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Optimization of an Absorption-Based Biogas Upgrading and Liquefaction Process

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The present work proposes a methodology for optimization of a liquefied biomethane (LBM) production plant. The LBM production plant comprises amine-based absorption upgrading followed by a single expander refrigeration cycle. The processes were modeled using Aspen HYSYS® and optimized through a Sequential Quadratic Programming algorithm. Any changes in the operating conditions of the upgrading process will affect the cooling demand in the liquefaction, while the opposite is not true. Based on this, a sequential optimization approach starting with the upgrading process is proposed. In order to accommodate the connection between the processes, different objective functions were formulated for the sequential optimization approach. The results from the sequential approach were compared with an overall optimization approach, where the entire LBM plant was optimized simultaneously. The results indicate that the same solution was obtained both for the sequential approach and the simultaneous approach. For the sequential approach, however, the best result was observed when the interaction between the upgrading and liquefaction processes was accounted for by considering the effect of the upgrading process on the exergy requirement in the liquefaction process.

1. Introduction

Mitigation of CO_2 from the transportation sector is challenging as fossil fuels are still dominant. Facilitating the use of alternative fuels characterized by higher energy density increases the share of sustainable energy in this sector (REN21, 2018). As an alternative fuel for heavy-duty vehicles, liquefied biomethane (LBM) produced from biogas has gained much interest because it can replace liquefied natural gas (LNG). However, production of LBM involves two energy intensive processes: biogas upgrading and liquefaction.

In biogas upgrading, the amount of CO_2 and trace compounds is reduced in order to produce high quality biomethane. Amine-based absorption is a widely used technology for gas separation in various industrial applications that can also be applied for biogas upgrading. As opposed to alternative upgrading methods such as membrane separation, pressure swing adsorption or water scrubbers, biogas upgrading through amine-based absorption can satisfy the specific purification requirements in LBM production (i.e. CO_2 content below 50 ppm (Bauer et al., 2013)) without additional polishing steps (Hashemi et al., 2019).

The energy supply to an LBM production plant with absorption upgrading consists of compression work and heat for amine regeneration. Law et al. (2017) optimized the energy and CO_2 removal efficiency of an absorption unit, observing large reductions in operating cost. Maile et al. (2017) conducted experiments regarding biogas upgrading through amine-based absorption. They showed that the CO_2 removal from the biogas mixture increased as the temperature in the absorber increased. Dara and Berrouk (2017) indicated that the trade-off between solubility of the CO_2 in the chemical solvent and the kinetic of chemical reaction determined the optimum temperature of the lean amine solution for maximum CO_2 removal. Øi et al. (2014) minimized the energy use of different chemical absorption configurations, for which the best result was observed in a vapor recompression configuration.

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Once high quality biomethane is obtained from the upgrading, biomethane is liquefied through a refrigeration cycle. Refrigeration cycles are well studied in literature in terms of not only process design but also energy optimization (Austbø et al., 2014). However, studies regarding the combination of a refrigeration cycle with other processes, such as biogas upgrading, has received limited attention in literature. The present work aims to develop an optimization methodology in order to minimize the exergy supply for a LBM production plant comprising amine-based absorption upgrading and a single expander refrigeration cycle. The processes are simulated using Aspen HYSYS® and optimized using a Sequential Quadratic Programming (SQP) algorithm.

2. Process description

A detailed LBM production plant layout is presented in Figure 1. The plant consists of an amine-based absorption upgrading process followed by a single expander refrigeration cycle. These two processes are connected through high quality biomethane and CO_2 streams. A detailed process description is available in the work by Hashemi et al. (2019). Biomethane and CO_2 in liquid form are considered as final product and byproduct, respectively, from the plant.
Once raw biogas is compressed in the compression unit, it enters the bottom of the absorber column and interacts with lean amine solvent from the top of the column in order to obtain high quality biomethane. Rich amine solvent from the bottom of the absorber column is depressurized through an expansion valve. After precooling a recycled lean amine solvent stream, the low-pressure rich amine solvent enters a stripper column where amine is regenerated by adding heat in the reboiler at the bottom of the stripper. The top product of the stripper column is high purity CO_2, whereas regenerated amine from the bottom of the stripper column is recycled to the absorber column. In order to compensate water and amine losses in columns, a make-up unit is considered. Moreover, cooling water is used to reduce the temperature of the lean amine solvent before it enters the absorber. Here, methyl diethanolamine (MDEA) is used as solvent.
In order to avoid ice formation during liquefaction, water is removed from the high quality biomethane and CO_2 streams in dehydration units before being sent to the liquefaction process. Here, a single expander refrigeration cycle with nitrogen as working fluid is considered. After liquefaction, the LBM stream is expanded to atmospheric pressure. The work and heat requirements in the plant are implemented independently without considering the potential energy integration.

