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Energy recovery from process purges: steam turbine integration and operation optimisation in biogas upgrading within SEMPRE-BIO project

Filippo Bisotti[a]*, Matteo Gilardi[a]*, and Bernd Wittgens[a]*

[a] SINTEF Industry – Process Technology – P.O. Box 4760 Torgarden, 7465, Trondheim, Norway
* Corresponding Authors: filippo.bisotti@sintef.no, matteo.gilardi@sintef.no, bernd.wittgens@sintef.no

ABSTRACT

The SEMPRE-BIO project tested and validated three different innovative technologies and pilots within the Horizon Europe framework. One of the pilots is commissioned in Belgium. The proposed technology purifies biogas from manure anaerobic digestion and delivers simultaneously pure biomethane and food-grade CO_2, conversely to conventional purification technologies such as absorption and adsorption. Due to the severe cryogenic conditions, energy recovery for purge and waste streams becomes relevant to improve the energy demand of the process. The present work will show an effective solution to reduce the electricity demand of the process. Biomethane slip and other purge stream are valorised in a steam boiler and a two-pressure-level steam turbine to deliver both middle pressure steam as utility in distillation reboilers and produce electricity. The analysis will propose a simple, but rigorous methodology to maximise the steam turbine loop and the net power. The present work is based on solid and concrete assumptions for the design of the steam turbine. The analysis shows that the process allows for recovering up to 12% of the total electricity and around 55% of the total energy demand for the refrigeration loop to preserve the cold box of the bio-CH_4/CO_2 distillation.

Keywords: Turbines, Energy Efficiency, Combustion Heat and Power (CHP), Energy recovery, Biogas upgrade

INTRODUCTION

Biogas appears as a promising feedstock for combustion heat and power (CHP) applications [1, 2] as well as for replacing fossil methane in chemical production [3]. Biogas is a wet mixture of biomethane (bio-CH_4) and biogenic CO_2 (bio-CO_2) with a variable ratio of the two main components. Generally, bio-CH_4 ranges from 45 vol% to 75 vol% [4]. The remaining is mainly wet bio-CO_2. Prior to any application and transport, biogas undergoes deep dehydration and purification/upgrading by removing bio-CO_2 [5]. In addition, bacteria releases H_2S and NH_3 in traces during the anaerobic digestion of biological wastes (food residue, sludges, wastewater, crops, and manure) [4, 5]. These species are harmful and corrosive. Hence, biogas purification is energy intensive [6]. The bulk capture of CO_2 and successive dehydration of bio-CH_4 consumes heat, cooling water, and electricity [7]. This work will introduce a novel concept for biogas upgrading patented by Cryo Inox S.L. and tested within SEMPRE-BIO, an EU project under the EU's Horizon Europe Program. The site capacity and biogas productivity are sensitive. The analysis shows that valorising side streams covers part of the electricity energy and satisfy the total demand for thermal energy of the plant.

MATERIALS AND METHODS

Process block flow diagram (BFD)

Figure 1 depicts the three different process layouts considered in the analysis. The proposed BFDs differ for the "cold box" and how refrigeration is performed. The remaining of the biogas upgrading is kept unchanged. Regardless of the process configuration, manure (S1-01) is digested resulting in the slurry digestate (S1-04) and raw biogas (S1-07). The produced biogas is purified (H_2S and NH_3 removal) while dehydration and compression are carried out. The compressed biogas (S1-19) still contains

