 10 MW, the specific $CAPEX_{Meth}$ can be reduced to 790 €/kW. It can be seen that the specific $CAPEX_{Meth}$ of the Troia methanation plant is slightly higher than the specific $CAPEX_{Meth}$ of the optimized Falkenhagen demo site. Due to the innovative reactor design of the plant and the relatively small plant size regarding SNG output, the investment costs are estimated relatively high. The reactor costs for the milli-structured reactor were calculated according to the same pattern as for the reactor in Falkenhagen. In addition, only one reactor was needed in Troia, but a compressor was needed for the recycling of the membrane's permeate. Due to the innovative character of the Troia plant and a large cost reduction expected for the milli-structured reactor, strong reduction of the $CAPEX_{Meth}$ is assumed in future. Also, a further scale-up of the plant can be conducted. Since the LNG output of the demo site is relatively low, no further scaling of the LNG output had been performed for the $CAPEX_{Meth}$ calculations.

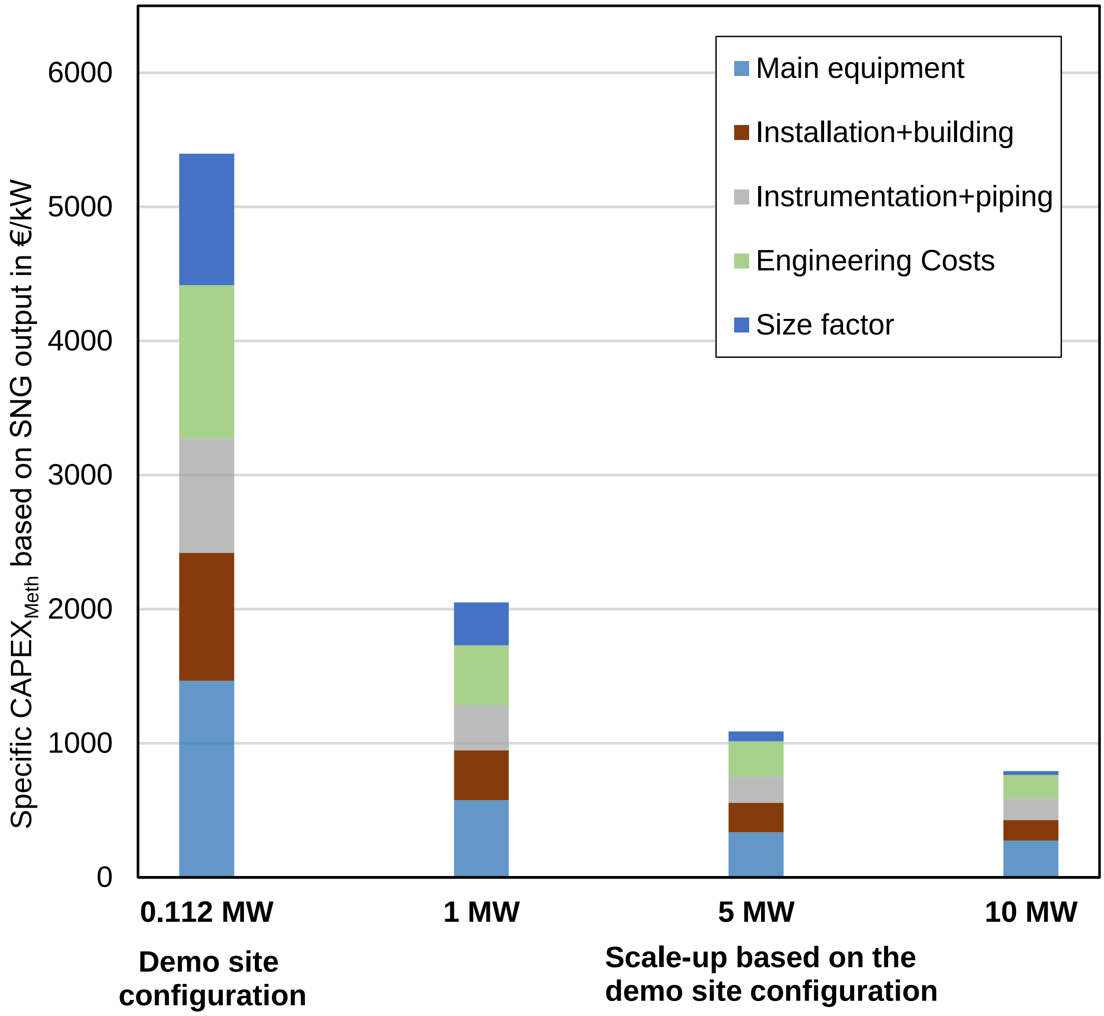

Figure 5-4: Calculations of the $CAPEX_{Meth}$ of the methanation unit of Troia using the Add-on factor method.Future Capital Costs Development

In the section above, the current $CAPEX_{Meth}$ of the methanation units and of the scaled plants is shown. To estimate the future costs development of the three power-to-gas technologies, learning curves must be applied to the calculations. Learning curves help to take into account the development potential of a new technology and calculate the impact on the cost reduction. As part of the STORE&GO project, Energieinstitut Linz (EIL) carried out a future costs estimation of general power-to-gas technologies based on learning curves (more detailed results are reported in Deliverables D7.5 and 7.7). Thereby, the learning curves are based on defined scenarios. Assumptions must be made about the entire installed capacity of power-to-gas plants in the EU. On this basis, the total amount of power-to-gas plants needs to be estimated. The determined learning curves can be used to evaluate the future $CAPEX_{Meth}$ of the methanation units until 2050, see Figure 5-5, Figure 5-6 and Figure 5-7. For Falkenhagen, the learning curves were applied on the $CAPEX_{Meth}$, calculated after the second optimization step, see Figure 5-2. This includes the improved compression of the volumetric inlet flow and halving the catalyst costs. As example, a $CAPEX_{Meth}$ reduction by about 60 % is forecasted for a 5 MW plant based on the optimized Falkenhagen plant design. Hence, the specific $CAPEX_{Meth}$ will be 380 €/kW in 2050. After this calculations, in WP 2 further optimization potential concerning the reactor design had been investigated, see Figure 5-1 (increased GHSV). Considering these further optimizations regarding the reactor design, even lower specific investment costs can be expected. The specific $CAPEX_{Meth}$ are estimated to 350 €/kW for a 5 MW methanation unit based on Solothurn plant design. This corresponds to a reduction of 61 % in 2050. For the Troia methanation unit based on 5 MW LNG output, the specific investment costs will decrease to 520 €/kW in 2050, which means a reduction of 52 %. Further cost reductions are also expected here due to the high optimization potential of the plant.

Furthermore, a higher H_2 content in the gas grid is currently discussed. If the restrictions on H_2 content in the gas grid were eased, the plant design of the two demo sites could be adapted and the CAPEX reduced. For example, for the Falkenhagen plant only one reactor would be sufficient to reach the methane content. In Solothurn, the volume of the reactor could be further reduced causing a CAPEX reduction.

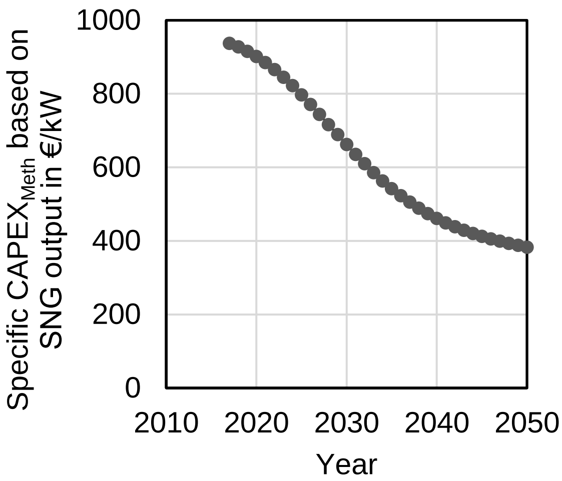

Figure 5-5: Future $CAPEX_{Meth}$ development based on the Falkenhagen methanation unit with an SNG output of 5 MW.

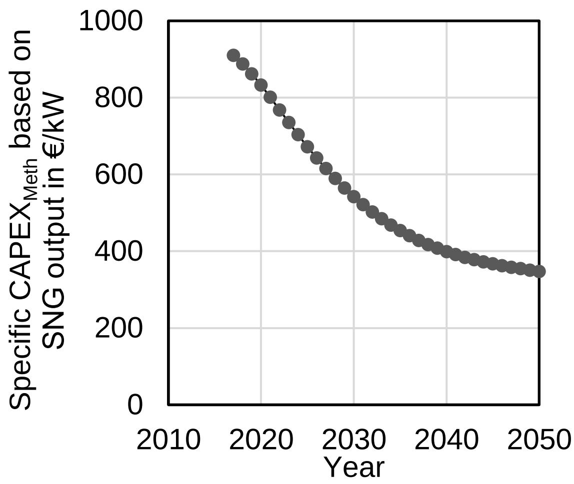

Figure 5-6: Future $CAPEX_{Meth}$ development based on the Solothurn methanation unit with an SNG output of 5 MW.

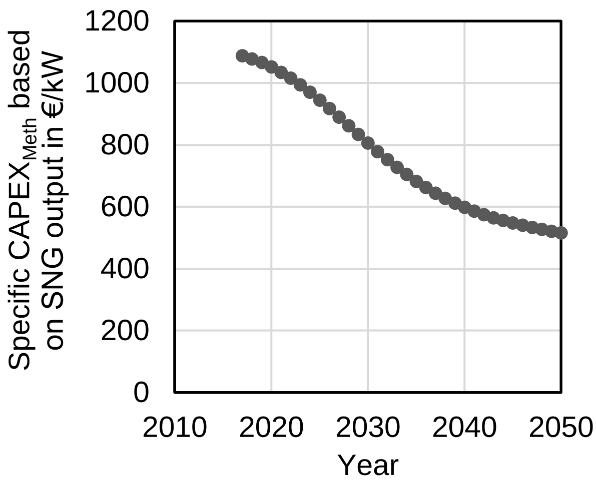

Figure 5-7: Future $CAPEX_{Meth}$ development based on the Troia methanation unit with an SNG output of 5 MW.

5.2 Investment Cost for the Different Power-to-Gas Plants

In the previous section, the calculations of the $CAPEX_{Meth}$ of each demo site have been shown. Furthermore, one goal within this project was the $CAPEX_{PtG}$ reduction of an industrial power-to-gas plant by 15 % compared to the state-of-the-art technologies. This corresponds to a reduction by 200 €/kW – 400 €/kW based on electrolyser power rating. Therefore, the capital expenditures of the entire PtG plant ($CAPEX_{PtG}$) must be determined for an industrial scale. The $CAPEX_{PtG}$ summarizes the investment costs of each process unit and includes following the investment cost: electrolyser ($CAPEX_{Ely}$), CO_2 conditioning/capture ($CAPEX_{CO2\text{-}Con}$), optimized methanation unit ($CAPEX_{Meth}$), injection/liquefaction ($CAPEX_{Inj/Liq}$). Table 5-1 gives an overview of the optimized $CAPEX_{Meth}$ and of the $CAPEX_{PtG}$ of the entire process chain for each demo site.