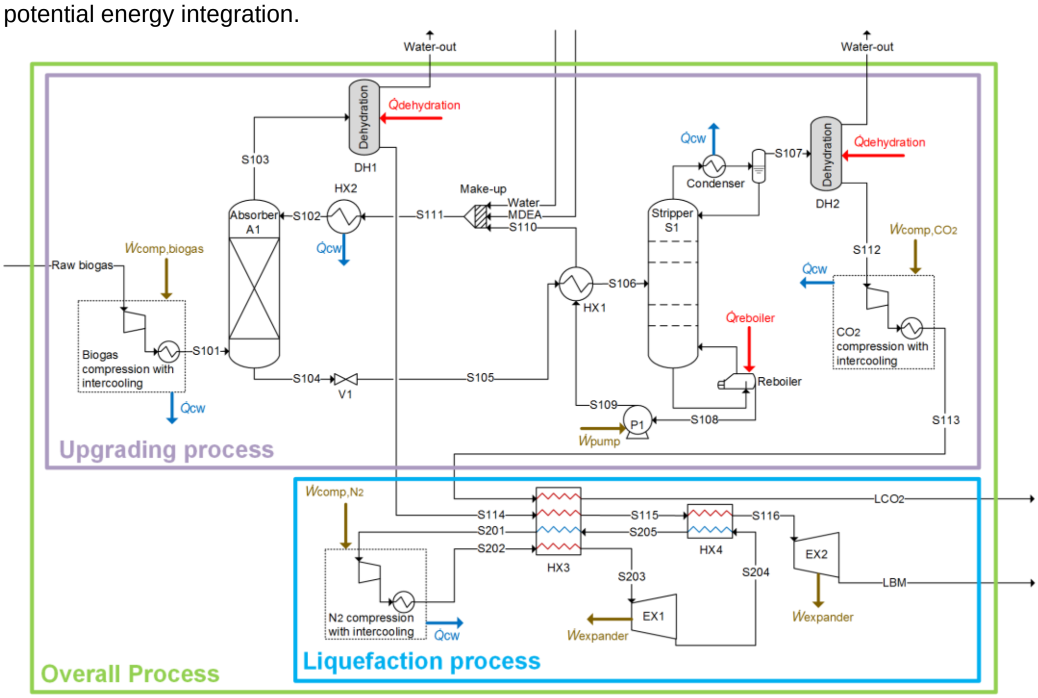

Figure 1: LBM production plant layout and different process boundaries

3. Methodology

3.1 Process modeling

The LBM production plant was simulated with Aspen HYSYS® (Aspen Technology Inc., V9.0). The Soave-Redlich-Kwong (SRK) equation of state was employed for the biogas mixture in the compression units and the

refrigeration cycle, whereas the "Acid gas – chemical solvent" package was used for the absorption process (AspenTech, 2017). The raw biogas stream contained 60 mol% CH_4, 39.9 mol% CO_2 and 0.1 mol% H_2S, with a molar flow rate of 1000 kmol/h at 35 °C and atmospheric pressure. It was assumed that the LBM was produced at atmospheric pressure with CO_2 content below 50 ppm. The LCO_2 comprised all the H_2S from the raw biogas, at 35 °C and 110 bar, which are suitable conditions for CO_2 pipeline transportation (Yousef et al., 2016).
In order to ensure satisfying the CO_2 content specification, the absorber and the stripper had 25 and 20 theoretical trays, respectively (Hashemi et al., 2019). The lean amine solvent was introduced at the 9th stage from the top of the stripper column. For the stripper, a reflux ratio of 1.25 and 95 mol% of CO_2 in the top stream were specified, which differs from what were considered in the work by Hashemi et al. (2019). MDEA with a concentration of 45 wt% was considered. In order to improve the kinetics of the chemical reaction between CO_2 and MDEA, the inlet temperature of lean amine solvent to the absorber was 10 °C higher than the temperature of the compressed raw biogas (Lange et al., 2015). The inflow streams entered the absorber column with identical pressure.
All the water present in the biomethane stream from the absorber and the CO_2 stream from the stripper was removed in the dehydration units. The dehydration units were based on tri-ethylene-glycol (TEG) absorber/regeneration columns, where the TEG regeneration temperature was assumed to be 200 °C and the outlet temperature of the dehydration units 35 °C. The heat requirement in the reboiler of the TEG regeneration column was calculated according to Hashemi et al. (2019). Furthermore, the following assumptions were taken into account in model simulations:

- Gas compression units were treated as four-stage compressors with identical pressure ratio and intercooling to 35 °C
- The cooling required in the condenser of the stripper column was provided by cooling water with inlet and outlet temperature of 20 and 25 °C, respectively
- Pressure drops in heat exchangers, columns and dehydration units were neglected, along with heat losses and gains
- Isentropic efficiency of 80 % was assumed for the compressors and the expander, while the pump had 85 % isentropic efficiency

3.2 Process evaluation

The thermodynamic performance of the LBM production plant was evaluated using exergy analysis. Exergy is supplied to the LBM production plant in the form of work ($\dot{E}_x^W$) and heat ($\dot{E}_x^Q$), which are calculated as

$$\dot{E}_x^W = \sum_i \dot{W}_i, \tag{1}$$

$$\dot{E}_x^Q = \sum_i \dot{Q}_i \cdot \left(1 - \frac{T_0}{T_i}\right). \tag{2}$$

Here, T_0 and T_i denote the ambient temperature and the temperature at which the heat ($\dot{Q}_i$) is transferred, respectively. Moreover, $\dot{W}$ refers to the amount of work supplied or extracted from the plant. In this study, the exergy supply associated with heating was provided by saturated steam at 3.5 bar. The exergy of material streams was calculated by means of a Visual Basic code in Aspen HYSYS® flowsheet according to the methodology described by Kotas (2012). In this methodology, the exergy of matter is split into physical exergy and chemical exergy. Neglecting kinetic and potential energy, the physical exergy ($\bar{\varepsilon}_x^{phy}$) can be expressed as

$$\bar{\varepsilon}_x^{phy} = \left(\bar{h} - \bar{h}_0\right) - T_0 \cdot \left(\bar{s} - \bar{s}_0\right), \tag{3}$$

where $\bar{h}$ and $\bar{s}$ are the molar enthalpy and entropy of the material stream in the actual state (T, p), respectively. The subscript "0" denotes that the specific enthalpy and entropy are calculated at environment state (T_0 = 25 °C, p_0 = 1 atm = 1.01325 bar). The chemical exergy ($\bar{\varepsilon}_x^{chem}$) for an ideal mixture can be expressed by

$$\bar{\varepsilon}_x^{chem} = \sum_i x_i \cdot \bar{\varepsilon}_{x,i}^{std} + T_0 \cdot \bar{R} \cdot \sum_i x_i \cdot \ln x_i, \tag{4}$$

where x_i and $\bar{\varepsilon}_{x,i}^{std}$ are the molar fraction and standard chemical exergy of component "i" in the mixture, respectively. $\bar{R}$ is the universal gas constant. The standard chemical exergy of each component was obtained in reference tables provided by Szargut et al. (1988). However, the standard chemical exergy of MDEA in liquid phase was estimated according to the group contribution method proposed by Szargut et al. (1988). In this method, the standard chemical exergy of MDEA (with molecular formula of $C_5H_{13}NO_2$) was estimated to $3.386 \cdot 10^6$ kJ/kmol.

3.3 Process optimization

The objective of the optimization was to maximize the thermodynamic performance of the LBM production plant, for given inlet and outlet conditions (temperature and pressure), which is equivalent to minimizing the exergy supply to the plant. Due to a large number of degrees of freedom and challenges associated with convergence of unit operations and recycles, optimizing the overall plant is challenging. Any changes in pressure, flow rate or composition of the streams leaving the upgrading process affect the operating conditions of the liquefaction process. However, changes in the liquefaction process will not influence the upgrading process. Hence, an alternative approach in which the upgrading and liquefaction processes are optimized sequentially, starting with the upgrading process, is proposed. In this case, however, the objective functions should be formulated such that they account for the effects of changes in the upgrading process on the liquefaction process.

Three different objective function formulations for the upgrading process are given in Table 1. In the first objective function (Obj1), the upgrading and liquefaction processes are optimized independently, considering only the exergy supply in the upgrading process. The purpose of the liquefaction process is to remove the heat required for liquefaction of the biomethane and CO_2 streams. Therefore, the sum of the exergy supply in the upgrading process and heat removal in the liquefaction process ($\dot{Q}^{liq}$) is minimized in the second objective function (Obj2). Likewise, in the third objective function (Obj3), the sum of the exergy supply in the upgrading process and the exergy of the heat removed in the liquefaction process ($\dot{E}_x^{Q,liq}$) is minimized. In all formulations, the liquefaction process is optimized by minimizing the net work supply.

