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Modelling synthetic methane production for decarbonising public transport buses: A techno-economic assessment of an integrated power-to-gas concept for urban biogas plants

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ABSTRACT

The integration of power-to-gas (PtG) technology into existing urban anaerobic digestion (AD) plants could be an interesting concept to recycle biogenic CO_2 and increase CH_4 production as renewable fuel to further decarbonize public transport buses (PTB). However, such implementation is challenging for several reasons, including power restrictions during peak load, physical and temporal availability of CO_2 from AD plants, and the need for expensive intermediate gas storages to avoid mismatch between the constrained synthetic CH_4 production and the variable fuel demand. To investigate whether synthetic CH_4 could be a feasible alternative for buses currently powered by fossil fuels, a dynamic model was built for discrete-event simulations of PtG technology integrated into an urban AD plant designed to supply biomethane as fuel for bus fleets. Different scenarios were assessed, including variations in power availability to run a proton exchange membrane electrolyser as well as variations in the production scale of synthetic CH_4 based on *ex-situ* biological methanation. The results show that a constrained power utilization (maximum of 12 h per day) increased the production cost of synthetic CH_4 by 20%. In contrast, an increase in PtG production capacity from 0.75 MW_{th} to 2.25 MW_{th} decreased costs by 16%. From the PTB operators' perspective, the total cost of ownership (TCO) increased in all analysed scenarios when replacing diesel buses by gas buses powered by synthetic CH_4. However, when using synthetic CH_4 as drop-in fuel to replace natural gas in existing gas bus fleets, the TCO could be reduced up to 4.4% depending on the PtG plant configuration and the assumed fossil fuel price. Furthermore, our results show that a carbon tax on fossil fuels has only a limited effect on promoting synthetic CH_4 as alternative fuel for PTB, and additional incentives should be put in place to prioritize a fuel switch, especially for existing gas bus fleets.

1. Introduction

The conversion of electricity into hydrogen (H_2) and/or methane (CH_4) has been proposed as a measure for long-term energy storage, as fuel for mobility and agriculture, as coal substitute in steelmaking, as a building block for sustainable chemicals production, among others [1–4]. This so-called power-to-gas (PtG) concept has been mooted as a key element to tackle climate change by decarbonising sectors where direct electrification is technically unfeasible or economically less competitive [5,6]. For public transportation, especially buses running on longer distances, H_2 could become useful as a range extender in hybrid electric buses (battery + fuel cell) or when synthesized with carbon dioxide (CO_2) to produce renewable CH_4, in particular for already existing gas bus fleets (drop-in fuel).

For synthetic CH_4 production, the CO_2 source is an important aspect to consider as the energy required for capturing and conditioning the CO_2 can be significant for sources with low CO_2 concentration, thus directly affecting the levelised cost of CO_2 capture [7]. For this reason, the integration of PtG technology into anaerobic digestion (AD) plants has been previously proposed, because the process of upgrading biogas to biomethane potentially yields a concentrated CO_2 side-stream that is suitable for biological methanation (BM) with green H_2 produced from water electrolysis ($4H_2 + CO_2 \rightarrow CH_4 + 2H_2O$; $\Delta H = -165.1$ kJ/mol) [8,9].

In cities where municipal solid waste and sewage sludge are treated

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Nomenclature

List of abbreviations

AD	anaerobic digestion
BM	biological methanation
BoP	balance of the plant
CAC	carbon abatement cost
CAPEX	capital expenditures
CCU	carbon capture and utilisation
CH_4	methane
CNG	compressed natural gas
CO_2	carbon dioxide
EEG	renewable energy act (*Erneuerbare-Energien-Gesetz*)
EAC	equivalent annual cost
EU ETS	European Union Emissions Trading Scheme
FLH	full load hours
GPC	gas production cost
H_2	hydrogen
HHV	higher heating value
OPEX	operational expenditures
PEM	proton exchange membrane
PTB	public transport buses
PtG	power-to-gas
PtX	power-to-X
RED II	renewable energy directive II
RFNBO	renewable gas of non-biological origin
STP	standard temperature and pressure
TCO	total cost of ownership

List of symbols and units

BM_{FLH}	annual FLH of BM (h)
BM_i	BM operation mode in each hour i
$CAPEX_{PtG}$	capital expenditures for the PtG plant over the lifespan of the project (€)
$CAPEX_{bus}$	capital expenditure for gas and diesel buses over their lifespan (€)
C_{cold}	yearly costs to keep the electrolyser on cold standby (€)
$C_{el,i}$	costs associated with electricity use in each hour i (€)
$C_{comp,i}$	costs associated with gas compression for each hour i (€)
C_{ff}	fuel consumption of the different buses running on fossil fuels (L/year or kg/year)
C_{total}	total yearly electricity costs for the PtG plant (€)
D	average distance travel by the buses (km/year)
E_i	electrolyser operation mode in each hour i for the next 24 h
E_{FLH}	annual FLH of the electrolyser (h)
$E_{FLH,24h}$	electrolyser FLH in the day-ahead scheme (h)
EF_{ff}	emission factor of the different fossil fuels (kg CO_2/L or kg CO_2/kg)
$Fuel_y$	fuel expenses of buses powered by gas (CNG or synthetic CH_4) and diesel consumption in each year y (€/km)
k	discount rate of the PtG plant (%)
$m_{CH_4,i}$	synthetic CH_4 production for each hour i
m_{CH_4,i_s}	CH_4 storage level at the day-ahead bid time (kg)
$m_{CH_4,max}$	maximum CH_4 production capacity (kg/h)
$m_{CH_4,storage}$	CH_4 storage capacity (kg)
$m_{CO_2,i}$	CO_2 production from the biogas upgrading unit in each hour (kg)
$m_{H_2,i}$	H_2 production in each hour i (kg)
$m_{H_2,max}$	maximum H_2 production rate at full load (kg/h)
Ma_y	maintenance costs for gas and diesel buses in each year y (€/km)
$OPEX_y$	fixed operational expenditures in each year y (€/year)
ρ_{H2}	H_2 density at STP (kg/m^3)
P_N	electrolyser's nominal rated power (MW)
$Power_y$	variable operational expenditures due to electricity used for synthetic CH_4 production in each year y (€/MWh)
SMP_y	synthetic CH_4 production and delivery in each year y (kg CH_4/year)
TCO_{CH4}	total cost of ownership of synthetic CH_4 buses based on different PtG plant scenarios (€/km)
TCO_{ff}	total cost of ownership of buses running on different fossil fuels (€/km)
T_{grid}	fixed tariff for grid-based power (€/MWh)
$T_{spot,i}$	day-ahead spot market price in each hour i of the electrolyser operation (€/MWh)
W_{cold}	power consumption during electrolyser cold standby (kW)
W_{comp,CO_2}	electricity consumption for CO_2 compression (kWh/kg)
W_{comp,CH_4}	electricity consumption for CH_4 compression (kWh/kg)
W_{el}	hourly power consumption of the electrolyser on full load (MWh)
W_{H_2}	specific power consumption during electrolyser operation mode (kWh/m^3 H_2 at STP)
W_{safe}	power consumption for safety infrastructure (kW)

in AD plants and the produced biogas is used as vehicle fuel, the integrated production of synthetic CH_4 could play a complementary role in decarbonising public transport as well as in improving urban air quality thanks to lower emissions of pollutants like particulate matter compared to liquid fuels (e.g. diesel) [6]. However, the integration of PtG technology into urban AD plants is challenging for different reasons: (i) the electricity required for the process might not be available on demand (e.g., due to constraints in the power grid created by residential peak loads); (ii) the physical and temporal availability of CO_2 from an adjacent AD plant is not constant which in turn represents a limiting factor for the synthetic CH_4 production [10], and (iii) the operation of the PtG plant is likely to be driven by a variable vehicle fuel demand which require costly intermediate gas storages to avoid a mismatch between the constrained production and the fuel demand.

Different studies describing potential applications for PtG technology in combination with AD plants have been recently reported. For instance, small-scale implementations were investigated for different geographical locations including the possibility of cost reduction when oxygen from water electrolysis is recovered and power curtailment from variable renewable energy is minimised [11]. Combined with biogas plants, techno-economic assessments of *in-situ* and *ex-situ* BM have been performed based on different plant configurations and resource-based scenarios [12,13]. Also, the economic performance of BM with CO_2 derived from an amine scrubbing system was investigated in comparison with the option of direct methanation of raw biogas in an *ex-situ* bioreactor [14]. Furthermore, studies on the optimisation of plant components like H_2 storage have been conducted to cope with constraints related to catalytic methanation depending on the availability of solar, wind, and power obtained from spot markets [15,16].

Even though some of these existing studies in the literature performed comprehensive modelling of the PtG technology, this is in most cases related to grid injection of the produced synthetic CH_4. However, to the best of the authors' knowledge, no previous investigation has addressed the production of synthetic CH_4 according to the specific demand from public transport buses (PTB), in particular considering hourly-based refuelling times. A business model for CH_4 production specifically targeted to demand profiles of PTB fleets differ greatly from a more standard business model of CH_4 production for grid injection as the former require specifically designed plant configurations in terms of electrolyser/methanation capacities, intermediate gas storage as well as operating schedules which would directly influence the competitiveness of synthetic CH_4 in comparison to conventional fossil fuels [17].

Furthermore, previous studies do not explore fluctuations in the availability of CO_2 from AD plants. In fact, CO_2 availability is subject to seasonal fluctuations in the feedstock used for biogas production, dynamic operation of biogas upgrading units as well as their downtime for maintenance. Therefore, the use of real data from AD plants is of utmost importance to understand the impacts of CO_2 availability on the design of PtG plants as well as production scale which directly affects the cost of the produced synthetic CH_4.

In brief, the present study adds to the existing body of literature by assessing the operation of PtG plants dedicated to the refuelling of gas bus fleets on an on-demand hourly-basis, and by incorporating into the assessment existing constraints found in urban environments for the implementation of PtG technology, such as power supply during peak loads and limited CO_2 availability for a variable fuel demand.

For this purpose, a dynamic model was developed for discrete-event simulations of different power availability and synthetic CH_4 demand scenarios with the following objectives:

- To optimise the PtG plant configuration for each scenario analysed in terms of gas production cost (GPC) based on Monte Carlo simulations;
- To evaluate the influence of power availability on the dynamic operation of the PtG plant, including energy losses due to standby time;
- To investigate the competitiveness of synthetic CH_4 use in PTB fleets against fossil fuel use by comparing the total cost of ownership (TCO);
- To assess the carbon abatement costs (CAC) of substituting fossil fuel by synthetic CH_4 as renewable alternative in PTB.

2. Methodology

2.1. System description

In this study, the PtG plant system refers to a synthetic CH_4 production facility in which synthetic CH_4 is delivered on-demand for PTB operators with a fleet of intercity or regional buses that cover medium to large distances and are therefore less promising for direct electrification. Different types of buses powered by a mix of fuels like diesel, natural gas and biomethane are used in the absence of the PtG plant. To take advantage of the existing infrastructure, the PtG plant is integrated into an AD system used to treat sewage sludge and source-sorted organic fraction of municipal solid waste with a yearly energy output of approximately 35 GWh_{th}. Within the AD plant biogas is upgraded to fuel-grade biomethane resulting in a concentrated CO_2 side-stream (e.g. via amine scrubbing). After being upgraded, biomethane is compressed at 5 bar and injected into a dedicated gas grid that delivers fuel to a depot where PTB are periodically refuelled. In the absence of a PtG plant, biomethane is responsible for partly supplying the fuel demand of the PTB fleet, and CO_2 from biogas is vented to the atmosphere.

For the investigated integration of a PtG plant, H_2 is produced through a proton exchange membrane (PEM) electrolyser and further biologically synthesized with CO_2 derived from the AD system according to Eq. **(1–2)** [9].

$$H_2O \underset{electrolysis}{\rightarrow} H_2 + \frac{1}{2}O_2; \Delta H = +285.8kJ/mol \quad (1)$$

$$4H_2 + CO_2 \underset{biomethanation}{\rightarrow} CH_4 + 2H_2O; \Delta H = -165.1kJ/mol \quad (2)$$

The electricity used during electrolysis, BM, and ancillary equipment like compressors is either obtained from the spot market of the Nord Pool power exchange in a day-ahead trading scheme (planned purchasing for H_2 production and conditioning) or from the regulated market (purchasing smaller volumes during system downtime). Deionized water is considered according to the specifications of the technology assessed. As the BM rector is operated at 10 bar, synthetic CH_4 storage is operated at the same pressure of the existing gas grid dedicated to transport biomethane from the AD plant to the bus depot. This is an important integration aspect to avoid unnecessary electricity consumption during gas compression. For the same reason, CO_2 derived from the biogas upgrading unit of the AD plant is compressed and stored at a maximum pressure of 15 bar. The system does not consider H_2 storage since the electrolyser is operated based on the CH_4 storage level, and due to the fast ramp-up time of BM reactors [18]. Nutrients required for BM are obtained from the digestate of the existing AD plant, thus no additional expenses are incurred. Even though by-product recovery from electrofuels production could represent an additional source of revenue for PtG plants, the recovery of low-temperature waste heat (60 °C) and oxygen from the electrolyser as well as low-temperature waste heat from the thermophilic BM (55 °C) are disregarded in this study [4,19]. Table 1 and Fig. 1 show the overview of the different characteristics of the PEM electrolyser, BM reactor, gas compressor as well as the process flow diagram of the PtG technology integrated into the existing AD plant.

Table 1
Specifications of the PEM electrolyser, BM reactor and gas compressors.

