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# SEMPRE-BIO project: comparison of three innovative scaled up and optimised technologies for biomethane production and its purification

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## ABSTRACT

This work presents the scale-up and detailed analysis, including comparison of relevant key performance indicators (KPIs) and energy analysis, of three innovative technologies for producing biomethane and its subsequent upgrading. The SEMPRE-BIO project tested and validated three different innovative technologies and pilots within the Horizon Europe framework. The three pilots are in Spain (biomethanation of biogas from wastewater treatment fermentation), France (biomethanation of syngas from biomass gasification), and Belgium (purification of biogas from manure anaerobic digestion). The three case studies will be presented and discussed. The analysis will dive into the layout of the optimised process layout of the scale-up plants as well as an exhaustive comparison, presenting advantages, shortcomings and bottlenecks of each technology, accounting for the main KPIs, i.e., electricity and steam demand, consumables including cooling water and others specific to the technologies.

**Keywords**: Biogas, Biomethanation, Biomethane, Gasification, Waste valorisation, Carbon circularity, Energy transition

## INTRODUCTION

Biomethane (bio-$CH_4$) is gaining attention for its application as fuel and feedstock in chemical synthesis [1-4]. Currently, replacing natural gas with bio-$CH_4$ is part of the EU's decarbonisation strategy. The implementation of a large and integrated biogas network should increase the EU's energy independence and resilience to market shocks and shortages [5]. Biogas is produced via anaerobic fermentation. It contains mainly wet bio-$CH_4$ and biogenic $CO_2$ (bio-$CO_2$), but, in minor content, also pollutants, such as $NH_3$ and $H_2S$ [5, 6]. As for natural gas, biogas needs purification to meet specifications for use and/or transport. The purification of biogas, i.e., dehydration, cleaning, and removal of bio-$CO_2$, is the most straightforward strategy [7]. Direct biomethanation of biogas [8] or (bio)syngas [9] is becoming appealing to boost bio-$CH_4$ yield and site productivity, but challenges are to be fixed [10]. SEMPRE-BIO project, funded under the EU's Horizon Europe program, aims to secure domestic production of cost-effective bio-$CH_4$. For this reason, pilots for three technologies have been run at different locations using different feedstocks. The general scope is increasing the European portfolio, adding more options for bio-$CH_4$ production and unlocking the biogas economy. The proposed technologies covered either biomethanation with downstream cleaning or biogas purification. For all case studies, the focus was on validating the concepts and debottlenecking their deployment. This work presents the three case studies (CSs), the results from process simulations, and compares the technologies regarding to the most relevant key performance indicators (KPIs).

## MATERIALS AND METHODS

### Case studies presentation

This section presents the case studies (CSs) and the corresponding processes. For each demo site, the block flow diagram (BFD) and model assumptions are reported. All process simulations have been performed in COCO COFE V3.10 using both conventional units and custom-based solutions.