Table 1: Objective function formulations for optimization of LBM production plant

	Simultaneous optimization	
Overall	$\min\left(\dot{W}_{net}^{overall}+\dot{E}_x^{Q,reboiler}+\dot{E}_x^{Q,dehydration}\right)$	
	Sequential optimization	
	Upgrading	Liquefaction
Sequential Obj1	$\min\left(\dot{W}_{net}^{upg}+\dot{E}_x^{Q,reboiler}+\dot{E}_x^{Q,dehydration}\right)$	$\min\left(\dot{W}_{net}^{liq}\right)$
Sequential Obj2	$\min\left(\dot{W}_{net}^{upg}+\dot{E}_x^{Q,reboiler}+\dot{E}_x^{Q,dehydration}+\dot{Q}^{liq}\right)$	$\min\left(\dot{W}_{net}^{liq}\right)$
Sequential Obj3	$\min\left(\dot{W}_{net}^{upg}+\dot{E}_x^{Q,reboiler}+\dot{E}_x^{Q,dehydration}+\dot{E}_x^{Q,liq}\right)$	$\min\left(\dot{W}_{net}^{liq}\right)$

For this study, a limited number of degrees of freedom was examined. The chosen decision variables and inequality constraints for the upgrading and liquefaction processes are listed in Table 2. A minimum temperature difference of 2 °C was considered for the heat exchangers and the CO_2 content of the LBM stream was limited to 50 ppm. Equality constraints such as mass and energy balances were handled by the process simulator. It is worth mentioning that the selection of variable bounds is particularly important for the upgrading process due to nonlinearity of constraints and issues regarding column convergence in Aspen HYSYS®. In order to avoid convergence issues, secure variable bounds were determined through several simulation runs prior to optimization, although the optimization problem was limited to a certain domain. For the liquefaction process, the lower and upper pressure levels were set to 1 and 140 bar, respectively. Moreover, the upper pressure of the stripper was limited by the temperature of the reboiler, which should not exceed 127 °C (Lange et al., 2015). The proposed nonlinear optimization problem was solved using the Hyprotech SQP solver from Aspen HYSYS®. Based on experience, all convergence tolerances were set to 10^{-6} both for the optimizer and the unit operations. In order to reduce the likelihood of getting trapped in local optima, each objective function was examined with 30 random starting points. When optimizing the liquefaction process in the sequential optimization approach, the best result obtained for the upgrading process was used. The study was performed on a 2.67 GHz Intel® Xeon® X5650 CPU with 192 GB RAM.

4. Results and discussion

Variable values for the best solution obtained for each objective function formulation are given in Table 3, with corresponding objective function values in Table 4. In Table 5, the exergy supply to the two processes is given, along with the cooling demand in the liquefaction process and its corresponding exergy demand.

As expected, all the inequality constraints are active. The results indicate that the same solution is obtained for the simultaneous approach and the sequential approach with Obj3. Similar results are obtained also for Obj1 and Obj2, but with slightly larger exergy supply. However, the best solution obtained from the present work is different from the previous work provided by Hashemi et al. (2019). In the previous work, the optimization was performed based on an exhaustive search method considering sequential optimization of the absorber and the stripper. Therefore, the CO_2 content constraint dominated the optimization, resulting in lower absorber pressure and higher amine flow rate. In addition, the variable bounds for the reboiler temperature and the high pressure level in the liquefaction process have been changed due to practical considerations.

Table 2: Decision variables and constraints of optimization problem for upgrading and liquefaction

Decision variables	Unit	Variable range	Constraints
Upgrading process			
Absorber pressure (p_{S101})	bar	50 – 70	$x_{CO_2,LBM} \leq 50$ ppm
Stripper pressure (p_{S106})	bar	1 – 2	$\Delta T_{min,HX1} \geq 2$ °C
Stripper inlet temperature (T_{S106})	°C	75 – 90	
Lean amine flow rate ($\dot{n}_{S102}$)	kmol/h	5000 – 8000	
Liquefaction process			
Refrigerant flow rate ($\dot{n}_{S201}$)	kmol/h	1600 – 4000	$\Delta T_{min,HX3} \geq 2$ °C
Low pressure (p_{S201})	bar	1 – 7	$\Delta T_{min,HX4} \geq 2$ °C
High pressure(p_{S202})	bar	80 – 140	
Intermediate temperature (T_{S203})	°C	−50 – 30	