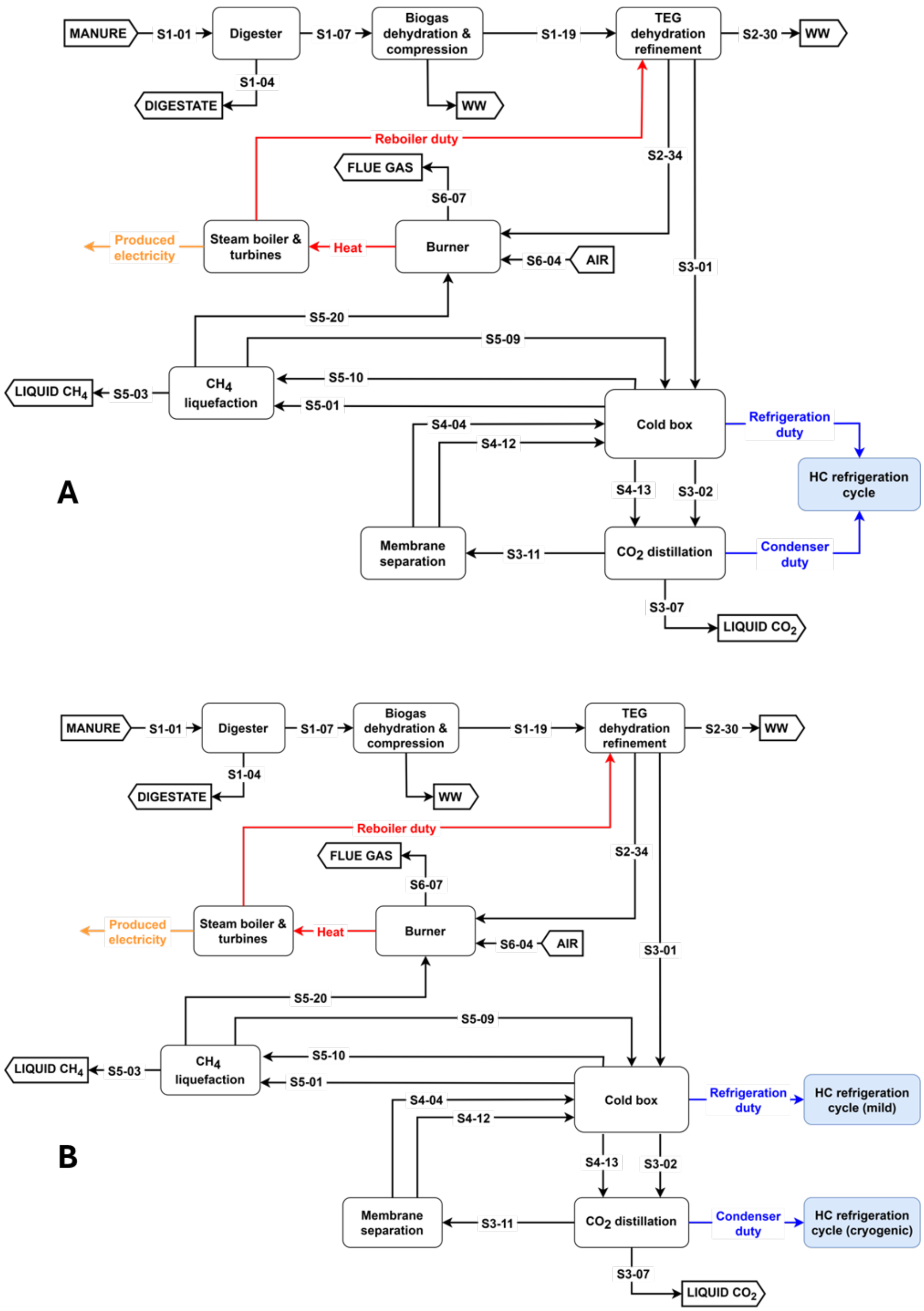

Figure 1: Process BFDs for three different configurations of the analysed process. The processes differ for the cold box configuration and how cooling (blue arrows) is managed within the process. The remaining part of the process remains the same. WW stands for wastewater.

a relevant residual moisture. The moisture content is refined through a conventional triethylene glycol (TEG) absorption process to keep the residue water content below 3 ppm (vol). The dry biogas (S3-01) leaves the absorber. As the absorber is run at high pressure and low temperature to abate residue moisture, bio-CH_4 and bio-CO_2 accumulates in the solvent [8, 9], which are easily released as heat is supplied. TEG regeneration is run