Characteristics		Value	Unit	Source
PEM	Electricity consumption	4.9[a]	kWh/m^3 H_2	[19–21]
	Conversion efficiency at nominal load	72.3[b]	%	Calculated
	Ramp-up time	10	min	[22]
	Operation pressure	50	bar	[22]
BM	Electricity consumption	4[c]	% of total output	Operation experience
	Conversion efficiency	78.2[a,d]	%	Calculated
	Ramp-up time	15[c]	min	Operation experience
	Operation pressure	10[c]	bar	Operation experience
Compressor	Electricity consumption for CO_2 compression[c]	0.185	kWh/kg CO_2	Own experience
	Electricity consumption for CH_4 compression[e]	0.34	kWh/kg CH_4	[23]

Note:
[a] Based on higher heating value (HHV) and standard temperature and pressure (STP) values (0 °C and 101.325 kPa).
[b] Based on the HHV of 39.4 kWh/kg H_2 ÷ (4.9 kWh/m^3 H_2 ÷ 0.08988 kg/m^3) = 72.3%.
[c] Electricity consumption (average value), ramp-up time (maximum value) and operation pressure (maximum value) of the BM reactor as well as the electricity consumption for CO_2 compression are derived from own operation experience of the PtG demonstration site at Solothurn, Switzerland. Compression of CO_2 from 1 bar(a) to 13.5 bar(a) is based on a single-state water-cooled compressor.
[d] 1 kg H_2 = 2 kg CH_4 (2*15.4 kWh/kg CH_4 ÷ 39.4 kWh/kg H_2) = 78.2%.
[e] CH_4 compression is required for refill the gas buses at 200 bar.

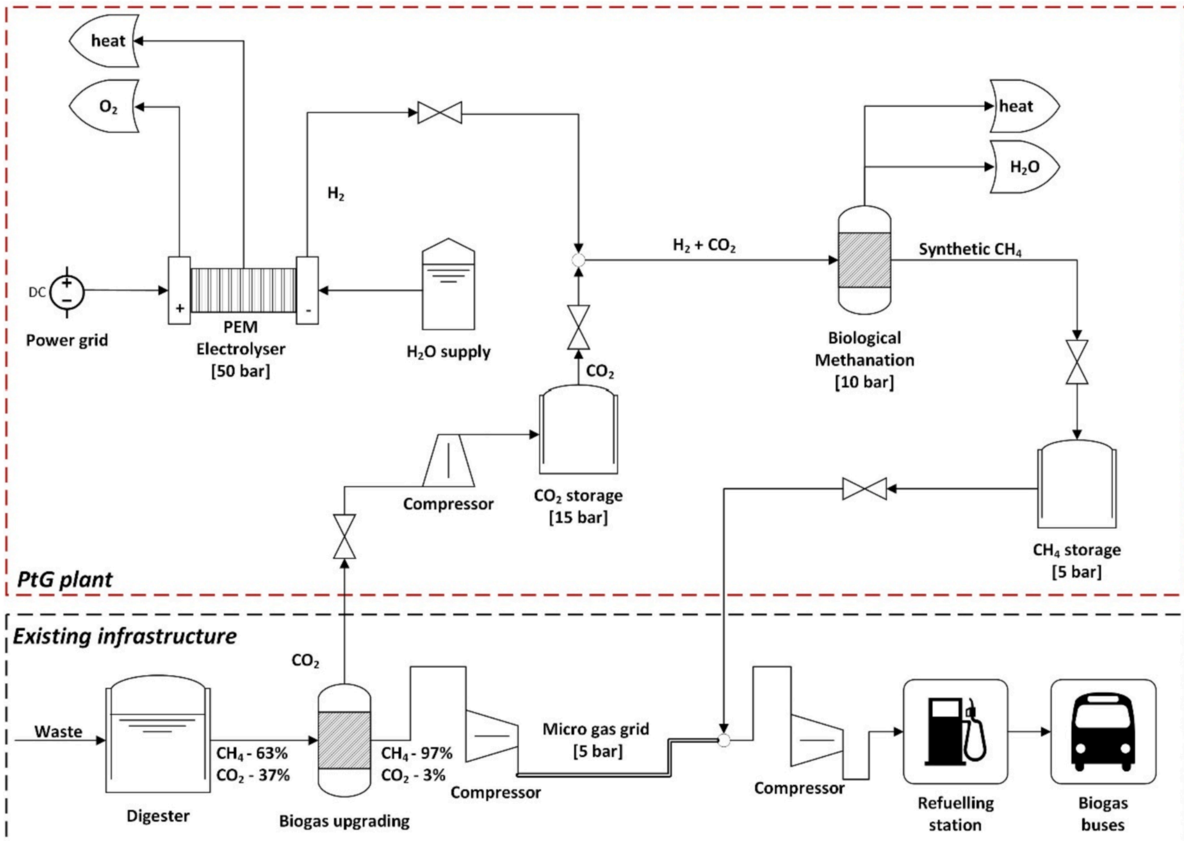

Fig. 1. Process flow diagram of the PtG plant integrated into the existing AD plant infrastructure.

2.2. Dynamics of the PtG plant operation

The operation of the PtG plant is based on hourly time-step ($t = 1\ h$) in which all individual components like electrolyser, methanation unit and compressors are run simultaneously. As the PtG plant aims to supply on-demand fuel for mobility, the demand for synthetic CH_4 from PTB dictates whether the electrolyser and methanation reactor should operate or not. Therefore, every day at the bidding time of the day-ahead market (i.e. 12:00 noon), the number of electrolyser full load hours (FLH) needed to fill up the CH_4 storage is calculated and used for placing a price-independent order in the power auction. This purchasing strategy is only possible if the decision is assisted by a price forecasting method which our group has previously described in detail [17]. Thus, whenever the calculated FLH are higher than zero and lower than 24, the electrolyser is scheduled to operate in the cheapest forecasted hours. However, when the calculated FLH are higher or equal to 24, the electrolyser is scheduled to operate on full load the entire day no matter the forecasted price. In case of the calculated FLH being equal to zero, the electrolyser is put on cold standby. The ramp-up time of the PEM electrolysers from cold standby is estimated at approximately 10 min [22]. During this time, power is consumed on full load by the electrolyser but no H_2 is produced. Due to the ramp-up time, approximately 16.6% less H_2 is produced than expected during steady operation within one time-step of 1 h.

For the methanation reactor to be able to produce highly concentrated synthetic CH_4, an adequate mixture of H_2 and CO_2 should be respected [24]. Based on the methanation stoichiometric reaction **(Eq. (2))**, for each kg of H_2 to be methanised, 5.5 kg of CO_2 is required. However, for BM processes additional CO_2 is needed since carbon is a major element in microbial cell composition. For this reason, 6% of CO_2 was assumed to be needed for microbial growth, resulting in 5.83 kg of CO_2 per kg of H_2 methanised [25]. In case this criterion is not met in any time-step of the PtG plant operation, the reactor is put on standby, and H_2 and CO_2 are not utilised. After each standby event, a penalty time of 15 min is applied for the BM reactor ramp-up during which the output gas is flared due to low CH_4 concentration. For this reason, 25% less synthetic CH_4 is produced than expected during steady operation within one time-step of 1 h. In contrast, synthetic CH_4 is produced at part or full load of the BM reactor depending on the methanation capacity and the amount of CH_4 required to fill up the storage. Finally, synthetic CH_4 is delivered for PTB operation when the CH_4 storage level is equal to or higher than the fuel demand. In case synthetic CH_4 is not delivered on demand, the CH_4 storage displays negative values which are used to identify plant configurations that don't meet the basic criteria of on demand production. Fig. 2 shows a flow chart of the PtG plant simulation.

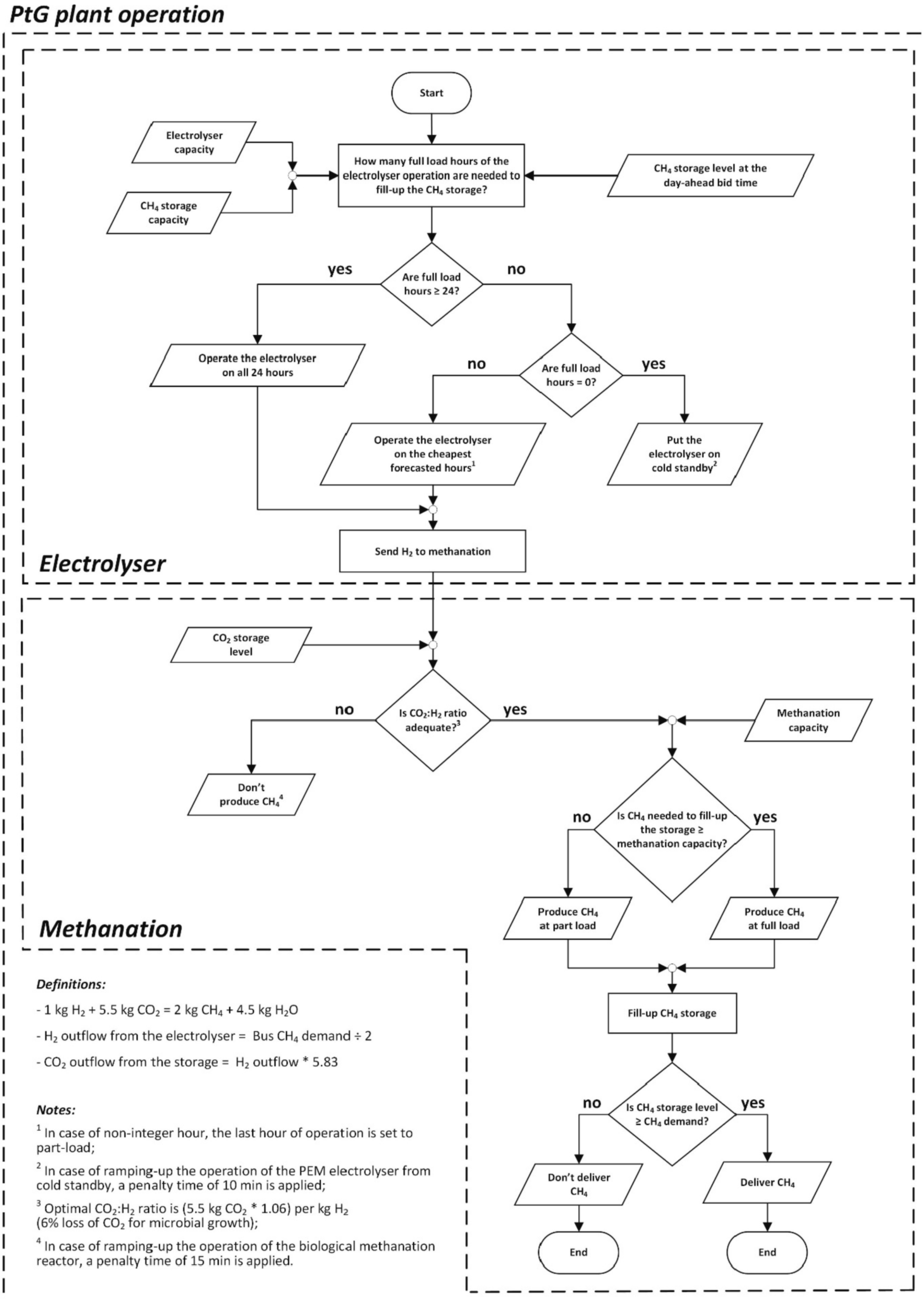

Fig. 2. Flow chart to simulate the PtG plant operation.

2.3. Electricity prices, CO_2 availability, and synthetic CH_4 demand

Hourly values from the day-ahead spot market of the Nord Pool power exchange for the region SE3 in 2018 were used to calculate the electricity costs to run H_2 production in the PtG plant [26]. Even though electricity prices can vary significantly across different years, the year 2018 was chosen since the average electricity price found for this year is relatively high, which results in a more conservative assessment for the production costs of electrofuels. In addition, as the mentioned study described higher forecasting errors for the same year, this allows to test the robustness of the current model in terms of misleading operations due to a mismatch between real and forecasted prices.

Biogas production from an existing AD plant treating industrial and municipal solid waste was obtained and used to estimate the CO_2 availability for synthetic CH_4 production. In this case, the hourly mass of biogas (63% CH_4 and 37% CO_2; v/v) upgraded to biomethane (97% CH_4 and 3% CO_2; v/v) was used to calculate the hourly mass of concentrated CO_2 suitable for BM. This approach allows simulating the PtG plant under real-world constraints such as seasonal fluctuations in substrate input for AD, dynamic operation of the biogas upgrading unit as well as its downtime for maintenance.

The synthetic CH_4 demand for PTB was modelled according to the hourly biomethane demand of an existing depot in Sweden dedicated to over 100 tri-axle intercity/regional buses powered by different fuels. This assumption is based on the fact that both biomethane and synthetic CH_4 buses use the same powertrain technology resulting in similar refuelling times and demand profiles. The hourly distribution of the day-ahead electricity price, CO_2 availability, and synthetic CH_4 demand throughout the simulated year are shown in Fig. 3.

2.4. Economic assessment

The economic performance of synthetic CH_4 production and use as fuel in PTB was assessed based on three economic indicators, namely the GPC of synthetic CH_4 (€/MWh), the TCO (€/km), and the CAC (€/tCO_2). While the GPC is used to optimize the design of the PtG plant in terms of electrolyser and methanation capacities as well as CO_2 and CH_4 storage sizes, the TCO is used to assess the feasibility of substituting conventional diesel and compressed natural gas (CNG) by synthetic CH_4 from the bus operators' perspective. The climate mitigation cost of displacing fossil fuel by renewable gas is assessed by calculating the CAC for different PtG plant configurations and fossil fuel price scenarios.

