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Design and Optimization of Processes for the Production of Liquid Biomethane

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Biogas is considered as a good alternative to natural gas as an environmentally-friendly source of energy. It can be turned into Liquid Biomethane (LBM) through an energy-intensive process that consists of two steps: biogas upgrading, which aims at removing CO_2, and biomethane liquefaction. Among the available upgrading technologies, low-temperature separation by distillation represents a promising solution for LBM production as the synergy of the two processes results in a low energy consumption. This work focuses on the design and optimization of a liquefaction process to be operated downstream of the upgrading step performed by means of the Ryan-Holmes low-temperature extractive distillation process. To this purpose, the most suitable liquefaction technology has been determined by parameters optimization that has been performed by integrating the Genetic Algorithm toolbox available in MATLAB® with Aspen HYSYS® V11 through the ActiveX technology. The less energy-demanding liquefaction schemes have turned out to be the dual pressure reverse nitrogen cycle and the Claude cycle, which are characterized by an energy consumption of 7.36 kWh/kmol$_{LBM}$ and 7.18 kWh/kmol$_{LBM}$, respectively.

1. Introduction

Natural gas has been playing a key role in the energy transition era (Spatolisano and Pellegrini, 2021) and biogas is considered as a good alternative to it as an environmentally-friendly source of energy. Biogas can be turned into Liquid Biomethane (LBM), which has been established to be a valid alternative to conventional fossil fuels when directly used in the heavy transportation sector. In light of these considerations, recent energy policies have been developed for promoting its production. LBM production is a costly and energy intensive operation that encompasses two key processes: upgrading, where biogas is converted into biomethane (BM) through CO_2 removal, and refrigeration, which transforms it into a liquid form. The CO_2 content in the biomethane to be liquefied cannot exceed 50 ppm to avoid freeze-out problems (De Guido and Spatolisano, 2021). Cryogenic upgrading stands out as one of the most promising technologies for biogas upgrading due to its ability to produce biomethane at desired liquefaction purities while minimizing methane losses. Moreover, the cooling requirements involved in the purification phase of biomethane are synergistic with its liquefaction process and are about half of those involved in conventional upgrading technologies (Pellegrini et al., 2018) typically based on chemical absorption into aqueous amine solutions (Moioli et al., 2024). Another advantage is represented by the high purity and pressure at which the CO_2 by-product is obtained, which enables its selling to the food and beverage industry or its use as a pre-cooling refrigerant to be integrated in the liquefaction phase without addition of any energy expenditure. Despite the importance cryogenic biogas upgrading has been gaining, refrigeration cycles are optimized to treat a biomethane stream that has undergone upgrading by a conventional process, which is available at ambient conditions. Therefore, there is the need to optimize the biomethane stream produced by these novel upgrading technologies. The objective of this work is to perform this optimization for different refrigeration cycles mainly based on nitrogen expansion and on the Claude cycle taking into account the biomethane liquefaction duty.

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2. Methods

Finding the optimal parameters of liquefaction cycles is of fundamental importance for the liquefaction gas industry. In the following, the adopted optimization method is described.
In general, gradient-based solvers such as the Newton method or gradient method are typically valuable tools for tackling optimization problems. However, they do encounter challenges when faced with highly non-linear objective functions, as is the case in liquefaction cycle optimization (Mitchell, 1998). To overcome this problem, the choice of the optimization method moved towards one of the optimization algorithms based on global search techniques. In this work, the widely recognized Genetic Algorithm (GA) has been chosen due to its prevalent usage in the literature for similar objectives (Abdelhamid et al., 2017). The algorithm has been applied thanks to the toolbox available in MATLAB®, while the simulation of the cycles has been performed in Aspen HYSYS® V11 (AspenTech, 2019). The link between MATLAB® and Aspen HYSYS® has been established through the ActiveX technology (COM), which creates a dialogue window through which MATLAB® and Aspen HYSYS® are capable to exchange variables (De Guido and Pellegrini, 2019).
Since the goal is to minimize the energy input required by the refrigeration cycles responsible for providing the necessary cooling duty for biomethane liquefaction, the objective function to be minimized has been defined according to Eq(1):

