

Carbon recovery from biogas through upgrading and methanation: A techno-economic and environmental assessment

Original

Carbon recovery from biogas through upgrading and methanation: A techno-economic and environmental assessment / Minardi, M., Marocco, P., Gandiglio, M.. - In: JOURNAL OF CO2 UTILIZATION. - ISSN 2212-9820. - ELETTRONICO. - 78:(2023). [10.1016/j.jcou.2023.102632]

Availability:

This version is available at: 11583/2984333 since: 2023-12-04T10:58:53Z

Publisher:

Elsevier

Published

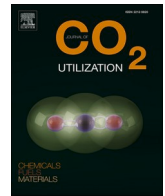
DOI:10.1016/j.jcou.2023.102632

Terms of use:

This article is made available under terms and conditions as specified in the corresponding bibliographic description in the repository

Publisher copyright

(Article begins on next page)



Carbon recovery from biogas through upgrading and methanation: A techno-economic and environmental assessment

Matteo Minardi, Paolo Marocco, Marta Gandiglio*

Department of Energy, Politecnico di Torino, Corso Duca degli Abruzzi 24, Torino 10129, Italy

ARTICLE INFO

Keywords:

Biogas
Carbon dioxide
Methanation
Biomethane
Hydrogen
System modelling

ABSTRACT

Reducing the use of fossil fuels is an essential measure to counteract the rise in greenhouse gas emissions. In this context, biofuels and e-fuels make an important contribution to achieving climate neutrality targets, especially if their distribution can take place within existing infrastructure, as in the case of methane.

The aim of this work is to carry out a techno-economic and environmental assessment of the combined production of biological and synthetic methane in a wastewater treatment plant (WWTP). Methane yield from biogas, usually associated only with biogas upgrading, is enhanced by recovering CO₂ to produce additional synthetic natural gas (SNG) through a methanation process. The analysis is applied to a medium-sized WWTP in Italy, whose biogas production profile is known throughout the year.

In the current scenario, SNG is not competitive on the gas market. The investment costs of the technologies and the electricity price are then varied in order to better investigate the profitability of SNG production. The results show that, considering long-term cost projections and an electricity price of about 50 €/MWh, SNG can become competitive, with a production cost of 1.4 €/Sm³. Finally, the environmental competitiveness of SNG (direct and indirect CO₂ emissions) with respect to fossil natural gas is investigated: results are shown as a function of the carbon intensity of grid electricity and the share of local renewable energy. To make SNG environmentally sustainable, the renewable share must increase to 46% or, alternatively, the carbon intensity of grid electricity must decrease to 187 g_{CO2eq}/kWh.

1. Introduction

The share of fossil fuels in the global energy mix has been consistently high for decades and accounted for around 80% in 2021 [1]. This has led to the release of massive amounts of greenhouse gases (GHG) emissions into the atmosphere, which are responsible for climate change. Policies and agreements – such as the Kyoto Protocol (1997), the Paris Agreement (2015) and the Glasgow Climate Pact (2021) – have been adopted to counteract this phenomenon.

In this context, bio- and e-fuels, when employed in place of fossil fuels, can contribute to the achievement of climate neutrality targets by avoiding GHG emissions [2]. Another compelling reason to invest in these fuels is that they are not only a clean energy source, but also a secure one, as they can be produced locally, which increases energy

security by reducing dependence on imported fossil fuels. Among alternative fuels, biomethane and synthetic methane are experiencing a strong momentum since they can be transported and exploited by means of the same gas infrastructure and end-use technologies already in place for natural gas. The European Union has recognised their potential as renewable fuels and has set targets for their production, as well as implemented some initiatives aimed at developing new technologies and increasing production, such as EU's Horizon 2020 research program (2014–2020), Horizon Europe (2020–2027), "Fit for 55" package (2021), BIOMETHAVERSE project (2022), REPowerEU plan (2022). According to the REPowerEU plan, biomethane production must reach a target value of 35 billion of cubic meter per year by 2030 [3]. Biogas and biomethane production in Europe has grown steadily over the last 10 years, reaching 4.5% of the European Union's gas consumption in 2021

Abbreviations: AD, Anaerobic digester; BG, Biogas; BNG, Biological natural gas; BT, Battery; EF, Emission factor; EL, Electrolyser; GHG, Greenhouse gas; HHV, Higher heating value; LCOP, Levelised cost of product; LHV, Lower heating value; MT, Methanation unit; NG, Natural gas; NTP, Normal temperature and pressure; NPC, Net present cost; O&M, Operation & maintenance; PEM, Proton exchange membrane; PV, Photovoltaic; RES, Renewable energy sources; SNG, Synthetic natural gas; SOC, State of Charge; UP, Upgrading unit; WWTP, Wastewater treatment plant.

* Corresponding author.

E-mail address: marta.gandiglio@polito.it (M. Gandiglio).

<https://doi.org/10.1016/j.jcou.2023.102632>

Received 17 July 2023; Received in revised form 17 November 2023; Accepted 20 November 2023

2212-9820/© 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

[4]. A good opportunity to increase their production comes from sewage sludge generated in wastewater treatment plants (WWTP) as part of the water cleaning process [5,6].

There are several articles in the literature on the production of biomethane and the methanation process, but usually these processes are considered as separate arguments.

Both Qyyum et al. [7] and Angelidaki et al. [8] provided a comprehensive overview of the available technologies and future prospects for biomethane production. Specifically, the first study focused on biogas production, cleaning technologies, current upgrading technologies and possible biomethane liquefaction technologies, while the second study showed the state-of-the-art for biogas upgrading with particular attention to the emerging biological methanation processes. Other authors also examined the environmental and economic aspects of the main upgrading solutions in a life cycle perspective. For example, Ardolino et al. [9] carried out a techno-economic and environmental analysis based on data obtained mostly from existing Italian plants and the results showed the complete sustainability of the upgrading process.

Many other studies only investigated the methanation process [10]. Hidalgo et al. [11] described current research and future trends on the methanation topic and also gave a general overview of power-to-methane plants in Europe. Other works focused more on the analysis of specific processes, comparing different innovative solutions and performing both a techno-economic and an environmental assessment [12–14]. In addition, recent studies have also focused on an innovative approach that involves the direct methanation of biogas. Gutiérrez-Martín et al. [15] explored three different configurations based on carbon dioxide (CO₂) methanation, direct biogas methanation and syngas methanation. Canu et al. [16] carried out a thermodynamic analysis of the process, while Gutiérrez-Martín et al. [17] also considered the economic side of the process and showed that the total cost of synthetic natural gas production is still high compared to natural gas.

Instead, there is little literature on the upgrading of biogas to biomethane and on the methanation of the biological CO₂ recovered from the upgrading section. Collet et al. [18] studied methane production from sewage sludge by combining anaerobic digestion and power-to-gas technology. They performed a techno-economic analysis and a life cycle assessment for different configurations of the process and considered different technologies. A similar work was proposed by Ghafoori et al. [19], who estimated the production costs of biomethane from landfill biogas in both a standard biogas upgrading plant and an innovative plant with CO₂ valorisation and methanation.

In this context, the present work aims at evaluating the combined production of biological and synthetic methane from sewage gas, with the objective of an almost complete recovery of the carbon content from biogas. The analysis relies on a real case study (WWTP) with an hourly biogas production profile over the year. Starting from this specific case study, the results are then generalised through a sensitivity analysis on both environmental (carbon intensity of grid electricity, renewables share) and economic (cost of the components) parameters to provide a comprehensive overview of the boundary conditions that make CO₂ methanation feasible from a techno-economic and environmental perspective, both in current and future scenarios.

The paper is structured as follows: Section 2 defines the case study and the available biogas hourly profile, while Section 3 describes the methodology for the techno-economic and environmental analysis and shows all the input data to the model. Results are presented and discussed in Section 4 and main conclusions are summarised in Section 5.

2. Case study description

The selected case study is a wastewater treatment plant (WWTP) located in the municipality of Collegno, near Turin, in Italy (Fig. 1). It is a medium-sized plant that serves around 200,000 equivalent inhabitants, treating an incoming wastewater inflow of 59,000 m³ per day [20] and producing about 500,000 Nm³ of biogas per year. The present



Fig. 1. Aerial view of the wastewater treatment plant located in Collegno (Italy) and managed by SMAT S.p.a.

work is based on the real hourly biogas production profile of the WWTP for the year 2019 (Fig. 2). The average biogas production rate is 57 Nm³/h, while peaks reach 145 Nm³/h during the winter season. The biogas flow rate follows a seasonal pattern throughout the year with a minimum value in summer (holiday season) due to lower availability of wastewater: this trend is typical for medium-sized WWTPs where most of the incoming wastewater is of residential origin [21]. In contrast, the biogas production is usually constant during the day.

3. Methodology

Biological and synthetic methane production in the WWTP is investigated from a techno-economic and environmental point of view, with a focus on the potential to increase methane yield by recovering the CO₂ waste stream from the upgrading process. Indeed, synthetic natural gas can be generated in a methanation reactor by combining the recovered CO₂ with green hydrogen (which is generated on site through an electrolyser powered by local renewable energy sources). In this way, the plant produces both biomethane from the upgrading process (biological natural gas, BNG) and synthetic methane (synthetic natural gas, SNG) from the methanation process.

3.1. Plant description

The layout of the energy system analysed in this work is shown in Fig. 3.

Biogas is produced from the anaerobic digestion of sewage sludge. The biogas processing plant can be divided into two sections: 1) the biomethane production section, which includes the upgrading unit, and 2) the SNG production section, which includes the methanation unit and the electrolyser to produce green hydrogen from the on-site photovoltaic (PV) system.

In the first section, biomethane is produced by converting the biogas in the upgrading unit, where the CO₂ is removed from the biogas and electrical energy is consumed. In the second section, much of the recovered CO₂ is used to produce synthetic methane by reacting it with H₂ in a methanation reactor. The hydrogen is generated by an electrolyser that is supplied with electricity from a photovoltaic system and the electrical grid. As shown in Fig. 3, no compression is required for the streams at the inlet of the methanation unit (i.e. CO₂ and H₂), as it is assumed that all components (upgrading, methanation reactor and electrolyser) operate under pressurised conditions (see Section 3.6.1) [10]. In addition, to better exploit the PV system and reduce the electrical energy imported from the grid, a battery storage system is also considered.

The grey blocks depicted in Fig. 3 includes all the auxiliary components required for the operation of the units (e.g. water pumping system, cooling circuit and separators for the electrolysis unit).

