Cost benefits of optimizing hydrogen storage and methanation capacities for Power-to-Gas plants in dynamic operation

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HIGHLIGHTS
- Optimization tool is developed for dimensioning Power-to-Gas components.
- Detailed Power-to-Gas cost analyses are made for different operational environments.
- 6–17% reduction in gas production costs was achieved via component dimensioning.
- Sensitivity analyses show impacts of key parameters on plant operation.
- Optimal configurations are highly dependent on the electricity source being used.

ABSTRACT
Power-to-Gas technologies offer a promising approach for converting renewable electricity into a molecular form (fuel) to serve the energy demands of non-electric energy applications in all end-use sectors. The technologies have been broadly developed and are at the edge of a mass roll-out. The barriers that Power-to-Gas faces are no longer technical, but are, foremost, regulatory, and economic. This study focuses on a Power-to-Gas pathway, where electricity is first converted in a water electrolyzer into hydrogen, which is then synthetized with carbon dioxide to produce synthetic natural gas. A key aspect of this pathway is that an intermittent electricity supply could be used, which could reduce the amount of electricity curtailment from renewable energy generation. Interim storages would then be necessary to decouple the synthesized part from hydrogen production, to enable (I) longer continuous operation cycles for the methanation reactor, and (II) increased annual full-load hours, leading to an overall reduction in gas production costs. This work optimizes a Power-to-Gas plant configuration with respect to the cost benefits using a Monte Carlo-based simulation tool. The results indicate potential cost reductions of up to 17% in synthetic natural gas production by implementing well-balanced components and interim storages. This study also evaluates three different power sources which differ greatly in their optimal system configuration. Results from time-resolved simulations and sensitivity analyses for different plant designs and electricity sources are discussed with respect to technical and economic implications, so as to facilitate a plant design process for decision makers.

1. Introduction

In the context of a transition towards a sustainable energy system, the European Council [1] has proclaimed that the share of renewable energy is steadily increasing. However, the Intergovernmental Panel on Climate Change (IPCC) report [2] states that carbon dioxide (CO₂) emissions will only decrease with near-term mitigation efforts. Strong local positive and negative residual loads will occur, owing to the fluctuations of renewable energy sources (RES) such as photovoltaics (PV) or wind power. Today, such discrepancies between supply and demand are compensated for by shifting the loads of electricity producers and consumers (demand side management).

Abbreviations: AEL, Alkaline electrolysis; BoP, Balance of Plant; CAPEX, Capital expenditures; CH₄, Methane; CO₂, Carbon dioxide; H₂, Hydrogen; PEM, Polymer electrolyte membrane; PtG, Power-to-Gas; PV, photovoltaics; RE, renewable energy; RES, renewable energy source; SCR, Secondary control reserve; SNG, Synthetic natural gas; SOEL, Solid oxide electrolysis

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To avoid renewable energy curtailments it is essential to increase flexibility in all parts of the energy system. Different approaches for providing flexibility possess significant technical and economic potential [3]. One technology for the flexible use of renewable electricity and ancillary services is Power-to-Gas (PtG). PtG uses electrical energy to produce hydrogen (H₂) by water (H₂O) electrolysis. The H₂ molecules can further be converted into methane (CH₄) via the Sabatier process by adding CO₂. The product gas (synthetic natural gas, SNG) can be fed into natural gas grids, as it has almost the same physical properties as natural gas itself. The gas grids can be used for seasonal energy storage taking advantage of the large underground storage capacities existing in Europe [4]. By synergetic integration of the PtG process chains, the inter-sectoral coupling of energy, industry, mobility, domestic and commercial sectors can become reality [5].

Breyer et al. [6] states that PtG can be profitable in the case of a flexible operation mode offering electricity grid services. Götz et al. [7] and van Leeuwen and Zauner [8] noted the high investment costs, low full-load operating hours, and high electricity costs. Nevertheless, costs can change rapidly when implementing new support mechanisms and may trigger learning curve effects for storage options on a rapid time scale.

Another possibility for reducing gas production costs (GPC) is the optimization of the system concept and the operation. Different dynamics of the electrolysis and methanation processes can be exploited [9]. Whereas electrolysis reacts to changes in the electrical input energy within seconds, methanation takes several minutes to adjust the production rate while maintaining the SNG quality [10]. H₂ and CO₂ storages are mostly used to provide sufficient suction volume in front of compressors and to avoid the transmission of pulsation after the compressor. If the storage tanks are designed to be larger, methanation can be conducted independently of electrolysis. In Audi’s PtG plant [10], a hydrogen storage tank was designed for half an hour of independent operation. By optimizing the methanation capacity and the size of the hydrogen storage, the investment costs, and therefore the methane production costs can be reduced.

To make full use of the available electricity, the PtG plant must be prepared for power peaks and load changes by design. This study examines the potential of reducing methane production costs by determining optimal capacities of intermediate hydrogen storage and methanation for three electricity supplies.

2. Power-to-Gas

PtG is a term for technologies for converting electrical energy into a gaseous chemical energy carrier. In the following, PtG refers only to the process of using (excess) electrical energy from predominantly renewable sources to produce synthetic CH₄ via the intermediate product H₂ from water electrolysis and CO₂. The PtG products serve as fuels for non-electric energy markets or seasonal energy storage, and are substitute for their fossil-based analogue with an aim of decarbonization. Fig. 1 shows the main components of a PtG plant in a block flow diagram. The first production step is always electrolysis of water using (excess) electrical energy. In the case of methanation plants, the second stage converts the H₂ into CH₄ and H₂O via the Sabatier reaction, by adding CO₂. The heat released from the exothermic reaction can be used within the process or externally. The produced gas needs to meet the gas quality requirements for the natural gas grid. In methanation the chemical efficiency is directly connected to the conversion starting with 100% at 0% conversion and 78% (83%) at 100% conversion, based on the upper (lower) calorific value [11]. Higher conversion help to keep effort for upgrading small.