Table 5-1: Overview of the optimized $CAPEX_{Meth}$ of each demo site and the entire $CAPEX_{PtG}$ of the PtG plant compared to the project goals

	Falkenhagen	Solothurn	Troia
Individual process units			
$CAPEX_{Meth}$ of an optimized plant with 5 MW SNG output	720 €/kW	870 €/kW	1090 €/kW
$CAPEX_{Ely}$ of the electrolysis for a 5 MW (SNG output) PtG plant based on literature (state-of-the-art) [11]	1860 €/kW	1860 €/kW	1890 €/kW
$CAPEX_{CO2\text{-}Con}$ of the CO_2 supply for a 5 MW (SNG output) PtG plant based on literature	456 €/kW [12]	342 €/kW [13]	1620 €/kW[13]
$CAPEX_{Inj/Liq}$ of the injection/liquefaction for a 5 MW (SNG output) PtG plant based on literature	56 €/kW [4]	56 €/kW [4]	601 €/kW[10]
Overall PtG plant			
$CAPEX_{PtG}$ of an optimized plant with 5 MW SNG output	3100 €/kW	3130 €/kW	5200 €/kW
$CAPEX_{PtG}$ of an optimized plant with 5 MW SNG output; specific costs referred to electrolyser power (8.4 MW)	1832 €/kW	1848 €/kw	3027 €/kW
CAPEX reduction compared to the state-of-the-art (2660 €/kW based on electrolyser power)	31 %	31 %	none

[10] Data based on Hysytech

5.3 Evaluation of the SNG/LNG Current Productions Costs

To evaluate the entire production costs of the three power-to-gas plants, the operating costs were included. The current production costs were calculated using the annuity method as described in section 3.7, and refer to 2017. The future development of the production costs is discussed in Deliverables D7.5/D7.7 from EIL. The $CAPEX_{Meth}$ was calculated and optimized within this project (described above in section 5.1). The production cost calculations are based on the $CAPEX_{PtG}$ for the entire PtG plant. The $CAPEX_{PtG}$ includes the optimized $CAPEX_{Meth}$ of the different methanation units. Hence, for Falkenhagen the inlet flow's compression and the catalyst's cost could be reduced. At the demo site Solothurn, the reactor size could be minimized, due to an improved MFR. The CAPEX of the additional process units as electrolyser $CAPEX_{Ely}$, CO_2-conditioning/capture $CAPEX_{CO2\text{-}con}$, and injection or rather liquefaction $CAPEX_{Inj/liq}$ are based on literature. The following evaluation of the production cost considers many different costs. Table 5-2 lists the assumptions made for the calculations of a 5 MW SNG/LNG plant. Thereby, the efficiencies and the energy input or output refer to the high heating value (HHV).

Table 5-2: Parameters and assumptions of a plant scaled to 5 MW based on SNG/LNG output for the OPEX calculations.

	Parameter			Unit
	Falken-hagen	**Solothurn**	**Troia**	
Amortization period	20	20	20	a
Calculatory interest rate	6.865	6.865	6.865	%
Heat price	0.01	0.01	0.01	€/kWh
Electrolyser				
Electrolyser type	AEL	AEL	AEL	
Efficiency η [11]	76	76	76	%
CAPEX based on H_2 output [11]	1100	1100	1100	€/kW
Maintenance costs (without stack replacement costs) [11]	20	20	20	€/(a kW)
H_2 output at full load for 5MW SNG output	6450	6540	6560	kW
Power input electrolyser	8460	8570	8590	kW
Stack lifetime [11]	57000	57000	57000	h
Stack replacement costs	40 % from $CAPEX_{Ely}$			
CO_2 supply				
CO_2-Separation type	Absorption (amine scrubbing)	Membrane	DAC	
CO_2 Volume flow at full load	460	460	460	m³/h
$CAPEX_{CO2\text{-}con}$ based on CO_2 output	2830 [12]	3700 [13]	17500 [13]	€/m³/h

OPEX based on CO_2 output [13]	8.9	8.3	4 % of the CAPEX	€-ct/m³
Electrical demand based on SNG output	1.0	1.3	3.95	kWh/m³
Methanation				
Maintenance costs based on $CAPEX_{Meth}$	3	3	3	%
Insurance based on $CAPEX_{Meth}$	0.5	0.5	0.5	%
Contingencies based on $CAPEX_{Meth}$	0	0	0	%
Catalyst costs	277740	0	544514 (10 % of CAPEX)	€
Lifetime catalysts	24000	endless	24000	h
Personnel costs	65814	65814	65814	€/a
Electrical demand	0.29	0.44	1.9	kWh/m³
Gas grid injection/liquefaction				
$CAPEX_{liq}$ Liquefaction	0	0	3005 400	€
Maintenance	0	0	4 % of CAPEX	
Injection grid	Transport grid (ONTRAS)	Transport grid (ONTRAS)		
Injection pressure	63	63	-	bar
Electrical demand	0.10	0.12	0.59	kWh/m³

As can be seen from the table above, unlike the current plant configuration, the same type of electrolyser with same performance indicators was assumed for all three demo sites. Since previously installed electrolysers were selected for the project, the performance indicators did not correspond to the current state-of-the-art technologies. For the cost calculations, a state-of-the-art AEL electrolyser was chosen. A large proportion of the electrolyser cost are the stack replacement cost. The stack life time for this electrolyser was specified as 57,000 h. Accordingly, the stack replacement refers to the operational hours. As example, if the plant runs at full load (8000 h/a), the stacks must be replaced every seven years. Thereby, for the calculations of the production cost, the stack replacement costs were treated as interim investment cost. Further, no energy costs for the hot stand-by of the electrolyser were considered. Depending on the operational hours and the operation mode, the electrolyser must normally be kept in hot stand-by to guarantee dynamic operation.

The CO_2 treatments differ between the demo sites. In Falkenhagen, a CO_2 separation from a biogas plant by absorption was planned, whereas for Solothurn the CO_2 comes from a waste water treatment plant and was separated by membranes. In Troia, the CO_2 was provided by a direct air capture (DAC) plant from Climeworks.

For the methanation unit, the annual maintenance expenses were assumed to be 3 % of the $CAPEX_{Meth}$. As described in chapter 4, it was assumed that the electrical demand of the methanation unit could be reduced for larger plants. In the following calculations of the production costs, an electrical demand of 25 kW/MW was assumed. In case of Solothurn, the electrical consumption of the agitator must be added to the electrical demand of the unit. The differences of the costs between the plants were the catalyst costs. For the demo sites Falkenhagen and Troia, catalyst life time and replacement costs were indicated in dependence of the operating hours. The catalyst replacement costs were assumed as an interim investment such as the stack replacement costs. For the demo site in Solothurn, the biocatalyst (Archaea) cost can be neglected. Once the biocatalyst is added, the microorganisms adapt to their environment and reproduce themselves. However, in Solothurn nutrients costs must be considered, see Table 5-3. In order to guarantee the production rate of the reactor, nutrients need to be fed into the reactor. The needed amount of nutrients, and the associated costs, depend on the operational hours. Since the used amounts of nutrients were very low and the media ‘100x’ and ‘SeW1000x’ are not commercially available, the specific costs are high. It will be expected that the nutrients costs can be strongly reduced, on the one hand by implementing a nutrient recovery system, and secondly by larger plants. Hence, in the following evaluation of the Solothurn demo site, a reduction of the nutrients costs by 50 % is considered.

Table 5-3: Consumption and costs for nutrients in Solothurn (Deliverable D3.4).

Chemicals/product	Volumetric flow in L/h	Costs in €/L
Antifoam	0.45	3.29
NH_3	6.46	2.71
100x	3.69	7.70
SeW1000x	0.37	8.38
Na_2S	4.31	4.60

In Falkenhagen and Solothurn, the produced SNG was injected into the gas grid, whereas in Troia the SNG was liquefied. In Falkenhagen, the SNG was injected into a transport gird (ONTRAS) at a maximum pressure of 63 bar. In Solothurn, the gas was injected into a distribution grid at a pressure level of about 4 bar. Nevertheless, for the calculations of the entire process costs, it was assumed that the SNG of the Solothurn plant was injected into the same transport grid as the SNG of Falkenhagen.

5.3.1 Evaluation of the Production Costs of Falkenhagen

As described in sections 3.7, the production costs of the entire process chain of the demo sites had been calculated using the annuity method, taking into account the assumptions listed in Table 5-2 and Table 5-3. Based on the assumptions above and the optimized $CAPEX_{Meth}$ (see Figure 5-5), the production costs of a 5 MW plant with 8000 h/a operational hours are 0.124 €/kWh based on the day-ahead market electricity price in Germany, see Figure 5-8 (basis). Similarly to the $CAPEX_{Meth}$ optimization, also the OPEX can be optimized. Therefore, the heat usage can be optimized. In the Basis scenario, only the heat usage delivered to the veneer mill is included (based on measurement data 1.7 kWh/m³, see Table 4-2. It is assumed that the excess heat is sold to 0.01 €/kWh, see Table 5-2. With regard to the temperature of the oil circuit, the heat dissipated at a temperature level lower than 180 °C is not used in the current process design. By further cooling down the product gas stream after the reactor to a temperature level of 60 °C, the heat usage can be improved to 3 kWh/m³. However, to use heat on a temperature level of 60 °C, an appropriate heat sink is required. As an example, for district heating a low temperature level could be suitable or a heat pump is used to reach a higher temperature level of the excess heat. At the current location of the demo site Falkenhagen, the infrastructure is not available, so a heat usage at 60 °C is only theoretically feasible. The additional investment costs, which would be necessary to use the heat, are not included in the production costs.

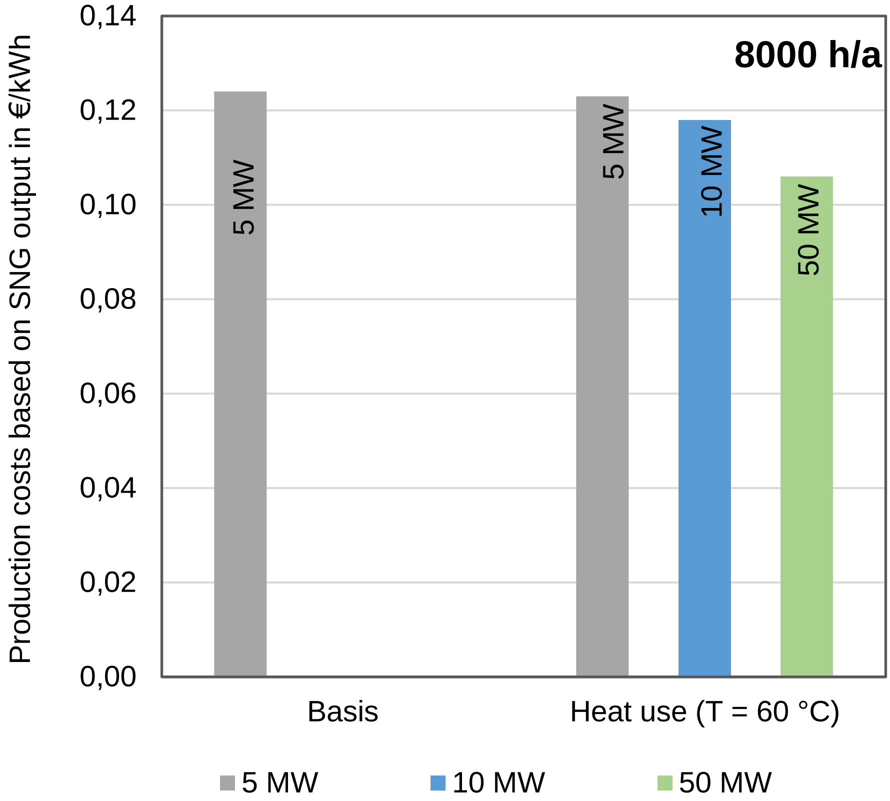

Figure 5-8: Production costs of the demo site Falkenhagen for 8000 h/a operational hours and included optimization potential, based on day-ahead electricity prices of Germany (see Table 3-2) .