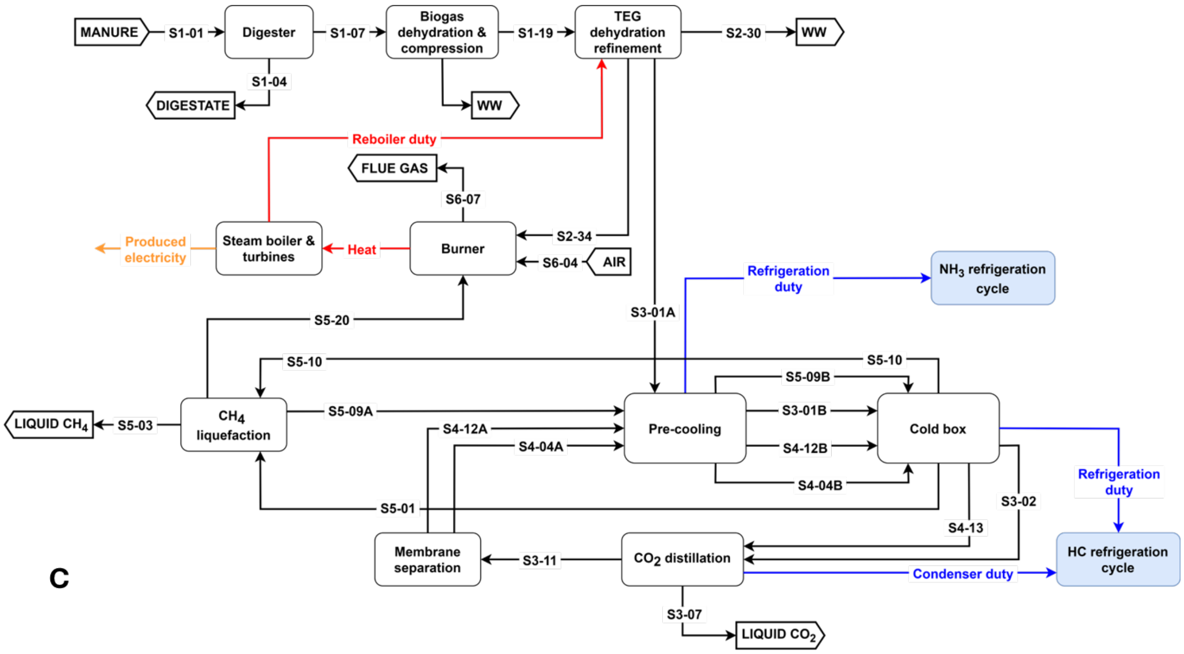

Figure 1. (continues):

thermally, and the process require steams. However, the temperature in the reboiler of the TEG regeneration is kept below 207°C to prevent it from degrading. To keep a minimum temperature approach ~10°C and avoid formation of hotspots in the reboiler, steam must be supplied around 219-221°C. A bio-CH_4-rich stream is released from the top of the stripper of the TEG regeneration. This stream is post-treated in a membrane to recover part of bio-CH_4. Pure methane is recycled back to the stripper after recompression. Injecting hot methane as stripping gas in TEG regeneration column is an industrial practice to increase the TEG purification from 98 to 99.9+ wt%, while lowering the thermal demand for the regeneration and reducing the temperature at the bottom [10, 11]. This is reflected in a minimum TEG degradation and liquid-to-gas ratio in the absorber to reduce the moisture content in the gas phase below 3 ppm (vol). The unrecovered bio-CH_4 (S2-34) is a purge and valorised in the burned to deliver steam for the steam turbine.