2.1. Applications for Power-to-Gas plants

PtG can play several roles in a future energy system. The literature reviews by Lewandowska-Bernat and Desideri [12] and Mazza et al. [13] discuss four main fields of application:

- Large-scale and long-term storage of renewable energy
- Services to balance the loads in electricity networks
- A considerable source of clean fuel for heating or transportation
- Contribution to emission reduction targets

Luo et al. [14] provides an overview of the current application potential of PtG and other storage technologies in power system operation. A PtG plant can basically be operated in two ways. In input-oriented operation, the CH₄ production is determined by the availability of the reactants (mainly electrical energy, and where necessary, CO₂ and H₂O). In contrast, in output-oriented operation, CH₄ needs to be produced according to a defined production volume. This study analyzes input-oriented operation.

PtG can have advantages for electricity grid operation if the PtG system runs in the input-oriented mode and absorbs excess renewable electricity from solar power [15] or wind [16]. Guandalini et al. [17] investigated additional management principles for a gas turbine and a PtG plant balancing the system. All three papers concluded, that curtailments of fluctuating RES can be reduced by absorbing surplus energy with operating PtG plants. The German Bundesnetzagentur [18] declared curtailments of 5,518 GWh of renewable energy production in 2017, owing to overloads in the electricity grid. The excess electricity could have been used by flexible consumers, such as in PtG processes. The integration of PtG into an energy system reduces the overall carbon footprint [19,20]. Qadrdan et al. [21] showed that an overall energy system cost can be reduced by the use of PtG.

Another input-oriented operation of PtG is in direct coupling with RES, and without connections to the public electricity grid. Feasibility studies were performed by [22,23], and [24]. Norway [25], Scotland [26], and Spain [27] tested direct coupling in demonstration projects. One advantage of direct coupling is that there is no need to pay network usage fees, and thus operating costs can be reduced. However, the resulting lower number of operating hours is a disadvantage.

A PtG system can be designed to offer control reserve. Control
reserve is used to compensate for mismatches between production and consumption in the electricity grid that lead to deviations in the grid frequency. The aim of control reserve is to keep the frequency within certain a tolerance range (approximately 50 Hz), and to eliminate possible regional deviations of the power balance from its target value. Dynamic producers and consumers are necessary to provide control reserve. The control reserve is divided into primary, secondary, and tertiary control reserve according to dynamic and temporal requirements. Guinot et al. [28] investigated a case where the electrolysis process provided primary reserve to a French TSO. The authors assumed that an electrolyzer operator would not benefit from participating in the frequency regulation market, given the technical and economic assumptions made. Simulations show that a theoretical benefit, in that bidding on the secondary control reserve (SCR) market can obtain low hydrogen production costs of 1.1 €/kg [29].

2.2. Dynamic operation and optimization of Power-to-Gas plants

One technical challenge of input-oriented PtG operation is coping with the different load dynamics of the electrolyzer and methanation sub-systems. Methanation may not be able to process the produced hydrogen instantly while maintaining the product gas quality. If PtG systems face a highly fluctuating load profile, the two sub-systems must be decoupled and operated separately. Independent sub-system operation leads to more continuous production, which in turn can positively influence the CH₄ yield. Furthermore, the maximum hydrogen processing rate of the methanation reactor can be lower than the maximum production rate of the electrolysis. The hydrogen storage can help to maintain a load interval and load change rates that maintain the gas quality. The size of the interim hydrogen storage needs to be optimized for the operation strategies on a case-by-case basis [30].

2.2.1. Dynamic characteristics of water electrolysis technologies

Today, three water electrolysis technologies are on the market for medium- to large-scale projects: alkaline electrolysis (AEL), polymer electrolyte membrane electrolysis (PEM), and solid oxide electrolysis (SOEL). Butler and Sliethoff [31] identified twenty manufacturers of AEL and twelve manufacturers of PEM. The SOEL technology is only represented by one supplier. According to the manufacturer’s specifications, PEM electrolysis by Siemens can vary between 0% and 100% of the nominal load, and requires less than 10 s from cold standby to the nominal load [32]. The AEL specifications for load flexibility are between 20% and 100% of the nominal load and a start-up time between 1 and 5 min. The Falkenhagen project [33] showed that a start-up time of 1–2 min is possible with current-controlled AEL. In general, a load change rate of at least 20%/min can be assumed for alkaline electrolyzers. This corresponds to the minimum requirement for the provision of SCR in Germany [34]. According to the tender documents of the national grid company Swissgrid in Switzerland, the SCR is activated and deactivated with a power change of 0.5% of the nominal power per second. In a case of primary control power, complete power is called up in 30 s (ca. 3.3%/s) and after a further 30 s the power must be within a tolerance limit of ± 2.5% of the nominal power [35].

The suitability of PtG for auxiliary services has been proven in practice. The Audi PtG plant in Werlte has successfully completed the qualifications for offering SCR using AEL [10]. The same qualification was obtained by the PEM electrolysis system in Energiepark Mainz [36].