If a scale-up to an SNG output of 10 MW and 50 MW is performed based on the optimized cost assumptions, the production costs are 0.118 €/kWh and 0.106 €/kWh, respectively.

Based on the optimized costs, in Figure 5-9 the production costs are shown for a 5 MW plant split into the $CAPEX_{PtG}$ and the fixed and the variable OPEX. In addition, the costs are presented in dependence of the operational hours as well as for the different day-ahead electricity prices of Germany, Italy and Switzerland. It can be seen that the electricity price only has an impact on the variable OPEX. Due to the low electricity price in Germany, the production cost are most affordable with 0.123 €/kWh for 8000 h/a. While the highest electricity prices are dedicated in Italy, the production costs are with 0.028 €/kWh (22 %) more expensive. By varying the electricity prices, the CAPEX's and the fixed OPEX's share of the production costs remains the same. As a result, the lower the operational hours of the plant, the higher the CAPEX's shares in the production costs. Considering 8000 h/a, the costs of the CAPEX are 0.046 €/kWh for each plant. This corresponds to about 33 % of the total costs, whereas for 1500 h/a operational hours the CAPEX accounts for 67 % (0.205 €/kWh) of the production costs.

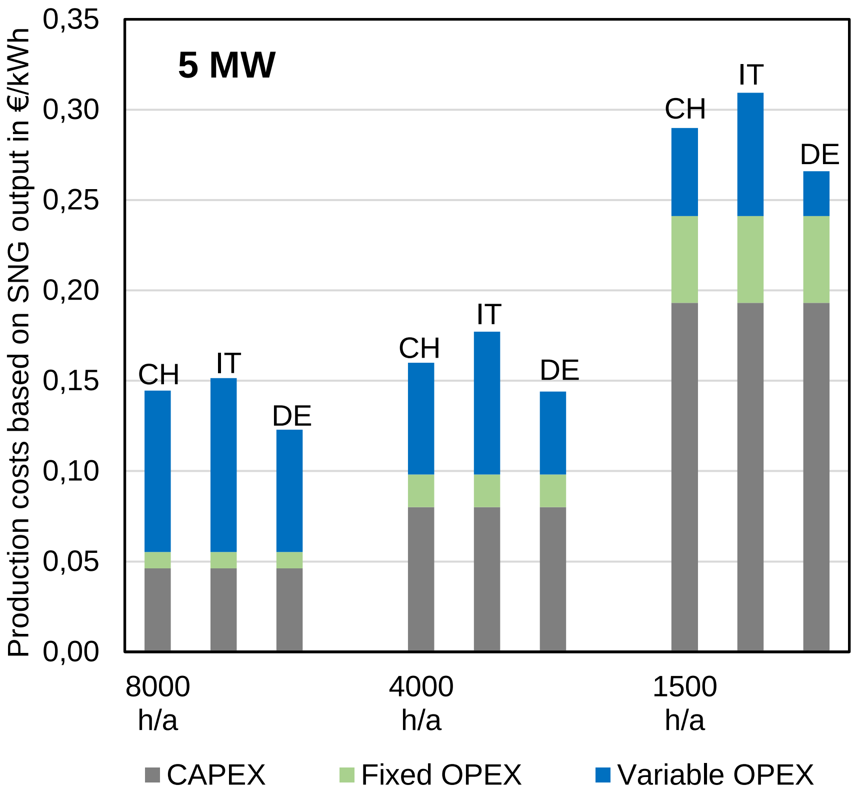

Figure 5-9: Production cost for a 5 MW plant based on the optimized Falkenhagen demo site in dependence of the operational hours and the different electricity prices for Switzerland, Italy and Germany, see Table **3-2**.

Also, the production costs of 8000 h/a are slightly lower than for 4000 h/a operational hours. Based on the German day-ahead electricity prices, the production costs are 0.144 €/kWh for 4000 h/a. Corresponding to 8000 h/a, the costs are 0.021 €/kWh more expensive. A further reduction of the operational hours per year leads to a strong increase of the production costs. Therefore, it can be concluded that the optimum amount of operational hours are between 8000 h/a and 4000 h/a.

5.3.2 Evaluation of the Production Costs of Solothurn

In the same procedure as above for Falkenhagen, the production costs for the demo site in Solothurn had been calculated. The production costs were calculated based on the assumptions and the optimized $CAPEX_{PtG}$ (see Figure 5-3) for a 5 MW plant with 8000 h/a operational hours. For Solothurn, the productions costs are 0.125 €/kWh based on the day-ahead electricity price in Germany, see Figure 5-10 (Basis). In a second step, the costs were optimized as in the $CAPEX_{Meth}$ calculations. In Figure 5-10 the different optimization steps are shown for a plant with 5 MW SNG output. First, due to the chosen location of the demo site, the costs of the CO_2 separation and the injection could be neglected. The CO_2 was a waste product of a waste water treatment plant located about 2.5 km away. In addition, at the current demo site, the SNG was injected into a distribution grid instead of a transport grid as previously calculated. Due to the low pressure level of the distribution grid at about 4 bar and the operating pressure of 11 bar, there are no injection costs. If these two optimization potentials were considered, the production costs are reduced to 0.111 €/kWh. Further, the dissipated heat could be used even on a low temperature level, due to an existing heat pump. Including the excess heat of the electrolyser and the methanation, 5.9 kWh/m³ could be used by a district heating network. Since the required technical facilities, as example heat pump etc., were already available at the location, no additional investment costs must be included in the calculations. Considering the heat usage with a heat price of 10 €/MWh, the production costs can be further decreased by 0.6 €-cent/kWh. A closer look on the total expenses shows that the nutrients cost have high influence on the SNG productions costs. As mentioned above, the nutrients were not commercially available resulting in high specific costs. It was assumed that the nutrients costs will strongly decrease by about 50 % due to increasing plant size. In addition, the measurement data of the required nutrients amount is based on data from Deliverable D3.4, where the nutrient recovery system had not yet been implemented.

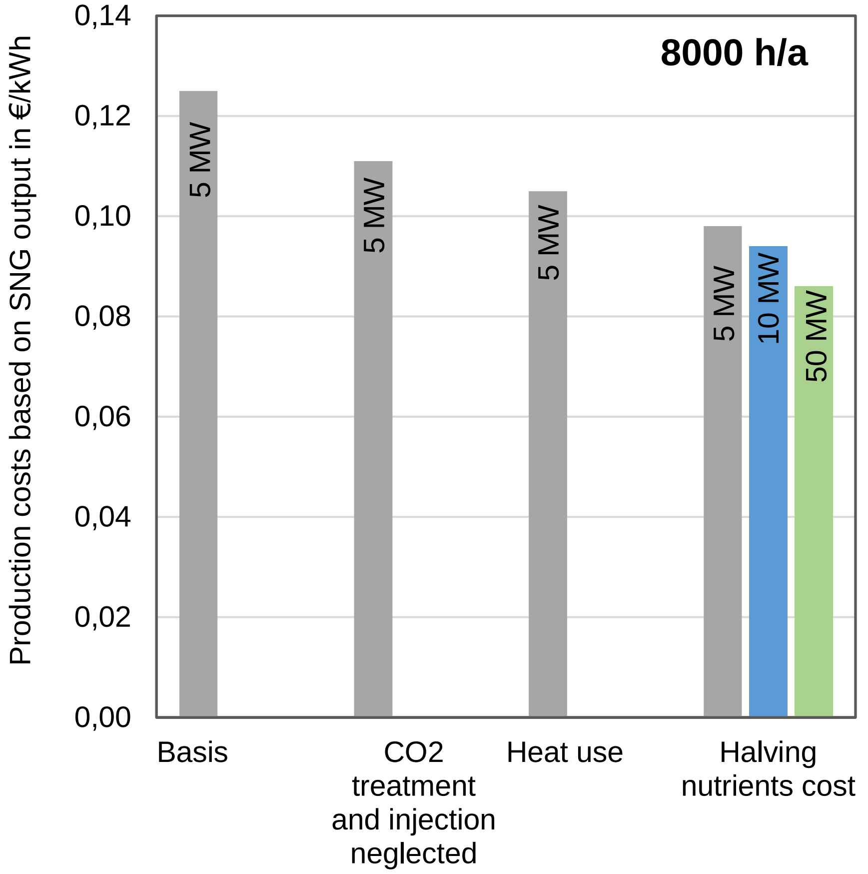

Figure 5-10: Production costs of demo site Solothurn for 8000 h/a operational hours and included optimization potential, based on day-ahead electricity prices of Germany, see Table 3-2.

The nutrient recovery system was implemented in month 45. Initial tests showed that about 80 % of the nutrients could be recaptured and be recycled into the reactor. Including this result in the calculations, the production cost can be decreased to 0.098 €/kWh considering the German day-ahead market electricity price and a 5 MW plant based on SNG output. Scaling the plant to an SNG output of 50 MW, the cost even reduces to 0.086 €/kWh.

Table 5-4: Influence of the heat price on the production costs for the demo site Solothurn scaled to 5 MW SNG output and the German day-ahead-electricity price, see Table 3-2.

	Production costs in €/kWh		
Heat price in €-ct/kWh	**8000 h/a**	**4000 h/a**	**1500 h/a**
0.5	0.101	0.107	0.167
1	0.098	0.104	0.164
2	0.093	0.099	0.159

As seen in Table 5-2, the revenues from excess heat were assumed to be 10 €/MWh. Since the legal and regulatory framework is not regulated, the prices for excess heat can deviate. Hence, a sensitivity analysis was conducted for the heat prices, see Table 5-4. For 8000 h/a operational hours and the German day-ahead market electricity price, the production costs vary between 0.101 €/kWh for a heat price of 50 €/MWh and 0.093 €/kWh for a heat price of 20 €/MWh.

After considering the optimization potentials in the calculations, the impact of the operational hours as well as the electricity price on the production costs is demonstrated in Figure 5-11. It should be noted that the following results of the production costs always include the optimization as shown in Figure 5-10. In the diagram below, the production costs are given depending on the operational hours for a 5 MW plant. It can be seen that the costs are in the same range for 8000 h/a and 4000 h/a operational hours. Based on the day-ahead market electricity price in Germany, the production costs only differ by 0.06 €/kWh between 8000 h/a and 4000 h/a. At significantly lower operational hours (1500 h/a), the production cost increases to 0.164 €/kWh. This implies that the optimal amount of full load hours per year is likely between 8000 h/a and 4000 h/a. Furthermore, the diagram shows the shares of the CAPEX and the fixed and variable OPEX in the total production costs for the various electricity prices in Switzerland, Italy and Germany.

