

# Exergy Analysis, Optimization Approach and Transient Mode Operation Study of Non-Flammable Biomethane Liquefaction Process

Imane Oudghiri , Rodrigo R. Tinoco\*, Chakib Bouallou

Centre for Energy Efficiency of Systems (CES), Mines ParisTech, PSL Research University, Palaiseau, 91120, France  
 rodrigo.rivera\_tinoco@mines-paristech.fr

Since the advent of the industrial age, the planetary energy demand has substantially increased to improve daily comfort. In a view to counterbalance the growing energy consumption of fossil fuels, such as natural gas, various strategies have been deployed for increasing the share of renewable energy (i.e., solar, wind, biogas...) in the global energy mix. A particular interest is paid to the use of biogas as natural gas substitute. For producers geographically located far from natural gas grid infrastructures, biogas shall be liquefied to facilitate its transport.

Existing mixed refrigerant liquefaction units, operate with flammable hydrocarbons. However, increasingly stringent safety requirements impel to predict the inherent risks of handling hydrocarbons, in particular in biogas units close to farms. The use of non flammable alternative refrigerants is a potential solution currently barely considered. In this paper, a novel mixed refrigerant cycle for biogas micro-liquefaction process using a non flammable mixed refrigerant is proposed. The proposed cycle is first simulated in Aspen HYSYS® and further assessed by the exergy analysis method in order to evaluate process performance, to locate exergy destruction and to define key parameters for optimization approach. In attempt to improve the energy efficiency of the cycle, a link between Aspen HYSYS®, PIKAIA genetic algorithm tools (Charbonneau, 2002) and Microsoft Visual Basic was developed. In the proposed methodology the objective function consists on the maximization of the amount of the liquefied biomethane per unit of the required compressor work. The combination of exergy analysis and genetic algorithm optimization improved the proposed architecture performance leading to higher energy efficiency. Actually the operating parameters optimization contributed, in comparison to the reference case, to a 27 % reduction in electric power consumption and 26 % in exergy destruction. As a second step and with the aim of studying the transient-mode operation of liquefaction units, results obtained for a steady state operation are considered in order to assess the suitability of optimized solutions and robustness of systems against quick start-and-stop or partial load operating mode.

## 1. Introduction

With the increasing demand of natural gas, as a clean and affordable fossil energy resource, the existing reserves are depleting or leading to long distance transport of LNG. Consequently, and mainly for energy security, the use of biogas as a renewable version of natural gas (NG) is increasing. Biogas covers a wide variety of gases, mainly methane and carbon dioxide. The biogas has the advantage of producing energy in short circuits (farm-cities), with lower emissions compared to the use of heavy hydrocarbons and coal, and recoverable in very varied power units (cogeneration, engine-generators, boilers, power plants). The easiest way for using biogas is as fuel for producing electricity and heating, because besides water removal, no pretreatment is required (Hoo et al., 2014). Another possibility for using biogas is as a natural gas substitute. In this case, the methane is the only recoverable fraction, also called biomethane. CO<sub>2</sub> and other impurities must be removed. For producers located geographically far from natural gas grid infrastructure, the most profitable way of transportation for the biomethane is as liquefied biomethane (LBM).

Due to the similarities between the biogas and NG, NG liquefaction processes are considered as reference to build adapted processes for biomethane. Moreover, it is also considered the use of non-flammable refrigerants in order to reduce risks of explosion in biogas units. To date, several NG liquefaction technologies exist. Three groups are distinguished: Cascade cycle with pure refrigerant, Mixed Refrigerant (MR) and  $N_2$  expander processes (Venkatarathnam, 2010). Among them, the MR and  $N_2$  expander processes are the most suitable for biomethane liquefaction plants at micro-sale. In this work, the MR cycle is considered for its higher energy efficiency and relatively lower investment cost. There are three kinds of mixed refrigerant liquefaction processes, namely Single Mixed Refrigerant (SMR), Dual Mixed Refrigerant (DMR) and pre-cooling mixed refrigerant liquefaction process (e.g.  $C_3$ MR process) (He et al., 2018). The SMR is the most frequently employed process in the natural gas liquefaction industry at small scale because it is simple and it uses only one mixed refrigerant.