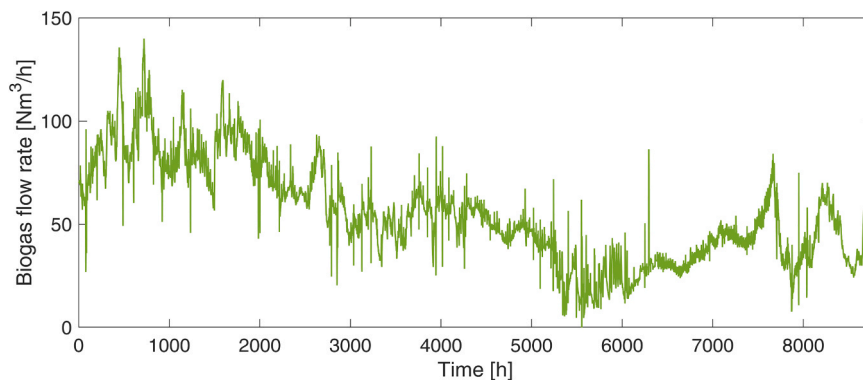


Fig. 2. Biogas flow rate (hourly resolution and time horizon of one year) for the chosen WWTP. Hour 0 corresponds to January 1st, 00:00. Data are referred to year 2019.

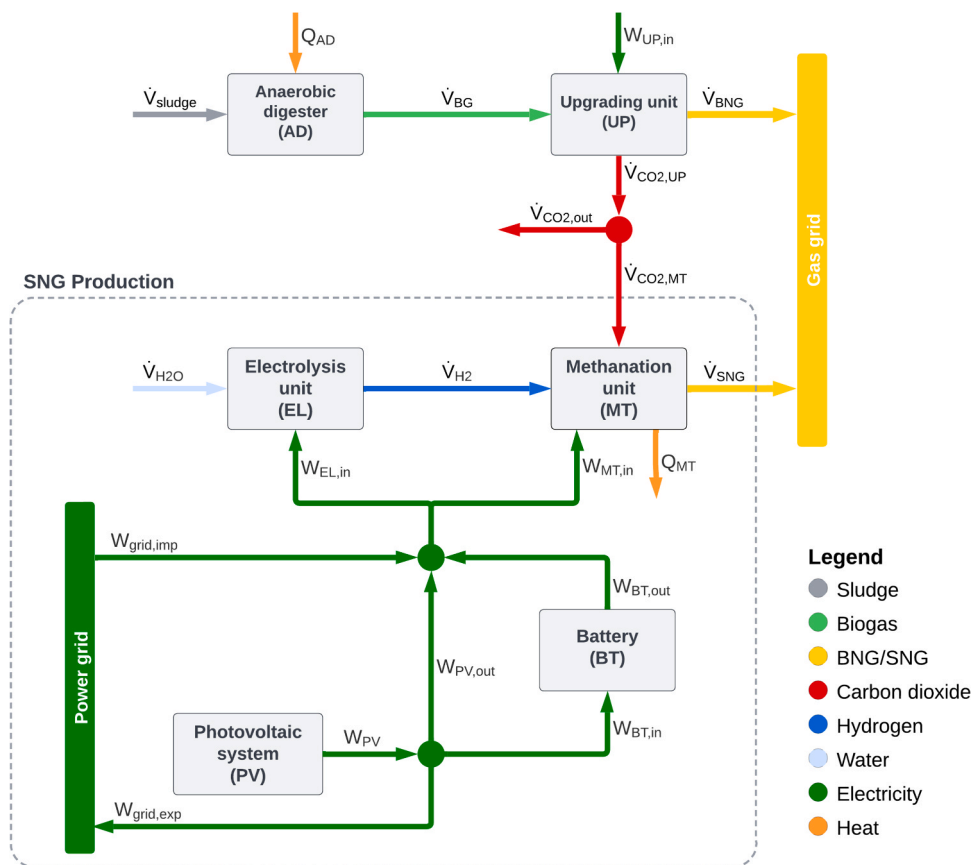


Fig. 3. Overall plant layout. The SNG production route is highlighted by the grey dashed line.

3.2. System modelling

The following section provides information on the modelling of the plant components. The main input data needed for the analysis can be found in Section 3.6.

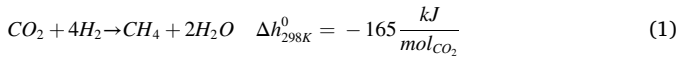
The present work is focused on the SNG production, which is highlighted by the grey dashed line in Fig. 3. The biogas upgrading section is taken into account to evaluate the available CO₂ stream (with hourly resolution) and the specific production cost of the BNG stream, but is not included in the techno-economic evaluation reported below, which indeed aims at assessing the SNG production cost. Anyway, details on the modelling of the anaerobic digester (AD) and the upgrading unit (UP) and on the evaluation of the BNG cost are shown in Appendix A.

It should be noted that the following sections are based on

volumetric flow rates. All volumetric flows and all the specific energy consumption values refer to normal temperature and pressure (NTP) conditions – i.e. at a temperature of 20 °C and a pressure of 1 atm – or, only for the SNG production cost, standard temperature and pressure (STP) conditions – i.e. temperature of 15 °C and a pressure of 1 atm [22].

3.2.1. Methanation unit

For this analysis, a catalytic methanation reactor is considered, specifically the one analysed in the EU project STORE&GO [23]. The main reaction that takes place is the hydrogenation of carbon dioxide [12]:



For the simulation, a stoichiometric ratio of 4 moles of H₂ to CO₂ is assumed [24]. The key technical parameters of the methanation unit are:

- the conversion rate of the inlet CO₂ ($y_{\text{CR},\text{CO}_2}$, in vol%), defined as the fraction of the inlet carbon dioxide to the methanation unit that is converted into CH₄.
- the specific electrical energy consumption of the methanation unit, which takes into account the electricity demand of the auxiliary components (w_{MT} , in kWh/Nm³_{SNG})
- the specific thermal energy that can be recovered from the methanation reactor (q_{MT} in kWh/Nm³_{SNG}) and exploited at a temperature of 184 °C [23].

As shown in Fig. 3, the CO₂ supplied to the methanation reactor ($\dot{V}_{\text{CO}_2,\text{MT}}$, in Nm³/h) is a fraction of the CO₂ produced by the upgrading system ($\dot{V}_{\text{CO}_2,\text{UP}}$). As discussed in Section 4.1, this fraction is chosen to maximise the CO₂ supplied to the methanation while keeping the SNG production cost close to its minimum value (i.e. without excessive oversizing of the electrolyser).

The amount of CO₂ entering the methanation unit that is converted into synthetic methane is defined by the conversion rate $y_{\text{CR},\text{CO}_2}$. The SNG stream leaving the methanation reaction (\dot{V}_{SNG} , in Nm³/h) is composed by the as-produced methane ($\dot{V}_{\text{SNG},\text{CH}_4}$, in Nm³/h), the unconverted CO₂ ($\dot{V}_{\text{SNG},\text{CO}_2}$, in Nm³/h) and the unconverted hydrogen ($\dot{V}_{\text{SNG},\text{H}_2}$, in Nm³/h), according to the following equations:

$$\dot{V}_{\text{SNG},\text{CH}_4}(t) = \dot{V}_{\text{CO}_2,\text{MT}}(t) \cdot y_{\text{CR},\text{CO}_2} \quad (2)$$

$$\dot{V}_{\text{SNG},\text{CO}_2}(t) = \dot{V}_{\text{CO}_2,\text{MT}}(t) \cdot (1 - y_{\text{CR},\text{CO}_2}) \quad (3)$$

$$\dot{V}_{\text{SNG},\text{H}_2}(t) = \dot{V}_{\text{SNG},\text{CO}_2}(t) \cdot 4 \quad (4)$$

$$\dot{V}_{\text{SNG}}(t) = \dot{V}_{\text{SNG},\text{CH}_4}(t) + \dot{V}_{\text{SNG},\text{CO}_2}(t) + \dot{V}_{\text{SNG},\text{H}_2}(t) \quad (5)$$

The electrical demand ($W_{\text{MT},\text{in}}$, in kW) and the thermal production (Q_{MT} , in kW) of the methanation unit are then evaluated based on the technical specifications of the reactor:

$$W_{\text{MT},\text{in}}(t) = \dot{V}_{\text{SNG}}(t) \cdot w_{\text{MT}} \quad (6)$$

$$Q_{\text{MT}}(t) = \dot{V}_{\text{SNG}}(t) \cdot q_{\text{MT}} \quad (7)$$

3.2.2. Electrolysis unit

A proton exchange membrane (PEM) electrolyser, which operates under pressurised conditions [25], is employed for the production of hydrogen. The fast dynamic response makes PEM electrolysers a suitable choice in case of fluctuating power supply (e.g. variable renewable energy sources) [26].

The specific electricity consumption of an electrolyser (w_{EL} , in kWh/Nm³) can be defined as the electrical power demand (of the stack and auxiliaries) per unit of hydrogen produced. This value depends on the operating point of the electrolyser. In the present work, a performance curve is therefore implemented within the model to express the specific electricity consumption of the electrolyser as a function of its operating power [27]. The electricity consumed by the electrolyser ($W_{\text{EL},\text{in}}$, in kW) can be then evaluated as:

$$W_{\text{EL},\text{in}}(t) = w_{\text{EL}} \cdot \dot{V}_{\text{H}_2}(t) \quad (8)$$

where \dot{V}_{H_2} (in Nm³/h) is the hydrogen flow rate produced for the selected operating point. The hydrogen required by the methanation section is derived from the stoichiometric ratio shown in Eq. (1) [24], according to the following expression:

$$\dot{V}_{\text{H}_2}(t) = 4 \cdot \dot{V}_{\text{CO}_2,\text{MT}}(t) \quad (9)$$

3.2.3. Battery storage unit

A Li-ion (lithium-ion) battery storage system is used to enhance the exploitation of the electrical energy generated by the PV plant. For each time step (t), the energy stored within the battery is evaluated by means of the following equation:

$$E_{\text{BT}}(t+1) = E_{\text{BT}}(t) + W_{\text{BT},\text{in}}(t) \cdot \Delta t \cdot \eta_{\text{BT},\text{ch}} - \frac{W_{\text{BT},\text{out}}(t) \cdot \Delta t}{\eta_{\text{BT},\text{dc}}} \quad (10)$$

where E_{BT} (in kWh) is the stored energy, $W_{\text{BT},\text{in}}$ (in kW) is the battery input power (charging), $W_{\text{BT},\text{out}}$ (in kW) the battery output power (discharging), $\eta_{\text{BT},\text{ch}}$ is the battery charging efficiency, $\eta_{\text{BT},\text{dc}}$ is the battery discharging efficiency and Δt is the duration of the time interval (1 h in this analysis).

To avoid premature battery degradation, the energy that can be stored in the battery is limited by a minimum and maximum state of charge ($\text{SOC}_{\text{BT},\text{min}}$ and $\text{SOC}_{\text{BT},\text{max}}$, respectively), which are expressed as a percentage of the rated capacity of the battery ($E_{\text{BT},\text{rated}}$, in kWh), as shown in Eq. (11).

$$E_{\text{BT},\text{rated}} \cdot \text{SOC}_{\text{BT},\text{min}} \leq E_{\text{BT}}(t) \leq E_{\text{BT},\text{rated}} \cdot \text{SOC}_{\text{BT},\text{max}} \quad (11)$$

3.2.4. Photovoltaic system

It is considered the installation of PV panels on the roofs of the WWTP buildings to provide renewable energy for the on-site production of green hydrogen.