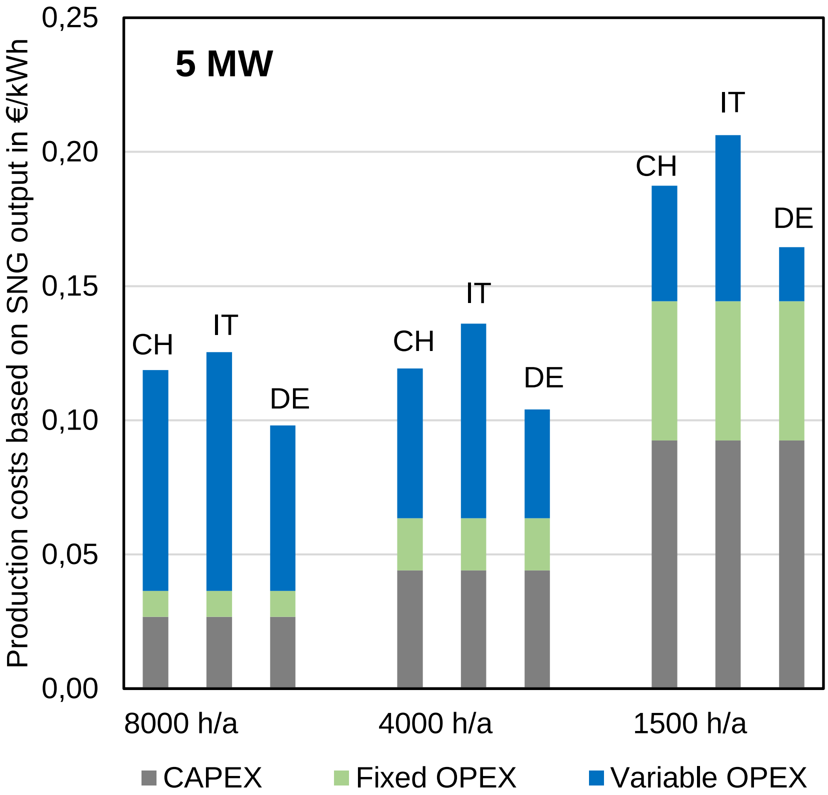

Figure 5-11: Production cost for a 5 MW plant based on the optimized Solothurn demo site in dependence of the operational hours and the different electricity prices for Switzerland, Italy and Germany, see Table 3-2.

By varying the electricity price, only the variable OPEX (blue bar) changes, whereas the share of the CAPEX (gray bar) and of the fixed OPEX (green bar) are equal for all three electricity prices. The production costs vary between 0.125 €/kWh (Italy) and 0.098 €/kWh (Germany) for a 5 MW plant and 8000 h/a operational hours. In Solothurn, electricity prices have a large impact on the production costs. Thus, it should be preferred to reduce the electricity consumption and to choose the location with regard to low electricity prices or network charges and fees.

5.3.3 Evaluation of the Production Costs of Troia

Based on the $CAPEX_{PtG}$ calculations and the energetically optimized plant configuration, the production costs were determined for the Troia plant. After calculating the productions costs for a basis scenario, further optimization potential was identified and taken into account in the production costs calculations. Figure 5-12 summarizes the results of the production cost of the basis scenario and the effects of the optimizations on the costs for a 5 MW plant, assuming 8000 h/a operational hours and the day-ahead market price for electricity in Germany. Furthermore, the optimized costs are shown for a plant scaled to 10 MW.

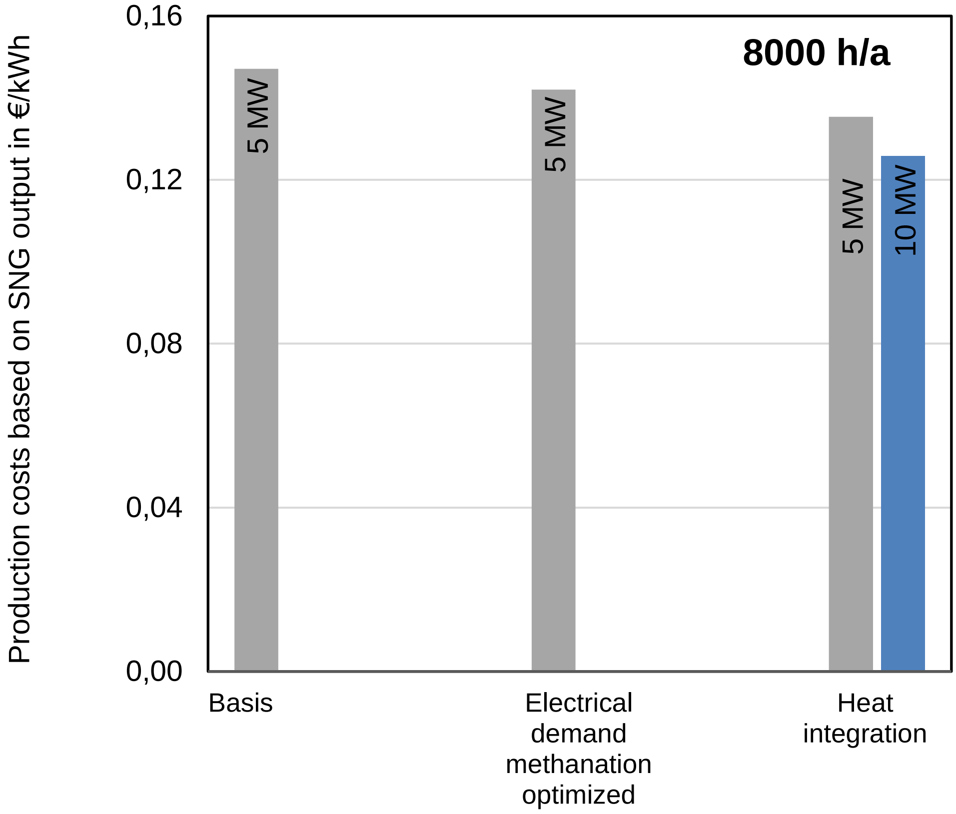

Figure 5-12: Production costs of demo site Troia for 8000 h/a operational hours and included optimization potential, based on day-ahead electricity prices of Germany, see Table 3-2.

Based on the Troia plant configuration, the production costs are 0.147 €/kWh for a 5 MW plant, assuming 8000 h/a operational hours and the day-ahead market price in Germany. The basis scenario relies on measurement data and the subsequent energetic optimization, presented in section 4.3.2. Thereby, the optimized electrical demand of the methanation unit was 1.9 kWh/m³. Compared to the catalytic methanation in Falkenhagen, which had an electrical demand of 0.29 kWh/m³, the electrical demand was still higher by 1.6 kWh/m³. In the energetic optimization of the methanation plant, the reactor's thermal insulation can be improved, see section 4.3.2. As an optimization potential for the production costs calculations, it was assumed that the electrical demand of the methanation unit can be further decreased due to the scale-up of the plant. Thus, the production costs are reduced to 0.142 €/kWh by assuming that the electrical demand of the methanation unit is 0.29 kWh/m³. A further optimization potential was detected by heat integration. The heat dissipated by the methanation reactor can be integrated into the DAC, which requires about 3 kWh/m³ at a temperature level of about 100 °C. Hence, 2.1 kWh/m³ of the methanation can be used for the DAC at a temperature of about 260 °C. Including the heat integration, the production costs can be decreased by 0.7 €-ct/kWh to 0.135 €/kWh. Finally, based on the optimized production costs functions, the plant configuration can be scaled to an LNG output of 10 MW. The production costs of a plant designed for a 10 MW

LNG output are 0.126 €/kWh, assuming 8000 h/a operational hours and the day-ahead market electricity price in Germany.

Based on the implemented optimizations, the influence of the electricity price on the production costs was calculated for Troia scaled to an LNG output of 5 MW. Figure 5-13 shows the production costs of a 5 MW LNG output plant for three different electricity prices and for 8000 h/a, 4000 h/a and 1500 h/a operational hours.

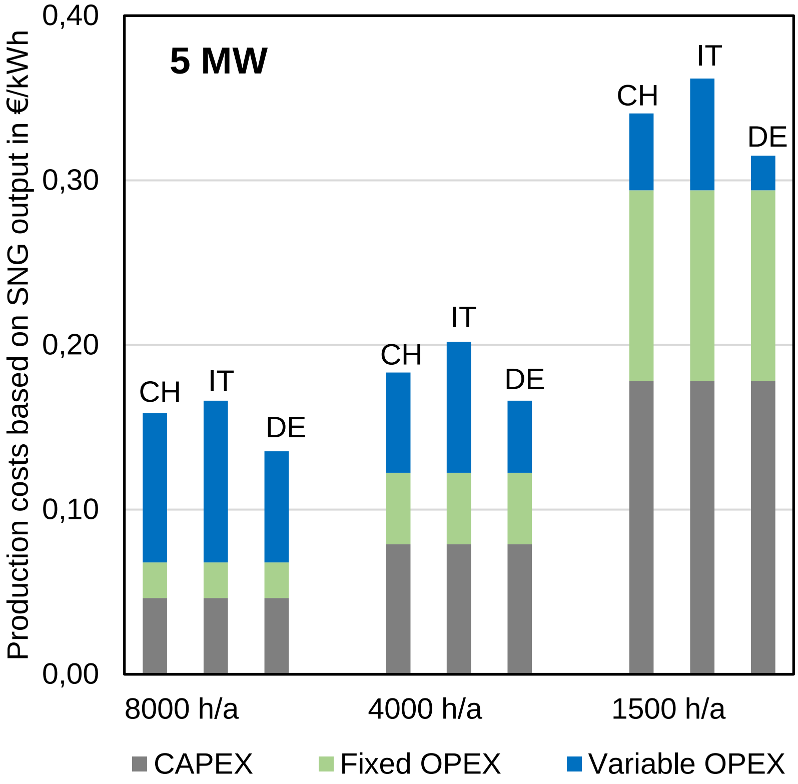

Figure 5-13: Production cost for a 5 MW plant based on the optimized Troia demo site in dependence of the operational hours and the different electricity prices for Switzerland, Italy and Germany, see Table 3-2.

Regarding the different day-ahead market electricity prices for Switzerland, Italy and Germany, it can be seen that the production costs for 8000 h/a vary between 0.135 €/kWh (Germany) and 0.166 €/kWh (Italy). Additionally, it can be seen that the electricity prices only affect the variable OPEX (blue bars). If the plant runs with less operational hours per year, the share of the variable OPEX on the production costs decreases. Therefore, the impact of the electricity price on the production costs falls as the operational hours of the plant decreases. While for 8000 h/a operating hours, production costs in Italy are 23 % more expensive than in Germany, for 1500 operating hours they are only 14 % more expensive. Furthermore, the graph shows that the lowest production costs can be achieved at full load (8000 h/a). Nevertheless, the CAPEX of the plant has a high impact on the total production costs. The plant in Troia is an innovative power-to-gas technology including DAC, a milli-structured reactor and a liquefaction of the produced methane. The process design is newly developed and is investigated on a pilot scale. Therefore, the investment costs are high for this process. In order to reduce production costs, it is more cost-effective to operate the plant at full capacity at 8000 h/a. In addition, the pilot plant has a relatively low LNG output. It is expected that the specific CAPEX will be further reduced by scaling the plant's LNG output, as can be seen in section 5.1.

6 Summary

The main focus of this report is the technical and economic evaluation of the three demo sites erected within the STORE&GO project. Therefore, the plant design of the three different demo sites is described in chapter 2. The Falkenhagen demo site contains an innovative honeycomb methanation reactor. The reactor consists of multi-tube channels, which are coated with a metallic catalyst. Whatever hydrogen and carbon dioxide has not reacted to methane in this honeybomb reactor, is converted to methane in the subsequent polishing reactor. In Troia, an innovative power-to-gas process chain consisting of a direct air capture (DAC) unit, a one stage milli-structured methanation reactor, and a liquefaction unit was implemented. In order to reach an overall conversion of nearly 100%, the H_2 and CO_2 is separated from the product gas and recycled. Whereas in Falkenhagen and in Troia a catalytic methanation concept is investigated, in Solothurn a biological stirred bubble column methanation is built. Hereby, the feed containing hydrogen and carbon dioxide in a stoichiometric ratio is converted into methane by a biocatalyst (Archaea).