The liquefaction unit operation is the most energy intensive step in NG value chain (Khan et al., 2017). Hence, several studies have been conducted for the optimization of NG liquefaction processes (Austbo et al., 2014). The optimization methods in related articles are classified into three categories namely, Stochastic, Deterministic and Hybrid methodologies (methods including both stochastic and deterministic approaches). Among them, Genetic Algorithms (GA) are the most adopted stochastic methods in optimization problems.

The robustness of NG liquefaction processes is evaluated by the system responses to disturbances including quick start-and-stop or partial load operation mode. The most of the research related to MR liquefaction processes consists mainly on the thermodynamic evaluation and the optimization of the process performances at steady state (Lee et al., 2015). Only a few studies have focused on transient operation mode of the NG liquefaction processes and all of them are using flammable refrigerants.

The objective of this study is to develop a non-flammable SMR for biomethane liquefaction at micro-scale and to further assess its performance upon load variations. In attempt to designing a high performance liquefaction process, an exergy analysis is performed as a preliminary diagnostic for the system. Then a new optimization methodology developed in a previous work is applied. Once the steady state simulation and optimization are achieved, a study of the behavior of the process at partial load operating mode is conducted.

## 2. Process description

The SMR liquefaction process for micro-scale LBM plant is illustrated in Figure 1. In general, the MR process employs a mixture of flammable refrigerant including light hydrocarbons and Nitrogen. In this study a non-flammable MR is considered. It consists of a mixture of three non-flammable refrigerants namely for confidentiality purposes as,  $R_1$ ,  $R_2$  and  $R_3$ . The process includes two blocks: the MR and the biomethane blocks.

