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Objective Function Evaluation for Optimization of an Amine-Based Biogas Upgrading and Liquefaction Process

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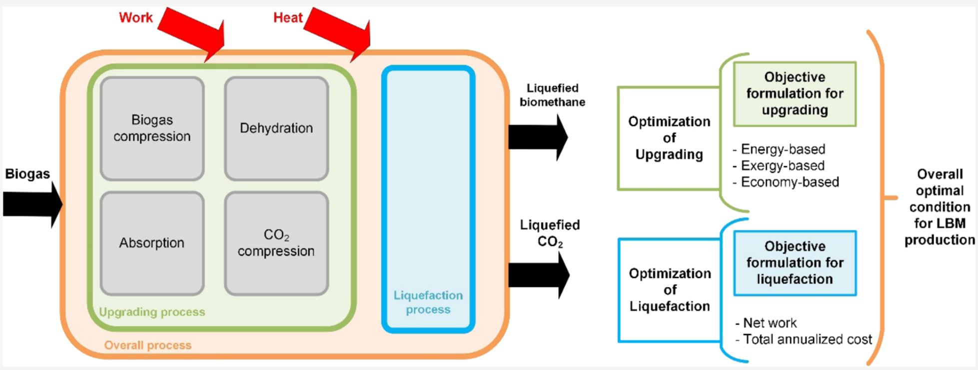

ABSTRACT: Conventionally, liquefied biomethane (LBM) is produced through biogas upgrading followed by a liquefaction process. In the present study, a detailed model for an LBM production plant including amine-based biogas upgrading and liquefaction was provided to compare thermodynamic and economic optimization for the biogas upgrading. In this context, multiple objective function formulations based on energy, exergy, and economy were examined. Furthermore, their impact on the exergy demand in the liquefaction process and the overall LBM production plant was investigated. The results indicated that optimization of the upgrading process based on exergy and total annualized cost would result in similar solutions, providing both the highest thermodynamic and economic performances, because the operating pressure was forced to be high to meet the strict CO_2 limitations for LBM. However, the results also indicated that the exergy demand for the overall LBM production plant would be approximately the same regardless of the objective function formulation used for the upgrading process, as exergy savings in the liquefaction process would compensate higher exergy demand in the upgrading process. Overall, thermodynamic and economic optima of the LBM production plant would be similar if the LBM production plant was optimized based on exergy supply or total annualized cost. It was also illustrated that the selection of a suitable refrigeration cycle would have more impact on the overall performance of the LBM plant than the formulation of the objective function for the optimization.

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1. INTRODUCTION

In the transport sector, renewable sources account for only 3.3% of the total energy use.[1] Moreover, in 2016, the transport sector accounted for approximately 23% of the global energy-related CO_2 emissions.[1] The penetration of the renewable energy sources in transport includes electrification using batteries and fuel cells and blending biofuels, hydrogen, and synthetic fuels with conventional fuels.[2] Because of their low energy density, batteries and fuel cells are mainly employed in light-duty vehicles,[2] while liquid biofuels and liquefied biomethane (LBM) produced from upgraded biogas are considered promising solutions for heavy-duty vehicles.[3,4] LBM shares similar characteristics to liquefied natural gas (LNG), which makes it suitable for long-range applications, but with a smaller carbon footprint. Pasini et al.[5] studied the utilization of biomethane for gas grid injection and LBM production. They confirmed that LBM production was economically feasible for long-distance transportation.

To produce LBM downstream of a biogas plant, two processes should be considered: upgrading and liquefaction. As the biogas results from the biological degradation of different biomass materials through anaerobic digestion, the composition of the biogas differs depending on the source of substrates and production method.[6] The biogas mainly consists of methane (CH_4) and carbon dioxide (CO_2) and minor compounds such as hydrogen sulfide, ammonia, and nitrogen. Biogas upgrading is required to remove CO_2 from the biogas mixture because the presence of CO_2 reduces the heating value

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of the biogas and increases the risk of solid CO_2 formation during liquefaction.

Common gas separation technologies such as chemical and physical absorption, temperature and pressure swing adsorption, membrane separation, and cryogenic separation can be employed in biogas upgrading.[6−8] The main difference between biogas upgrading and similar applications such as CO_2 capture in power plants is the inlet gas composition.[9] The CO_2 concentration is much higher in biogas than that for typical feed gas in pre or postcombustion CO_2 capture plants.[7] Therefore, the design and energy requirements of the gas separation technologies will be different for biogas upgrading.