First, the surface area of all roofs available for installing PV modules is determined. Then, using the PVGIS tool [28], the hourly production of the PV system (W_{PV} , in kW) is evaluated for the location under analysis (Collegno, Turin).

3.3. Energy management strategy

The energy simulation is performed over a reference year with an hourly resolution. For each time step, the input data are the biogas flow rate coming from the anaerobic digester and the hourly electrical energy produced by the photovoltaic system. The simulation starts with the biogas flow rate generated by the anaerobic digester. The biomethane produced and the CO₂ flow rate recovered through the upgrading process are then evaluated. Knowing the value of the CO₂ that is supplied to the SNG production section, the electrical energy demand of the electrolysis and methanation units are calculated.

It should be noted that, in the simulation, the PV and battery storage systems are only used to supply electricity to the electrolysis and methanation units, but not to the upgrading system. This choice is due to the aim of the work, which focuses on the SNG production section. The electricity demand is first supplied by the PV system, then by the battery (based on the available SOC) and finally withdrawn from the grid. A complete flowchart for the energy management strategy is shown in the [Supplementary Material \(S. 1\)](#).

3.4. Economic analysis

An economic analysis is carried out to understand the feasibility of the plant under investigation. Two economic indicators are employed to describe the economic performance of the energy system: the net present cost (NPC) and the levelised cost of product (LCOP). The analysis is presented here only for the SNG production section (see Fig. 3), as the focus of this work is to explore the economic and environmental performance of the SNG production route. However, in the Appendix A, the cost of the biological natural gas (BNG) from the upgrading unit is also evaluated for comparison with the SNG cost.

3.4.1. Definition of costs

The total investment cost (C_{inv} , in €), which occurs at the beginning of the analysis period, is given by the sum of the investment costs for all i -th components of the system (whose specific investment costs are reported in Table 2).

The annual net operating cost ($C_{op,t}$, in €/y), computed according to Eq. (12), can be divided into operation and maintenance costs ($C_{O\&M,i}$), replacement costs ($C_{rep,i}$), purchase of electricity from the grid ($C_{grid,imp}$) and incomes from the plant sub-products. The incomes include the revenues for the electricity sold to the grid ($R_{grid,exp}$) and the natural gas saving due to heat recovery within the WWTP (R_{heat}).

$$C_{op,t} = \sum_i (C_{O\&M,i} + C_{rep,i}) + C_{grid,imp} - (R_{grid,exp} + R_{heat}) \quad (12)$$

For each i -th component, the annual O&M costs ($C_{O\&M,i}$) are calculated as a percentage of its investment cost. Replacement costs ($C_{rep,i}$) are also computed as a percentage of the investment cost; they are due to the replacement of the electrolyser stack, the PV inverter and the battery modules over the plant lifetime. The $C_{grid,imp}$ term is due to the electricity imported from the grid ($W_{grid,imp}$) when the PV system and the battery storage cannot satisfy the total electrical demand. $W_{grid,imp}$ can be evaluated as:

$$W_{grid,imp}(t) = W_{EL,in}(t) + W_{MT,in}(t) - W_{PV,out}(t) - W_{BT,out}(t) \quad (13)$$

where $W_{EL,in}$ and $W_{MT,in}$ are the electrical power required by the electrolyser and methanation units, while $W_{PV,out}$ and $W_{BT,out}$ are the electrical power supplied by the PV and battery systems.

The revenues for the excess electricity ($R_{grid,exp}$) are estimated based on the amount PV electricity that is exported to the grid ($W_{grid,exp}$), which is defined according to the following expression:

$$W_{grid,exp}(t) = W_{PV}(t) - W_{PV,out}(t) - W_{BT,in}(t) \quad (14)$$

where W_{PV} is the total electrical power coming from the PV system, of which $W_{PV,out}$ is the share fed directly into the electrolyser and the methanation units and $W_{BT,in}$ is the share sent to the battery storage.

The savings (R_{heat}) for the heat recovered from the methanation section (computed through Eq. (7)) are calculated as the avoided natural gas that would have been required to produce the same amount of energy with a gas boiler (taking into account the efficiency of a traditional gas boiler). It is assumed that all recoverable heat is exploited within the WWTP, especially for the anaerobic digester heating, for which the thermal load is assessed according to the methodology shown in [29, 30].

3.4.2. Economic indicators

The final objective of the economic analysis is to evaluate two economic indicators: the net present cost of the system (NPC_{SNG} , in €) and the levelised cost of product ($LCOP_{SNG}$, in €/Sm³). The former is the present value of all the costs that the SNG production system incurs over its lifetime (minus the present value of all the revenues/savings) and is computed as follows:

$$NPC_{SNG} = C_{inv} + \sum_{t=1}^n \frac{C_{op,t}}{(1+d)^t} \quad (15)$$

where C_{inv} is the sum of the initial investment costs of all the components involved the system under analysis (i.e. for SNG production: PV system, battery storage, electrolyser, methanation unit, and heat recovery system), $C_{op,t}$ is net operating cost during the t -th year (derived through Eq. (12)), d (in %) is the discount rate, and n (in years) is the lifetime of the plant.

The $LCOP_{SNG}$ indicator represents the average cost per unit of the as-produced SNG. It is calculated as the ratio between the NPC_{SNG} indicator and the discounted sum of annual SNG production:

$$LCOP_{SNG} = \frac{NPC_{SNG}}{\sum_{t=1}^n \frac{V_{SNG,y}}{(1+d)^t}} \quad (16)$$

where $V_{SNG,y}$ (in Sm³/y) is the annual production of SNG. This term is obtained as the sum, over one year, of the hourly values derived from Eq. (5), converted from Nm³ to Sm³ (this conversion is done for comparison with the gas market price).

A similar approach is employed for the evaluation of the levelised cost of BNG, as discussed in Appendix A. A detailed investigation of the BNG production process is beyond the scope of this work but its production cost ($LCOP_{BNG}$) has been assessed and is presented in the Results section for comparison with the SNG route.

3.5. Environmental analysis

According to the Italian Ministerial Decree on biomethane [31], both biological and synthetic methane (if produced by the methanation of renewable hydrogen and CO₂ contained in the biogas) are considered as biofuels of biological origin and can access the available incentive. Furthermore, biogas is considered a near carbon-neutral energy source, as the carbon dioxide emitted during its combustion and use is equal to the amount of carbon absorbed during the growth of the biomass from which it was produced [32,33].

The environmental analysis is developed according to a Scope 2 approach that accounts for both direct (e.g. fossil fuels combustion) and indirect (e.g. purchase of grid electricity) emissions.

In order to evaluate the environmental sustainability of the SNG production route, the carbon ratio R_{CO_2} (-) is introduced to compare the emissions from the use of SNG and those from fossil natural gas (see Fig. 4). The carbon ratio can be defined as follows:

$$R_{CO_2} = \frac{M_{CO_2,SNG}}{M_{CO_2,NG}} \quad (17)$$

where $M_{CO_2,SNG}$ (in kgCO_{2eq}/y) is the CO₂ equivalent emissions associated with the SNG, from the waste CO₂ feeding the methanation unit to the use phase. This includes only the CO₂ equivalent emissions related to the electricity taken from the grid to power the electrolyser and the methanation units, since the CO₂ emitted during the combustion process has a net balance with the recovered one, which was already present in the atmosphere). The second term, $M_{CO_2,NG}$ (in kgCO_{2eq}/y), is the CO₂ equivalent emissions associated with fossil natural gas (i.e. upstream and combustion emissions related to an amount of fossil natural gas equal to the amount of SNG produced by the plant under analysis).

The $M_{CO_2,SNG}$ and $M_{CO_2,NG}$ terms (in kg) are computed according to Eqs. (18) and (19) respectively.

$$M_{CO_2,SNG} = \sum_{t=1}^{8760} W_{grid,imp}(t) \cdot \Delta t \cdot \varepsilon_{grid} \cdot 10^{-3} \quad (18)$$

$$M_{CO_2,NG} = \sum_{t=1}^{8760} \dot{V}_{SNG}(t) \cdot \Delta t \cdot \Delta h_{SNG} \cdot \varepsilon_{NG} \cdot 10^{-3} \quad (19)$$

where ε_{grid} (in gCO_{2eq}/kWh) is the carbon intensity of grid electricity, Δh_{SNG} (in MJ/Nm³) is the lower heating value of the synthetic natural gas (which depends on the fuel content of the produced SNG), ε_{NG} (in gCO_{2eq}/MJ) is the emission factor (upstream and combustion emissions) of the fossil natural gas, and Δt is the duration of the time interval (1 h in this analysis). The $W_{grid,imp}$ term (in kW) is estimated through Eq. (13), whereas \dot{V}_{SNG} (in Nm³/h) is derived according to Eq. (5).

A carbon ratio of 1 means that, when combusted, the bio-based SNG releases the same amount of CO₂ equivalent emissions as would be released by fossil natural gas. At values above 1, the SNG emits more CO₂ equivalent than fossil natural gas, making it unsustainable from an environmental point of view. On the contrary, for values below 1, the

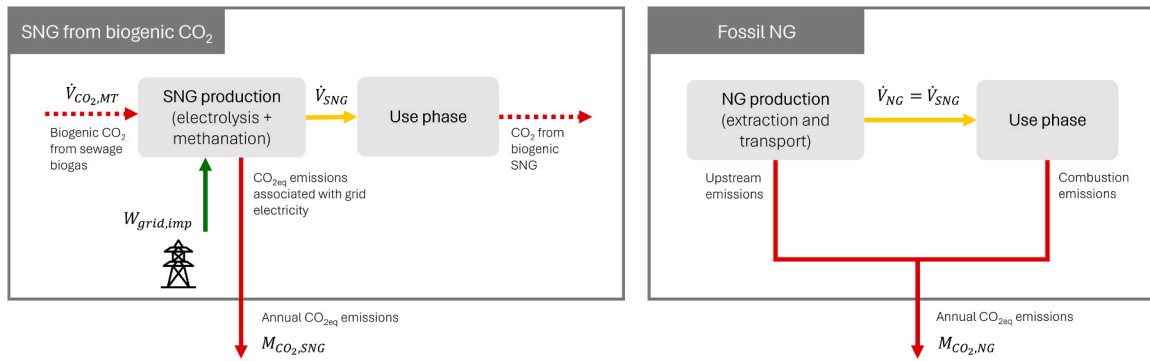


Fig. 4. Biogenic SNG and fossil NG control volumes employed for the environmental analysis. Dashed red lines are associated with the biogenic CO₂, which is not accounted in the carbon balance.

use of the SNG is responsible of a lower amount of CO₂ equivalent than the fossil alternative. As a limit scenario, a carbon ratio of zero corresponds to the case where the term $M_{CO_2,SNG}$ is zero. In this scenario, the electricity comes exclusively from renewable energy sources (local generation or guarantees of origin) and there are no CO₂ emissions (within the Scope 2 control volume) associated with the SNG.