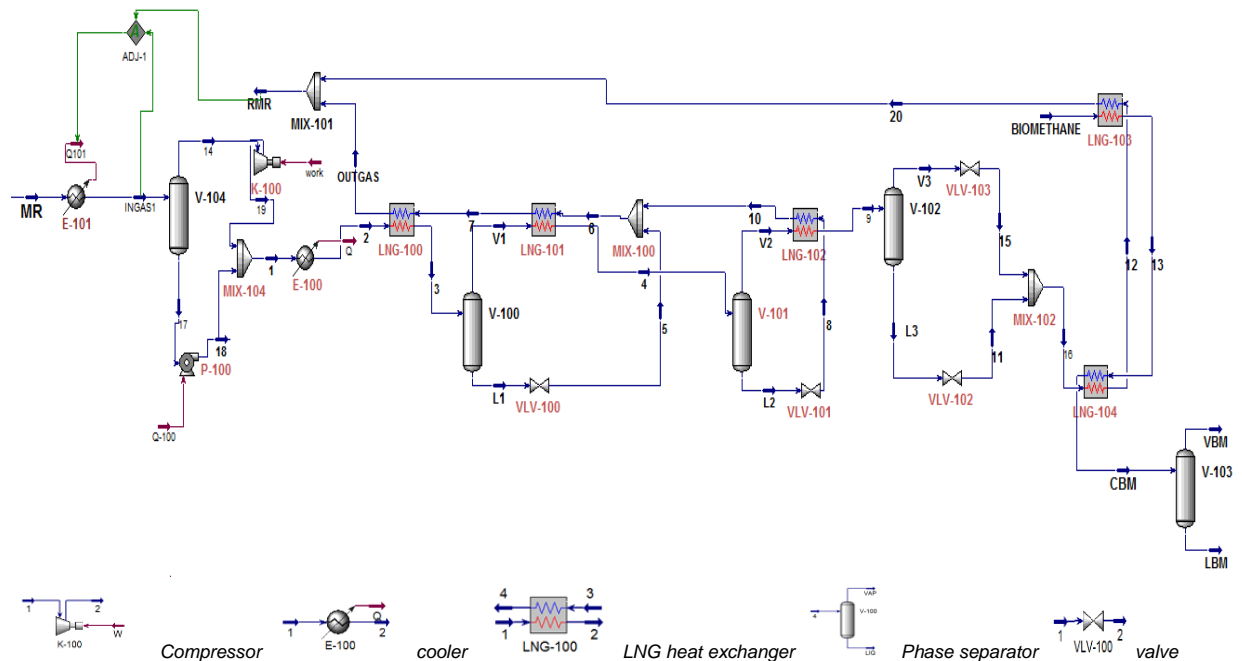


Figure 1: Flowsheet of the proposed SMR

In the first one, the MR is pressurized by the compressor to 11.25 bar(abs) and then pre-cooled in water cooler to 4 °C. After that, it is cooled in the first LNG heat exchanger (LNG-100). The outlet stream is a biphasic flow. Consequently, it enters a vapour liquid separator (V-100) to separate liquid and vapor MR. The liquid phase is throttled in the valve (VLV-1) and then returned back to the LNG-100. The vapor phase is conveyed to the second LNG heat exchanger (LNG-101) and then enters the separator V-101. The same operation occurs in the third heat exchanger (LNG-102). In the steady state simulation, all components of mixed refrigerant are condensed at this point. However in the transient mode, it is difficult to achieve this constraint. Hence another separator is added to the process. Both vapor and liquid phases are throttled and mixed and then sent to the second block.

The second block of the process includes two LNG heat exchangers (LNG-103) and (LNG-104). The first heat exchanger provides sensible heat for biomethane pre-cooling at 17 bar and the second one provides a latent heat for complete condensation.

### 3. Exergy analysis

The exergy of a given system is defined as the maximum shaft work that can be done by the system and at a specified reference environment (Dincer and Rosen, 2007). The specific exergy of a fluid is derived from the second law of thermodynamics and it is expressed in terms of specific enthalpy and entropy as expressed in Eq(1) (Figueroa-Jimenez et al., 2015).

$$ex = (h - h_0) - T_0 (s - s_0) \quad (1)$$

Where  $h_0$  and  $s_0$  refer to the enthalpy and the entropy of the fluid stream at the reference state (Venkatarathnam, 2010). In the current study,  $T_0 = 25$  °C and  $P_0 = 1$  bar have been considered as the reference state conditions for all exergy calculations.

The exergy analysis is a useful method for evaluating process performance and identification of irreversibilities in a system. In this study, it is applied to locate exergy destruction that occurs within the unit operations of the SMR process, and to identify decision variables for optimizing the process. Exergy destruction equations at each unit operation in the biomethane liquefaction process are listed in Table 1.

Table 1: Exergy destruction equations per unitary operation in the SMR process

Equipments	Exergy destruction (kW)
Compressor	$\Delta_{ex} = m(ex_{in} - ex_{out}) - W$
Cooler	$\Delta_{ex} = m(ex_{in} - ex_{out})$
LNG heat exchanger	$\sum_{i=1}^n m_i(ex_{i,in} - ex_{i,out})$
Phase separator	$\Delta_{ex} = m_{in}ex_{in} + m_{VAP}ex_{VAP} - m_{LIQ}ex_{LIQ}$
Throttle valve	$\Delta_{ex} = m(ex_{in} - ex_{out})$
Stream mixer	$\Delta_{ex} = m ex_{1,in} + m ex_{2,in} - m ex_{3,out}$

## 4. Simulation and optimization approach

### 4.1 Modeling and steady state simulation

The steady state simulation was performed in Aspen HYSYS® using Peng Robinson as thermodynamic model for the vapor liquid equilibrium calculation. The preliminary molar composition of the mixed refrigerant is considered to be 52.17 % R<sub>1</sub>, 17.39 % R<sub>2</sub> and 30.43 % R<sub>3</sub>. The molar composition, pressure and flow rate of the biomethane are considered to be 100 % Methane, 17 bar and 0.45 kmol/h.