Table 3: Variable values for the best solution obtained for each objective function

	p_{S101} (bar)	p_{S106} (bar)	T_{S106} (°C)	$\dot{n}_{S102}$ (kmol/h)	$\dot{n}_{S201}$ (kmol/h)	p_{S201} (bar)	p_{S202} (bar)	T_{S203} (°C)
Simultaneous	66.6	1.7	90	5000	1789	2.1	140	5.3
Sequential Obj1	61.7	1.7	90	5361	1848	2.3	140	1.8
Sequential Obj2	66.0	1.7	90	5038	1796	2.2	140	4.8
Sequential Obj3	66.6	1.7	90	5000	1789	2.1	140	5.3

Table 4: Assessment of different objective functions

	Objective function values				Avg. running time (min)	Avg. model evaluations (-)
	Overall (kW)	Obj1 (kW)	Obj2 (kW)	Obj3 (kW)		
Simultaneous	14664	8821	11112	10102	88	64
Sequential Obj1	14701	8807	11113	10112	45	36
Sequential Obj2	14667	8817	11110	10104	61	45
Sequential Obj3	14664	8821	11112	10102	56	43

The cooling requirement of the high quality biomethane decreases with increasing pressure level in the absorber (and thereby in the liquefaction process). Since the absorber pressure is lower in the solution obtained for Obj1 than for the other formulations, the net work in the upgrading process is lowest. Nevertheless, the exergy of heat supply in the upgrading process is higher due to larger amine flow rate. Still, the smallest exergy supply to the upgrading process is observed for Obj1 because of higher savings in work. However, as the cooling requirement of the liquefaction process is larger, the exergy supply to the liquefaction process and the overall exergy supply are larger than for the other objective formulations. This interaction is accounted for in Obj2 and Obj3, but with different weighting of the cooling demand.

As can be observed in Table 3, some of variables are on the bounds, which indicates that better solutions are likely to be found if the bounds are extended. In this case, the difference between different objective formulations is also expected to be larger. The simulation model must, however, be able to handle/avoid convergence issues related to the columns in the upgrading process.

In the plant studied here, the interaction between the upgrading and liquefaction processes is limited to the pressure level of the high quality biomethane stream after upgrading, as the temperature and composition are fixed. For the high quality CO_2 stream after upgrading, both the temperature and the pressure are fixed, and the variations in composition are negligible. This partly explains the similarity in results for the different formulations.

As can be observed in Table 4, the average number of model evaluations, and thereby the running time of the optimizer, is reduced when the optimization problem is solved sequentially. It is also worth mentioning that the same solution was obtained for liquefaction process regardless of the starting point, whereas the solution obtained for the upgrading process was highly dependent upon the starting point.

The results suggest that the sequential approach performs well for the optimization of the LBM production plant with amine-based absorption and a single expander refrigeration cycle for liquefaction.

Table 5: Exergy and energy supply for the LBM production for different objective functions

	Upgrading process			Liquefaction process		
	$\sum \dot{E}_x^Q$ (kW)	$\dot{W}_{net}^{upg}$ (kW)	$\sum \dot{E}_x^{upg}$ (kW)	$\dot{Q}^{liq}$ (kW)	$\dot{E}_x^{Q,liq}$ (kW)	$\dot{W}_{net}^{liq}$ (kW)
Simultaneous	2,847	5,974	8,821	2,291	1,281	5,843
Sequential obj1	2,906	5,901	8,807	2,306	1,305	5,894
Sequential obj2	2,854	5,963	8,817	2,293	1,287	5,850
Sequential obj3	2,847	5,974	8,821	2,291	1,281	5,843

5. Conclusions

An LBM production plant using amine-based absorption upgrading followed by a single expander refrigeration cycle was modeled in Aspen HYSYS® and optimized using an SQP algorithm. The objective was to minimize the exergy supply to the plant in terms of work and heat. Different problem formulations in which the upgrading and liquefaction processes were optimized sequentially have been proposed and compared with a conventional approach where the whole plant is optimized simultaneously.

The results indicate that the same solution was obtained for the sequential optimization approach and the simultaneous approach. However, the objectives should be formulated such that the interaction between the two processes is accounted for, i.e. the influence of the upgrading process on the exergy demand of the liquefaction process. In this study, only a limited number of degrees of freedom was used, with relatively tight variable bounds in order to avoid convergence issues in simulation. The results suggest that further studies on the sequential optimization approach should be conducted, especially as the complexity of the two processes is increased with more design variables and larger variable ranges. In this case, the choice of objective function formulations is expected to have larger impact. For future studies, more complex refrigeration processes (e.g. mixed-refrigerant cycles) and convergence challenges will be investigated.

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