It is noteworthy that the considerations derived from this analysis also apply to any waste CO₂ stream, further broadening the relevance and the applicability of the results.

3.6. Techno-economic and environmental input data

3.6.1. Technical data

The main technical input data are shown in Table 1.

For what concerns the PEM electrolyser, the specific electricity consumption (w_{EL}) is a function of the electrolyser operating point (which is expressed as the ratio between the inlet electrical power and the nominal power of the electrolyser). The w_{EL} term refers to the entire electrolyser system, including both the stack and the balance-of-plant components. The specific electricity consumption curve, retrieved from [34], shows a minimum of 3.9 kWh/Nm³ (LHV efficiency of 71%) for a load of 15%, while for higher loads the consumption increases to 4.7 kWh/Nm³ (LHV efficiency of 60%) at nominal load.

The chosen conversion rate for the methanation section is 99.5%: starting from an equilibrium concentration in the range 96%–98%, and employing multiple reactors in series (as shown in [35]), higher conversion rates (up to 99.5%–99.6%) can be achieved [10]. It is also

Table 1
Technical parameters for the energy system.

Parameter	Symbol	Value	Unit	Ref.
Methanation unit				
CO ₂ conversion rate	y_{CR,CO_2}	99.5	vol%	[10, 35]
Specific electrical energy consumption	w_{MT}	0.76	kWh/Nm ³ _{SNG}	[37]
Specific thermal energy recoverable from the methanation unit	q_{MT}	1.60	kWh/Nm ³ _{SNG} (at 184 °C)	[37]
Electrolysis unit				
Specific electrical energy consumption	w_{EL}	Efficiency curve	kWh/Nm ³ _{H₂}	[34]
Battery storage unit				
Charging efficiency	$\eta_{BT,ch}$	92	%	[39]
Discharging efficiency	$\eta_{BT,dc}$	92	%	[39]
Minimum SOC	$SOC_{BT,min}$	20	%	[39]
Maximum SOC	$SOC_{BT,max}$	100	%	[39]

supposed that the upgrading, methanation and electrolysis units operate at the same pressure (neglecting pressure drops in the components). A value of 14 bar is assumed, as it is consistent with the operating ranges of the various components reported in the literature [27,36,37].

The gas boiler efficiency (to estimate the savings from the recovered heat) is set equal to 95% [38].

3.6.2. Economic data

The values assumed for the main economic parameters are shown in Table 2. There is no cost for the CO₂ stream recovered from the upgrading unit, as it is a by-product of the upgrading process. The cost of the water for the electrolyser is also neglected, as clean water is available from the outlet stream of the WWTP. The price of energy vectors, such as the electricity imported from the grid, the excess PV electricity exported to the grid and the natural gas consumed by the plant, are estimated for the year 2019 for the Italian scenario (to avoid the price fluctuations of the 2020–2022 energy crisis). A sensitivity analysis is also performed to underline the effect of energy prices on the economic feasibility of the SNG production plant. The reference import prices of electricity and gas are retrieved from the Eurostat database and refer to

Table 2
Economic parameters for the energy system.

Parameter	Value	Unit	Ref.
General Assumptions			
Lifetime of the plant	20	years	Assumption
Discount rate	4.5	%	Assumption
Investment costs			
Electrolysis unit specific cost	1200	€/kW	[43]
Methanation unit specific cost	3138	€/kW _{SNG(HHV)}	[37]
Heat recovery system cost	5	% (of $C_{inv,MT}$)	Assumption
PV system specific cost (rooftop)	790	€/kW	[44]
Battery unit specific cost	355	€/kWh	[45]
Operating costs			
Electrolysis unit O&M cost (annual)	2.5	% (of $C_{inv,EL}$)	[46]
Methanation unit O&M cost (annual)	2.0	% (of $C_{inv,MT}$)	Assumption
PV system O&M cost (annual)	1.2	% (of $C_{inv,PV}$)	[44]
Battery unit O&M cost (annual)	2.5	% (of $C_{inv,BT}$)	[45]
Electrolyser stack replacement cost	40	% (of $C_{inv,EL}$)	[47]
Electrolyser stack lifetime	10	years	[47]
PV inverter replacement cost	20	% (of $C_{inv,PV}$)	[44]
PV inverter lifetime	12	years	[44]
Battery modules replacement	50	% (of $C_{inv,BT}$)	[38]
Battery modules lifetime	15	years	[48]
Energy prices			
Electricity import price (reference)	163.2	€/MWh	[41]
NG import price	36.0	€/MWh _(HHV)	[40]
Electricity export price	52.0	€/MWh	[49]

non-household consumers (excluding VAT and other recoverable taxes and levies) for Italy [40,41]. The revenues from the excess PV electricity sold to the grid are calculated assuming that the electricity is sold at a price equal to the average national electricity price (Prezzo Unico Nazionale, PUN) during 2019. The PUN indicator stands for the wholesale price of energy exchanged between producers and suppliers in the national market.

In order to explore the economic profitability of the SNG production route also in future scenarios, cost projections for the technologies are applied according to the data presented in Table 3. A reduction in the specific investment cost for the electrolysis, methanation and PV units are foreseen for the next decades (from 2030 to 3050). Regarding the methanation reactor, two cost projections are employed, as investigated during the STORE&GO project [42]: the first one (reference trend) shows the decrease in cost of a standard methanation reactor, while the second one (optimised trend) refers to an optimised reactor where improvements are assumed for what concerns gas compression, catalyst cost and required mass, and gas hourly space velocity.

3.6.3. Environmental data

The input data for the environmental analysis employs are the carbon intensity of the grid electricity (for Italy, 2021 [50]) and the emission factor of natural gas [51]. These values are both expressed as CO₂ equivalent emissions per unit of energy, as shown in Table 4. A sensitivity analysis on the electricity carbon intensity is also conducted (from 0 to 500 gCO_{2eq}/kWh) to evaluate its effect on the environmental performance of the analysed process.

4. Results

4.1. Energy assessment

The sizes and the results of the energy simulation are shown in Table 5. The nominal size of the solar PV system (920 kW) is defined so as to cover the entire available roof area with PV modules. The rated capacity of the battery is set at 2 MWh, as higher values do not bring any significant energy improvements. Specifically, the chosen battery size allows to maximise the electrical load covered by the PV, as indicated in the Supplementary Material (S.2).

The electrolyser size is chosen based on a sensitivity analysis, as show in Fig. 5 where the SNG production cost (namely, LCOP_{SNG}) and the processed CO₂ (i.e. the percentage fraction of the available CO₂ from the upgrading unit that is supplied to the methanation reactor) are displayed as a function of the electrolyser rated power.

A cost-optimal value (LCOP_{SNG} equal to 3.86 €/Sm³) is found for an electrolyser size of 290 kW. However, this point corresponds to a processed CO₂ of 71.6% (i.e. 28.4% of the available carbon dioxide from the upgrading unit is not sent to the methanation unit and thus not converted).

On the contrary, the CO₂-optimal point (with minimum cost) corresponds to an electrolyser size of 930 kW (which is able to capture all the carbon dioxide recovered from the upgrading unit) and leads to a production cost of 4.59 €/Sm³. In this scenario, 100% of the recovered

Table 3

Cost projections for 2030 and 2050 scenarios.

Parameter	Current	Short term	Long term	Unit	Ref.
Electrolysis unit specific cost	1200	900	320	€/kW	[43]
PV system specific cost	790	510	380	€/kW	[44]
Battery unit specific cost	355	192	156	€/kWh	[45]
Methanation unit specific cost (reference trend)	3138	2237	1269	€/kW _{SNG(HHV)}	[42]
Methanation unit specific cost (optimised trend)	1935	1379	782	€/kW _{SNG(HHV)}	[42]

Table 4

Environmental parameters for the energy system.

Parameter	Symbol	Value	Unit	Ref.
Carbon intensity of the electrical grid	ϵ_{grid}	234	gCO _{2eq} /kWh	[50]
Emission factor of natural gas	ϵ_{NG}	66	gCO _{2eq} /MJ	[51]

Table 5

Results of the sizing and energy simulation.

Parameter	Value	Unit
Plant design		
Electrolysis unit nominal size	550	kW _{el}
Methanation unit nominal size	0.3	MW _{SNG(HHV)}
PV system nominal size	920	kW _{el-DC}
Battery unit nominal size	2	MW _{h-el}
Energy simulation		
CO ₂ recovered from the upgrading unit	307	t/y
CO ₂ sent to the methanation unit	297	t/y
Processed CO ₂	96.6	%
BNG production	330	kNm ³ /y
SNG production	165	kNm ³ /y
Electricity demand of methanation and electrolysis unit	3031	MWh/y
PV annual production	1050	MWh/y
Electricity imported from the grid	2045	MWh/y
Electricity exported to the grid	32	MWh/y
Thermal energy recovered from the methanation unit	258	MWh/y
Thermal energy requested by the anaerobic digester	2824	MWh/y

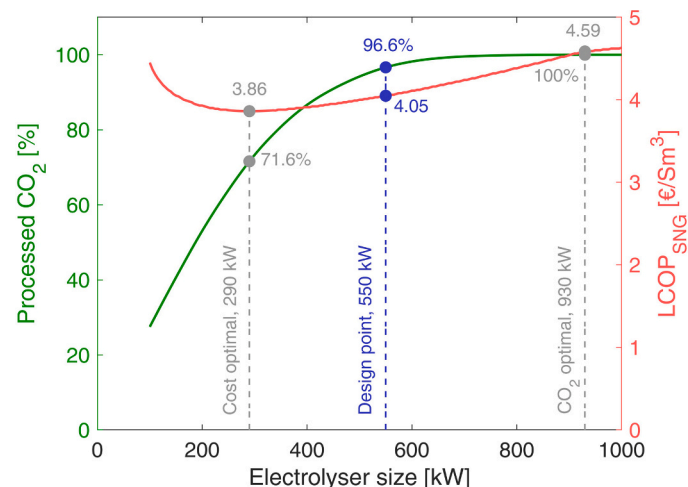


Fig. 5. SNG production cost (LCOP_{SNG}) and processed CO₂ as a function of the electrolyser size. The “CO₂ processed” term is the percentage fraction of the available CO₂ (i.e. recovered from the upgrading unit) that is sent to the methanation reactor.

CO₂ is fed to the methanation unit to be processed.