To determine the GPC, all production costs are considered over the lifespan of the PtG plant divided by the total synthetic CH_4 production output as follows:

$$GPC = \frac{\frac{CAPEX_{PtG} + \sum_{y=0}^{n} OPEX_y + Power_y}{(1+k)^y}}{\sum_{y=0}^{n} \frac{SMP_y}{(1+k)^y}} \tag{3}$$

Where:

- $CAPEX_{PtG}$ - capital expenditures for the PtG plant over the lifespan of the project ($n = 20$ years), including replacements (€);
- $OPEX_y$ - fixed operational expenditures in each year y (€);
- $Power_y$ - variable operational expenditures due to electricity used for synthetic CH_4 production in each year y (€);
- k - discount rate estimated at 6.5% per year based on onshore wind projects in the Nordic countries [27];

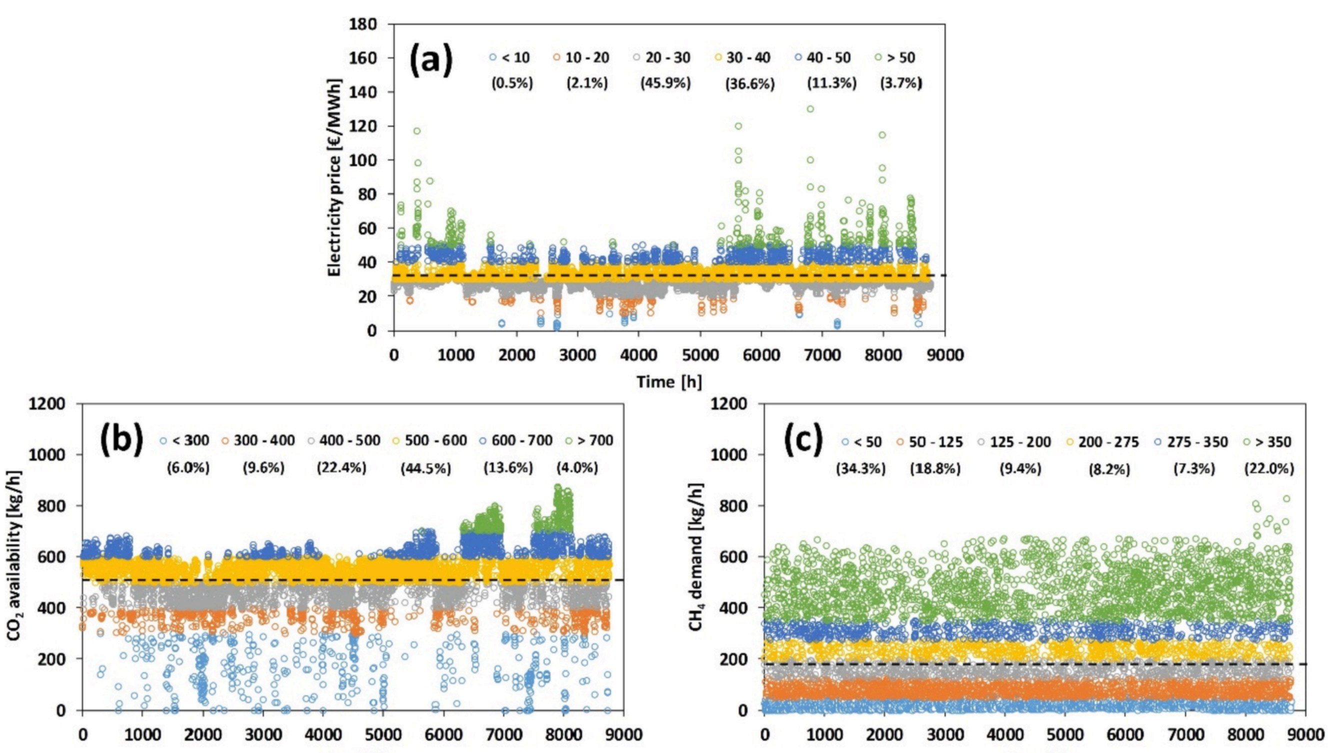

Fig. 3. Hourly distribution of the main input parameters used for modelling the PtG plant. (a) Day-ahead electricity prices; (b) CO_2 availability from the biogas plant; and (c) maximum allowed synthetic CH_4 production based on CO_2 availability.
Note: The dotted lines represent the yearly average values.

Table 2
Summary of the different operation scenarios.

Power availability scenarios	Fuel demand scenarios	Maximum daily FLH [h/d]	Synthetic CH_4 demand[a] [MW_{th}]	Scenario reference
Constrained	Low	12 h	0.75	Co_{low}
	Mid	12 h	1.50	Co_{mild}
	High	12 h	2.25	Co_{high}
Unconstrained	Low	24 h	0.75	Un_{low}
	Mid	24 h	1.50	Un_{mild}
	High	24 h	2.25	Un_{high}

Note:
[a] Corresponds to the utilisation of 26.4% (low), 52.9% (mid), and 79.3% (high) of total CO_2 available from the AD plant. Synthetic CH_4 demand values are based on HHV.

- SMP_y - synthetic CH_4 production and delivery in each year y (kg CH_4);

The TCO represents the expenses of the bus operator per distance driven according to **Eq. (4)**:

$$TCO = \frac{\frac{CAPEX_{bus}+\sum_{y=0}^{n}(Ma_y \cdot D)+(Fuel_y \cdot D)}{(1+k)^y}}{\sum_{y=0}^{n}\frac{D}{(1+k)^y}} \tag{4}$$

Where:

- $CAPEX_{bus}$ - capital expenditures for gas and diesel buses over their lifespan ($n = 10$ years; in €);
- Ma_y - maintenance costs for gas and diesel buses in each year y (€/km);
- $Fuel_y$ - fuel expenses of buses powered by gas (CNG or synthetic CH_4) and diesel consumption in each year y (€/km);
- D - average distance travel by the buses (km)

Finally, the costs to reduce carbon emissions by displacing fossil fuel by synthetic CH_4 are calculated as follows:

$$CAC = \frac{(TCO_{CH4} - TCO_{ff}) \cdot D}{D \cdot C_{ff} \cdot EF_{ff}} \tag{5}$$

Where:

- TCO_{CH4} - total cost of ownership of synthetic CH_4 buses based on different PtG plant scenarios (€/km)
- TCO_{ff} - total cost of ownership of buses running on different fossil fuels (€/km)
- C_{ff} - fuel consumption of the different buses running on fossil fuels (L/km or kg/km)
- EF_{ff} - emission factor of the different fossil fuels (kg CO_2/L or kg CO_2/kg)

The model considers electricity supply from the grid for the PtG plant. However, in this study, it is assumed that this power is certified as renewable, thus resulting in zero carbon emissions for the synthetic CH_4 production.

The lifespan of the PtG plant includes a 3-year commissioning phase, 20 years of operation (during which the electrolyser is replaced once), and one year for decommissioning. Capital expenditures (CAPEX) and operational expenditures (OPEX) values of the PEM electrolyser and the BM reactor as well as costs related to the different buses assessed in this study are shown in **Appendix 1** (Tables 1A-1B).

2.5. Scenarios

One of the biggest challenges for providing a business case for the integration of PtG technology into existing AD plants is the availability of renewable electricity at low costs. Therefore, two main scenarios were assessed for constrained and unconstrained power availability. Since electricity may not be available for electrolysis at any time in urban environments because of peak loads and grid constraints, a scenario was considered where the electrolyser operation of a PtG plant would be restricted to a maximum number of FLH of 12 h per day. Conversely, another scenario was developed where the electrolyser could be operated without any restrictions.

In addition, to allow for a better understanding of the effects of process upscaling, three different fuel demand scenarios for synthetic CH_4 production were considered, namely low (0.75 MW_{th}), mid (1.5 MW_{th}), and high (2.25 MW_{th}). These scenarios correspond to the utilisation of 26.4%, 52.9%, and 79.3% of total CO_2 available from the existing AD plant for which real data was obtained for this study. It can serve as a reference to a demonstration-scale, mid-scale and full-scale implementation. Table 2 shows a summary of the different operational scenarios analysed, and in **Appendix 2** (Fig. 2A) the scale-effect applied on CAPEX of both the PEM electrolyser and the BM reactor (Fig. 2A).

2.6. PtG model and optimisation procedure

The PtG model was implemented in the Matlab-based Simulink environment version R2019b (MathWorks, USA). Individual equations are discretized for a fixed step size (sampling time) of one hour. It is based on real and forecasted hourly values of day-ahead spot market price, variable CO_2 production from an existing AD plant, and synthetic CH_4 demand from PTB. The PEM electrolyser and the BM reactor were modelled in combination with compressed gas storage systems to assist synthetic CH_4 production and delivery on-demand. The model calculates synthetic CH_4 production and delivery, FLH of both the PEM electrolyser and the BM reactor, and total electricity costs (incl. gas compression, cold standby of PEM electrolyser, and safety infrastructure).

The decision of whether the electrolyser should operate or not is dependent on the CH_4 storage capacity, the CH_4 storage level at the day-ahead bid time, the electrolyser capacity as well as the number of FLH for the electrolyser allowed per day according to the chosen power availability scenario. To schedule the electrolyser's operation based on the day-ahead market, the number of FLH required for the electrolyser to produce enough H_2 for the PtG plant to fill-up its CH_4 storage is calculated once per day **(Eq. (6))**:

$$E_{FLH,24h} = \frac{m_{CH_4,storage} - m_{CH_4,i\_s}}{2 \cdot \dot{m}_{H_2,max}} \tag{6}$$

where:

- $E_{FLH,24h}$ - Electrolyser FLH in the day-ahead scheme (h)
- $m_{CH_4,storage}$- CH_4 storage capacity (kg)
- m_{CH_4,i_s}- CH_4 storage level at the day-ahead bid time (kg); where i_s indicates the start of the next 24 h of the day-ahead bid time
- $\dot{m}_{H_2,max}$ - Maximum H_2 production rate at full load (kg/h). It is calculated based on the specific power consumption of PEM electrolyser (4.9 kWh/m^3 at STP), H_2 density (0.08988 kg/m^3 at STP) and the hourly power consumption on full load (more information is found in **Eq. (7)**). This parameter is multiplied by 2 to account for the mass difference between H_2 and CH_4 according to the methanation stoichiometric reaction **(Eq. (2))**.

The individual hours of operation are derived from the forecasted electricity prices for the next day. Thus, the electrolyser's operation mode E_i for the next 24 h i at the day-ahead bid time are distributed to the cheapest hours according to the forecasted prices, as follows:

(i) Sort forecasted hourly prices for the next day
(ii) Run full load $E_i = 1$ for the cheapest FLH (integer values of E_{FLH})

(iii) Run part load in the (remaining) hour if $0 < E_{FLH} < 1$. Part load of the electrolyser is defined as the remaining load needed to produce sufficient CH_4 to fill-up the CH_4 storage of the PtG plant

The H_2 production for each hour is calculated based on the power consumption of the PEM electrolyser **(Eq. (7))**:

$$m_{H_2,i} = E_i \cdot W_{el} \cdot \frac{\rho_{H2}}{W_{H_2}} \tag{7}$$

where:

- $m_{H_2,i}$ - H_2 production in each hour i (kg)
- W_{el} - Hourly power consumption of the electrolyser on full load (MWh)
- ρ_{H2} - H_2 density (0.08988 kg/m^3 at STP)
- W_{H_2} - specific power consumption during operation mode (4.9 kWh/m^3 H_2 at STP)

In case the ratio of $m_{H_2,i}$ with CO_2 available at the storage ($m_{CO_2,i}$) reaches the value of 5.83 kg CO_2 per kg H_2, the gas mixture is considered suitable for BM. In this case, the BM reactor can operate at full or part load (BM_i), depending on its maximum CH_4 production capacity ($m_{CH_4,max}$) and the amount of CH_4 needed to fill-up the storage. The latter is defined as the difference between the CH_4 storage capacity ($m_{CH_4,storage}$) and the CH_4 storage level at the day-ahead bid time (m_{CH_4,i_s}). The calculation of BM reactor full- and part load (BM_i) is described in **Eq. (8)**:

$$BM_i = \begin{cases} 1 & if\left(m_{CH_4,storage} - m_{CH_4,i\_s}\right) \geq m_{CH_4,max} \\ \dfrac{\left(m_{CH_4,storage} - m_{CH_4,i\_s}\right)}{m_{CH_4,max}} & else \end{cases} \tag{8}$$

where:

- BM_i - BM reactor operation mode in each hour i

Synthetic CH_4 production for each hour i ($m_{CH_4,i}$) is calculated based on the load required for BM and the mass of H_2 used to produce CH_4 (0.5 kg H_2 for 1 kg CH_4 according to stoichiometric reaction **Eq. (2)**) **(Eq. (9))**:

$$m_{CH_4,i} = BM_i \cdot m_{H_2,i} \cdot 2 \tag{9}$$

The annual FLH of the electrolyser and BM reactor are defined as the sum of hourly operation events (i.e. full or part load) of the electrolyser and the BM reactor **(Eq. 10–11)**:

$$E_{FLH} = \sum\nolimits_{i=1}^{8760} E_i \tag{10}$$

$$BM_{FLH} = \sum\nolimits_{i=1}^{8760} BM_i \tag{11}$$

where:

- E_{FLH} - annual FLH of the electrolyser (h)
- BM_{FLH} - Annual FLH of BM (h)

The costs associated with electricity use during the electrolyser's operation ($C_{el,i}$) are based on the day-ahead spot market (W_{el}) and from the grid for safety infrastructure (W_{safe}) as follows:

$$C_{el,i} = T_{spot,i} \cdot W_{el} \cdot \frac{m_{H_2,i}}{\rho_{H2}} + T_{grid} \cdot W_{safe} \cdot \frac{P_N}{1.074} \tag{12}$$

where:

- $C_{el,i}$ - costs associated with electricity use during the electrolyser's operation (€)
- $T_{spot,i}$ - day-ahead spot market price for each hour i of the electrolyser's operation (€/MWh). A fixed value of 10 €/MWh is added to the day-ahead spot market price to account for grid transmission fees.
- T_{grid} - fixed tariff for grid-based power (100 €/MWh)
- P_N - electrolyser's nominal rated power (MW).
- W_{el} - power consumed from the spot market (MWh)
- W_{safe} - from the grid for safety infrastructure (MWh)

The yearly costs to keep the electrolyser on cold standby (C_{cold}) during non-operating hours is described in **Eq. (13)** as follows:

$$C_{cold} = T_{grid} \cdot W_{cold} \cdot \frac{P_N}{1.074} \cdot (8760 - E_{FLH}) \tag{13}$$

where:

- C_{cold} - annual costs to keep the electrolyser on cold standby (€)
- W_{cold} - power consumed from the regulated market during cold standby (MWh)

The power consumption during cold standby and for safety infrastructure is based on a 1.074 MW plant and is proportionally adjusted to each assessed electrolyser size [28]. Additionally, to allow for gas storage and refuelling, CO_2 ($W_{comp\_CO_2}$) and CH_4 ($W_{comp\_CH_4}$) are compressed at 15 and 200 bar, respectively. The costs associated with gas compression ($C_{comp,i}$) in each hour i are described in **Eq. (14)** below:

$$C_{comp,i} = \left(T_{spot,i} \cdot W_{comp\_CO_2} \cdot m_{CO_2,i}\right) + \left(T_{spot,i} \cdot W_{comp\_CH_4} \cdot m_{CH_4,i}\right) \tag{14}$$

where:

- $C_{comp,i}$ - costs associated with gas compression for each hour i (€)
- W_{comp,CO_2} - electricity consumption for CO_2 compression (0.185 kWh/kg CO_2)
- $m_{CO_2,i}$ - CO_2 production from the biogas upgrading unit for each hour (kg)
- W_{comp,CH_4} - electricity consumption for CH_4 compression (0.34 kWh/kg CH_4)

Finally, the total electricity costs for the PtG plant (C_{total}) are based on costs associated with the electrolyser's operation ($C_{elec,i}$), gas compression ($C_{comp,i}$), and the cold standby of the electrolyser (C_{cold}) **(Eq. (15))**:

$$C_{total} = \sum\nolimits_{i=1}^{8760} \left(C_{el,i} + C_{comp,i}\right) + C_{cold} \tag{15}$$

To determine the optimal plant configuration, a total number of 900 simulations based on a Monte Carlo approach were run for each power availability and fuel demand scenarios. Each simulation corresponded to a combination of electrolyser capacity between 500 kW_{el} and 15,000 kW_{el} (500 kW_{el} increments) and CH_4 gas storage between 500 kg and 15,000 kg (500 kg increments). As no intermediate H_2 storage is considered, variations in BM reactor capacity were performed simultaneously to the electrolyser. This approach avoided oversizing the BM reactor while ensuring that all H_2 production from the PEM electrolyser could be uptaken.