$$f_{ob}(\boldsymbol{x}) = \sum W_C - \sum W_T \tag{1}$$

where x is the vector of parameters to be optimized, while the two summations on the right-hand side, respectively, denote the overall power consumed by compressors and generated by turbines.
However, the optimal combination of decision variables must adhere to some feasibility constraints. One of them requires that the Minimum Temperature Approach (MITA) in heat exchangers exceeds a specified minimum threshold assumed to be 3 °C. Another constraint to be considered when dealing with inter-refrigerated compression consists in having a continuously increasing pressure across the compression process. The most suitable way to deal with this kind of constrained problems, especially when the optimization is operated through evolutionary algorithms, is to introduce a penalty function φ to be added to the original objective function, which consists in introducing a penalization to the original function whenever the specified constraint is violated. In this way, the optimization will be performed in the modified objective function, in which all constraints are included.
In this work, when dealing with the continuously increasing pressure constraint, a static penalty function with a constant value has been selected, as reported in Eq(2):

$$\varphi = C\delta \tag{2}$$

where δ=1 if the constraint is not met and δ=0 otherwise.
When considering the second constraint regarding the MITA, the penalty function φ(t) has been formulated (Eq(3)) in such a way that it could account for the "distance" from the admissible value since the objective is to keep the MITA as closer as possible to the minimum allowed one ($Minimum\ allowed\ MITA$ in Eq(4)).

$$\varphi(t) = \delta k t^{\theta} \text{ where } \begin{cases} \delta = 1\ if\ t > 0 \\ \delta = 0\ if\ t < 0 \end{cases} \tag{3}$$

$$t(x) = Minimum\ allowed\ MITA - MITA(x) \tag{4}$$

A work of calibration on the penalty function has been carried out through multiple optimizations on a simple reverse nitrogen cycle to determine the constants k and θ.
In this work, the liquefaction cycles have been all taken from the literature. However, since they were optimized for the liquefaction of a biomethane derived from conventional biogas upgrading, the reported optimized parameters were not suitable for the case analyzed in this work. At first, a simple reverse nitrogen cycle has been considered for optimization, since it has a low number of decision variables. Then, more complex cycles with a higher number of decision variables have been considered, as it will be shown in the following section.
The biomethane stream exiting the biogas upgrading section and fed to the liquefaction process has been considered as a methane-CO_2 binary mixture containing 50 ppm CO_2 at -87.4 °C and 40 bar (namely, the operating conditions it would be obtained by performing the upgrading step through the Ryan-Holmes extractive distillation process). Each refrigeration cycle is operated so that the produced LBM stream is available at -161.5 °C and atmospheric pressure.

3. Description and optimization of refrigeration cycles

3.1 Simple reverse nitrogen cycle

The simple reverse nitrogen cycle has been first analysed in this work due to its simplicity, though it has the poorest energy efficiency among the analysed cycles due to the high load assigned to a single compressor. With reference to the scheme illustrated in Figure 1, the following process decision variables have been considered for the optimization of this cycle: BM liquefaction pressure, P_2; maximum pressure in the refrigeration cycle, P_4; minimum pressure in the refrigeration cycle, P_7; refrigerant molar flow rate. In order to calibrate the penalty function and determine the values of k_1, k_2, θ_1, and θ_2 (to keep the MITA as closer as possible to the minimum allowed one in both the liquefier LNG-100 and the economizer LNG-101), several optimizations have been performed by changing the values of those parameters and checking whether the optimized result was respecting the constraint and how much the MITA of the two heat exchangers differed from the minimum allowed one. They have turned out to be: $k_1=10^7$, $k_2=10^{17}$, $\theta_1=3$ and $\theta_2=1$. The optimal value for the selected decision variables is reported in Table 1.

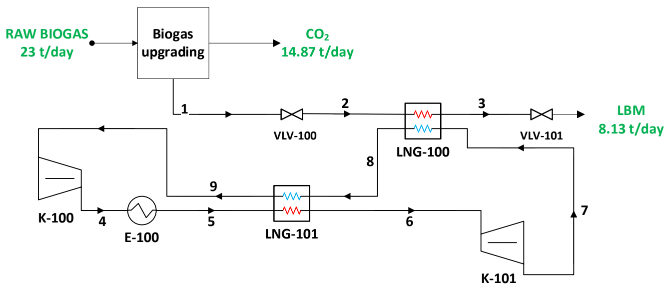

Figure 1: Scheme of the simple reverse nitrogen cycle

Table 1: Optimal decision variables for the simple reverse nitrogen cycle in Figure 1