The Thüga Group’s demonstration plant in Frankfurt qualified for primary control reserve with an PEM electrolysis system [37].

To react smoothly to time-variable dynamics the application of a model predictive control approach was tested in [38]. It was shown that the controls of the electrolyzer and hydrogen storage are capable of optimizing operation with respect to time-variable electricity prices, while operating within the limitations of the gas and electricity networks.

2.2.2. Dynamic characteristics of methanation technologies

Methanation shows undesirable changes in product gas composition when process conditions are affected. Rapid adaption of the input flow rates can lead to pressure fluctuations and temperature changes. Moderate adaption is necessary to keep the gas quality constant. The exothermic methanation reaction produces temperature gradients during start-up, which can cause mechanical stress or changes in the active centers of the catalyst, that deactivate the catalyst [39] in fixed bed reactors. Tests from the Zentrum für Sonnenergie- und Wasserstoff-Forschung Baden-Württemberg (ZSW) [40] have shown that the product gas quality of plate and tube bundle reactors can be kept constant for load changes between 100% and 70%, with a load change rate of ± 3%/min (see Fig. 20 in the Appendix A). It was shown that the temperatures in the reaction equilibrium and in the gas composition of the product gas remained almost constant during 900 cycles [41]. Although thermal deactivation cannot be generally excluded, the results indicate an expected lifetime of 10 years.

Biological methanation takes place in liquid phase at operation temperatures between 40 and 70 °C [42]. Lecker et al. [43] gives an overview of biological reactor designs and enhancement concepts. The minimum load of a biological methanation is not limited on biology [42]. However, the minimum load should clearly exceed the energy consumption of the stirrer [7]. Inkeri et al. [44] studied dynamic operation by analysis of the effects of load change, shut-down, and start-up. The reactor performed well in response to these changes in operating conditions, but control logics or buffer storage solutions were necessary to keep the CH₄ content above 95 vol% within the product gas stream.

2.2.3. Optimization of Power-to-Gas systems

Hydrogen storage systems are used to decouple electrolysis and methanation, as these sub-systems differ in dynamic behavior [9]. In the first commercial chemical methanation plant in an industrial environment, a medium-pressure hydrogen storage between electrolysis and methanation was expanded after commissioning, to further decouple the methanation from the electrolysis [10].

A suitable size of the hydrogen storage depends on the profile of the electrical input of the electrolyzer and the methanation capacity. As a result, the optimal storage size must be evaluated individually for each PtG plant. Well-balanced hydrogen storage and methanation capacities increase the annual full-load hours, and decrease CH₄ production cost.

3. System description

Fig. 1 shows the four system components studied in this work (green): power source, electrolysis, hydrogen storage and methanation. All remaining units are summarized as the Balance of Plant (BoP). The technical and economical parameters considered for the calculation of the SNG production costs are based on expectations for 2030. The economical parameters are the capital expenditures (CAPEX) and operational expenses (OPEX), for electrolysis, hydrogen storage, methanation, and the BoP. Approaches for validating the GPC include calculations of the present value of the total costs or the levelized costs of energy (LCOE) [45], also called the levelized costs of storage (LCOS) for energy storage applications [46]. In this publication the approach of LCOE is adopted for calculating the GPC of SNG (see Eq. (1) and Table 1 based on [45]) for the first year of operation. The CAPEX term uses a capital recovery factor to convert the total investment into periodic payments. The SNG term is simply the amount of SNG produced during the first operation year. The resulting GPC is about 5% larger than with the LCOE method. Furthermore, there is no energy cost in the basic calculations, only in Fig. 13. The GPC allow a cost comparison of different system configurations and modes of operation for producing SNG. A sensitivity analysis dedicated to the electricity cost is included in the work.
Table 1

Variables for the calculation of the gas production costs (GPC).

| Variable | Description |
|----------|-------------|
| CAPEX | Capital expenditure |
| OPEX | Operation and maintenance expenditure for one year |
| E | Electricity and heat costs for one year |
| SNG | Amount of SNG produced in one year |
| r | Interest rate |
| n | Component lifetime |
| i | Component index (for instance, electrolyzer, methanation unit) |

\[
GPC = \sum_{i=1}^{l} \frac{CAPEX_i}{(1+r)^n} + OPEX_i + E_i 
\]

3.1. Power source

Three data sources are used to derive the load curves for the electrolysis:

- Measurement data from a PV plant located in Switzerland, hereafter referred to as PV.
- SCR market data from Swissgrid in Switzerland, hereafter referred to as Control.
- Measurement data from a wind farm located in Northern Germany, hereafter referred to as Wind.

The load curve characteristics are listed in Table 2.

The PV profile was generated using 2016 solar radiation data from the city of Zurich, which were normalized to a peak power of 10 MW. The red dashed line in Fig. 2 shows the characteristic PV profile displaying a zero-production at least 50% of the time.

The installed capacity of the Wind power data was normalized to 10 MW as well. For wind farms, fluctuations of up to 25%/min are typical [47]. Compared to PV and Control, the Wind profile studied herein exhibits the highest dynamic requirements, with load change rates between −8.3 and +7.7%/min.

The load curve for Control is obtained from the Swissgrid for the year 2015 [48]. To simulate a case for SCR, calls from the control zones in Switzerland were accumulated and normalized to an output of 10 MWel. It is assumed that 5 MW of positive and 5 MW of negative SCR are offered. Therefore, the PtG plant has to operate at a nominal capacity of 5 MWel. Owing to this operation strategy, the Control profile provides the highest full-load hours (4459 h) as compared to PV and Wind.