Vilardi et al.[10] performed comprehensive energy and exergy analyses for biogas upgrading through pressure swing adsorption. They simulated the adsorption process using Aspen Plus and Aspen Adsorption and found that production of biomethane with a purity of 97% for injection to the gas grid at 70 bar required a specific energy use of 0.363 kWh/m^3 biomethane at STP (i.e., standard temperature and pressure, 0 °C and 1 bar). They showed that exergy losses within the adsorption process were mainly due to a high CH_4 loss (a CH_4 recovery of only 93.4%), while the main unavoidable irreversibilities were observed in pumps, compressors, and heat exchangers. In another study, Vilardi et al.[11] compared amine absorption, water scrubbing, and membrane separation for biogas upgrading. They reported that the lowest specific energy use for biogas upgrading could be achieved with amine absorption (0.204 kWh/m^3 biomethane at STP), while the specific energy use for water scrubbing and membrane separation was 0.475 kWh/m^3 biomethane and 0.940 kWh/m^3 biomethane at STP, respectively. Amine-based absorption has been applied in many large-scale industrial plants and can provide the targeted LBM specifications without requiring additional polishing steps.[12] Cavaignac et al.[13] performed a techno-economic assessment for medium- to large-scale biomethane production with different amine absorption alternatives. They illustrated that amine-based biogas upgrading was profitable for medium- to large-scale plants and showed that employing diglycolamine (DGA) was economically superior to a blend of methyldiethanolamine (MDEA) and diethanolamine (DEA). In comparison with other biogas upgrading technologies, Lombardi and Francini[14] indicated that biogas upgrading through amine-based absorption would result in the lowest global warming potential, because of a low CH_4 loss from the process.

It is worth mentioning that the majority of earlier studies mainly have focused on biomethane production with a quality suitable for gas grid injection applications, in the range of 90−98 mol % CH_4 depending on the specifications in different countries.[8] However, even higher purity is required in LBM production to avoid dry ice formation in the liquefaction process. No specific guidelines are given in the literature, yet considering the requirements in LNG production, the CO_2 concentration in the biomethane should not exceed 50 ppm.[15]

Many studies have strived to optimize amine-based CO_2 removal processes considering different objectives, including the thermodynamic performance of the process,[16−19] the economy of the process,[20−22] and multiobjective optimization considering both thermodynamic and economic performances.[23] Jassim[17] performed a sensitivity analysis for a CO_2 capture plant with MDEA as the solvent amine to determine the effect of the most influential process variables on the reboiler duty and CO_2 and H_2S recovery. The results indicated that the amine flow rate and amine concentration were the two main variables influencing the energy requirement. Rodriguez et al.[20] presented optimal operating conditions for minimum total annual cost (TAC) of an amine-based CO_2 capture plant using different alkanolamine solutions (DEA, MDEA, and MDEA-DEA mixture). The optimization tool available in Aspen HYSYS was used in their study, considering CO_2 lean loading, amine flow rate, and inlet temperature of the lean solution stream to the absorber as optimization variables. They highlighted that the determination of the optimal design of the plant could be a challenging task because of the nonlinear behavior of the process. Qiu et al.[23] conducted an optimization study to maximize the annual profit of a high-sulfur gas sweetening plant. They found that maximum annual profit would coincide with minimum operating costs and maximum treated gas yield. Despite the available studies for optimization of the amine-based biogas upgrading process, the difference between optimal operating conditions obtained from optimizing thermodynamic performance and economics has not been thoroughly investigated for LBM production plants. The strict CO_2 removal limitation in the LBM production plants may change the optimal operating conditions obtained from the thermodynamics or the economics.

The liquefaction process is another energy-intensive part of the LBM production plant. Many refrigeration cycles that have been developed for natural gas liquefication processes can be employed for LBM production.[24,25] However, the inlet gas stream is almost pure methane and does not include heavier hydrocarbons. This may influence the optimal operating conditions in the liquefaction process.[15] Because of the limited liquefaction capacity required in LMB production plants, N_2 expander and single mixed-refrigerant processes are often used for liquefaction. Capra et al.[15] evaluated the thermodynamic and economic efficiency of different refrigeration cycles for biomethane liquefaction and found a single mixed refrigerant (SMR) cycle with a Joule−Thomson expansion valve to be the best option for large-scale LBM production.

There are only a limited number of studies that cover the optimization and energy requirements of the entire LBM production plant. Pellegrini et al.[26] investigated the energy required to produce LBM, considering chemical absorption biogas upgrading and external refrigeration for the liquefaction. They reported that 57.4% of the total energy requirement in the LBM production plant was for the upgrading process. Baccioli et al.[27] compared the energy required for biogas upgrading through cryogenic separation and chemical absorption in a small-scale LBM production plant where a dual-expander refrigeration cycle was considered for the liquefaction. They found that the electrical energy required for cryogenic upgrading and liquefaction was 0.61 kWh/m^3 raw biogas, while for chemical absorption and liquefaction the electrical energy requirement was 0.72 kWh/m^3 raw biogas (in addition to heat requirements). Hashemi et al.[28] observed that the dependency between the upgrading and liquefaction processes was limited to only the pressure level of the produced biomethane from the upgrading process. This was because of the highly restricted composition and temperature of the produced biomethane before entering the liquefaction process. Hence, it was concluded that a sequential optimization approach would provide a solution near the solution obtained when optimizing both processes together.