After finding the combination of PEM electrolyser/BM reactor and CH_4 storage capacities that resulted in the lowest GPC, variations in CO_2 storage were performed to find the minimum storage size required to keep delivering synthetic CH_4 on demand according to the different power availability and fuel demand scenarios.

For each plant configuration thus obtained, specific CAPEX (€/kW), annual FLH, the average price paid for the electricity, and the GPC were calculated. To ensure that the PtG plant configurations were fulfilling the consumers' fuel requirement, the on-demand delivery of synthetic CH_4 was considered a mandatory criterion. The characteristic dependencies of different plant configurations on each performance indicator were visualized using Matlab 3-D contour plots (MathWorks, USA). For each scenario assessed, the combination of electrolyser/BM reactor capacities and CH_4 storage size that resulted in the lowest GPC and simultaneously fulfils CH_4 demand was considered as optimal plant configuration.

3. Results and discussion

3.1. Optimisation of the PtG plant

To identify the plant configurations that result in the lowest synthetic CH_4 production cost (i.e. GPC) for each scenario, a stepwise optimisation procedure was performed (see section 2.6). The interactions of specific CAPEX, annual FLH, electricity price, GPC, and synthetic CH_4 delivery are shown in Fig. 4 for the optimisation step in which the PtG plant capacity and the CH_4 storage size were varied**.** The results in Fig. 4 refer to the unconstrained high demand fuel scenario (Un_{high}) and serve as an example for the model's behaviour. The GPC for all scenarios can be found in **Appendix 3**.

As the investment costs for storing CH_4 compressed at 5 bar are relatively low (50 €/kg), variations in the PtG plant capacity resulted in a greater influence on the specific CAPEX compared to the CH_4 storage size. Furthermore, applying a scale effect to the main components of the PtG plant **(Appendix 2)** results in more expensive smaller plants **(**Fig. 4**a)**. When operated on-demand, such smaller plants can run with higher annual FLH compared to larger ones **(**Fig. 4**b)**. The effects of FLH on production costs have been extensively studied when electricity is obtained from the grid in spot markets with a limited number of low-cost hours throughout the year [4,29,30]. Essentially, it has been shown based on Irish, Japanese and Swedish electricity markets that at least 3000–5000 FLH are required to minimise H_2 production costs. Operating electrolysers at lower annual FLH would result in prohibitive costs, even at low electricity prices [17,29,30].

Therefore, finding a plant configuration that minimizes the GPC requires the optimisation of FLH and the price paid for the electricity under the pre-defined condition of fulfilling the fuel demand. For the Un_{high} scenario, the optimal PtG plant capacity was found to be 4500 kW_{el} with a CH_4 storage size of 11000 kg, which resulted in around 7700 FLH and an average electricity price of 54.49 €/MWh **(**Fig. 4**c-d)**. By reducing the CH_4 storage size, a marginal reduction in specific CAPEX could be noticed. However, in this case, the PtG plant wouldn't be able to deliver synthetic CH_4 on-demand anymore **(**Fig. 4**e)**.

After finding the optimal PtG plant capacity and CH_4 storage size, the CO_2 storage was optimised for each investigated scenario **(**Fig. 5**)**. While the amount of CO_2 required for BM doubles from the low to the mid fuel demand scenario and triples from the low to the high fuel demand scenario, the minimum CO_2 storage needed to allow delivery of synthetic CH_4 on-demand was 2.5 times higher for the mid fuel demand scenario compared to the low one and approximately 4 times higher for the high fuel demand scenario. As the needed CO_2 storage capacity is thus increasing more rapidly at higher synthetic CH_4 demands than the CO_2 input for the BM reactor, this could represent a challenge in urban contexts where space for compressed CO_2 storage systems is rather limited and/or expensive. In case the existing AD infrastructure offers a biogas upgrading system that does not result in a concentrated CO_2 stream (e.g. pressurized water scrubbing), different plant designs could be explored. For instance, raw biogas could be directly used for methanation as an alternative biogas upgrading system [31]. Such a concept would however directly interfere in the gas storage

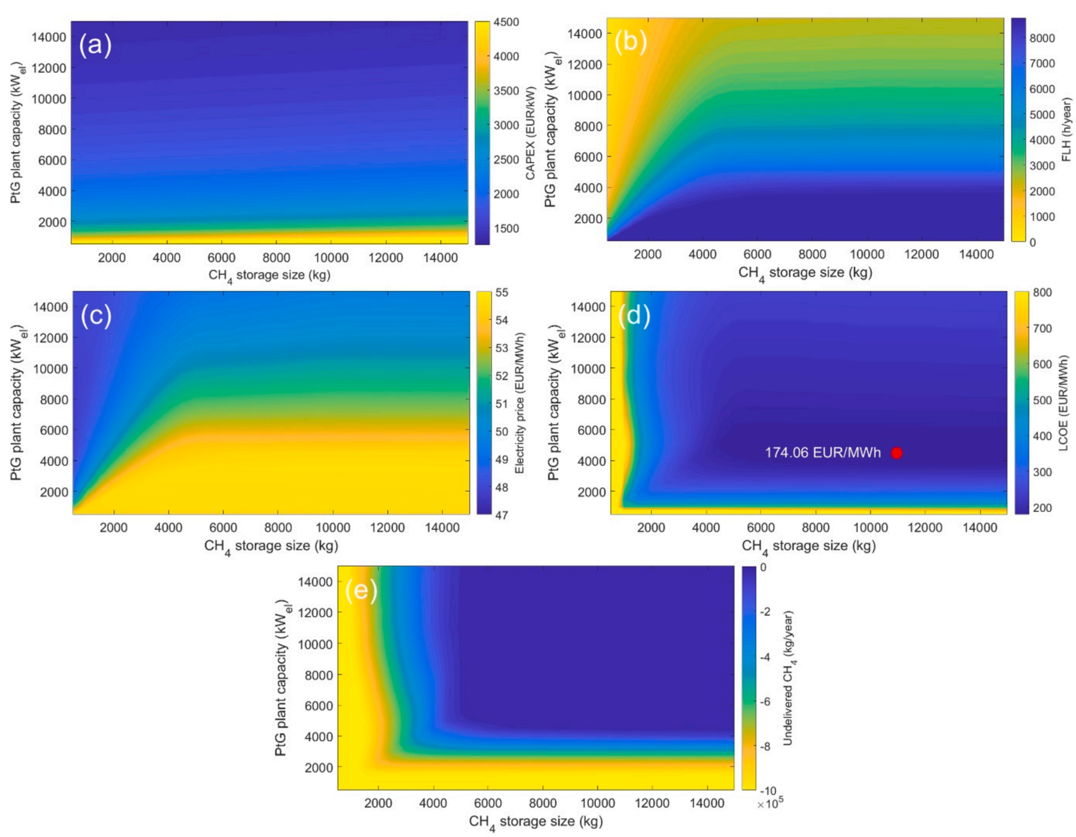

Fig. 4. Optimisation of PtG plant and CH_4 storage capacity for the unconstrained high demand fuel scenario (Un_{high}). (a) specific capital expenditures (CAPEX); (b) full load hours (FLH); average price paid for the electricity; (d) levelised cost of synthetic CH_4 production (GPC); (e) delivery check of synthetic CH_4.
Note: CO_2 storage size was kept at 30 ton during this optimisation step.

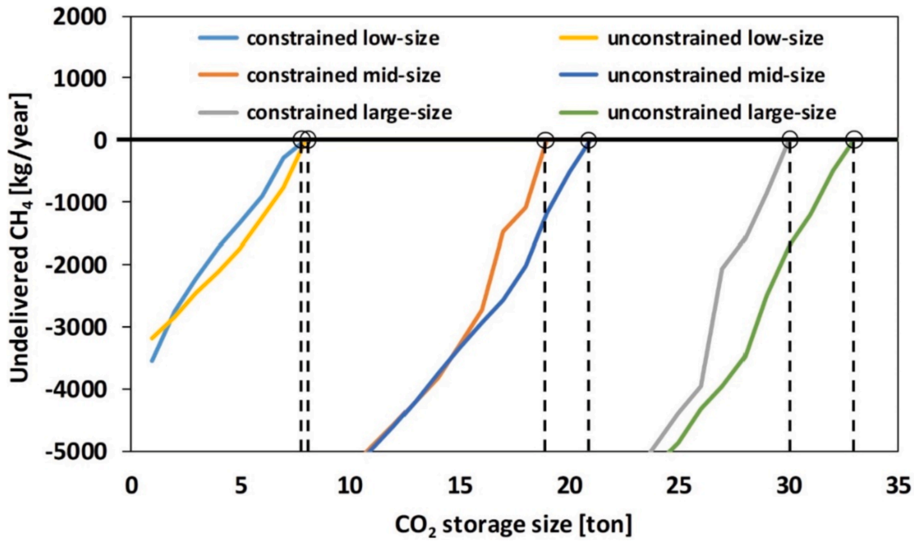

Fig. 5. Minimum CO_2 storage size required for the different PtG scenarios to deliver synthetic CH_4 on-demand.

requirements (CO_2 and CH_4) and size of the BM reactor. In addition, it would result in a mixed output of bio- and synthetic CH_4, which could make the differentiation under existing regulations and incentive schemes more complex for each type of renewable gas (more information is found in Section 3.5 – Summary for policymakers).

When comparing the different power availability cases, it is observed that the PtG plant capacity was 2 times higher by limiting the electrolyser operation to 12 h/day (constrained power availability) compared to unlimited electrolyser operation (unconstrained power availability). This characteristic directly influenced the annual FLH of both the electrolyser and the methanation unit as well as the price paid for the electricity, similarly as explained in Fig. 4. For this reason, the unconstrained power scenarios showed 18% lower GPC on average compared to the constrained ones. A summary of the optimal plant configurations found for all analysed scenarios is shown in Table 3.

Furthermore, when comparing across different plant capacities and price paid for electricity between constrained and unconstrained power scenarios, the breakdown of the GPC showed a lower average share of electricity costs in the constrained power scenarios (~47%) compared to the unconstrained ones (~60%). Conversely, CAPEX and fixed OPEX (based on a fraction of CAPEX), revealed to be more important to determine the GPC in the constrained power scenarios (~49%) compared to the unconstrained ones (~37%) (**Appendix 4**).

3.2. Dynamic operation of the PtG plant

This section presents the dynamic interactions of the different aspects affecting the optimal plant configuration (i.e. electricity price, power availability, and synthetic CH_4 demand).

3.2.1. Electrolyser operation

Based on the optimal plant configuration of both the constrained high demand fuel (Co_{high}) and Un_{high} scenarios, the dynamics of the electrolyser operation are shown in Fig. 6. The daily FLH operation limit of 12 h applied to the constrained power scenario resulted in a more frequent electrolyser operation between 10 pm and 5 am. As the operation scheduling of the PtG plant is based on day-ahead forecasted electricity prices, this period generally coincides with the cheapest prices throughout the day due to the lower overall load in the system overnight.

Similar behaviour is also observed for the unconstrained power scenarios, however, the fluctuations in daily electrolyser load were found to be less pronounced. With a more compact system, the unconstrained power scenario needed to compensate for its lower hourly H_2 production capacity by occasionally operating during daytime peak load hours.

The non-operating hours as well as the number of ramp-ups from cold standby increased when power availability was constrained. The

Table 3
Summary of the optimal plant configurations for the power availability and fuel demand scenarios.

Power availability scenarios	Fuel demand scenarios	PEM electrolyser [kW]	CO_2 storage [kg]	BM reactor [kg CH_4/h]	CH_4 storage [kg]	Electrolyser FLH [h/year]	Methanation FLH [h/year]	CAPEX [€/kW_{el}]	Price [€/MWh]	GPC [€/MWh]
Constrained	Low	3000	8000	110	5000	3905	3842	2309	51.39	234.42
	Mid	6000	19,000	220	7000	3902	3847	1895	51.35	207.14
	High	9000	30,000	330	10,500	3900	3828	1659	51.34	194.81
Unconstrained	Low	1500	8000	55	3500	7703	7685	3179	54.47	188.31
	Mid	3000	21,000	110	7000	7717	7698	2467	54.48	170.78
	High	4500	33,000	165	11,000	7720	7693	2164	54.49	162.34

Note: GPC refers to the levelised cost of synthetic CH_4 production.