3.2. Electrolysis

For the modeling of the PtG plant, an alkaline electrolyzer with a power input of 10 MWel and 30 bar operation pressure is assumed as a fixed input size for all cases. Smolinka et al. [49] predict the efficiency of a complete electrolyzer system for 2020–2030 to be in the range of 4.3–5.7 kWh/m³ (62–82%), and Butler and Spliethoff [31] predict 78.7%. This study assumes an overall efficiency of 75% (PAC/HHV H2), including cooling and BoP over the entire operating range. With a size of 10 MWel, most of the time the electrolyzer works far from the nominal operating conditions with the assumed power sources. This affects the efficiency, but for simplification, a constant efficiency is assumed. The electrolyzer can operate between 0 and 100% load [50], and a maximum load change rate of 20%/min (Table 3) is assumed. With this load change rate, the plant is able to follow the three previously-defined input power profiles. The lifetime of the electrolysis stack is up to 10 [51] years, or between 20,000 and/or 50,000 [49] and 120,000 [50] operation hours. The entire system has a lifetime between 20 [7] and 50 years, as stated for stationary operated AEL [31].

The produced hydrogen is fed into the storage, and is subsequently fed to the methanation process. If the electrolysis produces excess hydrogen, i.e., more than can fit in the storage, it is flared. Dismissing hydrogen should be the last option to fulfill a contract with an electricity supplier or ancillary services. In the simulations, it is assumed that H2 is always produced if electricity is available. The parameters of the electrolyzer for the year 2030 are based on literature and own calculations (Table 3). An economy of scale affects the CAPEX of an electrolyzer. The capital expenditure (CAPEX) is calculated based on the public results of the STORE&GO deliverable D7.7 [52].

3.3. Hydrogen storage

Hydrogen can be stored in gaseous, liquid or hydride form. To this day, only gaseous storages at various pressure levels are used in large-scale PtG plants. Some electrolyzers can provide hydrogen at pressures between 10 and 30 bar [50]. The hydrogen can thus be stored directly in pressure vessels by limiting the operation of storages to that pressure range. The upstream pressure for a methanation unit is assumed to be 10 bar.

Hydrogen can also be stored at higher pressures using additional compressors. High compression is preferred for applications where volume is sparse, such as in the mobility sector [8]. As it is more expensive, this study focuses on medium pressure storages, i.e., up to 30 bar. These reservoirs are highly standardized. According to internal
offers from manufacturers, the prices vary between 375 €/kg (50 bar; 33 €/m$^3$) and 490 €/kg (200 bar; 44 €/m$^3$). Van Leeuwen and Mulder [57] cite a cost range of 20–100 €/m$^3$ with no indication of maximum operating pressure. No operational constraints have been set for the dynamic operation of storages, i.e. any loading and unloading tasks are completed instantly and fully. As the chosen methodology in this work is based on a Monte Carlo optimization, it is necessary to define a range of valid hydrogen storage capacities from which samples are drawn. A range between 100 kg and 3000 kg was deemed practicable. Table 4 summarizes the parameters assumed for the hydrogen storage.

### 3.4. Methanation

Chemical-catalytic methanation in a cooled fixed-bed reactor is chosen for this publication. The theoretical energy efficiency with complete conversion of the reactants is 77.9%, based on the higher heating value (HHV) of the hydrogen and SNG [11]. Owing to thermodynamic limitations, pressure, and temperature dependency, the conversion is incomplete, and a CH$_4$ content of 80–93 vol% is achieved in the dried product gas [58]. The CH$_4$ content can be increased to > 96 vol% by additional membrane treatment. This work assumes a conversion rate of 100% of H$_2$ in the methanation reactor. The power consumption of the BoP, such as pumps and compressor, reduces the theoretical overall efficiency of 79% at 100% conversion to 69%.

SNG production can be interrupted by bringing the reactor to hot standby mode, where the reactor is held at a set temperature and typically flushed with hydrogen to prevent carbon deposits on the catalyst surface. In this work, whenever the methanation reactor is set in hot standby and restarted, the flushed hydrogen and the SNG out of the specification limits are considered to be flared, and have no value. Both the flushing and restarting stage are conservatively assumed to require 10 min. In the Audi plant, approximately 1/12 of the hydrogen requirement for a full-load operating hour is rejected by a start-up and shut-down cycle [41].

In the simulations, the pressure in the hydrogen storage controls the start and the load of the methanation. Between a pressure of 10 bar and the maximum pressure of 30 bar, the load varies linearly between 40% and 100%. The methanation reactor is implemented to enter hot standby mode if the hydrogen storage pressure drops below 10 bar. A minimum load of 40% is set for the methanation because of the increasing inaccuracy of measurement equipment with low flow rates. The maximum load change rate is set at 3%/min to keep the gas quality constant. The reactor starts only once the storage level rises above 18 bar, to prevent frequent on-off cycles. A sensitivity analysis is performed to assess the importance of the duration until reaching the required gas quality, as well as the storage pressure level before restarting the methanation.