Haider et al.[29] developed an integrated process for LBM production where they employed an imidazolium-based

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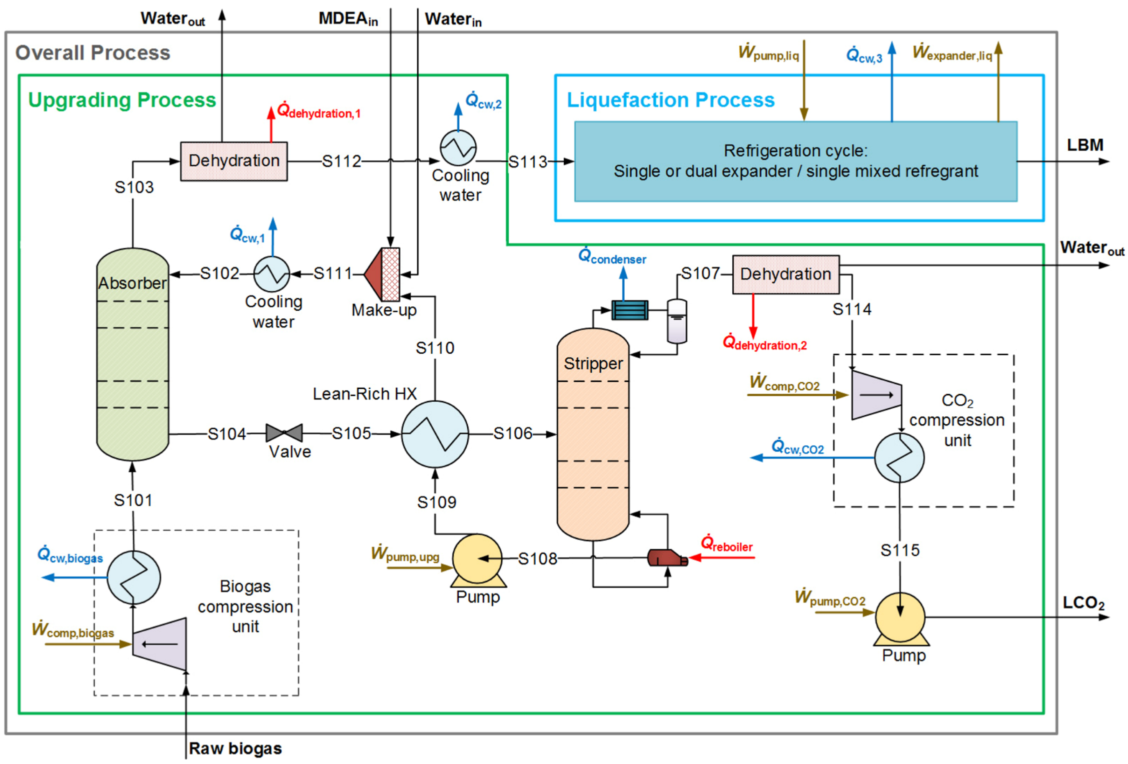

Figure 1. Schematic of the LBM production plant.

cationic ionic liquid for biogas upgrading instead of conventional amine-based absorption. For the liquefaction process, they considered a nitrogen expander process. They indicated that using a cationic ionic liquid as an absorbent provided great potential to produce biomethane with high CO_2 removal and low thermal energy requirement. They showed that the proposed integrated process would lead to an energy- and cost-efficient solution. This work is one of the few studies that considered both upgrading and liquefaction processes. However, they did not focus on optimizing such a process in their study.

Naquash et al.[30] developed an integrated upgrading and liquefaction process for LBM production. They used an antisublimation process for biogas upgrading, in which the CO_2 in the raw biogas was solidified within a specially designed cold box. The integrated process could provide advantages for upgrading and precooling the biomethane, reducing the cooling requirement in the liquefaction process. They showed that most exergy dissipated within the antisublimation upgrading process where further development should be considered to boost the practicality of the developed process.

Song et al.[31] proposed a novel hybrid system for biogas upgrading employing membrane and cryogenic separation technologies to produce LBM with purity up to 98%. CO_2 was removed through a polyimide membrane and liquefied using cold energy from the LBM employing flash drums to separate liquid and gas phases of the CO_2. They concluded that the proposed process could be a competitive alternative to conventional biogas upgrading processes. However, they did not consider the liquefaction process in their analysis.

Ghorbani et al.[32] developed an integrated process for LBM production. They considered a low-temperature CO_2 removal process using multiple flash drums and amine-based absorption for biogas upgrading. They used a mixed-refrigerant cascade process for the liquefaction that also provided cooling in the low-temperature CO_2 removal process. They showed that the exergy efficiency of the developed process was about 73%, with a specific power consumption of 0.48 kWh/kg biomethane.

Providing detailed economic calculations for the LBM production plant can be cumbersome because of the amount of equipment and the level of uncertainty involved in the calculations. This motivates examining alternative objectives that can provide solutions close to economically optimal but are easier to calculate. In the present study, alternative objective functions for optimization of an LBM production plant are formulated based on energy use, energy quality, and energy cost. The LBM production plant consists of an amine-based upgrading process using MDEA and a liquefaction process. Considering different objective function formulations would result in different thermodynamic and economic performance. This would provide insight into the difference between thermodynamic optima and economic optima. It should be noted that alternative objective function formulations are only considered for optimization of the upgrading process because the optimization of the upgrading process is more complex than for the liquefaction process, and because only work is required for the liquefaction process. Compared to the previous work by Hashemi et al.,[28] where the interaction between upgrading and liquefaction processes was evaluated, the present study aims to optimize the upgrading and liquefaction processes separately. Furthermore, challenges with the convergence of the plant flowsheet are solved and the number of variables and their ranges are increased. A detailed model for cost analysis of the upgrading process is provided to evaluate and compare the optimal solutions from different objective formulations based on the TAC. To evaluate the liquefaction process, multiple refrigeration cycles are optimized, and the impact of the pressure level of the