The dry biogas (S3-01) is cooled, and the resulting cold biogas (S3-02) is conveyed to the cryogenic distillation, whose operating conditions, namely, temperature and pressure, are sensitive. Part of the bio-CO_2 is recovered as pure liquid (>99.99 vol%) from the bottom, while bio-CH_4-enriched gas stream (S3-11 with CO_2 ~80 vol%) is sent to a membrane separation system where residue CO_2 selectively permeates and the gas stream is progressively purified until the outlet gas (S4-04) reaches a residue CO_2 content below 50 ppm (vol), which respects the specifications for grid injection. The membrane system works at ambient temperature. The permeated gas is enriched in CO_2 but still contains bio-CH_4 as membrane preferably permeate CO_2, but the separation is not sharp. Part of the permeated gas is recycled internally, while the purge (S4-12) is cooled and recycled to the cryogenic distillation to maximise both bio-CH_4 recovery and liquid bio-CO_2 production. Pure bio-CH_4 is cooled and conveyed to CH_4 liquefaction process (S5-01) where it is compressed and partially expanded to exploit Joule-Thomson effect and generate the frigories to liquefy part of the bio-CH_4 (S5-03). The not liquefied fraction (S5-20) is purged. The purged streams, namely S2-34 (from TEG dehydration) and S5-20 (from bio-CH_4 liquefaction), are transferred to a burned where excess air is co-fed. The combustion of purge streams is used to produce heat (steam) and power. Part of the produced steam is used for the regeneration of TEG solvent. The implementation and optimisation of combined steam boiler and turbine is described in the next sections.

Refrigeration strategies

Figure 1 shows three different layouts depicting three alternatives to manage the refrigeration and keep the cold box of the system. The basic process is described in the previous section, and it is kept equal regardless of the refrigeration strategy and configuration. The cold box satisfies the frigories demand in the cryogenic distillation (bio-CH_4 purification through bio-CO_2 liquefaction) and liquefaction of purified bio-CH_4. For the first, pre-cooling of the dry biogas and reaching cryogenic temperature in the top condenser of the distillation column (condenser duty in Figure 1) are needed. For the latter, cryogenic conditions are required to reduce the inefficiency. Indeed, if frigories are recovered exclusively

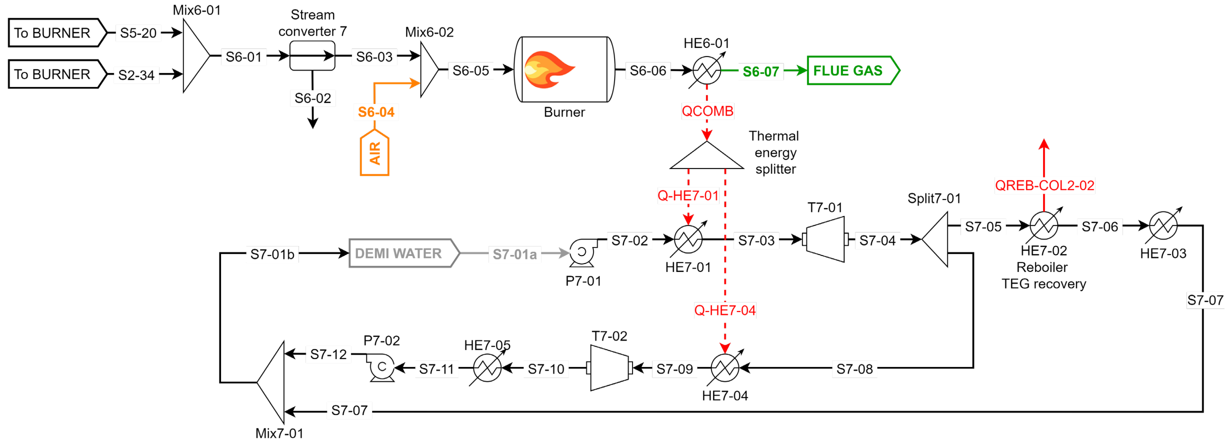

Figure 2. Combustion chamber and steam boiler with corresponding dual-pressure steam loop

using the liquefied bio-CH_4 without outsourced cooling duty, the net production of liquid bio-CH_4 would be reduced. Figure 1 distinguishes between the duty for the condenser of the cryogenic distillation and all the pre-cooling demand distributed around the plant. This distinction will be clarified later. Based on these considerations, three different layouts have been identified:

- 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 as working fluid but split into two independent refrigeration loops. In the first refrigeration loop (named as mild 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 here not reported. In the second refrigeration loop (named as cryo loop) all the streams are cooled to final desired temperature (sensitive). This option is justified by thermodynamic constraints. Warmer streams allow for having higher outlet temperature of the cooling fluid leaving the evaporator. Conversely, colder streams are more challenging to further refrigerate because of the minimum temperature approach in the condenser heat exchanger. Then, the milder loop would treat a lower amount of gas because the gas from the cryogenic distillation is treated and cooled only in the cryo loop.
- 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 refrigerant for the mild loop. The temperature is reduced to the final target value in a second refrigeration cycle still using the same HC mixture as for options A and B.