The chosen design point (blue dots in the figure) is a trade-off between the cost-optimal and the CO₂-optimal solutions and it is associated with an electrolyser size of 550 kW. In this configuration, the production cost is equal to 4.05 €/Sm³ (+4.8% compared to the minimum value) and the processed CO₂ is 96.6% (i.e. only 3.4% of the available CO₂ is not sent to the methanation unit). The goal of the chosen sizing approach is to convert as much CO₂ as possible while avoiding too large system oversizing (which would lead to an increase in the final SNG production cost).

Details on the energy simulation profiles during the year can be found in the Supplementary Material (S.3).

With this configuration (electrolyser size equal to 550 kW), the plant produces 330 kNm³/y of biological natural gas (BNG) and 165 kNm³/y

of synthetic natural gas (SNG), through the recovery of 297 t/y of CO₂ from the upgrading unit. The electrical energy required for the electrolysis and methanation unit is 3031 MWh/y, of which 67.5% is imported from the grid and the remaining 32.5% comes from the PV system (with support from battery storage system). The PV plant produces 1050 MWh/y, of which 97% is used for the electricity needs of the SNG production section and the remaining 32 MWh/y are exported to the grid. Finally, a heat recovery system enables to recover 258 MWh/y from the methanation unit, which can be used to meet about 9.1% of the thermal energy demand associated with the anaerobic digester.

4.2. Economic assessment

Fig. 6 shows the net present cost (NPC) and the breakdown of investment and operating terms for the current scenario and for the scenarios with the future cost projections. The results are evaluated here considering the reference electricity import price of 163.2 €/MWh (Table 2) and the reference trend for the methanation unit (Table 3). The economic analysis shows that the NPC amounts to 8.54 M€ in the current scenario (Fig. 6a). Specifically, operating costs account for the largest share (63.7%) and are dominated by the electricity withdrawn from the grid (Fig. 6c). Investment costs (Fig. 6b) make up for the remaining 36.3% and consist of the cost of the methanation reactor (30.9%), followed by the PV plant and the battery storage (23.4% and 22.9% respectively), the electrolyser (21.3%) and finally the heat recovery unit (1.5%).

Considering the future projections for the investment costs of the technologies (as defined in Table 3), the NPC could decrease by 16.9% (7.10 M€) in the short term and by 36.7% (5.94 M€) in the long term. As shown by the blue areas in Fig. 6a, the share of investment costs in the total NPC decreases in the future scenarios from 36.3% (current) to 29.1% (short term) and 20.9% (long term). The methanation unit is always the most expensive component and accounts for 31%–33% of the investment costs in all scenarios.

It is worth noting that the operating costs are the largest contributor to the NPC, mainly due to the costs associated with the large amount of electricity imported from the grid for the electrolysis process (“Energy” bar in Figure c). Their influence increases in the future scenarios due to the decrease in investment costs and accounts for 63.7%, 70.9% and 79.1% of the total NPC in the current, short-term and long-term scenarios, respectively (green areas in Fig. 6a).

The resulting value for LCOP_{SNG} is 4.05 €/Sm³ in the current scenario and decreases to 3.36 €/Sm³ and 2.72 €/Sm³ in the short- and long-term scenarios, respectively.

The NPC analysis discussed so far has highlighted the key role of operating costs, and in particular the electricity contribution, in the economic feasibility of SNG production. To better investigate this aspect, a sensitivity analysis on the price of electricity is then carried out, increasing it up to 350 €/MWh. Fig. 7 shows how the LCOP_{SNG} changes in the current scenario as a function of the electricity import price. It can be observed that the green area labelled “Energy” (which is dominated by the cost of the electricity purchased from the grid) decreases as the price of electricity decreases. Even at low electricity prices of about 50 €/MWh (which are representative for photovoltaic or wind power plants [52]), the LCOP_{SNG} is not economically competitive within the existing gas market, reaching a value of 2.62 €/Sm³. The EU gas market exhibited values above 1 €/Sm³ only during the 2022 energy crisis (with peaks up to about 3.3 €/Sm³ in August 2022) and the average price is usually below 1 €/Sm³ (e.g. average values of about 0.4–0.5 €/Sm³ in Q1–Q2 2023) [53]. As can be seen in Fig. 7, even if the price of electricity falls, there is still an important cost share (dominated by the investment costs), which makes the SNG production not competitive.

The production cost of biological natural gas (LCOP_{BNG}) is also shown in the graph (orange dashed line) and ranges from 0.24 to 0.35 €/Sm³ depending on the cost of the electricity. The current biological pathway of biomethane production is indeed already competitive within

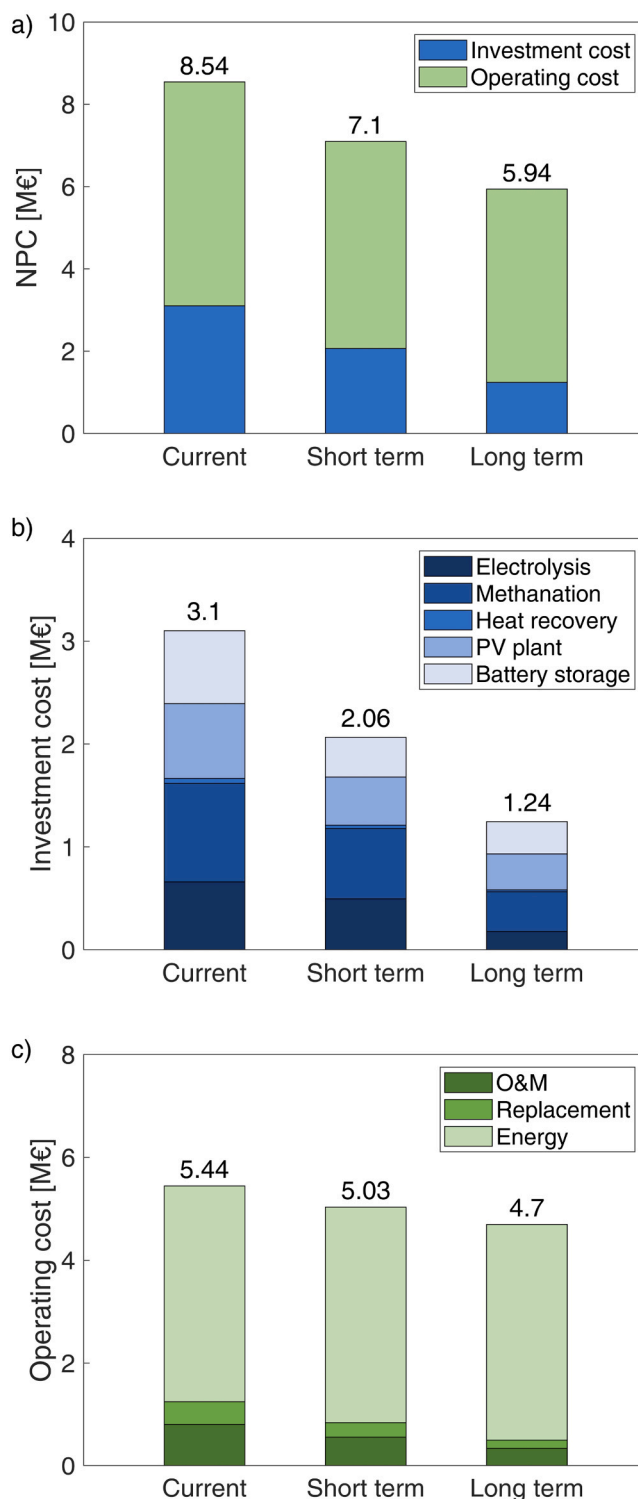


Fig. 6. NPC for the current, short-term and long-term cost projections (a), with contributions for the investment cost (b) and the operating costs (c). The “energy” term refers to the cost of imported electricity net of revenues and savings. A reference electricity import price of 163.2 €/MWh is considered.

the gas market, while more technological learning and incentives are needed to make SNG production viable. The costs shown in Fig. 7 are in line with those reported in literature for this type of plant. Böhm et al. [54] estimated the SNG cost to be between 3.14 and 8.37 €/Sm³, while Schlautmann et al. [55] found slightly lower values in the range 2.72–4.19 €/Sm³.

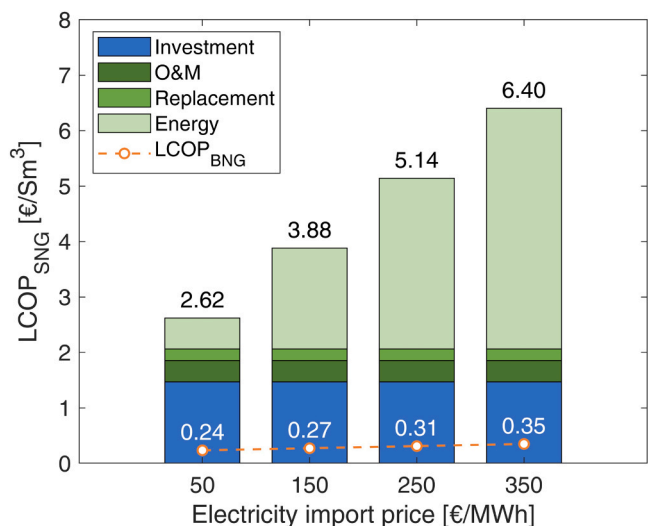


Fig. 7. Levelised cost of SNG (LCOP_{SNG}) as a function of the electricity import price (for the current scenario). The “energy” term refers to the cost of imported electricity (net of revenues and savings). The orange dashed line represents the cost for the production of BNG through the upgrading unit (LCOP_{BNG}).

The profitability of SNG production was also investigated for future cost projections (defined in Table 3) and is shown in Fig. 8. The graph compares the LCOP_{SNG} for the three scenarios (green lines) with the purchase price of biomethane in Italy in 2019 (orange dashed lines) and 2022 (yellow dashed lines) [56], both without incentives (darker colours, 0.17 €/Sm³ and 1.25 €/Sm³, respectively) and with incentives (lighter colours, 0.48 €/Sm³ and 1.56 €/Sm³, respectively). The incentives for biomethane injection into the natural gas grid, in Italy, are currently provided for both biological (from biogas upgrading) and synthetic (produced from biological CO₂) methane. According to [57], biofuels can be classified as “advanced” (usually from biological sub-products and waste streams) and “not advanced”, depending on the biomass source. The sewage biogas is included in the “advanced” biofuels. The incentives for grid injection consist of 375 € per CIC (“Certificati di Immissione in Consumo”), for a maximum period of 10 years. Each individual CIC is given for the injection into the national gas grid of a quantity of 5 Gcal of advanced biofuels (10 Gcal for not-advanced

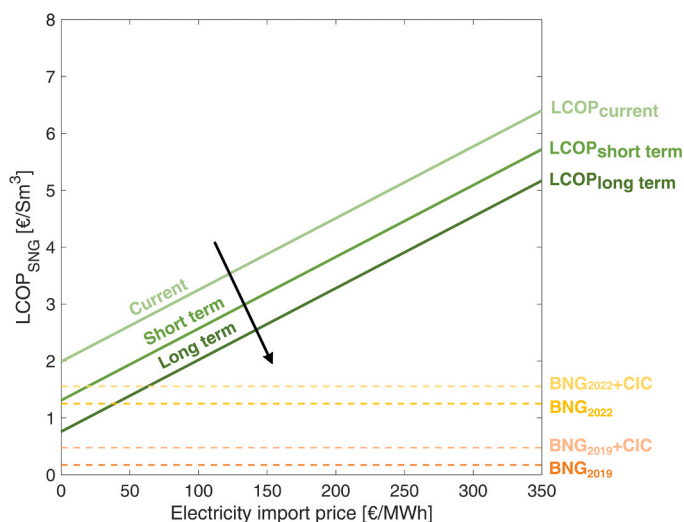


Fig. 8. Levelised cost of SNG (LCOP_{SNG}) as a function of electricity price for all cost scenarios (current, short-term and long-term). The dashed lines represent the market prices for biomethane in 2019 (orange) and 2022 (yellow), with and without incentives.

biofuels) [58].