The capacity of methanation is one of the optimization variables. The smallest theoretical methanation capacity is defined so that the reactor can completely process the annual hydrogen produced in the specific electricity profile, when operating the methanation at full load at all times. A capacity smaller than this would result in an annual H$_2$ overproduction. The theoretical maximum capacity for the reactor is achieved when the maximum rate of hydrogen production does not exceed the maximum rate of hydrogen conversion in the methanation reactor – even if the electrolysis operates at full load. In this study, the electrolyzer was always fixed to a capacity of 10 MW$_{el}$ and 75% efficiency, so the resulting maximum capacity of methanation is always 5.75 MW$_{el,SNG}$ (100% conversion rate, 55.66 MJ/kg). To find the minimum of the GPC, simulations were carried out with a range larger than this, from 0.1 to 7.5 MW$_{el,SNG}$ (HHV). The lifetime of a methanation reactor is assumed for 2030 from ENEA [59] with 20 years, whereas Moeller et al. [60] expect 30 years. For this work, a lifetime of 20 years is assumed. The cost estimates for 2030 are based on [8] and [55], and on our own calculations. The illustrated cost values include the cost of peripheral devices such as heat exchangers and compressors. Table 5 summarizes the relevant parameters used in simulations.

#### 3.5. CO2 capture and balance of plant

The CO2 required for CH$_4$ synthesis can originate from various sources, such as raw biogas, industrial point sources or ambient air. This study focuses on the separation of CO$_2$ from industrial point sources by means of amine scrubbing. After capture, the CO$_2$ is assumed to be compressed to 10 bar. The capture capacity is set to exceed the annual demand by approximately 23%, to guarantee sufficient CO$_2$ availability at all times. No buffer storage for CO$_2$ was modeled, as it would have introduced a new optimization parameter, and CO$_2$ was not considered as a focal point of this study. CO2 storage is less cost-intensive because four times less CO$_2$ is needed as compared to H$_2$, and ther are fewer material requirements.

It is expected that the operation costs of CO$_2$ capture from industrial waste gases will range from 25 to 135 €/tCO$_2$ [61]. The exact costs depend on the size of the plant and the composition of the exhaust gases. In this work, the chosen parameters result in an average capture cost of approximately 50 €/tCO$_2$. The same average price per ton of CO$_2$ is achieved for all scenarios by adjusting the capital expense. A fixed price is chosen, because CO$_2$ capture is assumed to obtain electricity from the grid.

### 4. Simulation specification

The objective of the Monte Carlo simulation was to find optimal capacities for the H$_2$ storage size and methanation that lead to the lowest possible levelized production cost for SNG. Monte Carlo methods allow detailed sensitivity analyses and deliver robust results, as instead of straightforward searching for an optimum value, a larger range of plausible input parameter values can be mapped. For instance, situations where different input configurations result in identical outcomes are readily observed. Monte Carlo simulations are especially useful when input values exhibit uncertainties, which often applies to assumptions for capital and operational expenses [62].

A MATLAB script from a previous work was utilized for performing the simulations [63]. Computations were performed in parallel, and over 2 million cases were simulated in total. Samples for the methanation capacity and storage size were drawn from a uniform distribution, where the lower and upper limits were set to represent practicable limits using a trial-and-error approach. For instance, it was found that increasing the hydrogen storage beyond 3 tons did not bring any significant benefits for the system. Likewise, the methanation capacity range can be fixed based on the theoretical investigation, as discussed in detail in Section 3.4. In practice, the performance of the system is evaluated at every imaginable condition between the lower and upper limit, and the economical optimum is identified. Naturally, the optimum is only valid for the current set of parameters and assumptions. For instance, variations in electricity profiles or cost parameters would...
lead to a new optimum.

The simulation process was divided into two phases. In the first phase, the hydrogen mass balance was tracked as it progressed through the PtG system. Hydrogen production was dependent on electricity profiles, and the consumption rate in methanation was calculated depending on the H₂ storage level at each time step. In the second stage, the costs of the system were calculated based on the input values and data obtained during the dynamic simulation phase.

### 4.1. Operation concepts

Three power sources (Wind, PV and Control) were selected to illustrate the optimization of the hydrogen storage size and the methanation capacity suitable for a 10 MWELAC electrolysis. In addition, two separate system schemes were distinguished: Standard and Full concept. The Full concept mandates that all produced H₂ must be converted into SNG, whereas the Standard concept allows some H₂ to be discarded.

For the Standard case, a Fixed variant was also analyzed. Here, the methanation capacity was fixed instead of treating it as an optimization variable. The numerical limit for the methanation capacity is 5.75 MW, as derived in Section 3.4. As the chosen capacity limit is at the theoretical maximum, the Fixed variant actually also fulfills the Full criterion, but most likely with a different configuration (see Table 6).

### 4.2. Sensitivity analysis

A sensitivity analysis was performed to estimate the importance of various input values. Specifically, the following input variables were investigated:

1. CAPEX of electrolyzer
2. CAPEX of hydrogen storage
3. CAPEX of methanation
4. STANDBY COST – Additional cost for maintaining the methanation reactor in standby mode
5. RESTART LEVEL – The storage level which triggers the methanation reactor to restart its production after a shutdown
6. RESTART TIME – The time it takes to reach the desired product quality after the methanation has been restarted – all SNG produced before the quality is met will be discarded

The sensitivity analysis focused on the results of Standard concept simulations using the Wind power source.

### 5. Results and discussion

In this section, the results for the Standard, Fixed and Full concept are presented using the Wind electricity supply profile. Subsequently, the results are compared to other power supply modes, and finally the sensitivity analysis is presented.

#### 5.1. Standard concept

Fig. 3 shows the SNG production costs as a function of the two optimization parameters. The figure presents the parameter pairs which were found to be, at most, 5 €/MWh more expensive than the lowest cost case. A broad interval was observed for the variation of feasible hydrogen storage and methanation capacities. The yellow area highlights all parameter pairs that allow for production costs between 128.1 and 128.9 €/MWh, ranging from approximately 3.0–4.3 MW for methanation capacity, and from 370 to 1180 kg for hydrogen storage.