Variable	Symbol	Optimal value
BM liquefaction pressure, kPa	P_2	1171
Max. refrigeration cycle pressure, kPa	P_4	1616
Min. refrigeration cycle pressure, kPa	P_7	343
Refrigerant molar flow rate, kmol/h	$\dot{n}_{N_2}$	165.4

3.2 Inter-refrigerated reverse nitrogen cycle

Inter-refrigerated reverse nitrogen cycle bears similarities to the simple reverse nitrogen cycle, except for the refrigerant compression part, where a 2-stage intercooled compression is used to decrease the overall compression power, as shown in Figure 2. With reference to this figure, the optimization variables for this case are the following ones: BM liquefaction pressure, P_2; maximum pressure in the refrigeration cycle, P_6; minimum pressure in the refrigeration cycle, P_9; intermediate compression pressure, P_4; refrigerant molar flow rate. In this case, when the intermediate pressure is higher than the maximum pressure of the cycle, a fixed value of 10^{16} has been given to the objective function as penalization (Eq(2)). The optimal value for the selected decision variables is reported in Table 2.

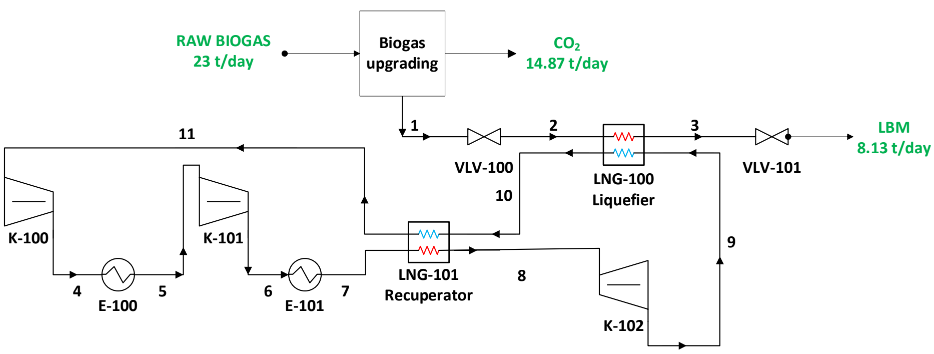

Figure 2: Scheme of the inter-refrigerated reverse nitrogen cycle

Table 2: Optimal decision variables for the inter-refrigerated reverse nitrogen cycle in Figure 2

Variable	Symbol	Optimal value
BM liquefaction pressure, kPa	P_2	3360
Max. refrigeration cycle pressure, kPa	P_6	6881
Min. refrigeration cycle pressure, kPa	P_9	564
Intermediate compression pressure, kPa	P_4	2000
Refrigerant molar flow rate, kmol/h	$\dot{n}_{N_2}$	87.58

3.3 Dual pressure reverse nitrogen cycle

The dual pressure reverse nitrogen cycle has the advantage to generate less irreversibility thanks to a greater matching between the cold refrigeration curve and the warmer one and a lower compression work since only part of the refrigerant flow rate is expanded to the minimum pressure of the refrigeration cycle. The scheme adopted for this technology for biomethane liquefaction has been taken from the work by Baccioli et al. (Baccioli et al., 2018) and is illustrated in Figure 3. The optimal value of the decision variables is reported in Table 3.