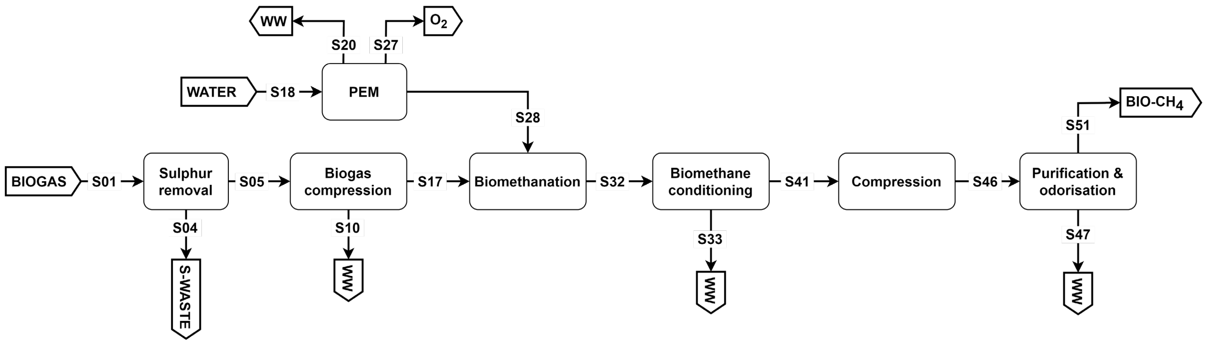


**Figure 1.** CS1 (Spain) - BFD

## Case Study 1 (CS1) - Spain

### Process description

Figure 1 depicts the BFD for the demo site in Spain to validate the direct biomethanation of cleaned biogas. The produced bio-$CH_4$ is expected to meet quality for fuelling public transport vehicles. The raw biogas comes from the fermentation of wastewater treatment sludge. The resulting raw biogas (S01) is desulfurised and the cleaned biogas (S05) compressed to 7 bar(a) and partially dehydrated (S17). The compressed biogas is sent to the biomethanation reactor, where $H_2$ from a proton-exchange membrane (PEM) module (S28) is fed and consumed. The relative ratio between $H_2$ and $CO_2$ is sensitive. The resulting gas mixture (S32), rich in bio-$CH_4$, is post-treated and conditioned to reduce its moisture content below the target value. Bio-$CH_4$ (S41) is compressed to the desired pressure based on storage and final user requirements. For this case, we considered 250 bar(a). The compression further refines the water content in the final product (S51).

### Model assumptions

For the simulation of this case study, the following assumptions have been adopted:

- Redlich-Kwong-Soave (RKS) cubic equation of state has been used to model both the liquid and gas phases.
- The desulphurisation unit has been designed as a splitter with assigned efficiency, whose value has been assigned based on the average value from literature and providers [6, 11].
- Compressors and pumps have been simulated with an assigned total efficiency of 75% (accounting for both mechanical and polytropic factors).
- The PEM electrolyser has been modelled as a simplified unit where cathode and anode reactions occur in a reactor at a fixed conversion of water. The conversion is a function of the operating temperature of the electrolyser. In addition, reaction efficiency has been considered based on the suggested value from the provider (ProPuls). This approach was already adopted in other studies [12, 13].
- Biomethanation reactor has been simulated as a reactor with fixed conversion for the limiting reactant (bio-$CO_2$). The average $CO_2$ conversion for thermophilic bacteria consortia has been retrieved from experimental evidence and the literature [14].

## Case Study 2 (CS2) - France

### Process description

Figure 2 shows the BFD for the demo site in France, for the biomethanation of biosyngas from woody biomass gasification. The chips (S01) are fed to the gasification. Steam is not supplied as the solid presents a high content of moisture, and steam is produced during the heating. The gasification chamber is run at 700-750°C under autothermal conditions. The solid (char) is discharged from the chamber, while the hot raw gas leaves the gasifier (S15). The sensible heat of S15 is recovered to sustain the gasification. The recovered heat is insufficient as gasification is highly endothermic. The remaining energy demand is covered with the combustion of a fraction of the produced biosyngas (S31). The raw biosyngas (S20) is conveyed to a cleaning unit to refine the tars and pollutants content. The cleaned syngas (S22) is split into two streams. One fraction (S31) is pre-mixed with air (S32) and burnt. The remaining (S23) is fed to a biomethanation reactor. Conversely to CS1 (biogas as feed), bio syngas contains $H_2$. Hence, two alternatives for the biomethanation have been considered: with co-feed of extra $H_2$ from an electrolyser (CS2-A) and without supplying outsourced $H_2$ (CS2-B). The resulting gas mixture (S28) is purified to deliver the best purity for bio-$CH_4$ (S30). During the cooling and purification condenses are generated. These streams are sent to wastewater treatment, not been considered in our simulations.

### Model assumptions

For the simulation of this case study, the following assumptions have been adopted:

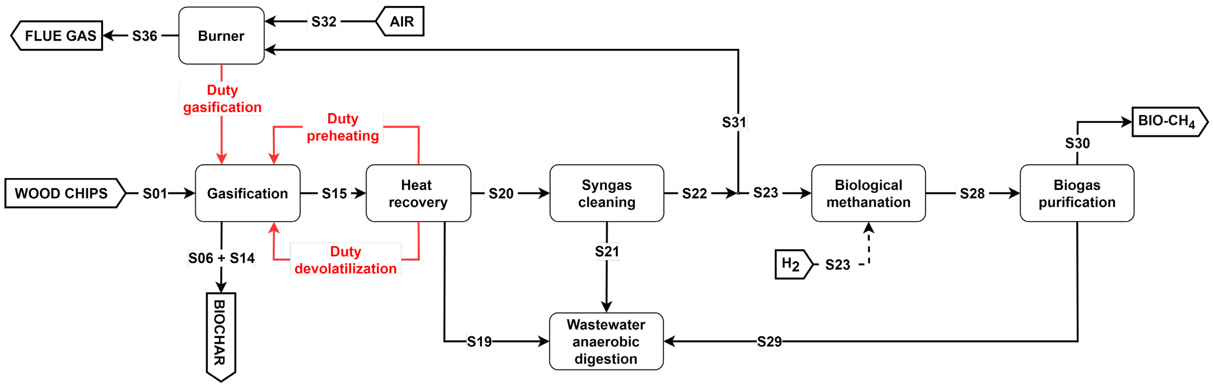


**Figure 2.** CS2 (France) - BFD

- The same equation of state has been used.
- Elemental and proximate analyses are considered to characterise the feedstock (sensitive).
- Gasification involves a complex chemistry with hundred thousand of species and reactions. The thermochemical process has been simulated considering the different steps of gasification, i.e., heating, devolatilization, pyrolysis, tar cracking, tar reforming, and secondary gas-phase lumped reaction. Details on model and preliminary validation are available [15].
- Syngas cleaning and biomethanation reactor have been simulated as for CS1.
- The fraction of burnt biosyngas is adjusted to reach autothermal conditions in the gasifier. Air is the comburent, in 10% excess. The flue gas leaves the burner at 850°C to mimic flame temperature and ensure that heat is supplied to the gasification chamber (operated at 700-750°C).

## Case Study 3 (CS3) - Belgium

### Process description

The process and the refrigeration strategies are described with more details in the paper *"Energy recovery from process purges: steam turbine integration and operation optimisation in biogas upgrading within SEMPRE-BIO project"*, which is the complement of this work. Figure 3 depicts the three process layouts considered in the analysis. The proposed BFDs differ for the "cold box". The remaining part of the process is kept unchanged. Manure (S1-01) is digested, resulting in the slurry digestate (S1-04) and raw biogas (S1-07). The produced biogas is purified from $H_2S$ and $NH_3$, then dehydrated and compressed (S1-19). Its residual moisture is reduced in triethylene glycol (TEG) absorption below 3 ppm vol. Dry biogas (S3-01) leaves the absorber, and a bio-$CH_4$-rich stream, the top of the stripper of TEG regeneration. This stream is post-treated in a membrane to recover part of the bio-$CH_4$. Pure bio-$CH_4$ is recirculated to the stripper. Injecting hot methane as stripping gas in TEG regeneration is an industrial practice to improve TEG purity from 98 to >99.9 wt%, without outsourcing heat. Unrecovered bio-$CH_4$ (S2-34) is burnt to generate steam for the turbines. The dry biogas (S3-01) is cooled and S3-02 conveyed to the cryogenic distillation: part of the bio-$CO_2$ is recovered as pure liquid (>99.99 vol%) from the bottom, while bio-$CH_4$-enriched stream (S3-11 with $CO_2$ ~80 vol%) is sent to a membrane cascade to purified until outlet gas (S4-04) reaches a residue $CO_2$ content below 50 ppm (vol), respecting the specifications for grid injection. The membranes work at ambient temperature. The permeated gas is enriched in $CO_2$ but contains bio-$CH_4$ as $CO_2$ permeation is favoured. Part of the permeate is recycled internally, while the purge (S4-12) is cooled and recycled to the cryogenic distillation. Pure bio-$CH_4$ is cooled and conveyed to the $CH_4$ liquefaction process (S5-01), where the expansion (Joule-Thomson effect) generates the frigories to liquefy part of the bio-$CH_4$ (S5-03). The not liquefied fraction (S5-20) is purged. The purges, S2-34 (from TEG dehydration) and S5-20 (from bio-$CH_4$ liquefaction), are burnt. The combustion of these streams produces heat for steam and power production. Part of the steam is used for the regeneration of TEG solvent (topic of the complementary paper).