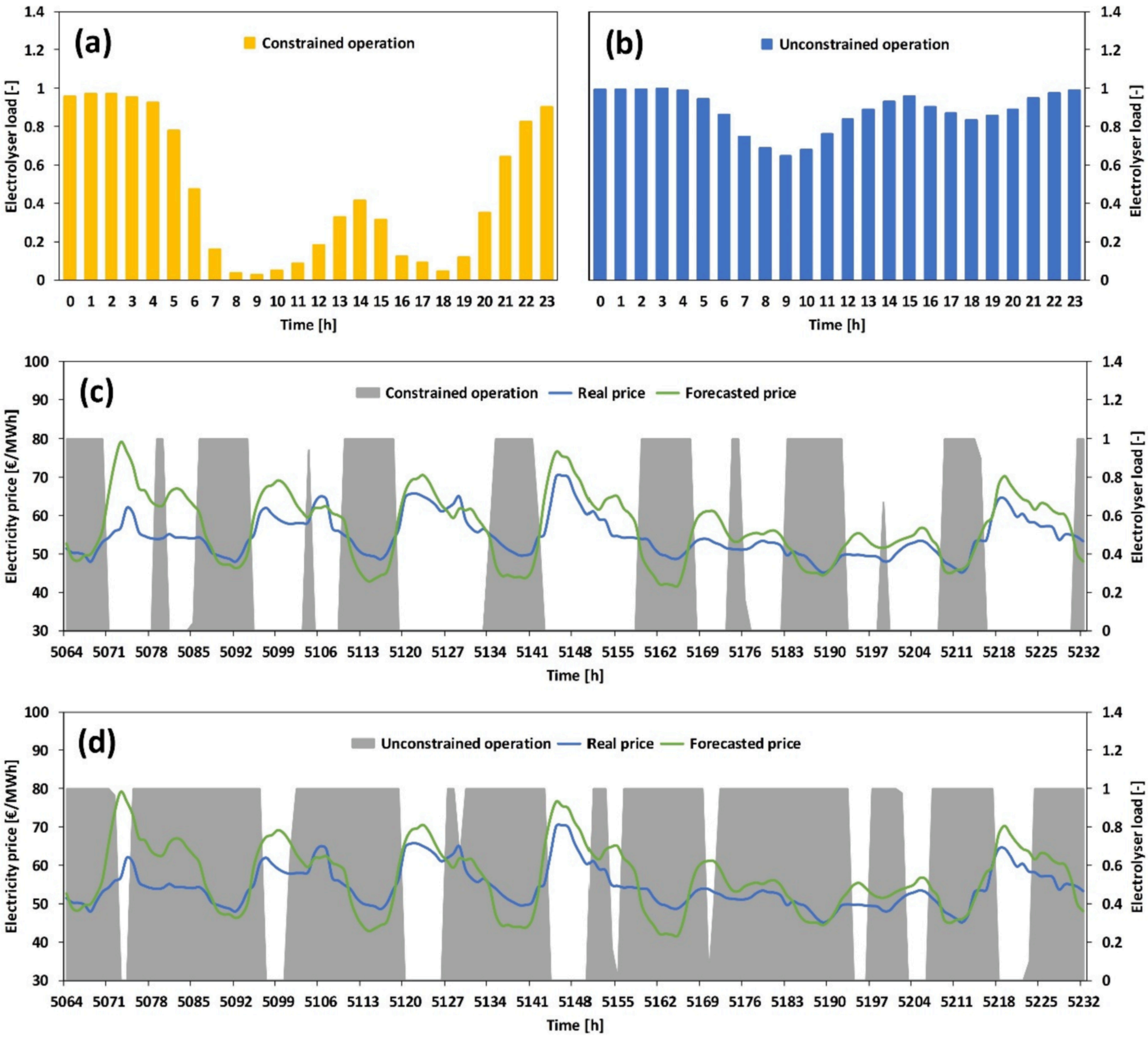

Fig. 6. Dynamic operation of the electrolyser under for the constrained and unconstrained power availability for the high demand fuel scenarios. (a) Annual hourly average electrolyser load for constrained power availability; (b) annual hourly average electrolyser load for unconstrained power availability; (c) One-week sample of the electrolyser operation under constrained power availability; and (d) One-week sample of the electrolyser operation without power availability constraint.

total electricity consumption per kg of produced H_2 therefore differed between constrained and unconstrained operation when taking into account the power used for safety infrastructure, during cold standby and ramp-up. The constrained operation resulted in approx. 2% higher total electricity consumption to produce H_2 (55.93 kWh/kg H_2) compared to the unconstrained operation (54.80 kWh/kg H_2), which in turn slightly reduced the overall efficiency of the process and thus increased production costs of the PtG plant under constrained operation.

Furthermore, the interactions between real and forecasted prices on the electrolyser operation can also be observed (Fig. 6**c,d**). The forecasted prices were useful to identify the cheapest hours in the day-head scheme and schedule the operation of the electrolyser on them. Such behaviour occurred independently of the mean absolute percent error of 14% found for the forecasted prices throughout the simulated year of 2018 [17]. Here the higher electrolyser capacity of the constrained power scenario showed to be an advantage to source the cheapest hours of the day since in the unconstrained power scenario an average higher electricity price of 6% was paid. Nevertheless, alone this aspect was not sufficient to result in a lower GPC for the constrained power scenario, in particular due to the relatively low annual FLH below 4000 previously highlighted.

To verify the influence of forecasting errors on the electrolyser's operation, real prices were used to simulate a zero error day-ahead electricity price forecasting. For the Un_{high} scenario, the price paid for electricity could be reduced by 10 ct EUR/MWh, however, with a negligible impact on the GPC. Meanwhile, zero error forecasting was able to reduce the price paid for the electricity by 62 ct EUR/MWh for the Co_{high} scenario, but again with a marginal reduction in the GPC by just 0.65%. Overall, this minor influence of forecasting errors in the GPC is explained by different reasons: (i) price forecasting is used to identify the cheapest hours of the day, therefore reducing the difference between forecasted and real values does not necessarily contribute as much as knowing when is cheap or expensive throughout the day is, (ii) even though the price paid for the electricity is the most important aspect when determining the GPC (**Appendix 4**), other aspects like CAPEX and fixed OPEX do not change as a function of price forecasts, therefore reducing the impact of forecasting errors on the production cost of synthetic CH_4.

3.2.2. Methanation operation

As the methanation capacity was simultaneously varied with the electrolyser capacity during the step-wise optimisation procedure, the operation of the BM reactor was directly connected with the electrolyser which in turn operated according to the amount of CH_4 available in the

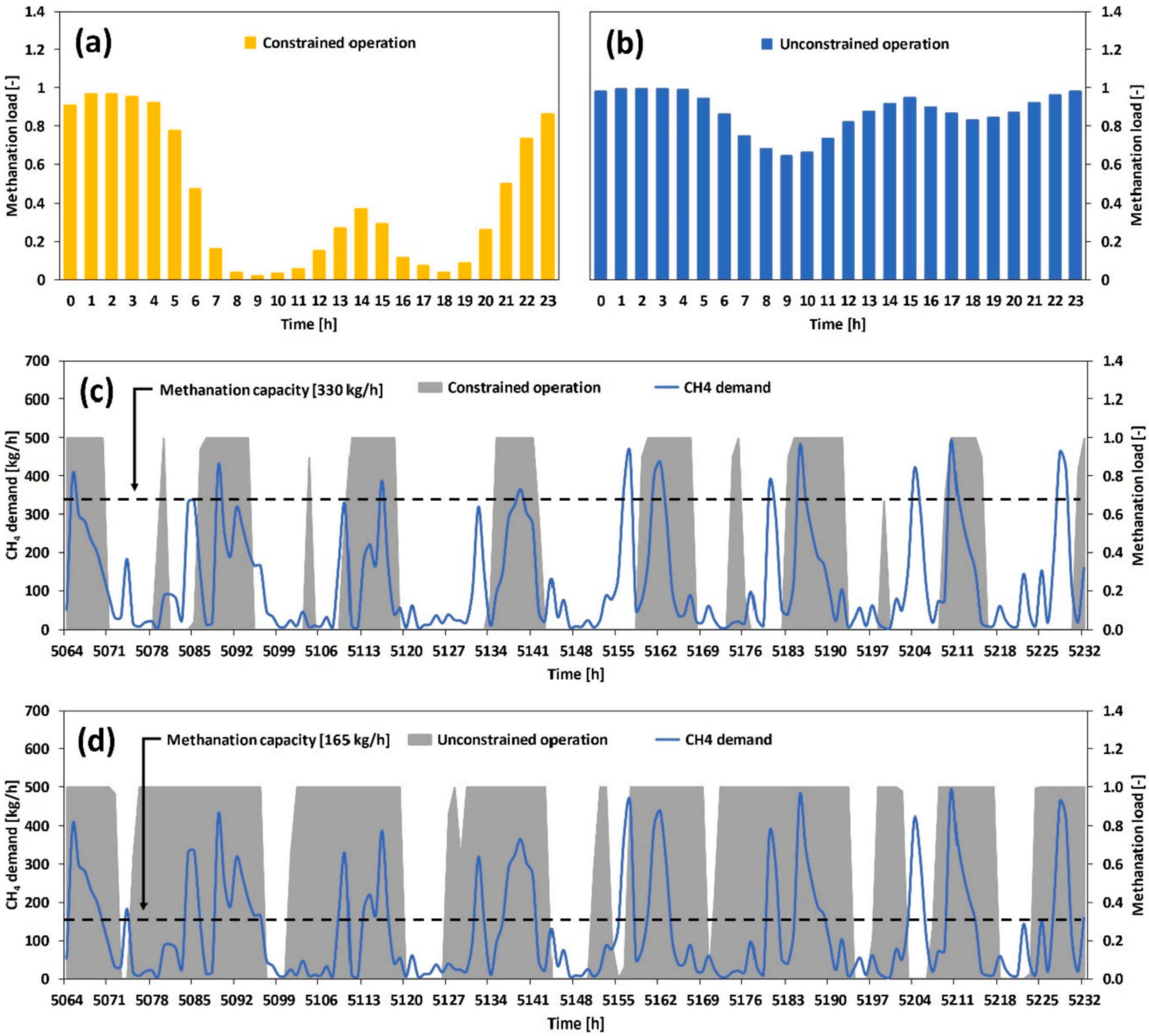

Fig. 7. Dynamic operation of the methanation reactor under constrained and unconstrained power availability for the electrolyser in the high demand fuel scenarios. (a) annual hourly average methanation reactor load for the constrained scenario; (b) annual hourly average methanation reactor load for the unconstrained scenario; (c) One-week sample of the methanation reactor operation for the constrained scenario; and (d) One-week sample of the methanation reactor operation for the unconstrained scenario.

storage. Thus, the maximum daily FLH allowed for the operation of the electrolyser was also reflected in the BM reactor operation. For the same reason, the constrained power setting showed a much higher number of ramp-ups of around 5 times higher (approx. 4780) than the unconstrained one (approx. 960) due to its higher production capacity. Similarly to the electrolyser, the BM reactor's ramp-up resulted in energy losses, this time due to unreacted input gases which could contribute to reducing the overall performance of the process. For instance, the Co_{high} scenario wasted 6.6 times more H_2 (2966 kg H_2/year) compared to the Un_{high} scenario (448 kg H_2/year). Despite being marginal, these losses contributed to reducing the overall efficiency of the PtG plant of 51.94% for the constrained power scenario and 53.36% for the unconstrained power scenario (based on HHV of synthetic CH_4). In fact, gas compression revealed to be one of the most important aspects for the energy balance of PtG plants which has often not been considered in previous studies. When neglecting the power used for compression of CO_2 and CH_4, the overall efficiency of the PtG plant would be 55.01% and 53.51% under the constrained and unconstrained power availability conditions, respectively **(Sankey diagrams are available in Appendix 5)**.

Furthermore, the larger methanation capacities of constrained power scenarios (110–330 kg/h) allowed a more flexible operation to supply the different synthetic CH_4 demands (average of 46–138 kg/h) compared to the unconstrained power scenarios (55–165 kg/h). Such aspect, however, did not influence the CH_4 storage capacities due to the PtG plant's need for purchasing power ahead in time to produce synthetic CH_4 and keep the storage full. The dynamics of the methanation reactor operation for both constrained and unconstrained power scenarios are shown in Fig. 7.

3.3. *Total cost of ownership*

The TCO was assessed as an economic indicator to compare the produced synthetic CH_4 with fossil fuels from a bus operators' perspective. The TCO calculations are based on CAPEX of different bus types (gas and diesel), maintenance costs, fuel consumption, and as well as driver salaries. The results of this analysis are separated into TCO for synthetic CH_4 buses displacing diesel **(**Fig. 8**a)** and CNG **(**Fig. 8**b)**. This approach is necessary because synthetic CH_4 is only a drop-in fuel for existing gas bus fleets running on CNG. In contrast, when diesel is to be

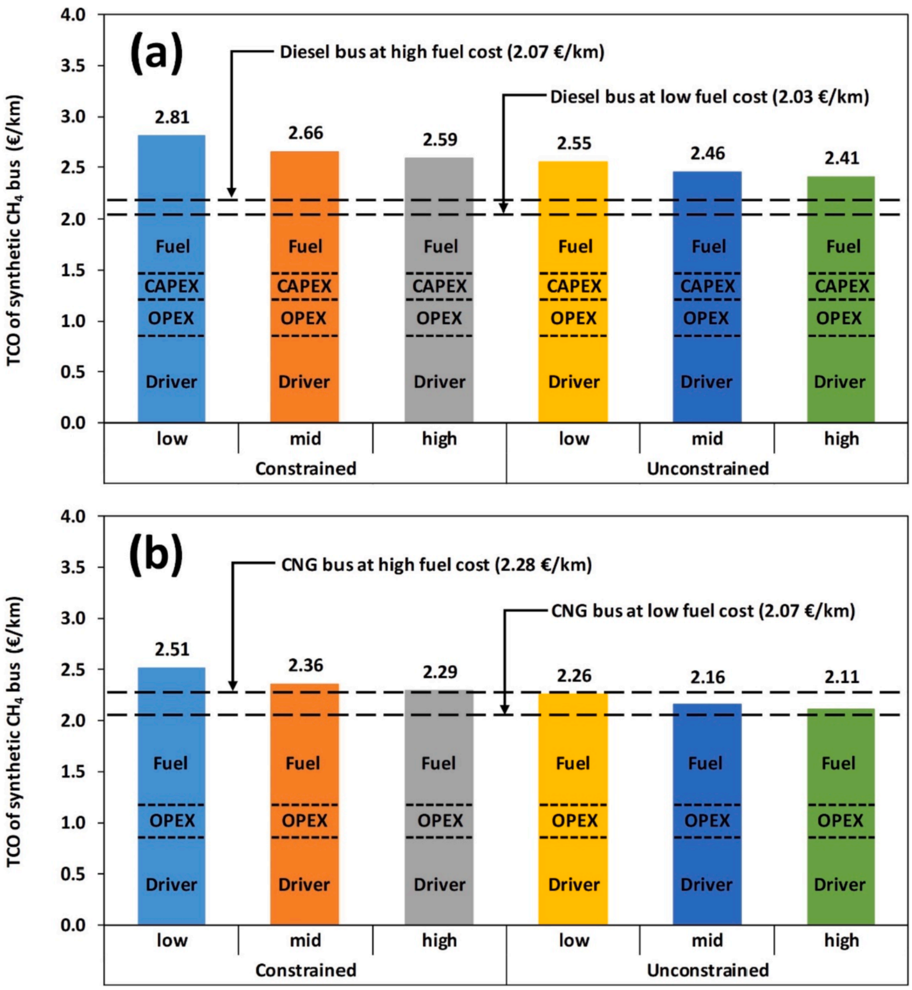

Fig. 8. Total cost of ownership (TCO) for synthetic CH_4 bus in the different PtG plant scenarios analysed. (a) synthetic CH_4 bus displacing diesel bus, and (b) synthetic CH_4 displacing CNG in a gas bus.
Note: Calculations do not consider refuelling station costs, bus depot overheads, insurance and vehicle residual value. Diesel and CNG prices were obtained from [32,33]. Operational expenditures (OPEX) refer to maintenance.

displaced, the buses need to be retrofitted or new buses need to be procured as synthetic CH_4 cannot be directly used in diesel-powered vehicles.