Calculations of the exergy destruction are performed by extracting enthalpy and entropy values from Aspen HYSYS® and then being sent to Excel via the VBA macro. Then the exergy destruction calculations are conducted following the equations given in Table 1.

### 4.2 Decision variables and constraints

The target is to maximize the amount of liquefied methane per unit of electric power supplied to the process. The function includes some penalties if calculations lead to unfeasible operating parameters. Such penalties are based on the constraints presented in Table 2 and have the following impact on the optimization:

- If the minimum temperature approach in the LNG heat exchangers is lower than the constraint, a penalty of 5 is considered.

- If the temperature of the pressure discharge stream is higher than the constraint, a penalty of 5 is considered.

The target function is expressed in Eq(2); it represents the flow of the biomethane divided by the electric power supplied to the process, minus the accumulation of penalties.

$$f_{\max} = \frac{\dot{m}_{\text{Biomethane}}}{W} - \sum \text{penalties} \quad (2)$$

Decision variables were identified referring to the exergy analysis results. The detailed method is presented in the work of Oudghiri and Rivera-Tinoco (2018). In the current optimization, the molar flow rate of each refrigerant, the compressor discharge pressure (HP), the compressor suction pressure (BP) and the outlet temperature of each LNG heat exchanger are considered as the most relevant parameters for the SMR process optimization.

Table 2: Constraints for SMR process optimization

Equipment/streams	Constraint	Penalty value
Compressor	Temperature of discharge stream lower than 120°C	5
	Temperature of the MR stream lower than the dew point	5
LNG heat exchanger	Minimum temperature approach equal to 2.8°C	5
Stream 20	Vapor fraction equal to 1	5
Stream 9	Vapor fraction equal to 0	5
Stream LBM	LBM vapor fraction equal to 0	1

### 4.3 The optimization methodology

For the process optimization a link between Aspen HYSYS®, Excel and Genetic Algorithm library (PIKAIA) is developed (Oudghiri and Rivera-Tinoco, 2018). The GA library creates a population of vectors. These vectors contain random values within the specified ranges of the decision variables. A VBA macro sends the vector one by one to Aspen HYSYS® and request simulation to be run. Once the simulation is achieved, the same VBA MACRO calls the results from Aspen HYSYS® and calculates the value of the target objective function for each vector. After that, results are sent to PIKAIA. Then, the PIKAIA subroutine ranks the results in decreasing order and proceeds to create the new population by crossing-over, mutation and keeping some few vectors from the former population.

### 4.4 Optimization results

Table 3 and 4 represent the operating parameters and resulting values for electric power needed and specific energy consumption for the SMR process.

Table 3: bases and optimized values for operating parameters of the SMR process

Variables	Reference values	Optimized values	Relative variation rate, %
HP (bar)	14.98	11.52	23.04
LP (bar)	2.01	2.025	-0.746
m <sub>R1</sub> (kmol/h)	0.5298	0.4822	8.98
m <sub>R2</sub> (kmol/h)	0.202	0.2651	-31.18
m <sub>R3</sub> (kmol/h)	0.2691	0.2527	6.09
T <sub>3</sub> (°C)	-8.7	-0.4	95.4
T <sub>4</sub> (°C)	-59.5	-61	-2.5
T <sub>9</sub> (°C)	-79.65	-82.48	-3.7

Table 4: Process performances results

Process performances	Reference values	Optimized values
Compressor power (kW)	3.9	2.812
Exergy destruction (kW)	3	1.96
kWh/kg of liquefied biomethane	0.57	0.42

As shown in Table 3 and Table 4, the optimization made a significant change to the composition of the MR, the HP and the T<sub>3</sub> after the optimization. The compressor pressure discharge (HP) rate was decreased by almost 23.04 %, the T<sub>3</sub> was increased by 95.4 % and the R<sub>2</sub> flow rate was increased by 31.18 %. Globally,

performance shows a reduction of 27 % relative of electric power and 26 % relative to the specific energy consumption (kWh/kg of liquefied biomethane). Exergy destruction is decreased by approximately 34 %. Figure 2 shows the distribution of exergy destruction per unitary operation.