One of the most important performance indicators is the efficiency of the overall power-to-gas process chain. The overall PtG efficiency takes into account the heat usage and the energy demands for the following process steps:

- CO_2 conditioning
- H_2 production via existing electrolyser
- Methanation unit
- Injection to a high pressure gas grid or liquefaction.

One main goal of the STORE&GO project was to demonstrate an overall PtG-efficiency higher than 75 % and to reach a methane content of y_{CH4} > 90 % in the product gas. In addition, a load flexibility of 20 – 100 % load and a load change rate of at least 5 %/min should be proven. Table 6-1 gives an overview of the energetic evaluation of the three demo sites. For the Falkenhagen plant, an overall PtG efficiency of 53 % was reached based on the measurement results. The methanation unit itself reaches a methanation efficiency of 85 % (including heat usage and electricity demand). The relatively low overall PtG efficiency arises from the poor efficiency of the existing AEL. Due to this fact, the biggest optimization potential in Falkenhagen is to use a state-of-the-art electrolyser. This would lead to an optimized overall PtG efficiency of 69 %. But the core technology in Falkenhagen (two-stage methanation unit) was capable of producing high quality SNG (y_{CH4} > 99 vol.-%) for a wide variation of the load. Also during load changes, the SNG quality fulfills the limits for injection of the gas.

Table 6-1: Overview of methane fraction after the methanation and the overall PtG efficiency and the optimized effi-ciency if all optimization potential are considered considered indicators of the energetic evaluation

	Project goals	Falkenhagen	Solothurn	Troia
Methane content of the product gas	y_{CH4} > 90 vol.-%	> 99 vol.-%	> 99 vol.-%	96 vol.-%[11]
Overall PtG efficiency based on measurements	$\eta_{\mathrm{PtG,HHV,ov}}$ > 75 %	53 %	76 %	29 %
Optimized overall PtG efficiency		69 %	89 %	46 %

[11] This is the methane fraction in front of the liquefaction. The gas quality is reached by gas separation and recycling the lean gas.

In Solothurn, the methanation unit reaches also a product gas quality with a methane content of y_{CH4} > 99 vol.-%. During the operation of the plant, the biocatalyst was slowly adapted to higher loads: At the end of the project, the plant was capable of operating at nearly 100 % load. The overall PtG efficiency is 76 %, which includes the usage of the low temperature (T_{use} < 60 °C) waste heat from the electrolysis. In Solothurn, the nearby 'Hybridwerk' was able to use this waste heat at relatively low temperature, since the heat was boosted via a heat pump for district heating. The energy demand for the CO_2 source was neglected, since the CO_2 stream to the plant was a waste product. Further optimization potential is the integration of the waste heat from the methanation reactor in the 'Hybridwerk', which was planned in the project but not tested. Another potential is the reduction of the electrical energy demand of the methanation unit. If both potentials would be realised, an overall PtG efficiency of 89 % could be reached. This shows that the efficiency of the overall PtG process chain is very dependent on the location of the plant.

Due to the innovative character of the overall process chain in Troia and the relatively small capacity of 0.1 MW SNG output, huge potential for energetic improvement exists. During the project, an overall PtG efficiency of 29 % was reached. It has to be considered that the DAC and the liquefaction of the SNG have a comparably high energy demand. Due to the recycle of lean gas to the front of the methanation unit, the overall conversion of CO_2 and H_2 is in the range of 99 %. A methane content of y_{CH4} = 96 % in front of the liquefaction was reached. It was also shown that the process could be operated dynamically from 20 – 80 % of load with a load change rate of 5 %/min. By heat integration and energetic optimization of the process units, an overall PtG efficiency of 46 % could be reached.

Beside a technical evaluation, an economic evaluation of the demo sites was also performed. The aim of the STORE&GO project was to achieve a cost reduction for industrial scale methanation plants by 15 % compared to state-of-the-art technologies. The economic evaluation of the three demo sites includes the calculations of the capital expenditure (CAPEX) for the methanation unit $CAPEX_{Meth}$, future expectations of the $CAPEX_{Meth}$ development until 2050, and the calculations of the production costs. Table 6-2 summarizes the results of the economic evaluation. In a first step, the $CAPEX_{Meth}$ of the demo sites' methanation units was determined using the Add-on factor method. Afterwards, the plant design was optimized, and the $CAPEX_{Meth}$ was calculated for the plants scaled to an SNG/LNG output of 5 MW, 10 MW and 50 MW. Thereby, the optimized specific $CAPEX_{Meth}$ for the three sites is in the same range, between 720 €/kW and 1090 €/kW for a plant scaled to 5 MW SNG/LNG output. The investment costs for the Solothurn plant with an SNG output of 5 MW are 870 €/kW. The relatively low costs can be explained since only one simple reactor is required, and since the biocatalyst reproduces itself. The optimized specific $CAPEX_{Meth}$ of Falkenhagen is 720 €/kW for a 5 MW plant. It must be considered that two reactor stages are necessary due to the higher restrictions for injection in Germany. In addition, there is a high potential to reduce the costs by optimizing the reactor design. Troia has the highest specific $CAPEX_{Meth}$ with 1090 €/kW for 5 MW LNG output. Partly, the technology of a milli-structured methanation reactor is relatively new and in part investigated on pilot scale for the first time. A huge potential is available in Troia to optimize the plant and thus reduce the CAPEX.

To estimate the potential of future cost reductions for methanation units, learning curves were implemented on the determined $CAPEX_{Meth}$ of the methanation units. Due to scaling effects and high technical development potential, the $CAPEX_{Meth}$ will strongly decrease. Based on the $CAPEX_{Meth}$ evaluations, the costs will be reduced to 380 €/kW in 2050, for the demo site Falkenhagen for an SNG output of 5 MW. The $CAPEX_{Meth}$ of the Solothurn plant will be reduced by 560 €/kW to 350 €/kW. The $CAPEX_{Meth}$ in Troia for an SNG output of 5 MW is reduced by more than half to 520 €/kW in 2050.

Table 6-2: Overview of selected results of the economic evaluation

	Falkenhagen	Solothurn	Troia
$CAPEX_{Meth}$ of an optimized plant with 5 MW SNG output	720 €/kW	870 €/kW	1090 €/kW
$CAPEX_{PtG}$ of an optimized plant with 5 MW SNG output	3100 €/kW	3130 €/kW	5120 €/kW
Methane production costs of an optimized plant with 5 MW SNG output (8000 h/a, German electricity price)	0.123 €/kWh	0.098 €/kWh	0.135 €/kWh

Based on the optimized $CAPEX_{Meth}$ calculations, the production costs for the entire process chain were calculated. In a first step, the CAPEX of the entire power-to-gas ($CAPEX_{PtG}$) plant must be determined. The $CAPEX_{PtG}$ includes the investment costs of following process units: the electrolyser $CAPEX_{Ely}$, the CO_2 conditioning/capture $CAPEX_{CO2\text{-}Con}$, the methanation unit $CAPEX_{Meth}$ and the injection/liquefaction $CAPEX_{Inj/liq}$. These data are partly based on literature and are summarized in section 5.3. For the calculations of the operational costs (OPEX), several assumptions must be made, due to the fact that a lot of operational experiences still need to be gathered. In addition, the production cost were determined for 8000 h/a, 4000 h/a and 1500 h/a operational hours. Since the network charges and taxes vary according to the different countries, only day-ahead market electricity prices were taken into account for the calculations. Thus, based on the day-ahead market electricity prices in Germany, the production costs are 0.123 €/kWh according to the Falkenhagen plant for an SNG output of 5 MW and 8000 h/a. These costs include, on the one hand, the optimization of the plant design taken into account in the $CAPEX_{Meth}$ calculations, and, in addition, further optimization potential with regard to operation was integrated. For Falkenhagen this means that the catalyst costs were further reduced and the heat usage was improved. In future, a high cost reduction of the honeycomb reactor is expected. For the Solothurn plant, the production costs are 0.098 €/kWh assuming an SNG output of 5 MW, 8000 h/a operational hours and the German electricity price. Due to the infrastructure of the ‘Hybridwerk’, in Solothurn heat usage at a low temperature level could be considered. This leads to high efficiencies and relatively low production costs. In addition, a strong drop of the nutrient cost for the biological reactor is assumed, if the nutrients are commercially available. For the same parameters (8000 h/a, 5 MW SNG output, German electricity price), the production costs of SNG amount to 0.135 €/kWh for the process chain in Troia. Compared to the other demo sites, the production costs are slightly higher. However, it should be noted that in addition to the newly developed milli-structured methanation reactor, also new technology like direct air capture (DAC) and small-scale liquefaction were used.

7 Bibliography

[1] Kost, C., Shammugam, S., Jülich, V., Nguyen, H.-T., SCHLEGL, T. Stromgehstehungskosten aus Erneurbaren Energien, März 2018.

[2] International Energy Agency. Natural gas information. Paris: IEA Publications, 2019. *IEA Statistics. ISBN* 978-92-64-49956-0.

[3] Deutsche Vereinigung des Gas- und Wasserfaches e.V. - Technisch-wissenschaftlicher Verein (DVGW). DVGW G 260, *Technische Regel. Arbeitsblatt G 260. Gasbeschaffenheit.* Bonn: Wirtschafts- und Verlagsgesellschaft Gas und Wasser mbH.

[4] Peters, M.S., Timmerhaus, K.D., West, R.E. Plant design and economics for chemical engineers. 5. ed., internat. ed. Boston: McGraw-Hill, 2004. *McGraw-Hill chemical engineering series. ISBN* 0071240446.

[5] Mosberger, E. Chemical Plant Design and Construction. In: *Ullmann's Encyclopedia of Industrial Chemistry.* Weinheim, Germany: Wiley-VCH Verlag GmbH & Co. KGaA, 2000. ISBN 3527306730.

[6] Konstantin, P. Praxisbuch Energiewirtschaft. Energieumwandlung, -transport und -beschaffung im liberalisierten Markt. 3rd ed. Dordrecht: Springer, 2013. *VDI-Buch. ISBN* 9783642372650.

[7] Zeymer, M., Herrmann, A., Oehmichen, K., Schmersahl, R., Schenider, R., Heidecke, P., He, L., Volz, F. Kleintechnische Biomassevergasung. Option für eine nachhaltige und dezentrale Energieversorgung. DBFZ Report Nr. 18. Leipzig, November 2013.

[8] Graf, F., Spörl, R., Eltrop, L., Colomar, D. Verbundvorhaben "Innovative Erzeugung von SNG und CNG aus biogenen Rest- und Abfallstoffen (Res2CNG)". Zwischenbericht.

[9] Bundesnetzagentur, B. *Er-mitt-lung der Netz-kos-ten* [online]. 2019. Verfügbar unter: https://www.bundesnetzagentur.de/DE/Sachgebiete/ElektrizitaetundGas/Unternehmen_Institutionen/Netzentgelte/Anreizregulierung/WesentlicheElemente/Netzkosten/Netzkostenermittlung_node.html

[10] Rindllisbacher, M., Hungerbühler, S. Leuchtturmprojekt Hybridwerk Aarmatt. Schlussbericht, 21. Dezember 2017.