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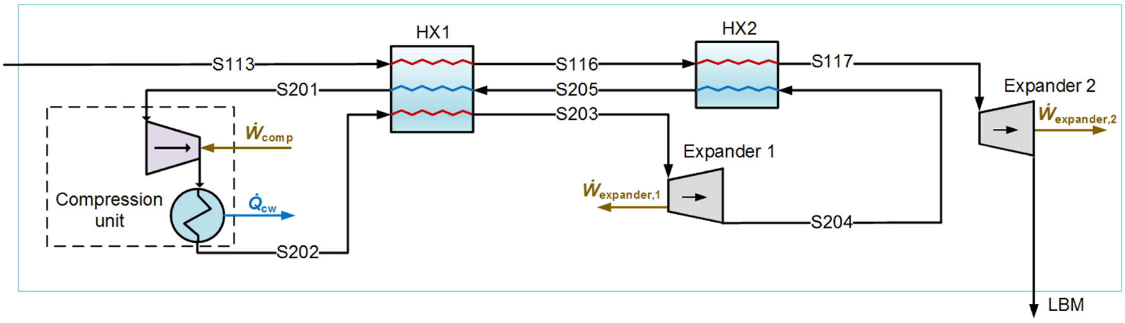

Figure 2. Schematic diagram of the single nitrogen expander cycle (SE).

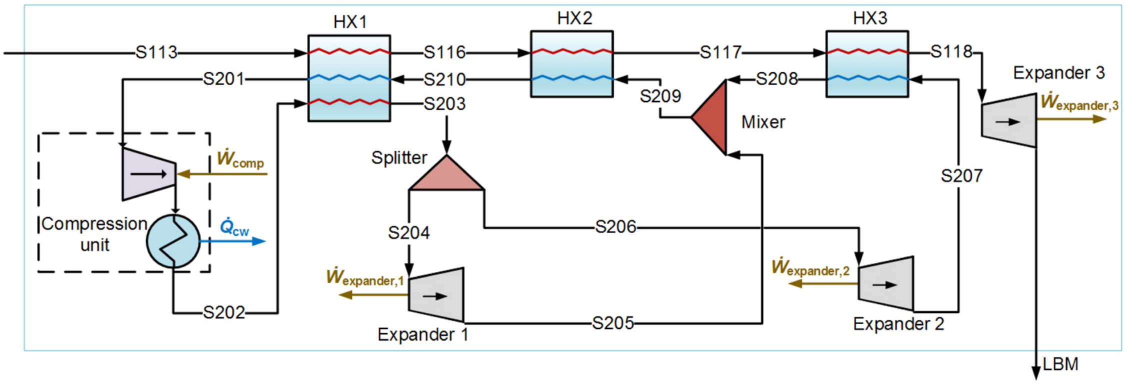

Figure 3. Schematic diagram of the dual nitrogen expander cycle (DE).

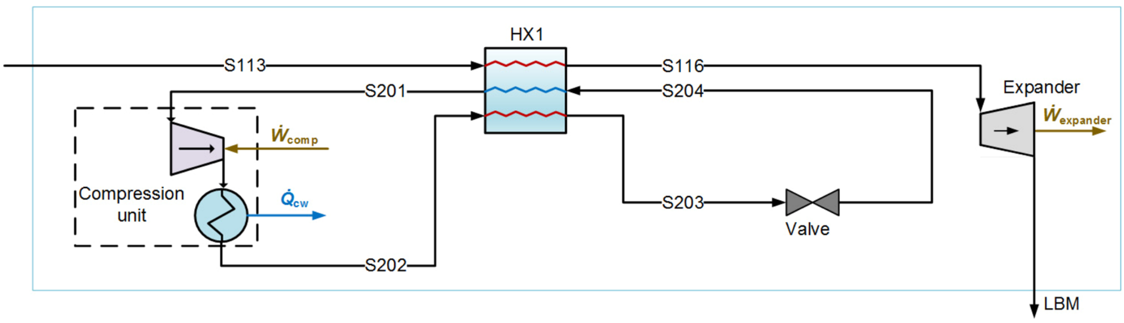

Figure 4. Schematic diagram of the single mixed-refrigerant cycle (SMR).

biomethane stream from the biogas upgrading process on the energy use in the liquefaction process is studied through a sensitivity analysis.

2. PROCESS DESCRIPTION

A schematic diagram of the studied LBM production plant is given in Figure 1. It is assumed that LBM and CO_2 in liquid form (LCO_2) are the final product and byproduct, respectively. The overall LBM production plant consists of a biogas upgrading process and a liquefaction process. These two processes are connected through a high-quality biomethane stream produced in the upgrading process (S113). A detailed process description for an amine-based biogas upgrading process can be found in the work by Hashemi et al.[33] Here, a brief description of the process and its main streams is presented.