The results showed that 8% lower TCO was found on average for the unconstrained power availability scenarios compared to the constrained ones. This value is smaller than the 18% difference observed in the GPC because fuel consumption represents only 40–53% of the TCO for buses driven with synthetic CH_4 depending on the PtG plant scenario assessed. However, as the driver salary is fixed, and CAPEX and maintenance do not differ significantly between gas and diesel buses **(Appendix 6)**, the fuel costs are still the most important factor from the bus operators' perspective.

Therefore, the TCO of buses powered by synthetic CH_4 showed to be higher than with conventional fossil fuels in most of the scenarios analysed. While diesel buses showed 14–28% lower TCO than synthetic CH_4, gas buses showed a different behaviour. In this case, depending on the PtG plant scenario and CNG price assessed, buses driven on synthetic CH_4 could be either more expensive or cheaper than fossil fuel. This is explained by lower production costs of synthetic CH_4 at larger plants specially operated without power constraint combined with geographical and temporal variations on CNG price.

Regardless the PtG scenario analysed, diesel (73–100 €/MWh) and CNG (83–129 €/MWh) always showed lower price than synthetic CH_4 (162–234 €/MWh). Previous studies on PtG based on high temperature electrolysis coupled with catalytic methanation reached similar conclusions even with a higher global efficiency of the system (71.9–75.1%) [34]. Such gap in fuel prices makes challenging to design climate mitigation policies supporting synthetic CH_4 as fuel in regional and intercity buses, most likely requiring a combination of different measures to incentivize them.

Overall, TCO were lower compared to usually found in the literature for PTB because in our case long-distance buses with a daily trip of 368 km were considered [35,36]. Should the daily distance trip be reduced, the TCO of all buses assessed would be increased favouring less capital intensive options [37].

3.4. Carbon abatement cost

The use of renewable CH_4 in PTB instead of fossil fuels could help to decarbonise intercity public transportation, where – due to the longer distances – direct electrification is currently not a reliable option. Based on the emission reductions of displacing fossil fuels (either diesel or compressed natural gas) and the cost difference between synthetic CH_4 and fossil fuels (considering both low and high fossil fuel prices), the CAC was calculated for a variety of scenarios. For the CAC analysis, the impact of different levels of carbon taxation on the consumption of fossil fuels was also considered. The CAC results are presented in Fig. 9.

Overall, producing synthetic CH_4 to displace diesel in PTB showed to be an expensive option (CAC > 250 €/tCO_2) regardless the power availability and the PtG plant size. Carbon taxation on diesel resulted in a limited effect on incentivising the adoption of synthetic CH_4 by PTB operators since even a relatively high carbon tax of 114 €/tCO_2 (current

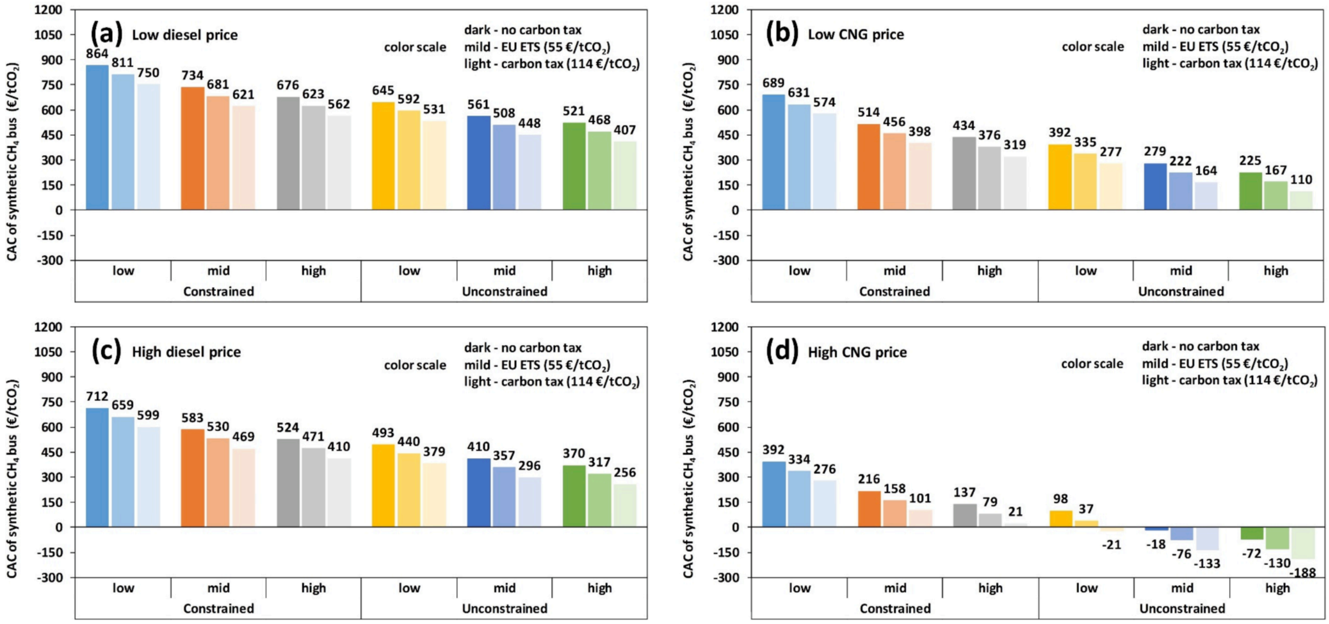

Fig. 9. Carbon abatement cost of fossil fuel substitution by synthetic CH_4 in the different PtG scenarios analysed. (a) substitution of diesel at low fossil fuel price, (b) substitution of compressed natural gas at low fossil fuel price, (c) substitution of diesel at high fossil fuel price, and (d) substitution of compressed natural gas at high fossil fuel price.

Note: Fuel prices (without carbon tax) were based on 0.94 €/kg (low CNG price), 1.61 €/kg (high CNG price), 0.80 €/L (low diesel price), and 1.20 €/L (high diesel price).

Swedish carbon tax) [38] would reduce the CAC just by 13–22% (Fig. 9**a,c**).

In contrast, synthetic CH_4 production to displace CNG showed a more nuanced picture (Fig. 9**b,d**): If the CNG price is low, carbon taxation would reduce the CAC at best by 51%, resulting in abatement costs of 110 €/tCO_2 for full-scale PtG plants with unconstrained power availability. This value is in the same order of magnitude as the CAC of biomethane in heavy-duty transportation [39]. Nevertheless, these results highlight the economic challenge of decarbonizing hard-to-abate sectors like heavy-duty vehicles, such as intercity buses, in a context of low fossil fuel prices. A carbon tax alone is not sufficient to incentivise a shift from diesel towards synthetic CH_4 in countries with low CNG prices.

However, if the CNG prices are high (like Sweden and Switzerland which fall with 1.61 €/kg and 1.87 €/kg in the upper range of CNG prices examined in this study [32]), a carbon tax could significantly contribute to reducing the CAC – even to the extent of reaching negative CAC levels (see Fig. 9**d**). While the CAC were already slightly negative for mid and high fuel demands in a high CNG price scenario with unconstrained electricity consumption, the carbon tax was able to amplify this effect and to assist PtG plants serving low fuel demands in reaching negative abatement costs as well.

3.5. Summary for policymakers

The present study investigates synthetic CH_4 as a renewable fuel option for PTB operators with bus fleets for intercity and regional transport, covering medium to large distances and being therefore less promising for direct electrification.

Our results show that synthetic CH_4 can already be competitive as alternative fuel option in the PTB sector, however only in limited cases of (a) no constraints in power availability for the PtG operation, (b) high prices for fossil fuels, (c) mid to high fuel demand, and (d) substituting CNG, i.e. not requiring investments in new or retrofitted buses. A high carbon tax demonstrated to have some effect on improving the business case of synthetic CH_4 as carbon–neutral PTB fuel **(see section 3.4)**. However, to incentivise a broader adoption of this kind of fuel, additional policy instruments should be envisaged. Such measures can tackle either the supply side (i.e. supporting the production of synthetic CH_4) or the demand side (i.e. incentivizing PTB operators to switch fuels and, where needed, retrofit or renew their fleets to be compatible with synthetic CH_4).

On the demand side, for instance, the European Commission's proposal for amending the Renewable Energy Directive (RED II; Directive 2018/2001/EU) [40], published in July 2021 as part of its Fit for 55 legislative package and currently under discussion in the Council of the European Union, introduces a target for renewable fuels of non-biological origin (RFNBO), which include synthetic CH_4 if produced based on renewable electricity, to meet 2.6% of all transport demand in 2030. Such a RFNBO target could guarantee some market for RFNBOs and thus support PtG plants in producing synthetic CH_4.

Other measures to incentivise PtG operation are to tackle the highest cost factor for production, i.e. electricity as shown by a breakdown of GPC and TCO (**Appendixes 4 and 5**), or to directly subsidise the PtG production. However, a flat subsidy per kg of synthetic CH_4 is not recommended as the analysis showed important variations in production cost and CAC depending on the plant configuration and scenarios. Flat subsidies would equally treat all potential production scenarios and uses, thus providing unnecessary funding to cases where CAC is already negative and can therefore be considered viable without public support. Instead, alternative options like (carbon) contracts for difference or competitive funding schemes should be explored, notably for first-movers who implement and run PtG plants at an early stage of the market ramp-up as a means of helping them to bring down costs for next generation PtG plants.

On the demand side, a multitude of policy instruments exists that could target the retrofitting and/or procurement of new buses that can be powered by CNG/ synthetic CH_4, such as purchase incentives or tax benefits. Both are common measures for stimulating electric vehicle sales and could also be applied to carbon–neutral buses that use renewable synthetic CH_4.

The present study is a technology modelling exercise that highlights the potential of flexible operation modes of PtG plants according to fluctuations in fuel demand over time. While the study does not present a systems analysis, the on-demand modelling of synthetic CH_4 production however can be interpreted in the broader scope of CCU and sector coupling. The utilisation of CO_2 from a biogas plant in close proximity to the PtG plant allows to recycle carbon that otherwise would be directly emitted to the atmosphere, thus improving the carbon footprint of the process. Such an industrial symbiosis further strengthens local economic clusters and sector coupling. In addition, thanks to its flexible operation design, the PtG plant can be beneficial for the electricity market by adjusting its production according to the grid load, thus offering a grid balancing service. However, as the analysis of constrained and unconstrained power availability showed, this flexibility comes at a cost, namely lower annual FLH that result in higher GPC.

The extend and timing of FLH for operating electrolysers and therefore also PtG plants is currently part of a broader sustainability debate for RFNBOs. In Germany, for instance, an ordinance to the Renewable Energy Act (Erneuerbare-Energien-Gesetz, EEG) grants an exemption from the EEG levy only for the first 5000 FLH of an electrolyser and sets further sustainability criteria for green hydrogen production. By 31 December 2021, the European Commission was expected to adopt a delegated act in accordance with RED II stipulations to establish a methodology that sets out the sustainability requirements for taking electricity from the grid for the production of RFNBOs. While the specific requirements are not yet published, the methodology will need to ensure that only electricity of additional renewable origin is used and that there is a temporal and geographical correlation between the electricity and RFNBO production units, thus avoiding power consumption of electrolysers in case of grid congestion. Depending on the final design of the sustainability criteria, the impact on electricity purchase costs can negatively impact the business case for synthetic CH_4 for PTB fleets.

4. Conclusion

A dynamic model was built for discrete-event simulations of synthetic CH_4 production aiming at the substitution of fossil fuel in medium- to long-distance intercity/regional public transportation buses (PTB). With this model, different scenarios for power-to-gas (PtG) integration into anaerobic digestion (AD) plants were assessed to consider real-world constraints such as limited power availability in urban settings as well as physical and temporal limitations in CO_2 supply from AD plants. PtG plants operated under constrained power supply required larger installed production capacities to provide the same synthetic CH_4 volumes as unconstrained PtG plants. This greatly influenced the optimal operation setting for the constrained PtG plants which in turn made synthetic CH_4 production more expensive when compared to unconstrained PtG plants. Also, variations in CO_2 supply from AD plants made the design of PtG plants more complex, in particular for full-scale applications which required large CO_2 intermediate storage capacities.