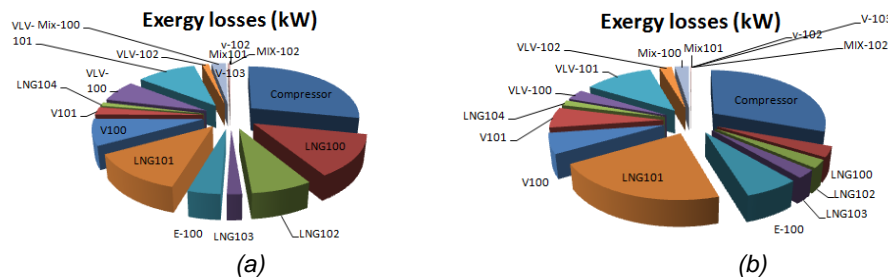


Figure 2: Exergy destruction shares for (a) the reference and (b) the optimized cases of the SMR proposed MR cycle

Once the optimized operating conditions are obtained from the VBA-Aspen-PIKAIA code, the process simulation is carried out only in Aspen HYSYS®. At this point, a sensitivity study is carried out to assess the performance of the process in partial load of methane is available.

## 5. Transient simulation

Natural gas liquefaction processes are prone to different types of disturbance, such as variations of feed gas temperature, pressure, flow rate and composition (He and Ju, 2016). In this section, the transient response of the system to biomethane flow rate variation is presented. The feed gas flow rate is decreased. The aim of this study is to evaluate the flexibility of the system at methane partial load without changing any equipment. In order to handle this variation, only compressor pressure discharge is manipulated. Figure 3 presents the transient response of the HP, the electric power consumption and the specific power consumption of the SMR cycle at saturation and sub-cooling points.

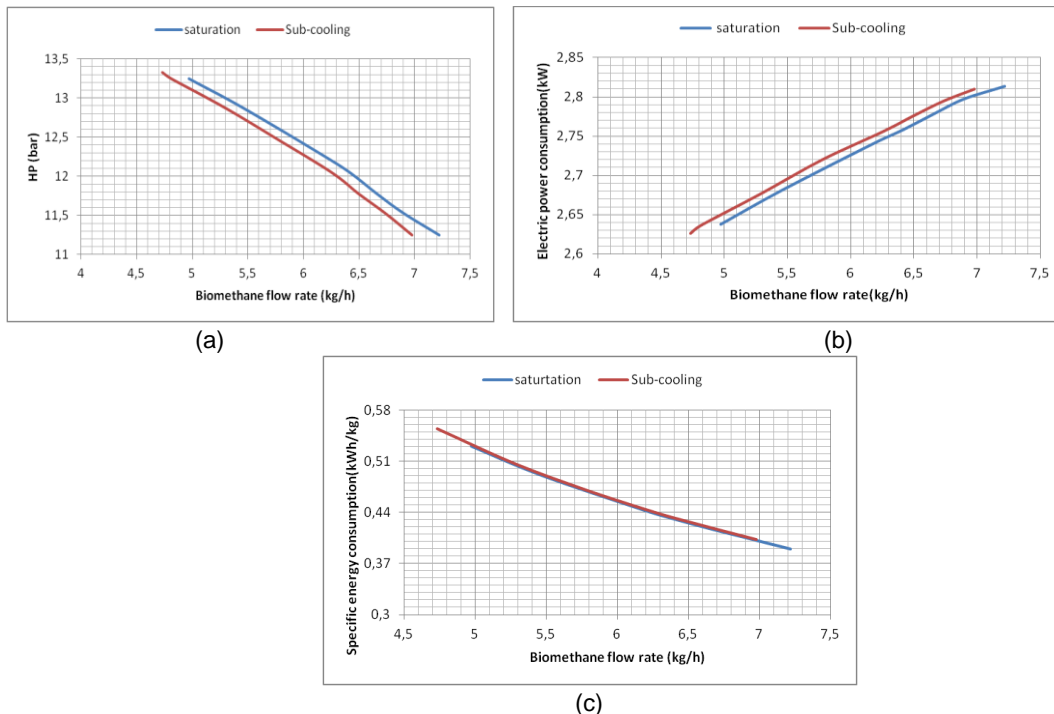


Figure 2: Transient response of the HP (a), the electric power consumption (b) and the specific energy consumption (c) of the SMR process at saturation and sub-cooling point