### Refrigeration strategies

Figure 3 shows three layouts depicting three alternatives to manage the refrigeration and keep the cold box of the system:

- Option A (single loop): the total cooling demand is covered using a hydrocarbon (HC) mixture as working fluid. The HC blend contains $C_2$-$C_3$ hydrocarbons. The relative ratio of $C_2$-$C_3$ has been tuned to keep the cold box minimum temperature (sensitive).
- Option B (double loop): the same mixture is used

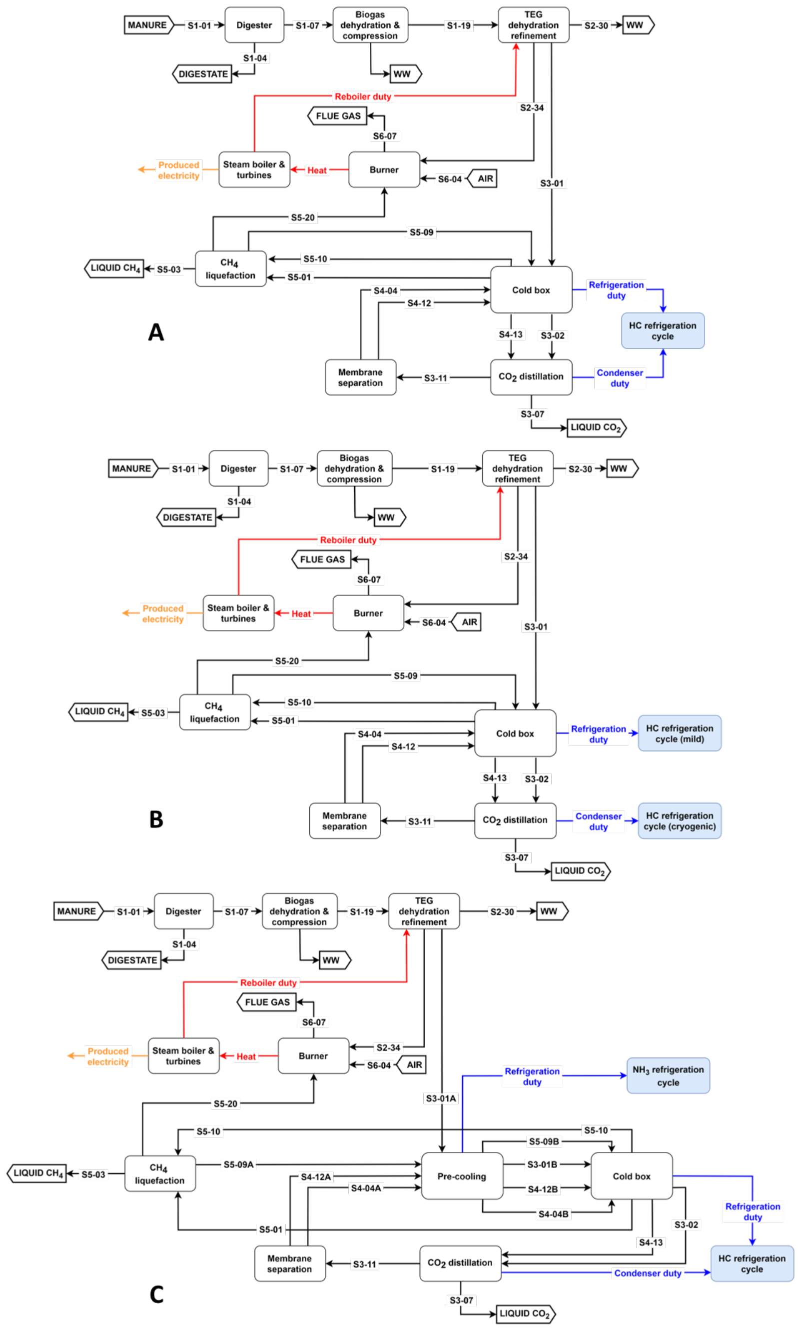


**Figure 3.** CS3 (Belgium) - BFDs

as working fluid but split into two independent mild refrigeration loops. In the first refrigeration loop (named as loop), all warm streams are precooled from ambient temperature (25-30°C) to reach the same intermediate temperature of the gas leaving the top of the cryogenic column. This temperature is sensitive and is not reported. In the second refrigeration loop (named as cryo loop), all the streams are cooled to the final desired temperature (sensitive).

- Option C (double loop with two refrigerants): is like Option B, but all warm streams are pre-cooled to the same intermediate temperature using ammonia for the mild loop. The temperature is reduced to the final target value in a second refrigeration cycle, using the same HC mixture as for options A and B.

## Model assumptions

The following assumptions are considered:

- Biogas flow and volumetric composition are assigned (sensitive).
- The same efficiency for pumps and compressors, $H_2S$ and $NH_3$ removal as for CS1 and CS2.
- The template for TEG has been retrieved from examples and ready-to-use flowsheets available in COCO COFE webpage [16].
- The membranes are modelled as a Python-based custom module integrated within the flowsheet. Permeance and selectivity for an industrial membrane are retrieved from the literature [17].
- Other model assumptions related to the refrigeration cycles and the steam turbine are available in the mentioned work.