Fig. 4 illustrates how the costs of the system are distributed between the different components. With the assumptions used in this work, the majority of the cost results from the capital expenditures of the electrolyzer, composing over 78% of the total costs. The operational expenditure (OPEX) is calculated without electricity costs.

The annual quantity of SNG produced depends on the storage and methanation capacity. Fig. 5 shows increasing gas production with increasing methanation and hydrogen storage capacities. With increasing storage capacity, less hydrogen is discarded, because the hydrogen storage reaches the maximum pressure less frequently.

### Table 6

The terms in bold refer to the shortened concept titles used in this paper.

| Concept               | Description                                                                 |
|-----------------------|------------------------------------------------------------------------------|
| **Standard** plant concept | The electrolysis capacity is fixed at 10 MWELAC and the electrolysis strictly follows the profile of electrical energy. Hydrogen can be discarded when the hydrogen storage reaches maximum pressure and the electrolysis produces more hydrogen than is consumed by the methanation unit. Optimal values are found for the methanation capacity and H₂ storage size. |
| Fixed methanation capacity | Methanation capacity is fixed at 5.75 MWth,SNG. Optimal value is found for the H₂ storage size. |
| Full use of H₂           | All produced hydrogen must be converted into SNG, i.e. the hydrogen must be temporarily stored and subsequently fed to the methanation plant in full. Hydrogen storage and methanation must be designed accordingly. Direct flaring of H₂ is not permitted, but SNG may still be flared during reactor start-up process. |
Increasing the hydrogen storage while decreasing the methanation capacity may lead to the same gas production cost. However, from an operational perspective, there might be drastic differences between different configurations. The plants differ in the number of shutdowns and operation principles. Fig. 6 shows the number of shutdowns the methanation reactor experienced in a year using the different configurations. A smaller H₂ storage size and larger methanation capacity clearly increased the number of shutdowns of methanation, and led to an intermittent production of CH₄.

Standard concept with fixed methanation capacity

In the Fixed variant of the Standard scenario, the methanation has a fixed capacity of 5.75 MWₘₐₜ₃, and the hydrogen storage is the only optimized parameter. The result depicted in Fig. 7 shows a minimum of 136 €/MWh found at 800 kg storage capacity. For a range of ± 45% oversize and undersize H₂ storage, less than a 1% increase of SNG production cost was observed.

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In the Fixed variant of the Standard scenario, the methanation has a fixed capacity of 5.75 MWₘₐₜ₃, and the hydrogen storage is the only optimized parameter. The result depicted in Fig. 7 shows a minimum of 136 €/MWh found at 800 kg storage capacity. For a range of ± 45% oversize and undersize H₂ storage, less than a 1% increase of SNG production cost was observed. With very small storages, the penalty incurred from frequent restarts significantly hinders production, as a larger portion of H₂ is lost in flushing the reactor. The lowest production cost identified in the Fixed configuration was approximately 6% higher than in the Standard case, whereas SNG production increased by approximately 5%. However, a portion of the increased yield was also lost in the restart cycles, resulting in a net yield increase of nearly 4%. Thus, the increase in SNG yield could not compensate for the rising investment cost of the PtG system as compared to the Standard case.

5.2. Full concept

The Full concept can be considered a restricted version of the standard configuration, as it enforces the additional constraint of mandatory complete use of H₂ for SNG production. In practice, this was achieved by implementing larger H₂ storage and methanation capacities. The optimum for the Full conversion concept was found at 5% larger SNG yield, but also a 6% higher production cost, owing to the larger methanation capacity and storage size. It was preferable in this case to radically increase the methanation capacity (+55% compared to standard), as opposed to increasing the storage size to very high levels. However, the storage size still increased moderately (by 20%) as compared to Standard. A larger methanation reactor allows the PtG system to more flexibly adjust to intermittent renewable electricity input.

The SNG production costs for various configurations in the Full concept are shown in Fig. 8. Alternative optimal configurations (having approximately the same production cost) can be found by simultaneously scaling the methanation up and the hydrogen storage down, or vice versa.

Even though hydrogen is typically considered a valuable feedstock, the results indicate that it can be acceptable and even favorable not to
blindly use all potentially available hydrogen if it is produced by a direct link to a variable electricity source. Design of the plant should focus on decent operation under normal conditions, whereas in rare situations suboptimal performance can be accepted. Such situations occur, for instance, when the methanation reactor is running at full capacity and the H2 storage is full, additional wind energy would still be available for producing H2. The extra investment required to adapt to such situations would exceed the potential gains from increased production.

5.3. Comparison of costs and viable configurations

Fig. 9 shows the calculated levelized SNG production costs for all three electricity supply scenarios as well as the three different operational concepts. The results are based on the most cost-efficient result for each simulation. The production costs do not include the cost of electricity. Instead, the effect of including electricity costs is shown separately, in Fig. 13. The lowest GPC were achieved for all power sources with the Standard case, where no constraint was applied concerning the percentage of H2 to be converted. Therefore, storage size and methanation capacity could be designed smaller. Owing to the lower investment required to reduce the hydrogen storage capacity, the methanation capacity was much closer to the theoretical minimum bound. This is attributed to the high dynamic fluctuation of the Wind power supply (Table 2): as the input profile is more chaotic, a larger methanation capacity is favorable so that it can more efficiently process sudden spikes in production (see Fig. 11).