The upgrading process (surrounded by a green border in Figure 1) contains a biogas compression unit, an amine-based absorption CO_2 capture unit, and a CO_2 compression unit. After compressing the raw biogas mixture in the biogas compression unit, the compressed biogas (S101) enters the bottom of an absorber column, where it interacts with the lean-amine solvent (S102) from the top of the column. High-quality biomethane (S103) leaves from the top of the absorber column, while the rich-amine solvent (S104) leaves from the bottom. The rich-amine solvent is depressurized through an expansion valve and heated by exchanging heat with a recycled lean-amine solvent stream in an intermediate heat exchanger (Lean-Rich HX) before entering a stripper column (S106). In the stripper column, CO_2 is separated from the amine again by adding heat through the reboiler at the bottom of the column. High-purity CO_2 (S107) leaves the stripper column from the top, while the lean-amine solvent from the bottom of the stripper is recycled back to the absorber column. A pump and a cooler are used to bring the pressure and temperature of the recycled stream back to the desired conditions in the absorber column. Moreover, water and amine solvent losses are compensated in a make-up unit after the intermediate heat exchanger.

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Dehydration units are located at the top of the absorber and the stripper to remove water from the high-quality biomethane and CO_2 streams. The dehydrated CO_2 (S114) is compressed and cooled through a CO_2 compression unit. As the CO_2 will be in liquid form after the compression unit, a pump is used to increase the pressure of the CO_2 stream up to a level suitable for CO_2 pipeline transportation. The dehydrated biomethane (S112) is cooled through a cooling-water heat exchanger before entering the liquefaction process (surrounded by a blue border in Figure 1) (S113), where the LBM is produced. The details of the liquefaction process are presented in the following, and in Figures 2−4.

Different refrigeration cycles can be considered for the liquefaction process. Here, single/dual nitrogen expansion cycles and an SMR cycle are studied. These cycles are selected because of their relatively simple design, which makes them suitable for the required train capacity. In the liquefaction process, cooling is provided by changes in the pressure level of the refrigerant through compression and expansion; the latter either through expanders (in the single/dual nitrogen expansion cycles) or expansion valves (in the SMR). Moreover, an expander is used to bring the pressure of the LBM to atmospheric pressure. Process configurations for the single and dual nitrogen expander cycles and the SMR are given in Figures 2−4, respectively. The working fluid in the liquefaction cycle differs depending on the refrigeration cycle (i.e., nitrogen for the single/dual expander cycles and a mixture of hydrocarbons in the SMR). The potential synergy from integrating heating and cooling requirements within the upgrading process and the liquefaction process is not considered in the present study.

3. METHODS

3.1. Process Modeling. The LBM production plant was simulated with Aspen HYSYS V9.0 (Aspen Technology Inc.). Equilibrium calculations and characterization of the phase behavior of mixtures were modeled using two different thermodynamic models from Aspen HYSYS; the "Acid gas−chemical solvent" package recommended by Aspen HYSYS was employed for the amine-based absorption CO_2 capture unit,[34] and the Soave−Redlich−Kwong equation of state was used in the compression units and the refrigeration cycles.

The inlet stream to the LBM production plant was a biogas mixture of 60 mol % CH_4, 39.9 mol % CO_2, and 0.1 mol % H_2S, representing a typical biogas composition,[35] with a molar flow rate of 1000 kmol/h at 35 °C and 1 atm. It should be noted that the flow rate is high compared to typical biogas production plants, yet consistent with previous studies.[28,33] The outlet streams from the LBM production plants were saturated liquid biomethane at 1 atm with a CO_2 content below 50 ppm and liquefied CO_2 at 35 °C and 110 bar containing all the H_2S from the biogas.

A survey in available literature indicates that MEA and activated MDEA are the most used amines in gas purification.[36] MEA is known for operating at low pressure in the range of 3−8 bar but requires a large amount of heat in the reboiler to regenerate the amine.[19] This means that the major energy requirement is heat to regenerate the amine in a stripper column. In contrast, MDEA provides the specified purity at an elevated pressure range between 45 and 70 bar, with lower heat demand for amine regeneration.[19] The high operating pressure for MDEA will also result in high biomethane pressure and thereby less work for liquefaction. In addition, MDEA is recommended in gas separation for mixtures with H_2S. These are the reasons MDEA was selected in the present study.

To ensure that the CO_2 content in the biomethane remained below 50 ppm for a wide range of operating conditions, absorber and stripper columns with 3 m diameter and 25 and 20 theoretical trays, respectively, were selected.[33] The rich-amine solvent entered the stripper column at the ninth tray from the top. Identical pressure was assumed for the streams entering the absorber column. Two independent variables are required to satisfy the degrees of freedom to solve the stripper column model. After a thorough examination of different variables, the condenser temperature and the CO_2 lean loading at the bottom of the stripper were selected to improve the convergence of the stripper column simulation. Assuming a water-cooled condenser with a minimum temperature approach of 10 °C, the condenser temperature was considered to be 30 °C. To minimize acid gas flashing in the lean-amine solvent recycling line and thereby avoid corrosion, the CO_2 lean loading should not be higher than 0.01 mol CO_2/mol MDEA.[37] Here, a CO_2 lean loading of 0.01 mol CO_2/mol MDEA was chosen, as a higher CO_2 lean loading results in a smaller heat supply to the reboiler of the stripper.[19] It was assumed that all the water in the biomethane stream (S103) and the CO_2 stream (S107) was removed in the dehydration units after the absorber column and the stripper column, respectively. This was done using tri-ethylene-glycol (TEG) absorber/regeneration columns. The TEG regeneration temperature was assumed to be 200 °C and the outlet temperature of the dehydration units was 30 °C. Details regarding the calculation of the heat requirements in the reboiler of the TEG regeneration columns can be found in the work by Hashemi et al.[33] The following assumptions were also considered for the LBM plant:

- Pressure drops in heat exchangers, columns, and dehydration units were neglected.
- An isentropic efficiency of 80% was assumed for the compressors and the expanders and 85% for the pumps.
- The cooling required in the condenser of the stripper column and the intermediate coolers was provided by cooling water with inlet and outlet temperatures of 20 and 25 °C, respectively.
- The gas compression units consisted of four-stage compressors with an identical pressure ratio and intercooling to 30 °C.
- The heat supplied to the plant was provided by saturated stream at 5 bar.