The attractiveness of SNG production is improved by reducing the investment costs for the technologies. In particular, competitiveness is achieved in the long-term scenario, where LCOP_{SNG} reaches a value of 1.39 €/Sm³ with an electricity import price of 50 €/MWh. This value is close to the gas prices in 2022, while still above gas prices before the energy crisis (2019). Methane is indeed a relatively cheap commodity, which makes it difficult for SNG to become economically convenient. Fig. 8 also shows that at very low electricity prices (below about 20 €/MWh), the SNG production cost could reach values similar to 2022 gas prices.

Even in future scenarios, the SNG costs are in line with those reported in the literature by Böhm et al. [54], who obtained a value of 1.57 €/Sm³. In contrast, Schlautmann et al. [55] estimated a more optimistic value of 0.94 €/Sm³.

Since the methanation unit is the component that has the greatest impact on the total CAPEX, the impact of an improved methanation reactor design is also assessed (i.e. optimised trend in Table 3). The results of the optimised trend and the comparison with the reference trend are included in the Supplementary Material (S.4). It is found that the cost projections of the optimised trend have a rather negligible impact on the economic assessment compared to the reference trend.

It should also be noted that no specific subsidy for CO₂ recovery is considered in this analysis, which would further improve the attractiveness of the SNG production process.

4.3. Environmental assessment

The results of the environmental analysis are summarised in Fig. 9. The map shows the value of the carbon ratio (R_{CO_2} , defined in Eq. (17)) as a function of the carbon intensity of the grid electricity (in g_{CO₂eq}/kWh) and the RES share (in %). The latter term is defined as the fraction of total electricity consumption of the SNG production process that is covered by local RES (for which the carbon intensity is zero).

The white point in the map refers to the analysed case study: the carbon intensity of the electrical grid in Italy is 234 g_{CO₂eq}/kWh [49] and the share of electricity covered by the on-site PV plant is 32.5%, as discussed in Section 4.1. The resulting carbon ratio for this case study is 1.25. This means that the SNG production process, if supplied with the on-site PV system (32.5%) and the electrical grid (67.5%), would result in more CO₂ emissions than those released by using the same amount of

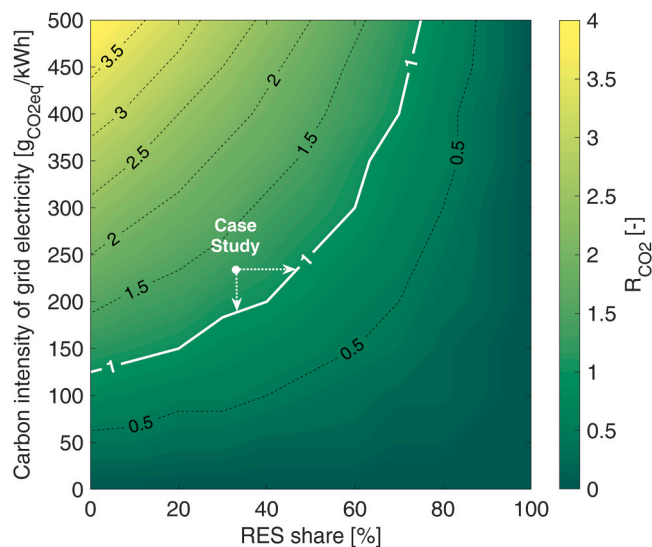


Fig. 9. Map of carbon ratio (R_{CO_2}) for different values of RES share and carbon intensity of grid electricity. “Case study” refers to the studied WWTP in Italy (carbon intensity of 234 g_{CO₂eq}/kWh and RES share of 33%).

fossil natural gas (+25%). To achieve a carbon ratio of less than 1 (i.e. the SNG-based route leads to a lower amount of CO₂ equivalent than the fossil alternative), the RES share must be higher than 46% (horizontal dashed arrow) or, alternatively, the carbon intensity of electricity must decrease to 187 g_{CO₂eq}/kWh (vertical dashed arrow).

Overall, the map shows the combinations of electricity carbon intensity and RES share that make the SNG production process environmentally sustainable ($R_{CO_2} < 1$) compared to fossil NG. It should be noted that, if only electricity from the grid is employed (RES share equal to zero), a carbon ratio lower than 1 is achieved for a carbon intensity of grid electricity below 127 g_{CO₂eq}/kWh.

5. Conclusions

The aim of this study is to investigate the techno-economic and environmental feasibility of the combined production of biological (BNG) and synthetic (SNG) methane in a wastewater treatment plant (WWTP).

The energy simulation shows that it is possible to increase the total production of methane within the plant by 50%: from 330 kNm³/y when only biomethane is produced through upgrading, to 495 kNm³/y by adding 165 kNm³/y of SNG, obtained from methanation of the recovered CO₂.

The economic analysis reveals that, in the current scenario, the operating costs account for the largest share (63.7%), primarily due to the electricity drawn from the grid for the electrolysis process. The remaining 36.3% of the total expenses are attributed to investment costs, which consists in the methanation reactor (30.9%), the PV plant and the battery storage (23.4% and 22.9% respectively), the electrolyser (21.3%) and finally the heat recovery unit (1.5%).

The levelised cost of synthetic natural gas (LCOP_{SNG}) is found to be 4.05 €/Sm³ in the current scenario. However, with future cost projections for the investment costs of the technologies, the LCOP_{SNG} can decrease to 3.36 €/Sm³ and 2.72 €/Sm³ in the short- and long-term scenarios, respectively. This study also underlines the importance of the electricity price for the economic feasibility of SNG production. Even at low electricity prices (which are representative of electricity production from local renewable energy sources), the levelised cost of SNG cannot compete with the NG market price in the current scenario. However, with a future reduction in the cost of technologies and low electricity prices (of about 50 €/MWh), the production cost of SNG becomes competitive, reaching a value of 1.39 €/Sm³ in the long-term scenario.

Regarding the environmental analysis, a carbon ratio (R_{CO_2}) of 1.25 is computed for the selected case study. This means that, when operated with local PV (sized according to the available roof area) and grid electricity, the SNG-based route results in 25% more CO₂ equivalent

emissions than the fossil alternative. To make the SNG environmentally sustainable compared to fossil NG (i.e. a carbon ratio below 1), the RES share must be above 46% or, alternatively, the carbon intensity of grid electricity must drop to 187 g_{CO₂eq}/kWh.

In summary, this study investigates the potential of combined biological and synthetic methane production from sewage biogas. While the current production costs of SNG cannot compete with natural gas, future cost projections and favourable electricity prices could make SNG production economically viable. Moreover, achieving a beneficial carbon footprint in the selected case study requires a higher RES share or a reduction in the carbon intensity of grid electricity. Future works will explore further case studies (e.g. other biogas sources) and the implementation of high temperature electrolysis to verify the advantages of a different technology for the hydrogen production process and its possible thermal integration into the biogas plant.

CRedit authorship contribution statement

Matteo Minardi: Conceptualization, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing, Software, Visualization. **Paolo Marocco:** Conceptualization, Methodology, Supervision, Writing – original draft, Writing – review & editing, Visualization. **Marta Gandiglio:** Conceptualization, Methodology, Supervision, Visualization, Writing – original draft, Writing – review & editing, Resources, Funding acquisition, Project administration.

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.

Data Availability

Data will be made available on request.

Acknowledgments

Project funded under the National Recovery and Resilience Plan (NRRP), Mission 4, Component 2 Investment 1.3 - Call for tender No. 1561 of 11.10.2022 of Ministero dell'Università e della Ricerca (MUR); funded by the European Union – NextGenerationEU. Project code PE0000021, Concession Decree No. 1561 of 11.10.2022 adopted by Ministero dell'Università e della Ricerca (MUR), CUP - to be indicated by each Beneficiary, according to attachment E of Decree No. 1561/2022, Project title "Network 4 Energy Sustainable Transition – NEST".

Appendix A

The following section shows the methodology and the input data for the modelling of the anaerobic digester (AD) and the biogas upgrading unit (UP).

The AD section is only investigated in terms of heat demand (following the methodology illustrated in [29,30] and validated for the specific case study) in order to verify the integration between the heat recovered from the methanation unit and the heat demand of the digester.

The modelling of the UP section is required to evaluate the CO₂ flow rate that can be recovered and the BNG production cost, but it is not included in the detailed techno-economic assessment of the SNG production route.

Anaerobic digester

The anaerobic digester is the component in which the sludge is stabilised, producing biogas and a solid biomass, called digestate. The organic matter remains in an anaerobic environment at a temperature of about 35–40 °C (mesophilic conditions) with an hydraulic retention time of about 20 days [59]. An external heating system is used to keep a constant temperature level inside the reactor [30]. Biogas can be considered as a sub-product of the anaerobic process in WWTPs since the main goal of the process is to stabilise the incoming sludges, which can then be used as secondary fuel in furnaces or landfilled.

The molar composition of the inlet biogas is assumed to be 65% CH₄ and 35% CO₂ (average value for the selected WWTP [20,21,59,60]).

Upgrading unit

Membrane separation is used for the upgrading unit. It is currently the most widely diffused technology for CO₂ capture both in Italy and Europe [4]. In this work, the upgrading unit is modelled as a black-box into which the biogas enters and two streams (i.e. biomethane and CO₂) exit. The electrical energy consumption of the unit is also estimated (e.g. due to compressors). The following parameters are needed by the UP model:

- the methane recovery ratio (R_{UP,CH_4} , in vol%), i.e. the fraction of the methane contained in the inlet biogas stream that is recovered through the upgrading process.
- the CO₂ loss ($f_{BG,CO_2,loss}$, in %), i.e. the carbon dioxide contained in the inlet BG stream that is not separated from the methane. It is expressed as a percentage of the inlet carbon dioxide.
- the specific electrical energy consumption of the upgrading system (w_{UP} , in kWh/Nm³_{BG}).