## Technology assessment: definition of key performance indicators (KPIs)

The three technologies have been assessed both individually and in comparison. The specific KPIs for each case study (CS) will be discussed in the dedicated section to contrast our results with pieces of evidence from prior literature. This analysis has been carried out to show the consistency of the implemented models. Regarding the comparison of the three technologies, the assessment has been structured to fairly compare them in terms of net production and other relevant indices. The demo sites validate different concepts with different solids starting feedstock, capacity, scale, and target. Hence, their direct comparison through extensive properties would be misleading. In addition, CS1 and CS2 include biomethanation, whereas purification of biogas is the core of CS3. For this reason, the analysis focuses on the net (dry) bio-$CH_4$ production per unit of both dry solid feedstock and volume of dry biogas or biosyngas. Here, biosyngas is considered because CS2 consider gasification of woody biomass and not biogas from solid waste fermentation. Remarkably, both bio-$CH_4$ and the reference solid/gas feedstock are considered on a dry basis. The bio-$CH_4$ purity, electricity demand, and cooling water consumption are other relevant parameters. Remarkably, the PEM electrolyser is considered for both CS1 and CS2A to supply green $H_2$. For these cases, the electrolyser demand is set from the simulation. The average demand for electrolysis is ~56 kWh/kg$_{H2}$, in line with literature and technical reports [18-20]. Regarding cooling water demand, the source is assumed at 20°C and the maximum allowable discharge temperature is set 30°C.

# RESULTS AND DISCUSSION

## Simulation results

This section will show and comment on the results from the simulations. In the first part, each case study will be discussed. Finally, an overview and comparison based on KPIs of the different case studies is addressed.

### CS1 results

The most relevant property of the compressed bio-$CH_4$ is its dew point ($T_{dew}$) at 250 bar(a). This is the storage pressure of the product for end-user usage (fuel for public transport). The dew point is directly proportional to the purity of delivered bio-$CH_4$ and the content of impurities. CS1 can deliver compressed bio-$CH_4$ with a purity of 98.45 vol%. The dew point of the gas mixture has been calculated based on the Redlich-Kwong-Soave model. It corresponds to -60.5°C, meaning that the level of other impurities (moisture, $H_2$, and $CO_2$) is kept below the threshold value. The estimated dew temperature respects the European standard EN 16723-2:2017 for compressed natural gas for the use of Bio-CNG (compressed natural gas) as an automotive fuel stored at 200–250 bar.

### CS2 results

Table 1 summarises the results related to the steam gasification of the woody biomass. The main KPIs are the gas yield, i.e, the amount of dry gas produced per unit of mass of dry solid, the volumetric composition of the syngas (still on a dry basis) limited to $H_2$ and $CH_4$, and the corresponding $H_2/CO_x$, defining the excess of $H_2$ compared to the sum of $CO_2$ and CO. In addition, the analysis considers the tar yield, the energy input, defined as the total amount of heat supply to the gasifier per unit of dry solid, and the heating values (both high and low). The results have been compared to experimental works and review papers referring to steam gasification [20-21]. The mentioned references confirm that the obtained values are aligned with experimental values and ranges for softwood biomass.

**Table 1:** Overview of CS2: KPIs for biomass pyrolysis (all values refer to dry product and feedstock)

| KPI | Value | References |
|---|---|---|
| Gas yield | 1.23 $Nm^3_{syngas}/kg_{solid}$ | [20, 21] |
| $H_2$ content | 50.9 vol% | [20, 21] |
| $CH_4$ content | 6.10 vol% | [20, 21] |
| $H_2/CO_x$ | 1.18 vol/vol | [20, 21] |
| Char yield | 0.25 $kg_{char}/kg_{solid}$ | [20, 21] |
| Tar yield | 17.7 $g_{tar}/kg_{solid}$<br>14.4 $g_{tar}/Nm^3_{syngas}$ | [20, 21] |
| Energy input/ demand | 1.02 $kWh_{th}/kg_{solid}$<br>or 3.66 $MJ_{th}/kg_{solid}$ | [20, 21] |
| HHW | 12.35 $MJ/Nm^3_{syngas}$ | [20, 21] |
| LHW | 10.60 $MJ/Nm^3_{syngas}$ | [20, 21] |

Finally, Table 2 gathers the main performance of the technology, including the biomethanation process. For CS2, two different configurations have been considered: with (CS2A) and without (CS2B) extra green $H_2$ supplied to the biomethanation.