3.2. Process Evaluation. In accordance with the previously mentioned observations by Hashemi et al.,[28] the biogas upgrading process and the liquefaction process were optimized separately in a sequential approach. Compared to the liquefaction process, optimizing the amine-based biogas upgrading process is challenging because of issues associated with the convergence of unit operations and recycles.[20] The upgrading process contributes significantly to the energy requirements and the investment cost of the LBM production plant. Therefore, the main aim of the present work was to optimize the amine-based biogas upgrading process examining potential objective function formulations for thermodynamic efficiency and cost. For the upgrading process, the thermodynamic performance was evaluated through energy and exergy analysis. The economic performance was evaluated based on either the TAC of the upgrading process or the operating cost alone. Furthermore, three alternative refrigeration cycles for biomethane liquefaction were optimized through minimization of the network requirement and the total annualized cost.

3.2.1. Exergy Analysis. Exergy is transferred to and from the plant in the form of work ($\dot{E}_x^{W}$) and heat ($\dot{E}_x^{Q}$):

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

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

Here, $\dot{W}$ refers to the power supplied to the compressors and pumps, or the power extracted from the expanders. Furthermore, $\dot{Q}_i$ is the heat flow transferred at temperature T_i, while T_0 denotes the environment temperature. The term in parentheses in eq 2 denotes the Carnot factor.

The exergy of material streams was calculated via Visual Basic code in Aspen HYSYS.[38] According to the methodology described by Kotas,[39] the exergy of material streams is a combination of physical exergy and chemical exergy. Assuming negligible kinetic and potential energy, the molar physical exergy ($\bar{\varepsilon}_x^{phy}$) can be expressed as

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

where $\bar{h}$ and $\bar{s}$ are the molar enthalpy and entropy, respectively, of the material stream in the actual state (T, p). Correspondingly, $\bar{h}_0$ and $\bar{s}_0$ are the molar enthalpy and entropy in the environment state (here T_0 = 25 °C and p_0 = 1 atm = 1.01325 bar). Assuming an ideal mixture, the molar chemical exergy ($\bar{\varepsilon}_x^{chem}$) can be calculated 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 $\bar{\varepsilon}_{x,i}^{std}$ refer to the molar fraction and standard chemical exergy, respectively, of component i in the mixture and $\bar{R}$ is the universal gas constant. The standard chemical exergy of each component was obtained from the reference tables provided by Szargut et al.[40] According to the group contribution method proposed by Szargut et al.,[40] the standard chemical exergy of MDEA with a molecular formula of $C_5H_{13}NO_2$ is $3.386 \cdot 10^6$ kJ/kmol.

3.2.2. Cost Analysis. For the cost analysis, the methodology described by Turton[41] was employed to calculate the TAC. Here, the TAC is the sum of the total annualized investment cost (AIC) and the annual operating cost (AOC):

$$\mathrm{TAC} = \mathrm{AIC} + \mathrm{AOC} \tag{5}$$

To calculate the AIC, the cost of equipment was calculated based on the bare module costing technique.[41] The AOC was estimated based on the sum of costs associated with capital investment (i.e., fixed cost), utility cost (UC), and labor cost. In this work, the cost of absorber and stripper columns, column packings, condenser, reboiler, lean-rich heat exchanger, coolers, pumps, compressors, expanders, and expansion valves were considered in the cost analysis. For the sake of brevity, the employed cost parameters and the detailed cost equations corresponding to equipment cost and operating cost are provided in the Supporting Information.

3.3. Process Optimization. The energy required in the amine-based biogas upgrading process is in the form of work and heat, while only work is required in the liquefaction process. The work supplied to the upgrading process consists of the work required in the biogas compression unit, the CO_2 compression unit, and the pumps. The heat supplied to the upgrading process includes the heat required in the reboiler and dehydration units. It is worth noting that the heat out of the process through cooling-water heat exchangers was not considered in the optimization.

The Hyprotech SQP optimizer in Aspen HYSYS V9.0 was used to optimize the processes. Each objective function was examined with 30 random starting points to reduce the chance of obtaining local optima. The best solution obtained from optimizing the upgrading process was considered when optimizing the liquefaction process. The study was performed on a 2.67 GHz Intel Xeon X5650 CPU with 192 GB RAM.

A sensitivity analysis was performed to assess key decision variables and to evaluate how different variables would influence the energy requirements in the upgrading process. This would provide better understanding of the interaction between variables, studying how altering one variable would change the optimal value for the others. The sensitivity analysis was performed for different values for the amine concentration and the lean-amine temperature, optimizing other variables (i.e., amine flow rate, amine concentration, absorber pressure, and stripper pressure) of the upgrading process using the total exergy supply as the objective function. As the aim of this work was to focus on the objective functions, the results from the sensitivity analysis are given in the Supporting Information for the sake of brevity. Furthermore, the impact of the biogas composition on the performance of the upgrading process is also included as part of sensitivity analysis in the Supporting Information.