Fig. 12 shows how the optimal storage and methanation capacities are distributed in the scenarios. The methanation capacity was quite small for the PV electricity supply, but it also had the largest storage to cope with the regular long time intervals that lack the PV power supply. The day and night cycle makes storages more cost-efficient for a PV-powered PtG system than one without H2 storage.

The methanation capacity was almost doubled between the Standard and the Full concepts for the Wind power supply. The capacities were also larger for the other power supply modes, but less drastic in difference. Again this difference can be attributed to the characteristics of the wind generation profile. There were a handful of high season periods, where the electrolyzer ran at a high load level for multiple hours, quickly filling the H2 storage up to its maximum level.

Fig. 8. Full concept results using direct connection of PtG with a 10 MW wind power source. Configurations inside the gray colored region would not convert all potential H2 and thus are not valid configurations for the full concept.

Fig. 9. SNG Production cost based on the best individual result for each scenario and plant configuration.

Fig. 10. H2 losses for the optimal case of each scenario and power source.
To prevent flaring of $\text{H}_2$, the methanation capacity was increased, as an investment in a larger methanation capacity was more cost effective than adjusting the hydrogen storage size. The optimal storage capacity for the Fixed variant scenarios is shown in the Appendix A.

Previously, GPC were calculated without electricity costs. In Fig. 13, the GPC for the Standard and Full cases are correlated to those of the Fixed scenario. Optimization of the methanation capacity reduces costs, especially when electricity prices are low.

When electricity costs are not accounted for, the GPC were approximately 6% (Wind) and 17% (PV) lower with an optimal system, as compared to the corresponding Fixed case. When the price of electricity was set at 50 €/MWh, savings of 2% (Wind) and 9% (PV) were still achieved.

As the price of electricity increases, the relative cost reduction decreases. One explanation for this can be derived from the quantity of hydrogen which remains unused in the scenarios. For instance, the cost difference between Standard and Fixed using Wind power diminishes, as the total costs of the system are allocated to a lower quantity of the product in the Standard case (or equivalently, a higher quantity of lost hydrogen, as in Fig. 10). Another explanation is a declining price difference in the PV Fixed case, where fewer hydrogen losses are occurring.

5.3.1. Duration of operation cycle

To minimize hydrogen discard during start-up and shutdown and to avoid costly standby losses of the methanation subsystem, the methanation should have the highest possible number of continuous operating hours. The size of the hydrogen storage and the methanation capacity are crucial parameters in this regard. As the Standard case achieved the lowest production cost regardless of the power supply mode, we focus on the Standard case in this section.

The Wind power profile most often led to short periods of hydrogen production, with an annual median of only approximately 0.5 h. However, very long individual cycles of 20–200 h were also observed. When coupled with a hydrogen buffer storage, the methanation reactor would typically operate in longer cycles, with an annual median of over 6 h. Interestingly, the methanation had a fewer number of very long operation cycles, and was most likely affected by the restriction of maintaining a minimum of 40% load with the reactor when in operation. No such restriction applied for the electrolyzer, and this aspect can slightly distort the statistics. A monthly summary of the operation cycles is shown in Fig. 14. As typical for wind energy, the longest production peaks occurred during the winter months. The total annual number of shutdowns was 297 for the methanation reactor, and 1559 for electrolyzer.

With PV as the source of electricity, there is a clear seasonal variation in cycle lengths, as seen in Fig. 15. The methanation reactor is clearly decoupled from the electrolyzer, as the methanation cycles are typically 2–10 (or more) times longer than the corresponding electrolyzer cycle. The annual medians are slightly above 12 and 20 h for electrolysis and methanation, respectively. The annual number of shutdowns was 365 for electrolysis, and 122 for methanation. The strong increase in the methanation cycle length can be attributed to the regular daily pattern of PV production, which is better suited for short term storage than the long and rare production spikes with a wind.

![Fig. 11. Boxes represent the optimal capacity adhering to scenario-specific limitations, whereas the circular markers are the minimum and maximum limits, as defined in Section 3.4.](image)

![Fig. 12. Optimal configurations for the standard and full concepts. Only a limited region of the optimal is presented to avoid overlap.](image)

![Fig. 13. Relative cost reduction of the cases Standard and Full in comparison to Fixed with the three investigated power supply modes while including electricity costs.](image)
The amount of hydrogen discarded also reduced (−168%), thus reducing the number of shutdown processes (−21%). Consequently, GPC were reduced by only 3%. Thus, the system was to some extent capable of maintaining a constant production cost by internally adapting to the external triggers.

Increasing the restart time of the system (Fig. 18a) resulted in a modest change in SNG cost (+2.4%), which can be explained by the system adjusting its hydrogen storage to be larger (+84%) to prevent shutdowns. The effect is strengthened by a parallel decrease in the methanation capacity (−11%).

The changes in restart level (Fig. 18b) were offset by methanation and storage capacities, so that there was no clear change in GPC.

When a standby cost was linked to the idle time of the methanation (Fig. 18c), the system compensated by using a smaller methanation reactor (−10% at 20€/h), which decreased the number of idle hours. To offset the reduced capability of the plant to produce SNG during a high season, the hydrogen storage capacity was slightly increased (+27%). Regardless, the SNG production decreased on an annual level (−16%).