[11] Smolinka, T., Wiebe, N., Sterchele, P., Palzer, A., Lehner, F., Jansen, M., Kiemel, S., Miehe, R., Wahren, S., Zimmermann, F. Industrialisierung der Wasser-elektrolyse in -Deutschland. -Chancen und -Herausforderungen für nachhaltigen Wasserstoff für Verkehr, Strom und -Wärme. Studie IndWEDe, 2018.

[12] Fachagentur für Nachwachsende Rohstoff e.V. *Faustzahlen Biogas* [online] [Zugriff am: 12. März 2020]. Verfügbar unter: https://biogas.fnr.de/daten-und-fakten/faustzahlen/.

[13] Schäffer, J., Ortloff, F., Lubenau, U., Imberg, C., Senner, J. Bewertung von Quellen und Abtrennungsverfahren zur Bereitstellung von CO2 für PtG-Prozesse. Abschlussbereicht, 16. April 2019.

8 Appendix

8.1 Definition of the Methanation Efficiency

In the following, the definitions for the methanation efficiency are given. The overall methanation efficiency is given in equation (3.3) in section 3.2.

Methanation efficiency:

$$\eta_{\mathrm{SNG,HHV}} = \frac{\dot{E}_{\mathrm{ch,SNG}}}{\dot{E}_{\mathrm{ch,H_2,in}}} \tag{8.1}$$

Methanation efficiency including heat usage:

$$\eta_{\mathrm{SNG,HHV,T_{use}}} = \frac{\dot{E}_{\mathrm{ch,SNG}} + \dot{E}_{\mathrm{th,use,SNG}}}{\dot{E}_{\mathrm{ch,H_2,in}}} \tag{8.2}$$

Overall methanation efficiency including heat usage, electricity demand:

$$\eta_{\mathrm{SNG,HHV,ov}} = \frac{\dot{E}_{\mathrm{ch,SNG}} + \dot{E}_{\mathrm{th,use,SNG}}}{\dot{E}_{\mathrm{ch,H_2,in}} + P_{\mathrm{el,Meth}}} \tag{8.3}$$

8.2 Testing Profiles

8.2.1 ST001

Stress test ST001 is done to have a reference for the following tests in this period. The test will be repeated in all periods to have reference profiles for detecting degradation over the whole project period. The sub-system electrolysis, methanation or the whole PtG system runs up to full load and runs in full load for 8 h. If full load over 8 h is not possible, choose one specific load and use the load for all monthly ST001.

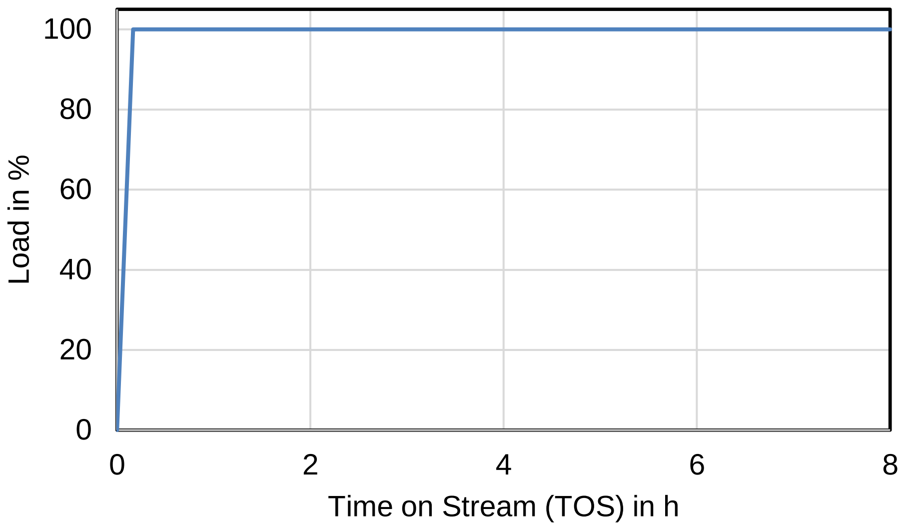

Figure 8-1: Load profile of the reference test.

8.2.2 ST008

Within stress test ST008, the maximum load change rate is defined. The minimum load change rate mentioned in the Grant Agreement is 5%/min for the methanation. After the first ramp-up to 50% load and a constant operation mode is achieved, the next ramp-up to 100% load should start. The load change should not reduce the methane concentration after the first reactor significantly, while H_2 and CO_2 are continuously fed to the reactor. The second ramp-up should start with the maximum allowed load change rate from 0% up to 100% load. The generic testing profile for ST008 is shown in the following graph.

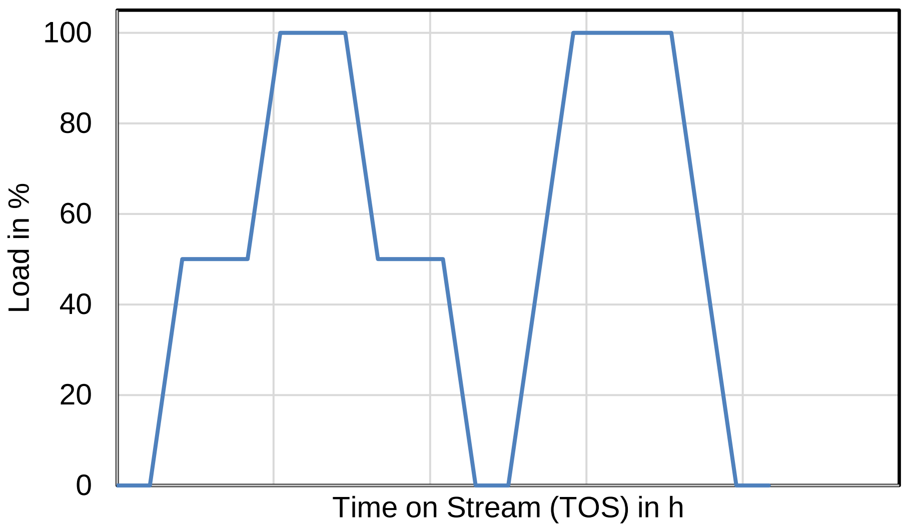

Figure 8-2: Load profile to define the maximum load change rate.

8.2.3 TC001 – Technical Characteristic Methanation

This test examines the efficiency of the methanation and the whole PtG plant in partial load as well as in full load. The specific energy requirement can be validated and compared with the manufacturer data. A constant operation mode regarding the product gas quality should be achieved after each ramp-up with a minimum of 20 minutes of operation at each load before switching to the next load. The main focus lies on the behaviour during load changes. The load profile is shown below:

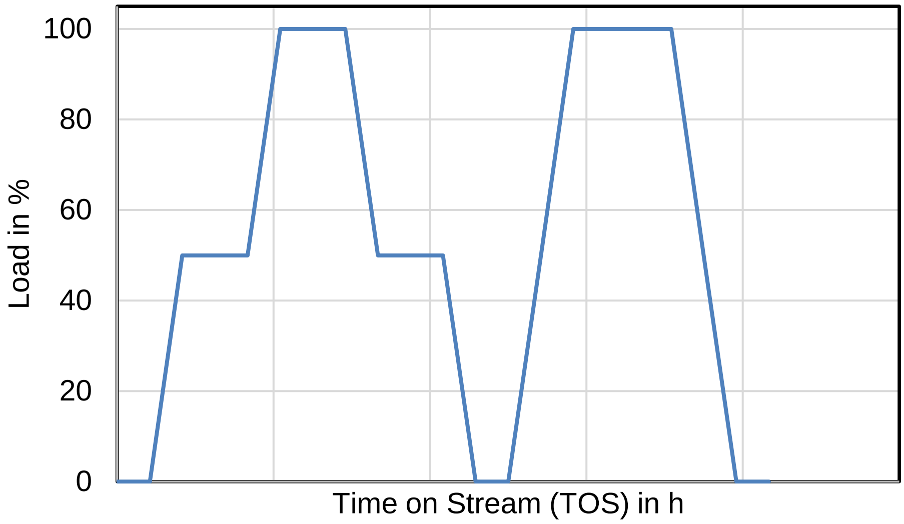

Figure 8-3: Test profile for testing the efficiency of the methanation at different loads.

8.2.4 TC002 - 72h test

The 72 h test aims to run the system over a long period with full load and check, if the quality of the output and the efficiency is always the same.