The different objective function formulations for optimizing the upgrading process and the liquefaction process are presented in Table 1. Because of having different forms of energy in the upgrading process, the objective function formulations for the upgrading process were based on energy (Obj Upg1−3), exergy (Obj Upg4), and economics (Obj Upg5 and 6). Two extreme cases were when the upgrading process was optimized based on either work or heat supply alone in Obj Upg1 and Obj Upg2, respectively, as these cases ignored the effect of the other form of energy. The total energy supply to the upgrading process was minimized in Obj Upg3, whereas the total exergy supply was considered in Obj Upg4. In other words, work and heat were weighted equally in Obj Upg3, while heat was weighted less in Obj Upg4 using the Carnot factor. To consider the energy prices, the cost of utilities was considered in the Obj Upg5. Furthermore, the

Table 1. Optimization Objective Formulations for the Upgrading and Liquefaction Processes

name	objective
upgrading process	
Obj Upg1	min ($\dot{W}_{upg}$)
Obj Upg2	min ($\dot{Q}_{upg}$)
Obj Upg3	min ($\dot{W}_{upg} + \dot{Q}_{upg}$)
Obj Upg4	min ($\dot{W}_{upg} + \dot{E}_x(\dot{Q}_{upg})$)
Obj Upg5	min (UC)
Obj Upg6	min ($\mathrm{TAC}_{upg.}$)
liquefaction process	
Obj Liq1	min ($\dot{W}_{liq}$)
Obj Liq2	min ($\mathrm{TAC}_{liq.}$)

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Table 2. Decision Variables and Corresponding Ranges for the Optimization of the Upgrading and Liquefaction Processes

	decision variable	unit	range	constraints
upgrading process	absorber pressure ($p_{absorber}$)	bar	30−90	$x_{CO2,LBM} \leq 50$ ppm
	stripper pressure ($p_{stripper}$)	bar	1−5	$\alpha_{rich} \leq 0.55$
	lean-amine temperature (T_{S102})	°C	40−60	$T_{reboiler} \leq 126.7$ °C
	rich-amine temperature (T_{S106})	°C	70−105	$\Delta T_{lean\text{-}rich\ HX} \geq 10$ °C
	amine molar flow rate ($\dot{n}_{MDEA@S102}$)	kmol/h	3000−15,000	
	amine concentration ($c_{MDEA@S102}$)	wt %	35−55	
liquefaction process	**single nitrogen expander cycle**			
	low-pressure level (p_{S201})	bar	1−20	$\Delta T_{HXs} \geq 2$ °C
	high-pressure level (p_{S202})	bar	40−120	
	nitrogen molar flow rate ($\dot{n}_{S201}$)	kmol/h	1000−8000	
	intermediate temp. (T_{S116})	°C	−80 − 0	
	dual nitrogen expander cycle			
	low-pressure level (p_{S201})	bar	1−20	$\Delta T_{HXs} \geq 2$ °C
	high-pressure level (p_{S202})	bar	40−120	
	nitrogen molar flow rate ($\dot{n}_{S201}$)	kmol/h	1000−8000	
	first intermediate temp. (T_{S116})	°C	−70 − 0	
	second intermediate temp. (T_{S117})	°C	−150 − −80	
	split ratio		0.001−0.999	
	single mixed-refrigerant cycle			
	low-pressure level (p_{S201})	bar	1−10	$\Delta T_{HXs} \geq 2$ °C
	high-pressure level (p_{S202})	bar	10−50	
	nitrogen molar flow rate ($\dot{n}_{N2}$)	kmol/h	10−800	
	methane molar flow rate ($\dot{n}_{CH4}$)	kmol/h	10−800	
	ethane molar flow rate ($\dot{n}_{C2H6}$)	kmol/h	10−800	
	N-propane molar flow rate ($\dot{n}_{C3H8}$)	kmol/h	1−100	
	N-butane molar flow rate ($\dot{n}_{C4H10}$)	kmol/h	10−800	

Table 3. Assessment of Different Objective Function Formulations

	$\dot{W}_{upg}$ (kW)	$\dot{Q}_{upg}$ (kW)	$\dot{W}_{upg} + \dot{Q}_{upg}$ (kW)	$\dot{W}_{upg} + \dot{E}_x(\dot{Q}_{upg})$ (kW)	UC (MUSD/y)	TAC (MUSD/y)
Obj Upg1	5052.2	16187.8	21240.0	9534.0	8.618	21.107
Obj Upg2	6585.9	5294.2	11880.1	8059.2	5.214	19.073
Obj Upg3	6239.8	5556.5	11796.4	7785.8	5.145	17.791
Obj Upg4	5769.4	6319.0	12088.3	7526.7	5.200	16.529
Obj Upg5	6166.1	5636.8	11802.9	7734.4	5.138	17.541
Obj Upg6	5734.2	6493.8	12228.0	7540.0	5.250	16.510

TAC of the biogas upgrading process was minimized in Obj Upg6. For the liquefaction process, two objective functions were considered: minimization of the network requirement (Obj Liq1) and minimization of the total annualized cost (Obj Liq2). Not considering cooling water, minimizing the energy supply is equivalent to minimizing the exergy supply in the liquefaction process.