Steam boiler and turbine

Process description

Figure 2 depicts the configuration of the burner and a conventional dual-pressure steam turbine system. Cold water (S7-01a) is pumped (P7-01), superheated (HE7-01) and conveyed to the first steam turbine T7-01. Expanded steam leaves the unit and it is split into two flows: S7-05 and S7-08. S7-05 is the steam supplied to the reboiler of the TEG regeneration. The resulting condensed steam (S7-06) is further cooled (HE7-03) and recycled back. The other stream from the split (S7-08) is heated a second time and sent to a second expander (T7-02). The low-pressure steam is then fully condensed (HE7-05), pumped (P7-02) and mixed with S7-07. The total condensed water is then recycled to the main pump (P7-01). This work aims to optimising the steam cycle to maximise the net work recovered from the combustion of bio-CH_4 rich purges defined as

$$W_{Net} = W_{T7-01} + W_{T7-02} - W_{P7-01} - W_{P7-02} \quad (1)$$

where W stands for the power generated (for turbines) and consumed (for pump) and consumed (for pump).

Input data and Degrees of Freedom (DoF) of the optimisation

The combustion of bio-CH_4 purges releases 96.04 kW_{th}. This is the total energy input to the steam cycle. The steam cycle is designed to deliver 39.12 kW_{th} to the reboiler of the TEG regeneration. As aforementioned, steam should be supplied saturated at 220°C, i.e., at 23.5 bar (absolute). The steam quality is stringent due to TEG thermal stability. The degrees of freedom of the system are: (1) the total flow of water/steam circulating in the cycle (S7-01a), (2) the upper pressure of the steam cycle,

(3) the thermal duty (Q-HE07-01) supplied to the first superheater (HE07-01), and (4) the discharge pressure of the second steam turbine (T7-02). These variables correspond to the optimisation parameters.

The system has been simulated in Aspen Hysys V12 using ASME steam thermodynamic model. The optimisation has been carried out using the sensitivity analysis tool in the simulation environment. For pump and turbines, a global efficiency, i.e., product of polytropic and mechanical efficiency, has been assumed. The following ranges have been investigated for the optimisation parameters:

Total mass flow: 95 – 115 kg/h with a step of 5 kg/h. Our investigation included 98, 107, and 112 kg/h to refine the domain and further optimise the process.

(1) Maximum/upper pressure of the steam cycle: ranges between 60 – 120 bar (absolute), typical for high-efficiency system for heat recovery and electricity production [12, 13]. 10 bar is the considered discretization step. The lower and upper limited has been identified based on steam turbine producers' suggestions and standard, i.e., not customised, units.

(2) Superheater Q-HE7-01 duty: ranges 90 – 96 kW_{th} and energy balances imposes that

$$Q_{HE7-04} = 96.04 - Q_{HE7-01} \quad (2)$$

(3) Discharge pressure of the second turbine T7-02: as been limited to 0.3 – 1.2 bar (absolute) with a step of 0.3 bar. The bottom and upper limits are retrieved from technical reports and producers [14]. It is unlikely to lower the discharge pressure below 30 kPa. It is possible to run the turbine above 1.2 bar absolute, however, this is normally done in district heating and not in steam boiler and turbine loops where the main target is the energy recover.