By knowing the values of the technical specifications (see Table A1), the methane content in the BNG stream at the outlet of the upgrading unit (\dot{V}_{BNG,CH_4} , in Nm³/h) and the required input power to the upgrading unit (W_{UP} , in kW) can be evaluated from Eq. (A1) and Eq. (A2):

$$\dot{V}_{BNG,CH_4}(t) = \dot{V}_{BG,CH_4}(t) \cdot R_{UP,CH_4} \quad (A1)$$

$$W_{UP}(t) = \dot{V}_{BG}(t) \cdot w_{UP} \quad (A2)$$

where \dot{V}_{BG,CH_4} (in Nm³/h) is the methane content in the biogas stream entering the upgrading unit and \dot{V}_{BG} (in Nm³/h) is the overall biogas feeding the upgrading unit (whose profile is displayed in Fig. 2 for the case study under investigation). As shown in Table A1, the methane recovery ratio (R_{UP,CH_4}) for the methane contained in the inlet biogas stream is assumed to be 100%.

From the model of the upgrading unit, the flow rate of CO₂ exiting the upgrading unit ($\dot{V}_{CO_2,UP}$, in Nm³/h) can be expressed as follows:

$$\dot{V}_{CO_2,UP}(t) = \dot{V}_{BG,CO_2}(t) - \dot{V}_{CO_2,loss}(t) \quad (A3)$$

which can be rearranged as:

$$\dot{V}_{CO_2,UP}(t) = \dot{V}_{BG}(t) \cdot y_{BG,CO_2} \cdot (1 - f_{BG,CO_2,loss}) \quad (A4)$$

where y_{BG,CO_2} (in vol%) is the fraction of carbon dioxide in the inlet biogas stream and $f_{BG,CO_2,loss}$ (in %) is the CO₂ loss within the upgrading section.

The CO₂ stream that is not separated within the UP unit is then removed by a final purification system before the BNG is injected into the grid. The biomethane fed into the grid is then evaluated as:

$$\dot{V}_{BNG}(t) = \frac{\dot{V}_{BNG,CH_4}(t)}{y_{BNG,CH_4}} \quad (A5)$$

where y_{BNG,CH_4} is the methane content (vol%) in the BNG outlet stream after the final purification step, and it is equal to 100%.

Finally, the levelised cost of product for the BNG stream ($LCOP_{BNG}$, in €/Sm³) can be assessed as follows:

$$LCOP_{BNG} = \frac{NPC_{BNG}}{\sum_{t=1}^n V_{BNG,y} \cdot (1+d)^{-t}} \quad (A6)$$

where NPC_{BNG} (in €) is the net present cost of the biogas upgrading section and $V_{BNG,y}$ (in Sm³/y) is the annual production of BNG. This last term is evaluated as the sum, over 1 year, of the hourly BNG flow rate values (derived in Eq. (A5), and converted from Nm³ to Sm³). The NPC_{BNG} term includes the investment cost for the upgrading section, the annual operating costs (O&M) and the cost of the electricity supplied to the unit.

Table A1

Technical and economic input parameters for the UP unit.

Parameter	Symbol	Value	Unit	Ref.
Technical parameters				
Methane recovered from biogas	R_{UP,CH_4}	100	%	Assumption
Methane content in biogas	y_{BG,CH_4}	65	vol%	[20,21,59,60]
Carbon dioxide content in biogas	y_{BG,CO_2}	35	vol%	[20,21,59,60]
Carbon dioxide loss in the UPG unit	$f_{BG,CO_2,loss}$	3.8	%	[36]
Methane content in BNG	y_{BNG,CH_4}	100	vol%	Assumption
Specific electrical energy consumption	w_{UP}	0.25	kWh/Nm ³ _{BG}	[36]
Economic parameters				
Upgrading unit specific cost	$C_{inv,UP}$	7500	€/Nm ³ _{BNG} /h)	[61]
O&M upgrading unit	$C_{op,UP,t}$	2	% $C_{inv,UP}$	Assumption
Electricity import price (reference)	$C_{grid,imp}$	163.2	€/MWh	[41]

Appendix B. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jcou.2023.102632](https://doi.org/10.1016/j.jcou.2023.102632).

References

- [1] IEA, World Energy Outlook 2022, Paris, 2022. (<https://www.iea.org/reports/world-energy-outlook-2022>).
- [2] EBA, The contribution of the biogas and biomethane industries to medium-term greenhouse gas reduction targets and climate-neutrality by 2050, Brussels, 2020. (<https://www.europeanbiogas.eu/the-contribution-of-the-biogas-and-biomethane-industries-to-medium-term-greenhouse-gas-reduction-targets-and-climate-neutrality-by-2050/#:~:text=Thebiogasandbiomethaneindustriessignificantandgrowingcontributors,em>).
- [3] European commission, REPowerEU Plan - COM/2022/230 final, (2022). (<https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=COM:2022:230:FIN>).
- [4] EBA, Statistical Report of the European Biogas Association 2022, Bruss, 2022. (https://www.europeanbiogas.eu/_trashed-3/).
- [5] N. Bachmann, Sustainable biogas production in municipal wastewater treatment plants - Technical brochure of IEA Bioenergy, 2015. (<https://www.ieabioenergy.com/publications/sustainable-biogas-production-in-municipal-wastewater-treatment-plants/>).
- [6] M. Gandiglio, A. Lanzini, A. Soto, P. Leone, M. Santarelli, Enhancing the energy efficiency of wastewater treatment plants through co-digestion and fuel cell systems, *Front. Environ. Sci.* 5 (2017), 70, <https://doi.org/10.3389/FENV.2017.00070>.
- [7] M.A. Qyum, J. Haider, K. Qadeer, V. Valentina, A. Khan, M. Yasin, M. Aslam, G. De Guido, L.A. Pellegrini, M. Lee, Biogas to liquefied biomethane: assessment of 3P's—production, processing, and prospects, *Renew. Sustain. Energy Rev.* 119 (2020), <https://doi.org/10.1016/J.RSER.2019.109561>.
- [8] I. Angelidaki, L. Treu, P. Tsapekos, G. Luo, S. Campanaro, H. Wenzel, P.G. Kougias, Biogas upgrading and utilization: current status and perspectives, *Biotechnol. Adv.* 36 (2018) 452–466, <https://doi.org/10.1016/J.BIOTECHADV.2018.01.011>.
- [9] F. Ardolino, G.F. Cardamone, F. Parrillo, U. Arena, Biogas-to-biomethane upgrading: a comparative review and assessment in a life cycle perspective, *Renew. Sustain. Energy Rev.* 139 (2021), <https://doi.org/10.1016/J.RSER.2020.110588>.
- [10] F. Salomone, P. Marocco, D. Ferrario, A. Lanzini, D. Fino, S. Bensaid, M. Santarelli, Process simulation and energy analysis of synthetic natural gas production from water electrolysis and CO₂ capture in a waste incinerator, *Appl. Energy* 343 (2023), 121200, <https://doi.org/10.1016/J.APENERGY.2023.121200>.
- [11] D. Hidalgo, J.M. Martín-Marroquín, Power-to-methane, coupling CO₂ capture with fuel production: an overview, *Renew. Sustain. Energy Rev.* 132 (2020), <https://doi.org/10.1016/J.RSER.2020.110057>.
- [12] F. Salomone, E. Giglio, D. Ferrero, M. Santarelli, R. Pirone, S. Bensaid, Techno-economic modelling of a power-to-gas system based on SOEC electrolysis and CO₂ methanation in a RES-based electric grid, *Chem. Eng. J.* 377 (2019), <https://doi.org/10.1016/J.CEJ.2018.10.170>.
- [13] Z. Uddin, B.Y. Yu, H.Y. Lee, Evaluation of alternative processes of CO₂ methanation: design, optimization, control, techno-economic and environmental analysis, *J. CO₂ Util.* 60 (2022), <https://doi.org/10.1016/J.JCOU.2022.101974>.
- [14] E. Giglio, A. Lanzini, M. Santarelli, P. Leone, Synthetic natural gas via integrated high-temperature electrolysis and methanation: part I-Energy performance, *J. Energy Storage* 1 (2015) 22–37, <https://doi.org/10.1016/J.JEST.2015.04.002>.
- [15] F. Gutiérrez-Martín, L.M. Rodríguez-Antón, Power-to-SNG technologies by hydrogenation of CO₂ and biomass resources: a comparative chemical engineering process analysis, *Int. J. Hydrog. Energy* (2019) 12544–12553, <https://doi.org/10.1016/J.IJHYDENE.2018.09.168>.
- [16] P. Canu, M. Pagin, Biogas upgrading by 2-steps methanation of its CO₂-thermodynamics analysis, *J. CO₂ Util.* 63 (2022), <https://doi.org/10.1016/J.JCOU.2022.102123>.
- [17] F. Gutiérrez-Martín, L.M. Rodríguez-Antón, M. Legrand, Renewable power-to-gas by direct catalytic methanation of biogas, *Renew. Energy* 162 (2020) 948–959, <https://doi.org/10.1016/J.RENENE.2020.08.090>.
- [18] P. Collet, E. Flottes, A. Favre, L. Raynal, H. Pierre, S. Capela, C. Peregrina, Techno-economic and life cycle assessment of methane production via biogas upgrading and power to gas technology, *Appl. Energy* 192 (2017) 282–295, <https://doi.org/10.1016/J.APENERGY.2016.08.181>.
- [19] M.S. Ghafouri, K. Loubar, M. Marin-Gallego, M. Tazerout, Techno-economic and sensitivity analysis of biomethane production via landfill biogas upgrading and power-to-gas technology, *Energy* 239 (2022), <https://doi.org/10.1016/J.ENERGY.2021.122086>.
- [20] M. MosayebNezhad, A.S. Mehr, M. Gandiglio, A. Lanzini, M. Santarelli, Techno-economic assessment of biogas-fed CHP hybrid systems in a real wastewater treatment plant, *Appl. Therm. Eng.* 129 (2018) 1263–1280, <https://doi.org/10.1016/J.APPLTHERMALENG.2017.10.115>.
- [21] M. Gandiglio, A. Lanzini, M. Santarelli, M. Aciri, T. Hakala, M. Rautanen, Results from an industrial size biogas-fed SOFC plant (the DEMOSOFC project), *Int. J. Hydrog. Energy* 45 (2020) 5449–5464, <https://doi.org/10.1016/J.IJHYDENE.2019.08.022>.
- [22] STP - Standard Temperature and Pressure and NTP - Normal Temperature and Pressure, (2023). (https://www.engineeringtoolbox.com/stp-standard-ntp-normal-air-d_772.html) (accessed July 1, 2023).
- [23] Z.A. Van Leeuwen C., Innovative large-scale energy storage technologies and power-to-gas concepts after optimisation. Roadmap and policy recommendations for power-to-gas in the EU up to 2050, (2020).
- [24] C. Dannesboe, J.B. Hansen, I. Johansen, Catalytic methanation of CO₂ in biogas: experimental results from a reactor at full scale, *React. Chem. Eng.* 5 (2019) 183–189, <https://doi.org/10.1039/C9RE00351G>.
- [25] M. Götz, J. Lefebvre, F. Mörs, A. McDaniel Koch, F. Graf, S. Bajohr, R. Reimert, T. Kolb, Renewable power-to-gas: a technological and economic review, *Renew. Energy* 85 (2016) 1371–1390, <https://doi.org/10.1016/J.RENENE.2015.07.066>.
- [26] G. Correa, P. Marocco, P. Muñoz, T. Falaguerra, D. Ferrero, M. Santarelli, Pressurized PEM water electrolysis: dynamic modelling focusing on the cathode side, *Int. J. Hydrog. Energy* 47 (2022) 4315–4327, <https://doi.org/10.1016/J.IJHYDENE.2021.11.097>.
- [27] M. Santos, R. Rodriguez, S. Garcia Sigrid Damman, U. Johansen, Techno-economic analysis of wind-hydrogen integration (Haeolus Project, Deliverable 5.3), 2019. (<https://www.haeolus.eu/wp-content/uploads/2023/03/D5.3.pdf>).
- [28] European Commission Joint Research Centre, Photovoltaic Geographical Information System (PVGIS Online Tool), JRC. (2001). (https://joint-research-centre.ec.europa.eu/pvgis-online-tool_en) (accessed April 20, 2023).
- [29] A.S. Mehr, M. Gandiglio, M. MosayebNezhad, A. Lanzini, S.M. Mahmoudi, M. Yari, M. Santarelli, Solar-assisted integrated biogas solid oxide fuel cell (SOFC) installation in wastewater treatment plant: energy and economic analysis, *Appl. Energy* 191 (2017) 620–638.
- [30] M. Gandiglio, A. Saberi Mehr, M. MosayebNezhad, A. Lanzini, M. Santarelli, Solutions for improving the energy efficiency in wastewater treatment plants based on solid oxide fuel cell technology, *J. Clean. Prod.* 247 (2020), 119080, <https://doi.org/10.1016/J.JCLEPRO.2019.119080>.
- [31] Gestore dei Servizi Energetici (GSE), Biometano - DM 02/03/2018, (2023). (<https://www.gse.it/servizi-per-te/rinnovabili-per-i-trasporti/biometano>) (accessed July 1, 2023).
- [32] E. Giglio, G. Vitale, A. Lanzini, M. Santarelli, Integration between biomass gasification and high-temperature electrolysis for synthetic methane production, *Biomass-- Bioenergy* 148 (2021), <https://doi.org/10.1016/J.BIOMBIOE.2021.106017>.
- [33] M. Gandiglio, F. De Sario, A. Lanzini, S. Bobba, M. Santarelli, G.A. Blengini, Life cycle assessment of a biogas-fed solid oxide fuel cell (SOFC) integrated in a wastewater treatment plant, *Energies* (2019), <https://doi.org/10.3390/en12091611>.
- [34] G. Eduardo, S. Maider, Protocols for demonstration of fuel-production strategy. Deliverable 8.3. Haeolus Project, 2021. (<https://www.haeolus.eu/?p=1073>).
- [35] E. Giglio, F.A. Deorsola, M. Gruber, S.R. Harth, E.A. Morosan, D. Trimis, S. Bensaid, R. Pirone, Power-to-Gas through High Temperature Electrolysis and Carbon Dioxide Methanation: Reactor Design and Process Modeling, (2018). <https://doi.org/10.1021/acs.iecr.8b00477>.
- [36] F. Bauer, C. Hultberg, T. Persson, D. Tamm, Biogas upgrading - review of commercial technologies (SGC Rapport 2013:270), Malmö (2013). (<http://www.sgc.se/ckfinder/userfiles/files/SGC270.pdf>).
- [37] F. Graf, Innovative large-scale energy storage technologies and power-to-gas concepts after optimisation. Roadmap and policy recommendations for power-to-gas in the EU up to 2050, Bonn (2019). (https://www.storeandgo.info/fileadmin/downloads/publications/STORE_GO_power_to_gas_roadmap.pdf).
- [38] P. Marocco, M. Gandiglio, M. Santarelli, When SOFC-based cogeneration systems become convenient? A cost-optimal analysis, *Energy Rep.* 8 (2022) 8709–8721, <https://doi.org/10.1016/j.egy.2022.06.015>.
- [39] P. Marocco, D. Ferrero, M. Gandiglio, M.M. Ortiz, K. Sundseth, A. Lanzini, M. Santarelli, A study of the techno-economic feasibility of H₂-based energy storage systems in remote areas, *Energy Convers. Manag.* 211 (2020), 112768, <https://doi.org/10.1016/j.enconman.2020.112768>.
- [40] Eurostat - Data Explorer, Gas prices for non-household consumers - bi-annual data (from 2007 onwards), (2022). (http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=nrg_pc_203&lang=en) (accessed June 8, 2023).
- [41] Eurostat - Data Explorer, Electricity prices for non-household consumers - bi-annual data (from 2007 onwards), (2022). (http://appsso.eurostat.ec.europa.eu/nui/show.do?dataset=nrg_pc_205&lang=en) (accessed June 8, 2023).
- [42] F. Mörs, R. Schlautmann, J. Gorre, R. Leonhard, Final report on evaluation of technologies and processes. Deliverable 5.9 Store&Go Project, 2020. <https://www.storeandgo.info/publications/deliverables/index.html>.
- [43] IRENA, IRENA (2020), Green Hydrogen Cost Reduction: Scaling up Electrolysers to Meet the 1.50C Climate Goal, International Renewable Energy Agency, Abu Dhabi, 2020. (<https://www.irena.org/publications/2020/Dec/Green-hydrogen-cost-reduction>).
- [44] Danish Energy Agency, Technology Data for Generation of Electricity and District Heating, (2019). (<https://ens.dk/en/our-services/projections-and-models/technology-data/technology-data-generation-electricity-and>) (accessed June 1, 2023).
- [45] W. Cole, A.W. Frazier, Cost Projections for Utility-Scale Battery Storage, Golden, CO, 2030. (<https://www.nrel.gov/docs/fy23osti/85332.pdf>).
- [46] L. Vidas, R. Castro, E. Cagno, M. Soltani, E. Poursmaeil, Recent developments on hydrogen production technologies: state-of-the-art review with a focus on green-electrolysis, *Appl. Sci.* 11 (2021) 11363, <https://doi.org/10.3390/AP112311363>.