Despite these challenges related to synthetic CH_4 production, it could be a climate-friendly option for displacing fossil fuels in medium- to long-distance PTB, specifically in case of existing gas bus fleets. However, the analysis of carbon abatement costs (CAC) showed that synthetic CH_4 is only competitive against fossil fuels in a limited number of specific scenarios and even high carbon taxation improves the business case only to some extent. Hence, additional measures would be required to promote a fuel shift in public bus transport towards renewable options such as synthetic CH_4, such as mandatory RFNBO targets on the supply side or purchase incentives for gas-powered busses on the demand side.

In addition to the GHG emission reduction that can be achieved when replacing fossil fuels by synthetic CH_4, the production of the latter can – when operated in a flexible manner as proposed in the present study – also improve the balancing of the electricity grid and in the current context of fuel price crises due to the Russian invasion of Ukraine in February 2022, the domestic production of synthetic CH_4 can reduce the dependence on fuel imports.

While CAC was addressed in the current study, further research should target the use of hourly-based marginal emission factors for PtG plant operations to provide a more nuanced picture of the process lifecycle emissions, including the attribution of CO_2 emissions from grid-power utilization without renewable energy certificates. Future studies could also extend the scope to the recovery of process by-products like oxygen and low-temperate waste heat to service local infrastructure (e.g. aeration of wastewater and sludge drying) while reducing the production costs of synthetic CH_4.

CRediT authorship contribution statement

Leandro Janke: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization, Project administration, Funding acquisition. **Fabian Ruoss:** Validation, Resources, Writing – review & editing, Funding acquisition. **Alena Hahn:** Validation, Writing – original draft, Writing – review & editing. **Sören Weinrich:** Software, Validation, Writing – review & editing. **Åke Nordberg:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix

Appendix 1: Costs related to the PtG plant.
(See Table 1A).

Table 1A
Capital expenditures (CAPEX), balance of the plant (BoP), operational expenditures (OPEX), gas storage and other costs related to the PtG plant.

Costs for the different components		Value	Reference
PEM electrolyser	CAPEX (€/kW_{el})	970	[19,21,41]
	BoP[a]	0.15	
	OPEX[a]	0.04	
	Replacement[a]	0.2	
BM reactor	CAPEX (€/kW_{th})	600	[21,41]
	BoP[b]	0.2	
	OPEX[b]	0.1	
	Replacement[b,c]	0.05	Own assumption
Gas storage	CO_2 storage at 15 bar (€/kg)	15	
	CH_4 storage at 5 bar (€/kg)	50	
Other	Water (€/m^3)	1.20	
	Land purchase	0.1[b]	

Note:
[a] Percentage of PEM electrolyser's CAPEX.
[b] Percentage of BM reactor's CAPEX.
[c] No catalyst replacement is required for biological methanation.

Appendix 2:. Scale effect on the capital expenditures.

(See Table 1B, Table 1B, Fig. 2A).

Table 1B
Costs and specifications of the fuels and buses assessed.

Description		Value	Reference
CNG bus[a]	CAPEX (€)	250000	[35]
	Maintenance (€/km)	0.36	[35]
	Fuel consumption (kg/km)	0.43	Personal communication[b]
	Fuel cost (€/kg)	0.94-1.87	[32]
	Emission factor (kg CO_2/kg)	2.252	
Diesel bus	CAPEX (€)	220000	[35]
	Maintenance (€/km)	0.30	[35]
	Fuel consumption (L/km)	0.45	Personal communication[b]
	Fuel cost (€/L)	0.80-1.75	[33]
	Emission factor (kg CO_2/L)	2.64	
Other	Trip distance (km/day)	368	Personal communication[b]
	Driver salary (€/year)[c]	57600	[35]

Note:
[a] CAPEX and maintenance of synthetic CH_4 bus are identical to CNG bus. However, fuel consumption of synthetic CH_4 bus (0.40 kg/km) is adjusted to account for differences in energy content.
[b] Obtained with the bus manager of Uppsala County, Sweden.
[c] Two drivers are required per bus.

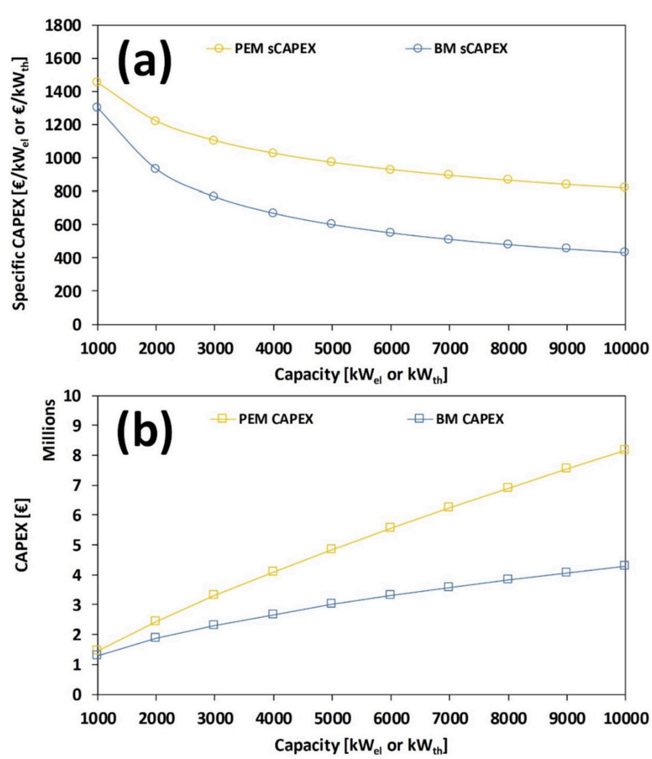

Fig. 2A. Scale effect for the capital expenditures of PEM electrolyser and BM reactor.
Note: Values are given in kW_{el} for electrolyser and kW_{th} for methanation unit according to STORE&GO project [41].

Appendix 3:. Optimisation of all cases assessed.

(See Fig. 3A).

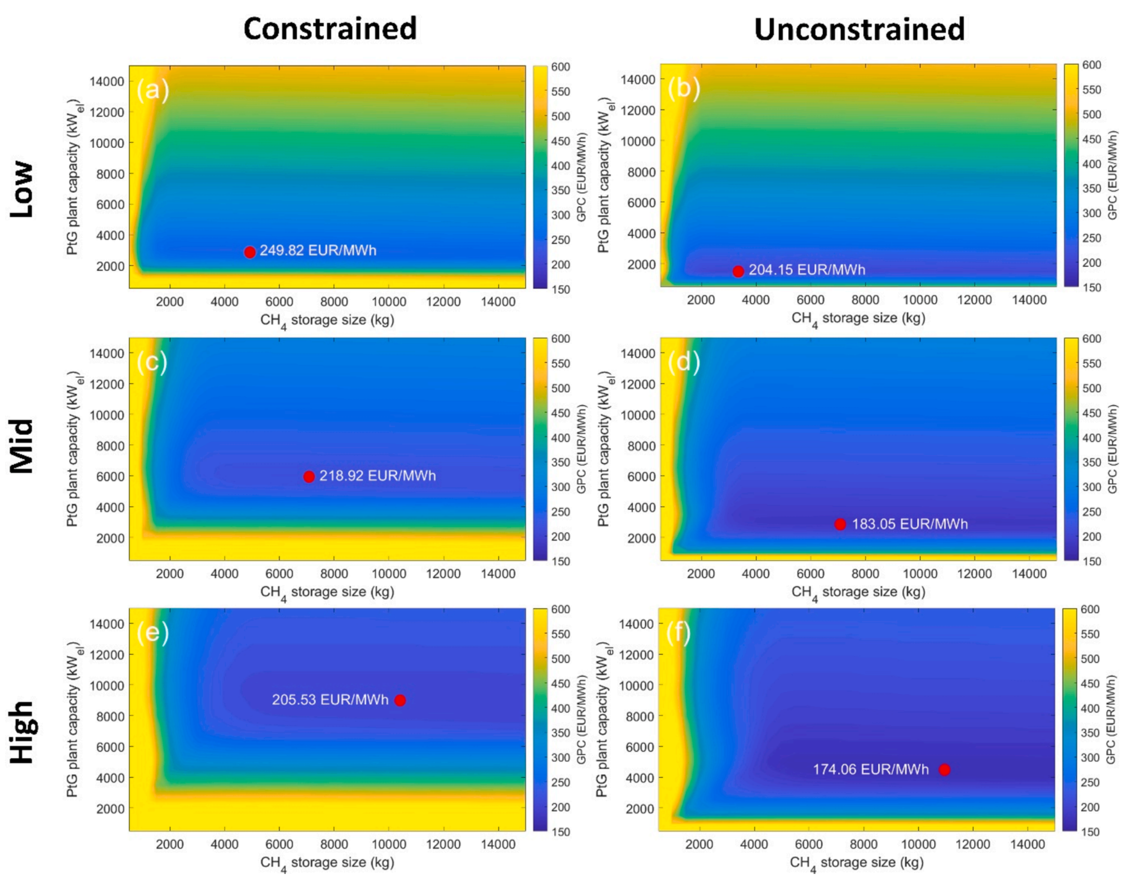

Fig. 3A. Optimisation of the PtG plant configuration in terms of electrolyser capacity and CH_4 storage size.
Note: Red dot shows the optimal plant configuration to minimise the levelised cost of synthetic CH_4 (GPC).

Appendix 4:. Breakdown of GPC.

(See Fig. 4A).

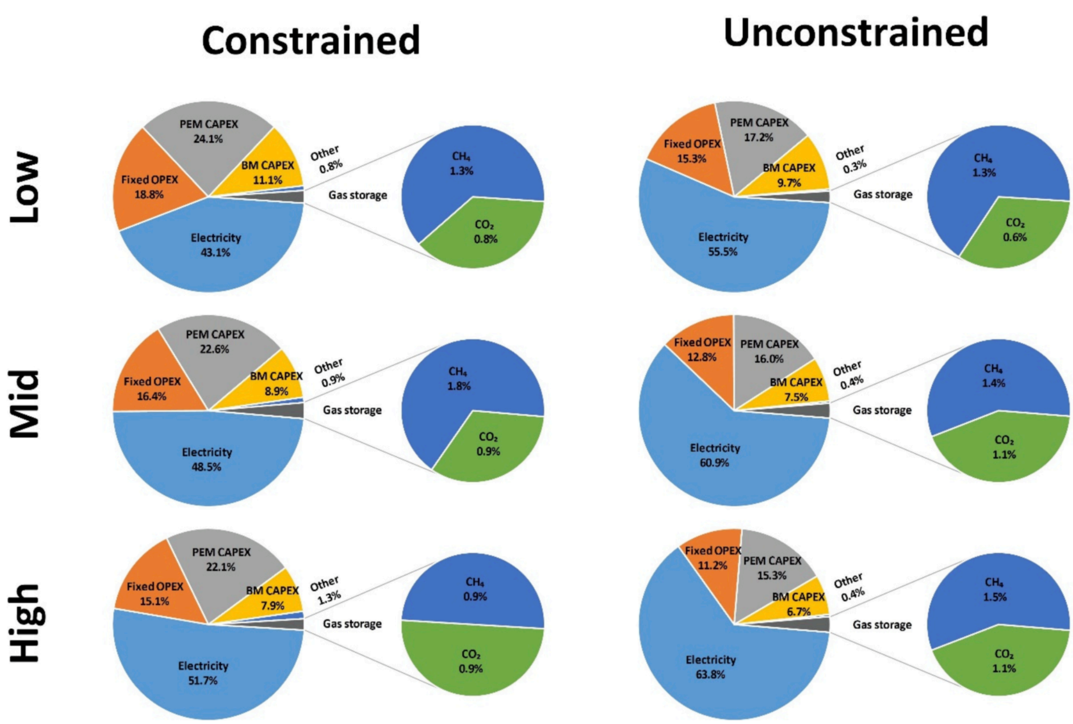

Fig. 4A. Breakdown of the GPC for all PtG scenarios assessed.

Appendix 5:. Energy balance of PtG plant according to the two power availability scenarios.

Note:

- The energy balance is related to the power obtained both from the spot- and regulated market.
- Unconverted power is the electricity used during electrolyser standby and for safety infrastructure.
- H_2 wasted is the H_2 used during ramp-up of BM reactor.

Appendix 6:. Breakdown of TCO.

(See Fig. 5A, Fig. 6A).

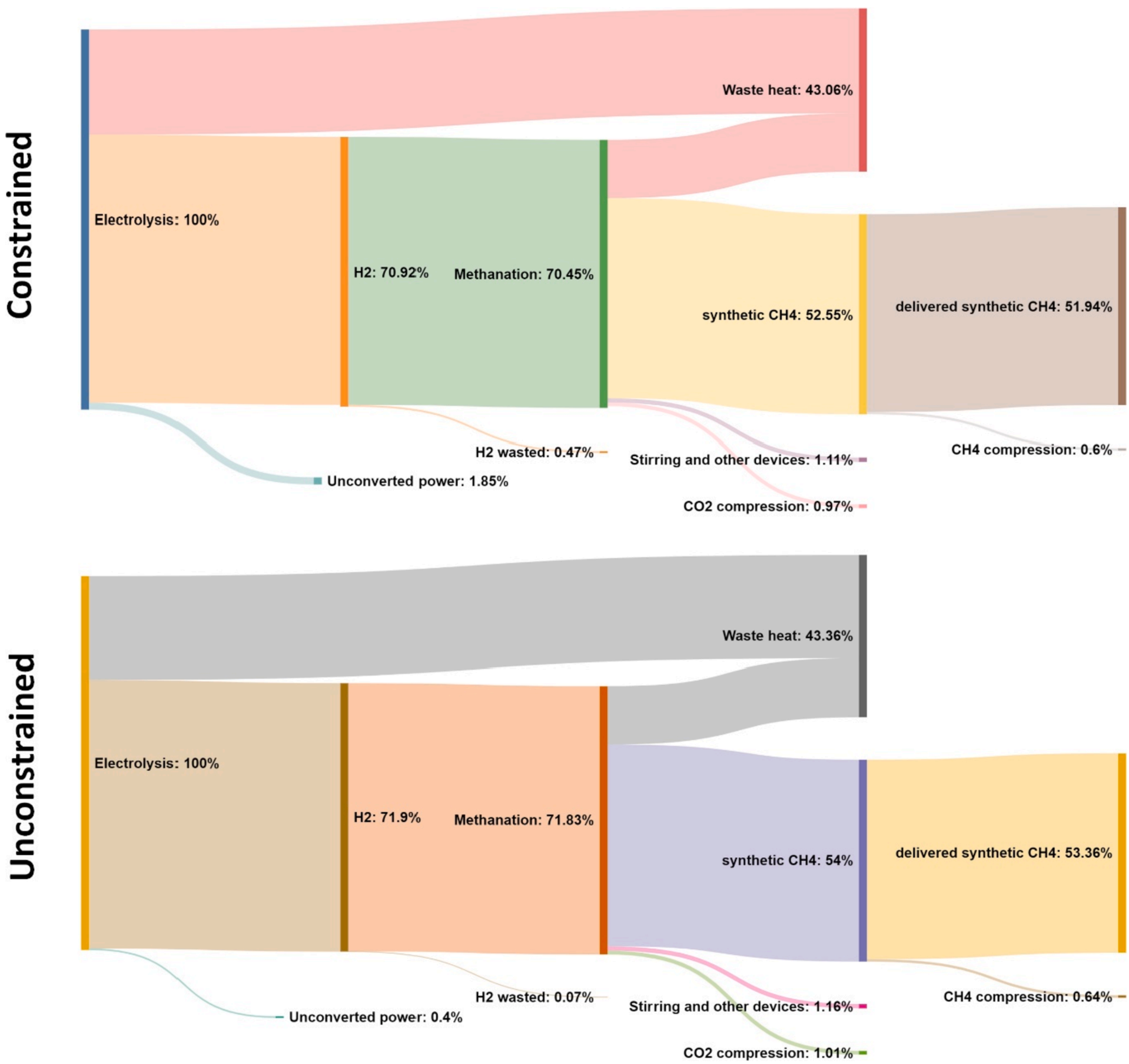

Fig. 5A. Sankey diagrams of the energy balance of the PtG plant according to constrained and unconstrained power availability scenarios.