Remarkably, the results show that the $H_2$ in the biosyngas is not sufficient to run the biomethanation to full depletion of $CO_x$. Hence, extra $H_2$ boosts the productivity of the system; thus, the purity and concentration of bio-$CH_4$ in the product mixture. The moisture content is still large for transport and use. Therefore, a deep dehydration step should be considered. Beyond this consideration, the analysis highlights the need to combine $H_2$ production with woody biomass gasification.

**Table 2:** Overview of KPIs of CS2 (all values refer to dry bio-$CH_4$ and solid feedstock)

| KPI | CS2A | CS2B |
|---|---|---|
| Productivity | 0.32 $Nm^3_{CH4}/kg_{solid}$ | 0.15 $Nm^3_{CH4}/kg_{solid}$ |
| $H_2$ addition | 1.91 $Nm^3_{H2}/Nm^3_{CH4}$ | Not available |
| $CH_4$ purity | 95.2 vol% | 45.0 vol% |
| Impurities | | |
| $CO_2$ | 0.03 vol% | 30.1 vol% |
| CO | 0.07 vol% | 20.6 vol% |
| $H_2$ | 0.34 vol% | < 100 ppmv |
| $H_2O$ | 4.34 vol% | 4.34 vol% |

## CS3 results

CS3 is the only demo site where direct biogas purification is performed. Compared to alternative processes such as amines [22], the technology validated in CS3 enables the delivery of both pure bio-$CH_4$ and bio-$CO_2$ (purity > 99.99 vol%). This is the main advantage of this new cryogenic system. Other relevant results refer to the integration of boiler and steam turbines, and how the selection of refrigerant and the refrigeration cycle layout affect the energy balance of the system. This paragraph will briefly summarise the main results. More details are reported in *"Energy recovery from process purges: steam turbine integration and operation optimisation in biogas up-grading within SEMPRE-BIO project"*. The analysis showed that keeping the cold box through a refrigeration cycle is energy-demanding. Compression of refrigerants accounts for 34 to 37% of the total electricity demand. The implementation of two independent refrigeration loops (Option B and C) lowers the electricity demand. Using ammonia as a refrigerant in the mild loop (Option C) further drops the electricity demand and valorising waste streams is needed to mitigate the energy demand of the overall process. The simulations highlight that the steam cycle allows for recovering up to 10% of the total electricity demand of the upgrading plant.

## Processes comparison

Table 3 highlights the main results of the global KPIs of the CSs, including all process configurations. Looking at the results, it is possible to draw the following conclusions:

- Woody biomass has the highest gas yield, meaning that it is the feedstock that releases the largest amount of gas (biosyngas) per unit of solid. Gasification indeed is an efficient technology to convert solids into syngas. Unfortunately, the same information is not available for wastewater sludge due to missing data related to the efficiency of the sludge fermentation.
- Apparently, biomethanation of biogas from sludge fermentation is the most productive route. It is worth remarking that for CS1 (but also CS2A), the total productivity is boosted due to the addition of green $H_2$ (in excess to the stoichiometric amount) to run the biomethanation. Conversely, for CS3, the productivity is the net productivity of the system because bio-$CH_4$ is recovered from biogas directly.
- The electricity consumption of the three technologies is completely different. Whenever biomethanation is part of the process, most of the electricity demand is concentrated in the green $H_2$ production. For CS3, the energy demand covers the compression of biogas and the energy for the refrigeration cycles (refrigerant compression). If PEM energy demand is disregarded, CS3 consumes almost 10 times more energy than other options.
- Directly from the previous comment, it is evident that biomethanation is an effective solution to produce bio-$CH_4$ from both biogas and biosyngas. The technology enables a substantial conversion of COx into CH4, thereby breaking the thermodynamic equilibrium and operating the system at thermophilic conditions (milder temperatures, e.g., ~50-55°C). However, despite this advantage, $H_2$ demand is still relevant and represents one of the bottlenecks of the technology.
- Cooling water demand is in the same order of magnitude for all the considered processes. This parameter is less relevant to the present analysis as it highly depends on the operative condition. Anyway, there are no significant differences in electricity demand.