Constraints and criteria

The results have been screened and filtered based on the following criteria (checkpoint list) and constraints:

1. The maximum inlet temperature of superheated steam to turbines (S7-03) is 600°C to avoid thermal stress [12, 15], i.e., periodic maintenance, and expensive construction materials [15]. This constraint links the duty to the first superheater (HE07-01) and the total water mass flow circulating in the loop. If the inlet temperature exceeds this threshold, i.e., T_{S7-03} > 600°C, the operating point is unfeasible. The same constraint holds for the steam conveyed to the second steam turbine (T7-02), hence T_{S7-09} < 600°C must be fulfilled.
2. S7-05 is the saturated steam for the reboiler of TEG regeneration. The maximum temperature of the steam is assumed 220°C to keep a minimum temperature approach of ~10°C in the reboiler and the avoid hotspots in the unit as TEG degrades above 207°C. Setting the steam pressure implies that the discharge pressure of T7-01 is 23.5 bar. Moreover, the reboiler duty for the TEG regeneration (QREB-COL02-02) is assigned; indeed

$$m_{S7-04} = \frac{Q_{REB-COL2-02}}{\Delta H_{EV}(T_{sat}, P_{sat}(T_{sat}))} \quad (3.1)$$

$$m_{S7-04} = \frac{39.12}{\Delta H_{EV}(220°C, 23.5\ bar)} \quad (3.2)$$

3. From the previous point it follows that the temperature of the superheated steam leaving the turbine T7-01 (S7-04) could be warmer than the one selected to deliver steam to the TEG reboiler. Hence, the optimal operational point should deliver steam as much as possible to the saturation point to avoid a kind of transfer line heat exchanger to remove the "excess" heat (Q_{excess}). This device would increase the inefficiency of the system.
4. The lower pressure of the loop, i.e., discharge pressure of the second turbine T7-02, is an optimisation variable and DoF of the system. The discharge pressure is constrained to the physical state of the steam leaving the unit. If steam starts condensing, the expander cannot be operated. Hence, the discharge pressure is selected as the lowest pressure at which only steam leave the unit, i.e., vapour fraction α = 1. This condition preserves the operation of the turbine while ensuring to recover the maximum work. It is straightforward, that the same condition must be satisfied also at the outlet of first turbine (T7-01). This condition is less stringent, unless to large water mass flow is processed in the cycle.

RESULTS DISCUSSION

Steam cycle optimisation

Table 1 gathers the results from the sensitivity analysis performed in Aspen Plus. Several feasible points have been identified; only the optimal one per each tested total mass flow of steam/water in the loop are reported. The optimal points are selected based on the maximum net work recovered from the loop. The trend of net work recovered from the steam cycle is depicted in Figure 3. The results show that the steam cycle can be operated only within a limited domain for the steam/water mass flows in the loop: between 98 kg/h (minimum) and 112 kg/h (maximum). Indeed, below 98 kg/h, the total fluid is too little and the inlet temperature of superheated steam either to the first or to second turbine violates the threshold value of 600°C. Under this condition, the combustion releases an excessive heat. Conversely, above 112 kg/h, the total fluid is too large: the first superheater requires a substantial fraction of the heat released from the combustion, but the remaining heat is insufficient to reheat the steam after the splitter.

Table 1: Optimal points for the steam cycle

Steam flow (kg/h)	P max (bar abs)	P min (bar abs)	Q_{HE7-01} (kW)	Q_{excess} (kW)	Inlet T (°C) T7-01 / T7-02	Net work (kW)
95	-	-	-	-	-	-
98	120	0.4	92.5	6.07	588 / 588	19.11
100	120	0.4	92.0	4.80	554 / 573	19.10
105	**120**	**0.4**	**90.0**	**1.46**	**468 / 566**	**19.04**
107	120	0.6	90.0	0.60	448 / 526	18.34
110	100	0.9	90.0	0.05	407 / 487	16.64
112	80	1.0	90.0	0.05	374 / 472	15.26
115	-	-	-	-	-	-