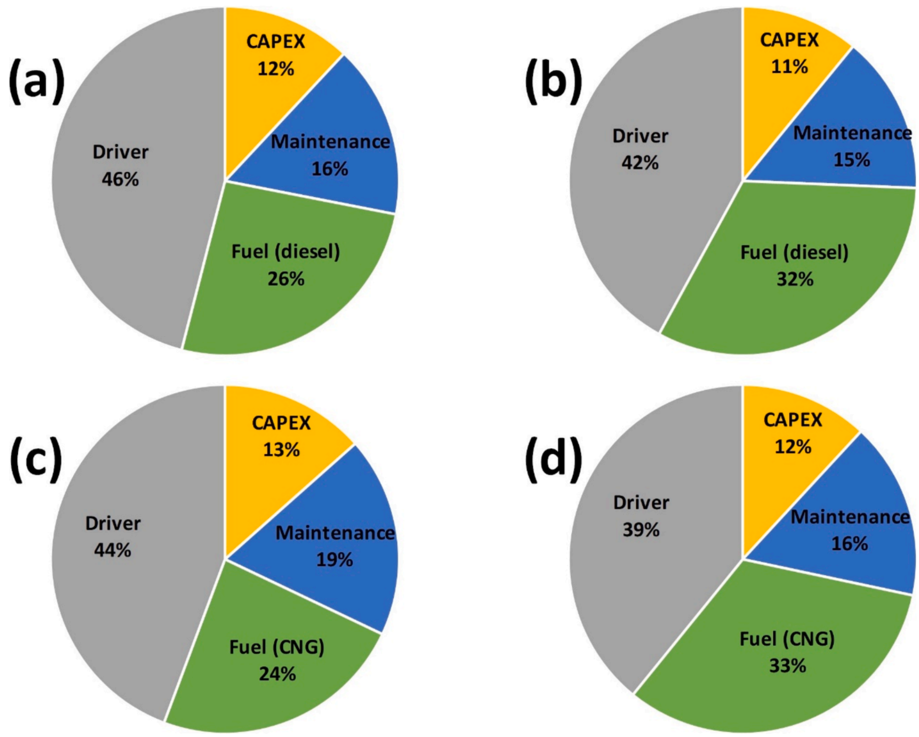

Fig. 6A. Breakdown of the TCO for the different types of buses running on fossil fuels. (a) diesel bus at low fuel price, (b) diesel bus at high fuel price, (c) CNG bus at low fuel price, and (d) CNG bus at high fuel price.

References

[1] Hanley ES, Deane JP, Gallachóir BPÓ. The role of hydrogen in low carbon energy futures–A review of existing perspectives. Renew Sustain Energy Rev 2018;82:3027–45. https://doi.org/10.1016/j.rser.2017.10.034.
[2] McDonagh S, Ahmed S, Desmond C, Murphy JD. Hydrogen from offshore wind: Investor perspective on the profitability of a hybrid system including for curtailment. Appl Energy 2020;265:114732. https://doi.org/10.1016/j.apenergy.2020.114732.
[3] Gorre J, Ortloff F, van Leeuwen C. Production costs for synthetic methane in 2030 and 2050 of an optimized Power-to-Gas plant with intermediate hydrogen storage. Appl Energy 2019;253:113594.
[4] Janke L, McDonagh S, Weinrich S, Nilsson D, Hansson PA, Nordberg Å. Techno-economic assessment of demand-driven small-scale green hydrogen production for low carbon agriculture in Sweden. Front Energy Res 2020;8:1–19. https://doi.org/10.3389/fenrg.2020.595224.
[5] Hannula I, Reiner DM. Near-term potential of biofuels, electrofuels, and battery electric vehicles in decarbonizing road transport. Joule 2019;3(10):2390–402. https://doi.org/10.1016/j.joule.2019.08.013.
[6] Gray N, McDonagh S, O'Shea R, Smyth B, Murphy JD. Decarbonising ships, planes and trucks: an analysis of suitable low-carbon fuels for the maritime, aviation and haulage sectors. Adv Appl Energy 2021;1:100008. https://doi.org/10.1016/j.adapen.2021.100008.
[7] IEA. Energy Technology Perspectives 2020 - Special Report on Carbon Capture Utilisation and Storage. 2020. https://doi.org/10.1787/9789264109834-en.
[8] Angelidaki I, Treu L, Tsapekos P, Luo G, Campanaro S, Wenzel H, et al. Biogas upgrading and utilization: current status and perspectives. Biotechnol Adv 2018;36(2):452–66. https://doi.org/10.1016/j.biotechadv.2018.01.011.
[9] Götz M, Lefebvre J, Mörs F, McDaniel Koch A, Graf F, Bajohr S, et al. Renewable Power-to-Gas: a technological and economic review. Renewable Energy 2016;85:1371–90. https://doi.org/10.1016/j.renene.2015.07.066.
[10] O'Shea R, Wall DM, McDonagh S, Murphy JD. The potential of power to gas to provide green gas utilising existing CO2 sources from industries, distilleries and wastewater treatment facilities. Renewable Energy 2017;114:1090–100. https://doi.org/10.1016/j.renene.2017.07.097.
[11] Morgenthaler S, Ball C, Koj JC, Kuckshinrichs W, Witthaut D. Site-dependent levelized cost assessment for fully renewable Power-to-Methane systems. Energy Convers Manage 2020;223:113150. https://doi.org/10.1016/j.enconman.2020.113150.
[12] Bedoić R, Dorotić H, Schneider DR, Čuček L, Ćosić B, Pukšec T, et al. Synergy between feedstock gate fee and power-to-gas: an energy and economic analysis of renewable methane production in a biogas plant. Renewable Energy 2021;173:12–23. https://doi.org/10.1016/j.renene.2021.03.124.
[13] Michailos S, Walker M, Moody A, Poggio D, Pourkashanian M. A techno-economic assessment of implementing power-to-gas systems based on biomethanation in an operating waste water treatment plant. J Environ Chem Eng 2021;9(1):104735.
[14] Vo TTQ, Wall DM, Ring D, Rajendran K, Murphy JD. Techno-economic analysis of biogas upgrading via amine scrubber, carbon capture and ex-situ methanation. Appl Energy 2018;212:1191–202. https://doi.org/10.1016/j.apenergy.2017.12.099.
[15] Gorre J, Ruoss F, Karjunen H, Schaffert J, Tynjälä T. Cost benefits of optimizing hydrogen storage and methanation capacities for Power-to-Gas plants in dynamic operation. Appl Energy 2020;257:113967. https://doi.org/10.1016/j.apenergy.2019.113967.
[16] Inkeri E, Tynjälä T, Karjunen H. Significance of methanation reactor dynamics on the annual efficiency of power-to-gas -system. Renewable Energy 2021;163:1113–26. https://doi.org/10.1016/j.renene.2020.09.029.
[17] Janke L, McDonagh S, Weinrich S, Murphy J, Nilsson D, Hansson P-A, et al. Optimizing power-to-H2 participation in the Nord Pool electricity market: Effects of different bidding strategies on plant operation. Renewable Energy 2020;156:820–36. https://doi.org/10.1016/j.renene.2020.04.080.
[18] Lardon L. Biocat project: A commercial-scale power-to gas facility and its capabilities to provide energy storage services to the Danish energy system. Available in: https://rekk.hu/downloads/events/Laurent%20Lardon%20Electrochae%20Biocat%20project.pdf 2019.
[19] McDonagh S, O'Shea R, Wall DM, Deane JP, Murphy JD. Modelling of a power-to-gas system to predict the levelised cost of energy of an advanced renewable gaseous transport fuel. Appl Energy 2018;215:444–56. https://doi.org/10.1016/j.apenergy.2018.02.019.
[20] Bucy J de, Lacroix O, Jammes L. The Potential of Power-To-Gas. vol. 33. ENEA Consulting; 2016.
[21] Schmidt O, Gambhir A, Staffell I, Hawkes A, Nelson J, Few S. Future cost and performance of water electrolysis: An expert elicitation study. Int J Hydrogen Energy 2017;42(52):30470–92. https://doi.org/10.1016/j.ijhydene.2017.10.045.
[22] Buttler A, Spliethoff H. Current status of water electrolysis for energy storage, grid balancing and sector coupling via power-to-gas and power-to-liquids: a review. Renew Sustain Energy Rev 2018;82:2440–54. https://doi.org/10.1016/j.rser.2017.09.003.
[23] Bauer F, Hulteberg C, Persson T, Tamm D. Biogas upgrading – Review of commercial technologies. SGC -Svenskt Gasleknisk Center AB 2013.
[24] Rusmanis D, O'Shea R, Wall DM, Murphy JD. Biological hydrogen methanation systems–an overview of design and efficiency. Bioengineered 2019;10(1):604–34. https://doi.org/10.1080/21655979.2019.1684607.
[25] Burkhardt M, Koschack T, Busch G. Biocatalytic methanation of hydrogen and carbon dioxide in an anaerobic three-phase system. Bioresour Technol 2015;178:330–3. https://doi.org/10.1016/j.biortech.2014.08.023.
[26] Nord Pool. Historical Market Data. Available in: https://www.nordpoolgroup.com/historical-market-data/. 2019.
[27] Grant Thorton. Renewable energy discount rate survey results - 2018. Available in: https://www.grantthornton.co.uk/globalassets/1.-member-firms/united-kingdom/pdf/documents/renewable-energy-discount-rate-survey-results-2018.pdf. United Kingdon: 2019.

[28] Frank E, Gorre J, Ruoss F, Friedl MJ. Calculation and analysis of efficiencies and annual performances of Power-to-Gas systems. Appl Energy 2018;218:217–31. https://doi.org/10.1016/j.apenergy.2018.02.105.
[29] IEA. The Future of Hydrogen: Seizing today's opportunities. International Energy Agency; 2019.
[30] McDonagh S, Wall DM, Deane P, Murphy JD. The effect of electricity markets, and renewable electricity penetration, on the levelised cost of energy of an advanced electro-fuel system incorporating carbon capture and utilisation. Renewable Energy 2019;131:364–71. https://doi.org/10.1016/j.renene.2018.07.058.
[31] Tsapekos P, Treu L, Campanaro S, Centurion VB, Zhu X, Peprah M, et al. Pilot-scale biomethanation in a trickle bed reactor: process performance and microbiome functional reconstruction. Energy Convers Manage 2021;244:114491. https://doi.org/10.1016/j.enconman.2021.114491.
[32] CNG Europe. Consumer price of compressed natural gas (CNG). Available in: http://cngeurope.com/. 2018.
[33] European Commission. Consumer price of petroleum products: automotive diesel oil. Available in: http://ec.europa.eu/energy/maps/maps_weekly_oil_bulletin/latest_Oil_Prices_DIE.pdf. Brussels: 2022.
[34] Ancona MA, Bianchi M, Branchini L, Catena F, De Pascale A, Melino F, et al. Numerical prediction of off-design performance for a Power-to-Gas system coupled with renewables. Energy Convers Manage 2020;210:112702. https://doi.org/10.1016/j.enconman.2020.112702.
[35] Grauers A, Borén S, Enerbäck O. Total cost of ownership model and significant cost parameters for the design of electric bus systems. Energies 2020;13(12):3262. https://doi.org/10.3390/en13123262.
[36] Hydrogen Valley, Solaris, Ballard, nel. Fuel Cell Electric Buses - An Attractive Value Proposition for Zero-Emission Buses in Scandinavia. 2017.
[37] Jones J, Genovese A, Tob-Ogu A. Hydrogen vehicles in urban logistics: a total cost of ownership analysis and some policy implications. Renew Sustain Energy Rev 2020;119:109595. https://doi.org/10.1016/j.rser.2019.109595.
[38] Government Offices of Sweden. Sweden's carbon tax. Available in: https://www.government.se/government-policy/swedens-carbon-tax/swedens-carbon-tax/. Stockholm: 2021.
[39] Frontier Economics. CO2 Emission Abatement Costs of Gas Mobility and Other Road Transport Options. 2021.
[40] European Parliament, Council of European Union. Directive (EU) 2018/2001 of 11 December 2018 on the promotion of the use of energy from renewable sources (recast). Official Journal of the European Union 2018.
[41] Zauner A, Böhm H, Rosenfeld DC, Tichler R. STORE&GO Report D7.7 - Analysis on future technology options and on techno-economic optimization. STORE&GO EU Project; 2019.