Saturation and sub-cooling point are both considered to identify the operating interval pressure of the cycle. By decreasing the biomethane flow rate, the compressor pressure discharge (HP) must be increased to handle temperature cross over in LNG-104 heat exchanger and to achieve the condensation of the biomethane flow rate. The performance of the process increased by decreasing the biomethane flow rate: in fact by increasing the HP, partial condensation is favored at each stage of the SMR cycle. This leads the flow returning back to the compressor to be colder and hence increasing its volumetric density. The result is lower consumption of electricity.

## 6. Conclusion

In this study, a novel process operating with non-flammable refrigerants for micro-scale LBM plant was developed. The process is a single mixed refrigerant for biomethane liquefaction cycle. The process is simulated by Aspen HYSYS® and assisted by the exergy analysis for irreversibility identification. For process optimization, a new optimization approach, developed in progress work, was applied. This methodology integrates PIKAIA Genetic Algorithm tools and Microsoft Visual studio. The objective function is to maximize the amount of liquefied biomethane per unit of electric power supplied to the process. After the optimization, the electric power consumption and the specific energy consumption was decreased respectively about 27 % and 26 % compared to the base case. As a regard the exergy destruction, a maximum of 34 % is reduced against the reference case. In order to evaluate the flexibility of the system to biomethane flow rate variation, a preliminary transient study was conducted. This analysis shows the responses of LBM discharge pressure, electric power consumption and the specific energy consumption after decreasing the biomethane flow rate by 34 %. It is noted that in order to handle the biomethane flow rate disturbance, the compressor discharge pressure must be adjusted leading to important variations in process performances.

## References

- Austbo B., Lovseth S., Gunderson T., 2014, Annotated bibliography-use of optimization in lng process design and operation, *Computers & Chemical Engineering*, 71, 391-414.
- Charbonneau P., 2002, An introduction to genetic algorithms for numerical optimization, NCAR Technical Note, USA.
- Dincer I., Rosen M.A., 2007, *Exergy: energy, environment and sustainable development*, El Sevier Science, ISBN-10: 0080445292.
- Figueroa-Jimenez S., Gamarra-Torres J., Bonilla-Correa D., Peralta-Ruiz Y., 2015, Evaluation of biodiesel production process from palm oil (eleais guineesis) using exergy analysis methodology, *Chemical Engineering Transactions*, 529-534.
- Venkatarathnam G., 2010, *Cryogenic mixed refrigerant processes*, Springer, Berlin, Germany.
- He T., Ju Y., 2016, Dynamic simulation of mixed refrigerant process for small-scale LNG plant skid mount packages, *Energy*, 97, 350-358.
- He T., Karimi I.A., Ju Y., 2018, Design and optimization of natural gas liquefaction processes for onshore and offshore applications, *Energy*, In Press.
- Hoo P.Y., Hashim H., Ho W.S., Tan S.T., 2014, Successful biogas implementation-a mini-review on biogas utilization, energy policies and economic incentives, *Chemical Engineering Transactions*, 1496-1500.
- Oudghiri I., Rivera-Tinoco R., 2018. Exergy analysis, optimization approach study of biomethane liquefaction processes, 31<sup>st</sup> International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems, Portugal.
- Khan M.S., Karimi I., Wood D.A., 2017, Retrospective and future perspective of natural gas liquefaction and optimization technologies contributing to efficient LNG supply: A review, *Journal of Natural Gas Science and Engineering*, 165-188.
- Lee C.J., Song K., Lee Y., Han C., 2015, A decomposition methodology for dynamic modeling of cold box in offshore natural gas liquefaction process, *Computers & Chemical Engineering*, 546-557.