8.3 Flow Sheet Falkenhagen

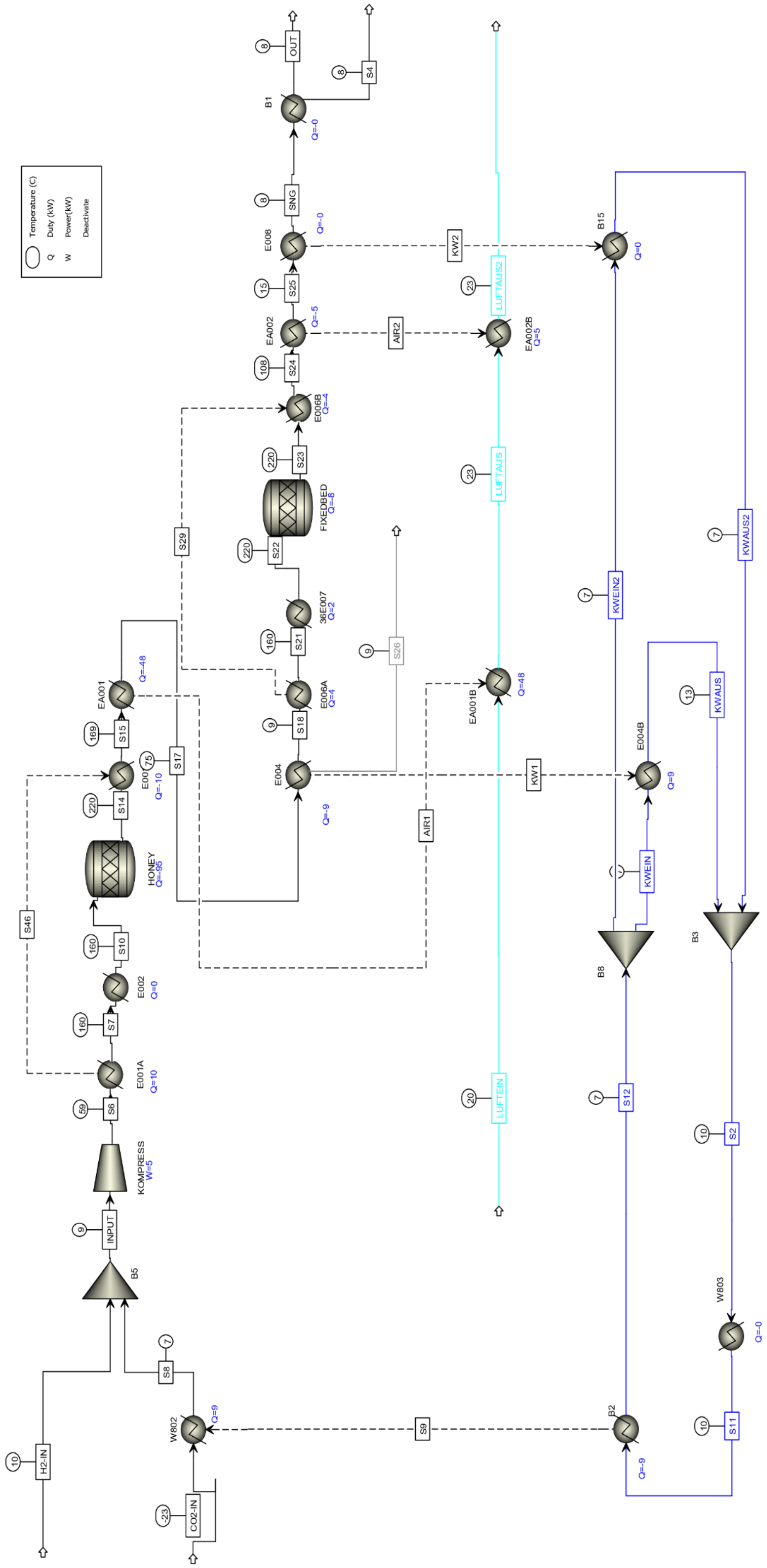

Figure 8-4: Flowsheet of Falkenhagen demo site in Aspen

8.4 Process Modelling based on ASPEN

In case of Falkenhagen, the measured stoichiometric ratio (y_{H2}/y_{CO2} = 4.35) deviated slightly from the ideal value of 4. Therefore, the measured inlet volumetric flow of H_2 ($\dot{V}_{H_2} = 200\ \mathrm{m^3/h}$) was set as input value. From this value, the inlet volumetric flow of CO_2 $\dot{V}_{\mathrm{CO_2}}$ was calculated using the ideal stoichiometric ratio of 4. Due to the assumption of ideal heat transfer, an ideal usage of heat sources can be required. Therefore, the oil circuit was not included in the ASPEN model, and for the calculation of the performance indicators (PIs) it was assumed that the total waste heat of the reactors could be used. The conversion rate of the respective reactors in Falkenhagen could be calculated with the measured methane fraction. In addition, an isothermal reaction temperature was assumed in the reactors. In agreement with the partner KIT, the reactor temperature is about 350 °C for the honeycomb reactor and about 240 °C for the fixed bed reactor. The electrical power demand for the demo site Falkenhagen was calculated based on measurement data. The measurement and control system of the plant in ASPEN was optimally set up so that all possible heat sinks and sources were used for heat integration. Therefore, no additional electrical heating was required except to heat the educt stream of the fixed bed reactor.

For the demo site Solothurn an almost ideal stoichiometric ratio of 4.05 was measured. Therefore, the input flows for the ASPEN model were only slightly adjusted to get an ideal stoichiometric ratio of 4.0. For the modelling of the biological methanation, the conversion rate for CO_2 was assumed to 98 %. This corresponds to the minimum conversion to achieve a methane content of more than 90 vol.-% in the product gas. The reaction temperature of the biological methanation was 62 °C. Due to the low heat level of the reaction, no heat usage was considered. The electrical power demand of the demo site was calculated to P_{el}=33.7 kW and includes e.g. the demand of the agitator, the CO_2 compressor and chiller.

The ASPEN model of the Troia demo site is only based on engineering data, see Deliverable 4.1 and 4.4. The input flows of CO_2 $\dot{V}_{\mathrm{CO_2}}$ and H_2 $\dot{V}_{\mathrm{H_2}}$ were in a stoichiometric ratio and were set to 10.2 m³/h and 40.8 m³/h, respectively. The milli-structured reactor was simulated as an equilibrium reactor. The reaction pressure inside the reactor was set to 4 bar. Equally to the demo sites Falkenhagen and Solothurn, the heat losses to the air were neglected. Hence, the reaction heat was considered for heat integration with the direct air capture unit (DAC) and the pre-heating of the educt stream. Additionally, the cooling of the product stream was identified as a possible heat source for heat usage, so it was assumed that the product stream was cooled to 80 °C. In Troia, the product stream was treated by membrane filtration. Therefore, SEPURAN membranes from Evonik were used at the demo site. The permeate (stream flows through the membrane) was recycled into the reactor. Due to missing measurement data, a two-component stream was assumed for the calculations of the composition of the recycle stream. The respective molar composition was calculated with help of the given selectivity of the membranes. Also, the electrical demand of the plant was estimated with help of Figure 4-5 of Deliverable D4.1 and was indicated as 44.5 kW. The demand includes the electricity demand needed for the DAC (minus the heat integration potential), for the BoP, and for the gas treatment. Due to the definition of the system boundaries, the electricity demand for the LNG unit was not considered.

8.5 Demo site history

Table 8-1: Overview of the scheduled activities during operation of the methanation plant in Falkenhagen from commissioning on 01/2019 until 02/2020.

Week	Planned/executed activities	Description
CW2 / CW3	SNG injection test	• Testing of the programming, the gas quality measurement in front of the injection, switch from SNG injection to flare • Testing of emergency stop scenarios
CW4	Approval for the injection in the ONTRAS gas grid	• Testing of programming and emergency scenarios • Injection testing • Approval by ONTRAS
CW7	Testing of all components and their functional interaction	• Testing of SNG injection and switch to flare • Adaption of the automatic educt stoichiometry controller • Emergency stop caused by programming issues
CW8	Free testing of all components and their functional interaction	• Test of educt stoichiometry controller and flare • Failure of the compressor for the injection to the gas grid. (caused by signal failure from ONTRAS grid, long time troubleshooting)
CW11	Free testing of all components and their functional interaction	• Mechanical failure of the clutch of the oil pump. • Only limited testing possible
CW12	Free testing of all components and their functional interaction	• Exchange of sensors and calibrations to improve educt stoichiometry controller • Testing of the switching moment to the flare • Test to reach maximum load (reached load only 82 % due to a to low pressure in front of the educt compressor)
CW13	Free testing	• Visit of HSR and DVGW (WP5) on site • Reference point measurements
CW16	Free testing of all components and their functional interaction	• Test of 100 % load. On 17.05.2019 approx. 95 % of load was reached (electrolysers were the limiting factor)
CW18-CW21	Modification of the methanation plant and extensive maintenance at the old system	

CW24	Free testing of all components and their functional interaction	• Error in the programming of the electrolyser system prevent the start of the methanation plant
CW27	Free testing of all components and their functional interaction	• At the end of the week, the failure in the PCS (Electrolysers) was fixed but unfortunately no testing of the site was possible
CW35	Free testing of all components and their functional interaction	• Test of start and stop sequences for the methanation plant • Implementation of the interface to the Dispatch Centre Düsseldorf
CW37	Free testing of all components and their functional interaction	• Test of start and stop sequences for the methanation plant • Implementation of the interface to the Dispatch Centre Düsseldorf
CW38	Testing of various emergency scenar-ios.	• test of flushing times with H_2 and N_2 • test of the SIL systems on the methanation
CW49	Test of PtG	• First test after re-assembling of the PtG plant (6 electrolysers + compressor unit)
CW50	Test of methanation	• General functional test of the methanation
CW2 2020	Free testing	• Collection of operating hours (on-site)
CW3 2020	Free testing	• Collection of operating hours (on-site)
CW4 2020	Free testing	• Collection of operating hours
CW5 2020	Free testing	• Collection of operating hours; visit from WP 5 (Test of 100 % load; test programme from WP5
CW7 2020	Free testing	• Collection of operating hours (on-site)
CW9 2020	Free testing	• Collection of operating hours (on-site) • Test of remote steering of the site from Düsseldorf and Potsdam

Table 8-2: Overview of the scheduled activities during operation of the methanation plant in Solothurn from commissioning on 1 September 2018 until 02/2020.

Month	Planned/executed activities	Description
01/19	Final adaptions agitator Completion of plant insulation	All leakage and control system tests completed
02/19	Inoculation of archaea Start-up of plant Plant shut-down	Reactor filled with archaea Initial start-up of plant Control system adaptions required, operation suspended Control system adaptions
03/19	No operation	Control system adaptions
04/19	No operation	Control system adaptions
05/19	Resuming operation	All systems checks performed, plant heated up, operation initiated
06/19	Operation	First SNG injected into grid
07/19	Operation	O_2 probe CO_2 compressor Haug defective, replaced initiated analyser calibration, problems with CH_4 analyser drift CH_4/CO_2 sensor defective, new part ordered leaking of chiller, defective rubber seal
08/19	Operation	leakage of cooler Chiller system pressure high alarm improved gas quality, reaching value > 96%
09/19	Operation	Problems electrolysers, CO_2 compressor solenoid valves Chiller failure Flare failure
10/19	Operation	Oil level low agitator Problems with control system 5000 h of operation reached
11/19	Operation	24 h operation for 6 days
12/19	Operation	Plant shut-down for the year, 17.12
01/20	Operation	Operation resumed 13.01.2020 Load tests performed Reaching 1000 h of plant operation
02/20	Operation	Final load tests performed Commissioning of membranes Final day of operation 27.02., plant shut-down at 16:20

Table 8-3: Overview of the scheduled activities during operation of the methanation plant in Troia from commissioning on 01 Jan 2020 until 02/2020.

Month	Planned/executed activities	Description
01/19	WE reactivation Individual tests	H_2 leakages in the buffer fixed, new PT installed and controlled to guarantee fix inlet pressure to the methanation system. SNG production from cylinders to the flare. LNG liquefaction from cylinders.
02/19	Individual tests	SNG production to the flare. LNG produced from cylinders.
03/19	Individual tests - not possible	Unavailability of N_2 at the plant. Definition of the commissioning tests. Implementation of the central SCADA.
04/19	General commissioning (technical)	First results with SNG production and LNG liquefaction with both the systems coupled. Recycle tested as well.
05/19	Individual tests	Tests on insulation in the methanation system. SNG production to the flare.
06/19	General commissioning (administrative)	Attainment of the last missing certification and documentation for the Fire Brigades (ending in August, with the validation of ATM's gas analyser). Operators found, training sessions planned.
07/19	Training session	Training of the local operators by ENG (SCADA, process, tests), BFP (fire and video surveillance) and CW (DAC unit)
08/19	Training session Shared test session	Training of the local operators by ATM (methanation system). SNG production to the flare. ST001 executed (no liquefaction).
09/19	Training session	Training of the local operators by HST (LNG unit). SNG production and LNG liquefaction with both the systems coupled. Recycle tested as well. Lack of nitrogen. Good quality tests.
10/19	Operational phase: Free operations	Restart of the plant after the unexpected administrative halt. Nitrogen provided. SNG production to the flare.
11/19	Operational phase: Free operations	SNG production to the flare. Extra maintenance on the methanation system needed for H_2 leakages in the reactor. Dismantling and welding of the reactor. Remote control of the plant made available through SCADA,
12/19	Operational phase: Free operations	SNG production to the flare. LNG liquefaction tested but not possible in a stale way for SNG unexpected non conformity. Check for the causes, fixed during the holidays.

01/20	Operational phase: Free operations Shared test sessions	Plant running in a stable way, with all devices coupled. SNG production and LNG liquefaction with recycles. ST001 and TC001 successfully performed. H24 operations and flare automatized by the SCADA. Very good quality of the tests.
02/20	Operational phase: Free operations Shared test sessions	Plant running in a stable way, with all devices coupled. SNG production and LNG liquefaction with recycles. Heat valorisation tested. ST001, ST006, ST008, TC001 and TC002 successfully performed. H24 operations and flare automatized by the SCADA. Safety loop perfectly functional. Very good quality of the tests.