Table 2: Comparison of the refrigeration strategies

Option	P_{tot} (kW)	$P_{tot-net}$ (kW)	Compression refrigerant (kW)	$Compr\%_{tot}$	Electricity Saving (ES)	ΔE_{tot}	ΔE_{net}
A	191.2	172.2	70.8	37.0%	-9.94%	Reference	Reference
B	186.2	167.2	66.1 (total)	35.5%	-10.2%	-2.6%	-2.9%
C	181.9	162.9	61.9 (total)	34.0%	-10.4%	-4.9%	-5.4%

This results in incipient condensation of the steam even above the atmospheric pressure (1.2 bar absolute has been considered as the upper limit). In other words, the system experiences a lack of thermal energy. This explanation justifies also the reduction of the upper pressure of the steam loop, which progressively reduces from 120 bar (maximum) to 80 bar. This is the result of the combination of saturation temperature (higher as pressure increases) and latent of vaporization (lower as pressure increases). Figure 3 displays an almost flat profile up 105 kg/h and then a sharp drop of the net work recovered. The average net work recovery is around 19 kW for the flat area. The reduction of the net work is substantial beyond 105 kg/h. Indeed, for an increment of the mass flow from 105 to 112 kg/h (+6.25%), the work recovery falls from 19 kW to 15.3 kW (-19%). The physics behind the system is straightforward and already commented above, when describing the boundaries of the feasible domain to run the steam cycle. The optimal operational point has been selected by globally accounting for the criteria listed and described in the dedicated section. In particular, the steam cycle run with 105 kg/h is considered the best option because of the limited excess heat, meaning that the steam withdrawn after the first expansion is almost close to saturation and no quench is required. In addition, in that configuration, superheated steam is fed to both turbines at milder temperatures, i.e., lower than the maximum allowable one (600°C), lowering the risk of mechanical failure.

Effect of refrigeration strategies and steam cycle integration in the biogas upgrading

In addition, the system has been analysed in terms of refrigerant demand and total energy demand to identify the best cold box configuration and the benefit associated with the addition of a steam turbine system in the base process. Based on the simulation of the whole process under the different options (Figure 1). The total cooling demand is around 49 kW_{th}, split into 19.3 kW_{th} for the condenser at the cryogenic distillation and 29.7 kW_{th} to provide frigories in other sections of the process.

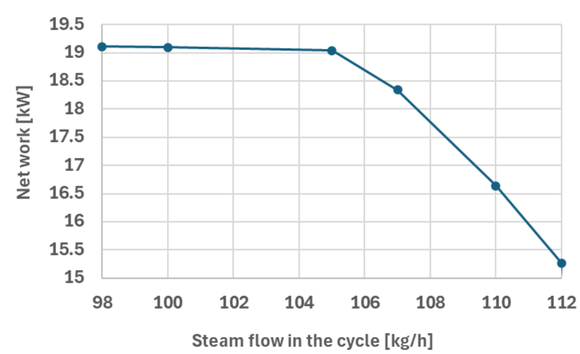

Figure 3. Locus of optimal points

Table 2 gathers the results from the simulation. The results show a progressive reduction in the fluid demand and the corresponding compression costs. Hence, the possibility to split the refrigeration into two independent loops is a benefit. Moreover, the possibility to run the mild loop with ammonia as refrigerant improves further the performance. Indeed, the pre-cooling with ammonia and reduces the total electricity demand to operate the refrigeration cycle. At this point, it is worth contextualising the role of the energy recovery and cold box optimisation in the plant for biogas upgrading and liquefaction.

Additional data are necessary to carry out this last assessment. Table 3 resumes the results for all the simulations. The total electricity, in Eq (4), power demand (P_{tot}) accounts for compressors and pumps and it has been estimated based on the simulations; while the net power ($P_{tot\text{-}net}$) is defined as the difference between the total power (P_{tot}) and the net total power/work recovered from the steam boiler and turbine system (W_{net}), as defined in Eq (1). All entities are expressed in kilowatt (kW).

$$P_{tot-net} = P_{tot} - W_{net} \tag{4}$$

It is important to highlight that the previous analysis concluded that the maximum net power (W_{net}) recovered from the turbine loop is around 19 kW (Table 1). The analysis considers the following key performance indicators (KPIs):

- Percentage of energy demand of compression of refrigerant(s) on the total electricity demand:

$$\mathrm{Compr\%}_{tot} = \frac{W_{refrigerant\ compression}}{P_{tot}} \tag{5}$$

where the numerator is provided in Tables 2 and 3.