- [47] J. Armijo, C. Philibert, Flexible production of green hydrogen and ammonia from variable solar and wind energy: case study of Chile and Argentina, *Int. J. Hydrog. Energy* 45 (2020) 1541–1558, <https://doi.org/10.1016/j.ijhydene.2019.11.028>.
- [48] R. Dufo-López, T. Cortés-Arcos, J.S. Artal-Sevil, J.L. Bernal-Agustín, Comparison of lead-acid and Li-Ion batteries lifetime prediction models in stand-alone photovoltaic systems, *Appl. Sci.* 11 (2021) 1099, <https://doi.org/10.3390/AP11031099>.
- [49] GME, Statistiche - dati di sintesi MPE-MGP, Gestore Mercat. Energ. (2023). (<https://www.mercatoelettrico.org/It/Statistiche/ME/DatiSintesi.aspx>) (accessed June 16, 2023).
- [50] European Environment Agency, Greenhouse gas emission intensity of electricity generation, (2023). (https://www.eea.europa.eu/data-and-maps/daviz/co2-emission-intensity-13/#tab-chart_4) (accessed September 10, 2023).
- [51] Directorate General for Energy, Directive (EU) 2018/2001 - Delegated regulation for a minimum threshold for GHG savings of recycled carbon fuels and annex, Eur. Comm. (2023). (https://energy.ec.europa.eu/publications/delegated-regulation-minimum-threshold-ghg-savings-recycled-carbon-fuels-and-annex_en) (accessed October 17, 2023).
- [52] L. Sens, U. Neuling, M. Kaltschmitt, Capital expenditure and levelized cost of electricity of photovoltaic plants and wind turbines – development by 2050, *Renew. Energy* 185 (2022) 525–537, <https://doi.org/10.1016/j.renene.2021.12.042>.
- [53] Trading Economics - Commodities Market, Natural Gas EU Dutch TTF, Trading Econ. (2023). (<https://tradingeconomics.com/commodity/eu-natural-gas>).
- [54] H. Böhm, A. Zauner, D.C. Rosenfeld, R. Tichler, Projecting cost development for future large-scale power-to-gas implementations by scaling effects, *Appl. Energy* 264 (2020), 114780, <https://doi.org/10.1016/j.apenergy.2020.114780>.
- [55] R. Schlautmann, H. Böhm, A. Zauner, F. Mörs, R. Tichler, F. Graf, T. Kolb, Renewable power-to-gas: a technical and economic evaluation of three demo sites within the STORE&GO project, *Chem. -Ing. -Tech.* 93 (2021) 568–579, <https://doi.org/10.1002/cite.202000187>.
- [56] GME, Statistical data Notices Min. Decree 2.03.2018 biomethane, Gestore Dei Mercat. Energ. (2023). (<https://www.mercatoelettrico.org/en/Statistiche/GAS/ComBiometanoNew.aspx>) (accessed June 1, 2023).
- [57] GSE, Incentivi biometano, Gestore Dei Serv. Energ. (2023). (<https://www.gse.it/servizi-per-te/rinnovabili-per-i-trasporti/biometano/incentivi>) (accessed June 1, 2023).
- [58] GSE, Obbligo di immissione in consumo, Gestore Dei Serv. Energ. (2023). <https://www.gse.it/servizi-per-te/rinnovabili-per-i-trasporti/obbligo-di-immissione-in-consumo#:~:text=L'OBBLIGODIIMMISSIONEIN,inatmosferadelsettoretrasporti>. (accessed June 1, 2023).
- [59] M. Gandiglio, Design and operation of an industrial size adsorption-based cleaning system for biogas use in fuel cells, *Energy* 259 (2022), 124941, <https://doi.org/10.1016/j.energy.2022.124941>.
- [60] A. Calbry-Muzyka, H. Madi, F. Rüsçh-Pfund, M. Gandiglio, S. Biollaz, Biogas composition from agricultural sources and organic fraction of municipal solid waste, *Renew. Energy* 181 (2022) 1000–1007, <https://doi.org/10.1016/j.renene.2021.09.100>.
- [61] T.R. Baena-Moreno, F.M. le Saché, E. Pastor-Perez, L. Reina, Membrane-based technologies for biogas upgrading: a review, *Environ. Chem. Lett.* 18 (2020) 1649–1658, <https://doi.org/10.1007/s10311-020-01036-3>.