8.6 Additional Measurement Data

8.6.1 Solothurn

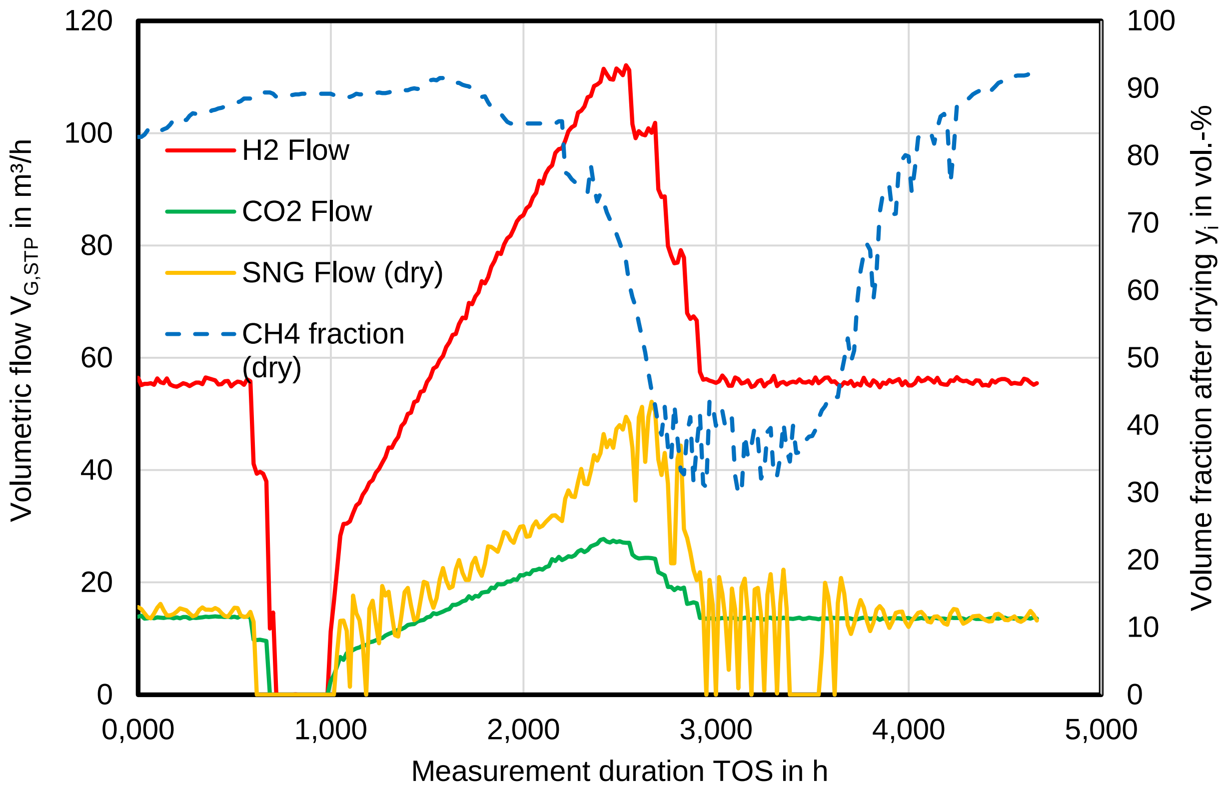

Figure 8-5: Measurement results from the demo site Solothurn (10/30/2019, 8:00 am – 11:40 am).

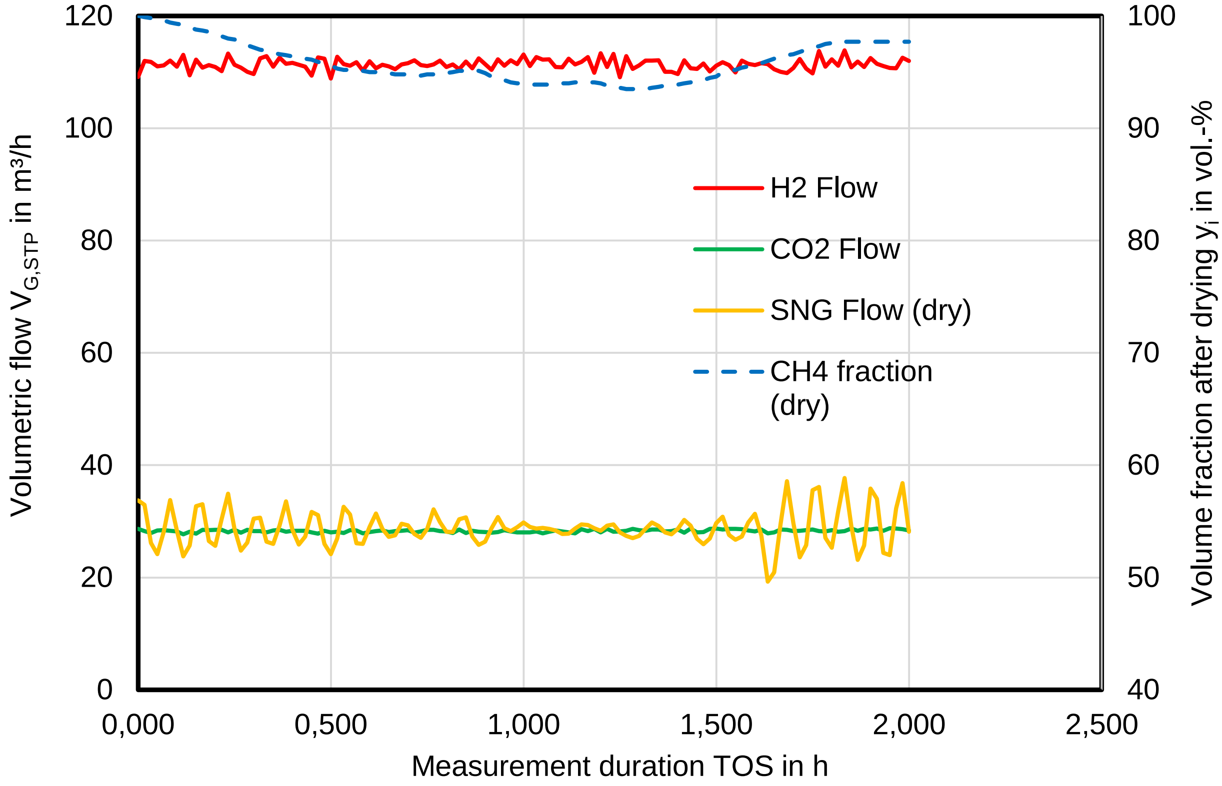

Figure 8-6: Measurement results from the demo site Solothurn (02/13/2020, 1:00 pm – 3:00 pm)

8.6.2 Troia

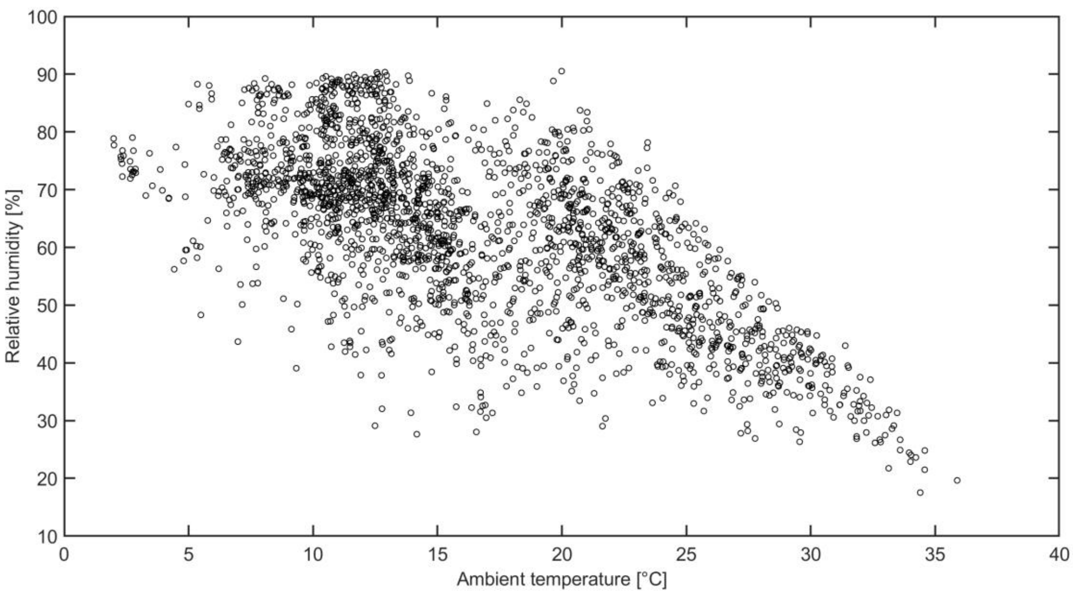

Figure 8-7: Weather fluctuations during the operation of the DAC in Troia

8.7 Additional Data for Economical Evaluation

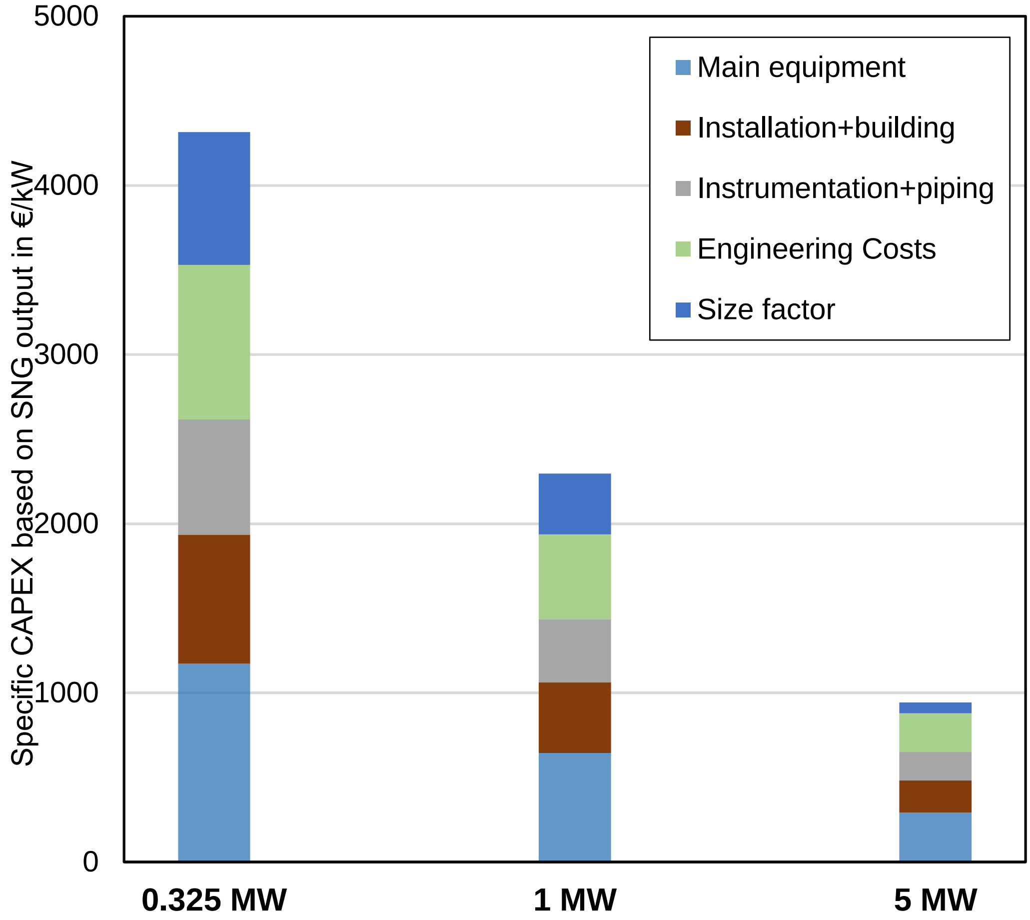

Figure 8-8: Specific CAPEX of the demo site Solothurn and of the plant scaled to an SNG output of 1 MW and 5 MW.