- The electricity saving defined as:

$$ES = \frac{W_{net}}{P_{tot}} \tag{6}$$

- Energy reduction factors (both using as reference the total and net power of Option A) calculated as:

$$\Delta E_{tot} = \frac{P_{tot} - P_{tot}(\text{option A})}{P_{tot}(\text{option A})} \tag{7}$$

$$\Delta E_{net} = \frac{P_{tot-net} - P_{tot-net}(\text{option A})}{P_{tot-net}(\text{option A})} \tag{8}$$

Table 3 shows that energy demand for the compression of refrigerant(s) represents one of the most substantial energy consumptions for running the purification of biogas. The implementation of a steam turbine enables to cover almost 10% of the total electricity of the system. In addition, Options B and C introduce additional improvements to the benchmark (Option A). These two configurations further reduce the total energy demand thanks to a better layout of the refrigeration loop (Option B) and selection of the refrigerants (Option C). The energy reduction with regards to the benchmark is still limited, but the electricity power demand drops up to -4.9% and –

5.4% if you consider the either the total or the net energy demand as baseline, respectively. Though the energy savings could sound limited, but the steam cycle delivers 100% of the steam demand for the entire process. The end user does not have to burn extra methane or outsource fuel, i.e., biomass waste, to integrate the amount of steam. Beyond the technical aspect and comments related to the energy analysis of the system, it is important to stress that also economic aspects must be considered to identify the best process configuration. However, this is out of the scope of the present paper.

CONCLUSIONS AND FUTURE WORKS

The paper presented a novel technology for the upgrading of the biogas. Remarkably, the system allows for purifying bio-CH_4, while producing bio-CO_2 with purity of both products 99.99+ vol%, meeting specs for injection in pipeline (methane) and carbon use, including food and beverage, or permanent storage (CO_2). The proposed technology, patented by Cryo Inox, represents an example of full electrification of biogas upgrading. The system includes the bio-CH_4 liquefaction. Liquefaction can be considered for small clusters of bio-CH_4 producers. Indeed, liquid bio-CH_4 can be transported. Hence, the proposed process is ideal whenever natural gas pipelines are far from the biogas site and/or connecting to existing infrastructure is not feasible. The work showed that the recovery and valorisation of waste stream coming from the process becomes relevant to improve the key performance of the system. For instance, the valorisation of purges containing bio-CH_4 can be exploited to generate steam and heat. The energy analysis highlighted that the integration of the combustion chamber with a steam boiler and a steam cycle can benefit to the upgrading process. The optimization of the fully integrated steam cycle enables to recover up to 10% of the total electrical energy demand of the system and satisfy 100% of the total thermal energy demand (steam). In addition, the paper also showed the optimisation of the refrigeration strategy, and its configuration have an impact on the overall

Table 3: Energy analysis of the three options.

Option	HC mixture (kg/h)	NH_3 (kg/h)	Refrigerant reduction	HC mixture compression (kW)	NH_3 fluid Compression (kW)	Compression reduction
A	800	NA	Reference	70.8	NA	Reference
B	392.5 (mild) 326.7 (cryo) 755.2 (total)	NA	-5.6%	37.5 (mild loop) 22.6 (cryo loop) 66.1 (total)	NA	-6.2%
C	637.0 (cryo)	34.0 (mild)	-16.1%	yo loop)	5.90 (mild loop)	-13.0%

indicators. Three configurations to supply frigories have been investigated: the selection of both the process layout and the proper refrigeration affects the performance. Beyond these technical aspects, it is worth underlining that the analysis should not be limited to "engineering" aspect, i.e., energy recovery and process optimisation, but integrate the economics assessment to identify the best options among the candidates and account for the impact of process configuration on the profitability. This is not included the present work, but it will be investigated within the SEMPRE-BIO project.

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