Regarding CS3, it is worth underlining that the electricity demand covers both the refrigeration cycle and the compression of biogas; more in general, the overall electricity demand to deliver both pure pressurised bio-CH4 and bio-CO2 (99.99+ vol% purity). Refrigeration covers from 34 to 37% of the total energy depending on refrigerant and cycle configuration. The process up to bio-CH4 purification, i.e., disregarding liquefaction of bio-CH4, consumes 1.70 $kWh_{el}/Nm^3_{biogas}$ and 0.15 $kWh_{th}/Nm^3_{biogas}$.

**Table 3:** Overview of KPIs of all CSs

| | Solid feed-stock | Biogas/ Biosyngas produced | Bio-$CH_4$ production[1] | Bio-$CH_4$ purity (vol%) | Electricity demand (w/o PEM)[2, 3] [$kWh_{el}/Nm^3_{CH4}$] | Cooling Water (CW) demand [$t_{CW}/Nm^3_{CH4}$] |
|---|---|---|---|---|---|---|
| CS1 | Sludge | Not Available | 0.709 $Nm^3_{CH4}/Nm^3_{biogas}$ | 98.5 | 11.9 (0.30) | 0.174 |
| CS2A (extra $H_2$) | Woody biomass | 1.23 $Nm^3_{syngas}/kg_{solid}$ | 0.260 $Nm^3_{CH4}/Nm^3_{biosyngas}$<br>0.320 $Nm^3_{CH4}/kg_{solid}$ | 95.2 | 10.1 (0) | 0.523 |
| CS2B (no extra $H_2$) | Woody biomass | As CS2A | 0.122 $Nm^3_{CH4}/Nm^3_{biosyngas}$<br>0.150 $Nm^3_{CH4}/kg_{solid}$ | 45.0 | 0 | 0.804 |
| CS3 Option A | Manure | 3.80 $Nm^3_{biogas}/t_{solid}$ | 0.591 $Nm^3_{CH4}/Nm^3_{biogas}$<br>0.09 $Nm^3_{CH4}/kg_{solid}$ | 99.99+ | 2.21 | 0.374 |
| Option B | Manure | As Option A | As Option A | As above | 2.15 | 0.207 |
| Option C | Manure | As Option A | As Option A | As above | 2.10 | 0.205 |

[1] Referred to dry biogas/biosyngas
[2] Assuming PEM consuming 56 $kWh_{el}/kg_{H2}$
[3] Energy recovered from turbine is included (only CS3)

Recent literature investigated several amine systems for upgrading biogas [22].

This work reports on average 0.15 $kWh_{el}/Nm^3_{biogas}$ and 0.28 $kWh_{th}/Nm^3_{biogas}$ but neglects the compression and purification of $CO_2$ and bio-$CH_4$. The purity of bio-$CH_4$ is poor, ranging 90-95 vol%; hence, further refinement is needed. This comment holds also for bio-$CO_2$ which can contain acid pollutants ($H_2S$) and degradation products. The purification and compression of $CO_2$ is expensive: values reported in the literature does not represent the actual energy cost for delivering bio-$CH_4$ matching market specification. The analysis is limited to the bulk separation of $CO_2$ from biogas. Moreover, the technology tested in the SEMPRE-BIO project (Cryo Inox's patent) allows for delivering both $CH_4$ and $CO_2$ pure and liquid to ease their transport. $CO_2$ liquefaction is always neglected in articles dealing with the biogas upgrading.

## CONCLUSION AND FUTURE WORK

This work presented the results of the process simulation and comparison for three technologies for bio-$CH_4$ production. The analysis showed that biomethanation of both biogas and biosyngas is a valuable opportunity to convert bio-$CO_2$ into bio-$CH_4$. However, this option is still under investigation and constrained to the availability of cheap green-$H_2$. Despite this potential limitation, it is worth noting that biomethanation can increase overall production of the site. Biogas upgrading is a well-established technology to purify bio-$CH_4$. The SEMPRE-BIO project validates a new option with cryogenic distillation. The analysis showed more efforts should focus on reducing the energy demand. This work is limited to the technical analysis of the three technologies. However, it is important to note that the analysis should be more holistic, accounting for integrated techno-economic and life-cycle assessments to address the economic and environmental aspects. Only a complete analysis encompassing all these aspects allows for leveraging advantage and identifying bottlenecks. This holistic analysis will be finalised later within the project.

## ACKNOWLEDGEMENTS

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