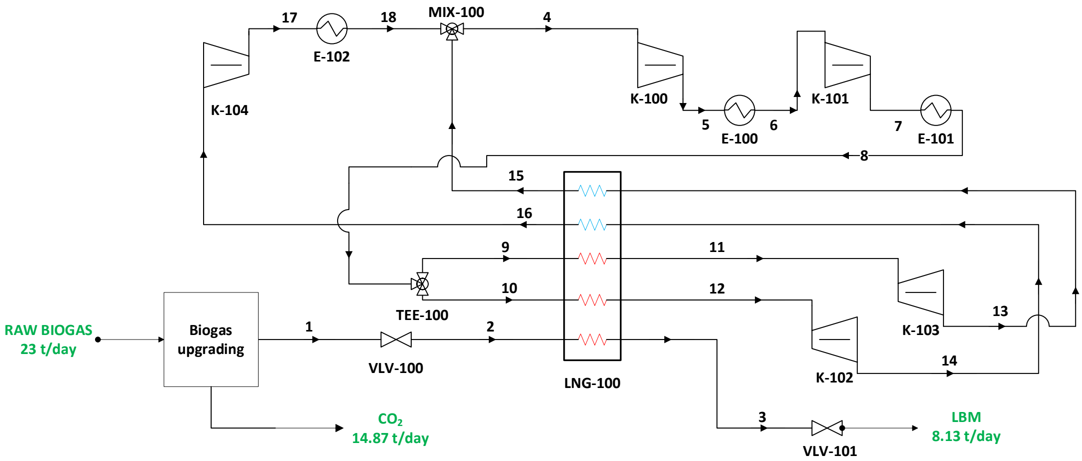

Figure 3: Scheme of the dual pressure reverse nitrogen cycle

Table 3: Optimal decision variables for the dual pressure reverse nitrogen cycle in Figure 3

Variable	Symbol	Optimal value
BM liquefaction pressure, kPa	P_2	3999.4
Max. refrigeration cycle pressure, kPa	P_7	7560.7
Min. refrigeration cycle pressure, kPa	P_{14}	935.20
Intermediate compression pressure, kPa	P_5	3848.50
Lower pressure in the high-pressure stream, kPa	P_{13}	1950.40
Temperature outlet after precooling, °C	T_{11}	-64.73
Temperature outlet after precooling, °C	T_{12}	-90.80
Temperature outlet of the low-pressure stream after heat exchange, °C	T_{16}	26.48
Split factor	TEE-100	0.3
Refrigerant molar flow rate, kmol/h	$\dot{n}_{N_2}$	72.73

3.4 . Claude cycle

The Claude cycle is a refrigeration system where cooling duty is given by the expansion of biomethane itself. The idea of analysing this cycle comes from the fact that the thermodynamic conditions of the biomethane generated in the upgrading step are theoretically favourable for its expansion since the gas is already at high pressure (i.e., 40 bar). The Claude scheme used for liquefaction has been adapted from the one presented in the work by Capra et al. (Capra et al., 2019) to account for the different temperature and pressure of BM at the refrigeration cycle inlet. Indeed, in this work, the recycled biomethane (stream #21 in Figure 4) and the upgraded biogas (stream #1 in Figure 4) are not at the same temperature and mixing the two streams would result in the generation of high irreversibility. Moreover, as the recycled gaseous biomethane stream has a higher flowrate than the upgraded one, a high amount of refrigeration power would be lost. Therefore, the cooling potential of the inlet stream has been exploited in the multi-stream heat exchanger LNG-100 before mixing it with the recycled stream, as shown in Figure 4. The optimal value of the decision variables for this cycle is reported in Table 4.