6. Conclusion/Outlook

In this work, a hydrogen storage size and methanation reactor capacity were economically optimized for a 10 MWel,AC electrolyzer. The synthetic natural gas production costs were calculated for three power sources (Wind, PV and Control). In addition, two separate system schemes were distinguished, i.e., Standard and Full concepts.

The supply profile of electricity affects the optimal capacity. The methanation capacity for PV-coupling is very small (1.72 MW_{el,SNG}) and the storage is rather large (1250 t), which works well in combination with the daily cycle of the PV. Wind power exhibits significantly more chaotic periods of production, so a larger methanation reactor (3.68 MW_{el,SNG}) with a smaller hydrogen storage tank (635 t) was favored. If complete utilization of the hydrogen is necessary, the methanation reactor (5.70 MW_{el,SNG}) and the hydrogen storage (770 t) would be even larger, resulting in a 5% increase in unit price. Higher capital expenditures were thus dominant over the increased product yield in this case. This result may not always hold true, because it is susceptible to changes in input parameters.
The analysis shows that synthetic natural gas production costs may be reduced in some situations by as much as 17% if the hydrogen storage size and the methanation capacity are optimized. One critical factor in this is the methanation capacity, which can be designed to be smaller than the electrolyzer output. The cost reduction benefit in this case arises mainly from the reduction in capital expenditures. For instance, a 5.75 MW\textsubscript{H2,SNG} methanation reactor (which roughly corresponds to the peak power of the electrolyzer) was reduced to 3.68 MW\textsubscript{H2,SNG} in the least cost case using wind power, reducing capital costs by 36%. The disadvantage of a smaller methanation plant is the potential need for a larger intermediate hydrogen storage tank. Cost optimization has shown that a tight design of the storage and methanation leads to lower gas production cost. The drawback of this optimization is the potential inability to utilize production spikes (excess electricity), which translated into a 5% net loss in the final product yield in the same case. The yield loss would otherwise be even higher, but it is compensated for by the more efficient use of hydrogen storage and the reduced hydrogen losses owing to fewer shutdowns cycles (~22%). Radically different downtimes and operational strategies are to be expected with different electricity supply modes. PV has predictable downtimes with short cycles and thus benefits from a hydrogen storage, whereas wind is more chaotic in nature and operates in longer cycles which impairs the use of hydrogen buffers. With secondary control reserve, there are hardly any downtimes, but the average price for electricity would probably also be higher.

The optimization of the H\textsubscript{2} storage and the subsequent methanation show high potentials for the reduction of gas production cost. The results support the claim that interim H\textsubscript{2} storages are best suited primarily for short-term storage, owing to their high specific cost of 490 €/kg. For instance, it was found to be economically beneficial to purposely avoid using all potentially-available hydrogen, as it would have required significant investments (either in methanation capacity or storage sizes).

As the production of H\textsubscript{2} is linked to the availability of electricity, the chosen electricity supply profile critically affects the importance of H\textsubscript{2} storage. In the case of direct coupling of the electrolysis to a PV field, the hydrogen storage should be designed so it can compensate for the day/night inequality. When the Power-to-Gas plant is directly coupled to a wind farm, the H\textsubscript{2} storage can be designed to be smaller, as the fluctuations are distributed over the entire day. If a balancing service such as symmetric secondary control reserve is offered, the H\textsubscript{2} storage may be dimensioned very small because the storage only has to compensate for short-term load peaks or load undercoverages.

It was shown that the use of an interim H\textsubscript{2} storage increases the flexibility of the electrolyzer and makes long and constant operation phases of the methanation reactor possible. When designing new plants, it is recommended to analyze the electricity supply characteristics on a case-by-case basis and preferably over a longer period of time, to

**Fig. 16.** Sensitivity analysis for the capital expenditure of electrolyzer and methanation, using the wind standard scenario. Default parameter values are highlighted with an asterisk.

**Fig. 17.** Sensitivity analysis for the H\textsubscript{2} storage cost (€/kg\textsubscript{H2}) in the wind standard scenario. Default values highlighted with an asterisk.
Fig. 18. Variation in SNG production cost for three parameters. Default parameter value is shown with an asterisk.

Fig. 19. Optimal storage size defined for the fixed variant of standard with all studied power sources for PtG operation. The optimal found storage sizes are 211 kgH₂ (Control), 500 kgH₂ (PV) and 790 kgH₂ (Wind). Red dots: The circular data markers highlighted in red colour mark the position of the cost optimum for each case.

Fig. 20. Gas composition and flow during start-up, shut-down and load change with the tube bundle reactor of the 250 kWel P2G® plant of the ZSW as a function of educt gas flow (T = 200–600 °C, p = 7 bara, SV = 1365 l_hydro/catalyst h) [40].
consider annual fluctuations and to obtain a more accurate input data for the optimization of the gas production cost. The production can have a high season for some time of the year, which should be acknowledged in plant design. Plant design should also consider the type of renewable energy to be used.

The smaller the number of full-load hours of the electrolyzer gets, the greater the influence of the reduction in investment costs becomes. The optimization of Power-to-Gas plants according to the expected electricity supply is crucial. Cost savings can be expected with low electricity prices. As the ratio of hydrogen production cost to total system cost decreases, so does the potential opportunity cost of unused seasonal resources. This work shows that there is no single optimal plant configuration, but a range in which capacities can be varied without significantly affecting the synthetic natural gas production cost. The robust results allow project developers and decision makers to choose plant configurations within certain ranges that lead to equivalent marginal costs. The optimal configuration should be designed based on typical conditions, and not the extremes.

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Appendix A

See Figs. 19 and 20.

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