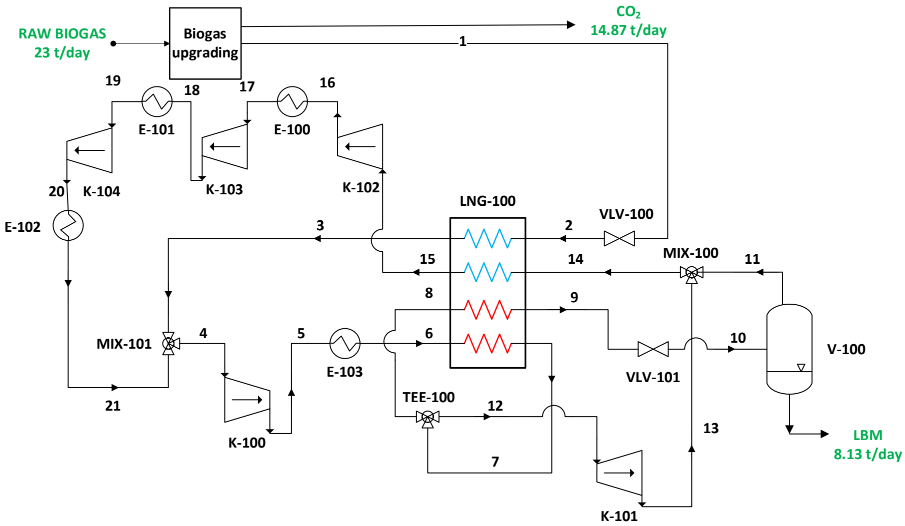

Figure 4: Scheme of the Claude cycle

Table 4: Optimal decision variables for the Claude cycle in Figure 4

Variable	Symbol	Optimal value
BM pressure at LNG-100 inlet, kPa	P_2	3761.30
Max. refrigeration cycle pressure, kPa	P_5	7438.70
Temperature after BM precooling, °C	T_7	-18.9
Temperature before VLV-101 valve, kPa	T_9	-157.40
Intermediate compression pressure, kPa	P_{18}	1100.00
BM molar flow rate to K-101, kmol/h	$\dot{n}_{12}$	37.56

4. Results

The bar chart in Figure 5 summarizes the specific energy consumption results of the analysed refrigeration cycles presented in the previous section. The energy consumption of the inter-refrigerated reverse nitrogen cycle demonstrates a reduction of approximately 50% compared to the value reported by Capra et al. (Capra et al., 2019) for the same technology, which was noted as 19.16 kWh/kmol. This energy saving is notable and aligns well with the reduction in energy required for liquefaction when comparing a conventionally upgraded gas to the Ryan-Holmes upgraded gas. The energy savings are even higher for the Claude cycle case, where Capra et al. (Capra et al., 2019) reports a cost-optimized energy consumption of 18.59 kWh/kmol. For the dual pressure reverse nitrogen cycle, Baccioli et al. (Baccioli et al., 2018) report an energy optimized value of 12.04 kWh/kmol$_{LBM}$ for the liquefaction of a conventionally upgraded BM, whereas the result obtained in this work is 7.36 kWh/kmol$_{LBM}$. Therefore, the obtained energy saving is approximately 39%. This is due to the cooling effect that can be obtained without the requirement of any further mechanical work.

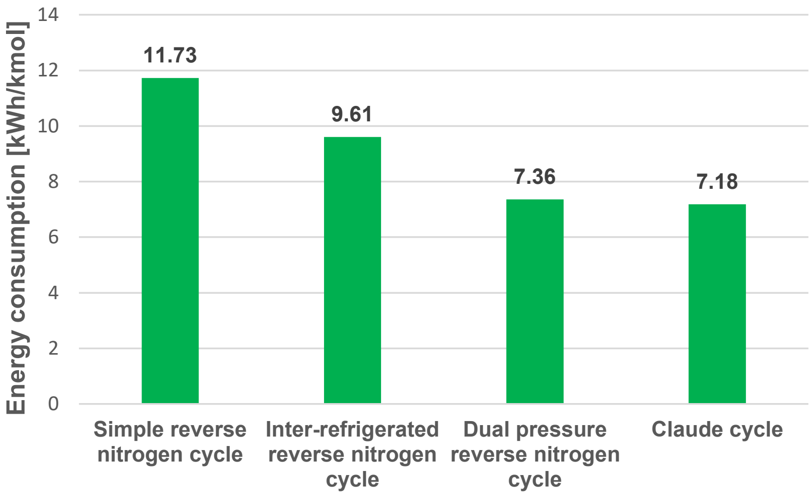

Figure 5: Specific energy consumption per kmol of LBM for all the analyzed refrigeration cycles

5. Conclusions

The technologies available for liquefaction of biomethane are typically optimized considering the need for treating a gas stream that has undergone upgrading by conventional processes based on chemical absorption. The increasing interest in low-temperature/cryogenic technologies for biogas upgrading requires to optimize liquefaction technologies in order to treat a gas stream available at lower temperatures. This work has focused on this optimization considering different refrigeration cycles mainly based on nitrogen expansion and on the Claude cycle. Their liquefaction duty has been minimized by means of the Genetic Algorithm. As a result, the less energy-demanding liquefaction schemes have turned out to be the dual pressure reverse nitrogen cycle and the Claude cycle, which are characterized by an energy consumption of 7.36 $kWh/kmol_{LBM}$ and 7.18 $kWh/kmol_{LBM}$, respectively. In a future work, these liquefaction processes will be integrated with the upgrading section in order to design and optimize a refrigeration process capable of supplying not only the cooling duty necessary for biomethane liquefaction but also the cooling duty required by the upgrading section.

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