Decision variables and their corresponding ranges to optimize the biogas upgrading process and different refrigeration cycles are given in Table 2. For the MDEA concentration, practical limitations were considered.[42] Furthermore, the selected range for the lean-amine temperature was based on considerations regarding MDEA degradation and CO_2 absorption capacity, in addition to a minimum temperature difference of 10 °C for the streams entering the absorber.[42] Wider ranges were selected for the remaining variables, adjusted to secure simulation convergence and make sure the strict limitations on the CO_2 content of the produced biomethane would be satisfied.

In a previous study by Hashemi et al.,[28] convergence of the process flowsheet was observed to be a limitation for the optimization of the amine-based biogas upgrading process, which led to considering narrow ranges for the decision variables. The issues associated with the convergence of the flowsheet were mainly due to the definition of the required specifications for the stripper column and unsuitable error tolerances for the unit operations and recycles. After examining different combinations of the specifications for the stripper column, it was concluded that selecting one variable from the top and one variable from the bottom of the stripper as specifications would improve the convergence of the stripper column. Furthermore, a convergence error tolerance of 10^{-8} was selected for all unit operations and recycles.

It was also observed that extreme changes in the variable values between each iteration during the optimization would deteriorate the convergence of the flowsheet. A smooth transition between each iteration improves convergence but also requires longer computational time. After preliminary examination of the optimizer setup, it was found that a step size of 0.01 and a perturbation value of 0.006 with an error tolerance of 10^{-8} provided consistent results for the optimization.

The optimization problems include sets of equality constraints (mass, energy balances, and correlations to compute physicochemical properties), which were handled by the Aspen HYSYS model. The inequality constraints

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considered for the optimization problems are listed in Table 2. As mentioned earlier, the CO_2 content (x_{CO2}) in the produced biomethane from the upgrading process was limited to a maximum of 50 ppm to avoid CO_2 ice-formation in the liquefaction process.[15] To avoid corrosion in the amine absorption process, the CO_2-rich loading (α_{rich}) should not exceed 0.55 mol CO_2/mol MDEA.[37] A maximum temperature of 126.7 °C was considered for the reboiler temperature in the stripper column to avoid degradation of the amine.[42] Furthermore, to limit the capital cost of the heat exchangers in the thermodynamics-based optimization, constraints on the minimum temperature approach of 10 and 2 °C were used for the lean-rich heat exchanger and the heat exchangers in the refrigeration cycles, respectively.

4. RESULTS AND DISCUSSION

4.1. Upgrading Process. Objective function values for the best solution obtained from the different objective function formulations are given in Table 3, with values of process variables and inequality constraints in Table 4. Furthermore, the energy distribution within the upgrading process is illustrated for the different solutions in Figure 5.

Depending on the objective function formulation employed in the optimization, the weighting between work and heat supply to the upgrading process, as well as equipment size in Obj Upg6, is different. Considering only the work or the heat supplied to the upgrading process (i.e., Obj Upg1 and Obj Upg2) is an extreme case because the unbalanced trade-off between work and heat will lead to the smallest work and the largest heat supply in Obj Upg1 (among the tested objective function formulations), and the opposite for Obj Upg2. Because the energy quality of work is higher than that of heat, the trade-off between work and heat is shifted toward less work and more heat when the sum of exergy supply in Obj Upg4 is considered rather than the sum of energy supply in Obj Upg3. Compared to the solution obtained from Obj Upg4, which provides the lowest exergy supply to the upgrading process, the solution obtained from Obj Upg5 (i.e., formulation based on the UCs) is shifted toward more work and less heat. This is because work is weighted more in Obj Upg5 than in Obj Upg4 (i.e., Carnot factor of approximately 0.25 in Obj Upg4 and utility price ratio between work and heat of approximately 0.66 in Obj Upg5). The solutions obtained from Obj Upg4 and Obj Upg6 are similar, with a slight shift toward less work and more heat for Obj Upg6.

As can be observed in Table 4, reduced work is obtained by a reduction in the absorber pressure. In return, a higher amine flow rate is required to satisfy the constraint related to the CO_2 concentration, which leads to an increase in the heat demand for the reboiler and the dehydration units. An increased amine flow rate is also accompanied by reduced amine concentration. Contrarily, reduced heat demand is obtained by the reduced amine flow rate and increased amine concentration and absorber pressure. As a consequence, the compression work increases. In the extreme cases of Obj Upg1 and Obj Upg2, hitting the bounds for process variables indicates that wider bounds for the amine flow rate, amine concentration, and absorber pressure would result in even more extreme solutions (see Table 4). Figure 5 demonstrates that the heat supplied to the upgrading process is mainly used in the reboiler because the amount of water removed in the dehydration units is small.

From an economic point of view, Obj Upg3 and Obj Upg5 provide similar solutions for the total UC; however, the total

Table 4. Variable Values and Values of Inequality Constraints for the Best Solution Obtained for Each Objective Function

	$p_{absorber}$ (bar)	$p_{stripper}$ (bar)	T_{S102} (°C)	T_{S106} (°C)	$\dot{n}_{MDEA@S102}$ (kmol/h)	$c_{MDEA@S102}$ (wt %)	$x_{CO2,LBM}$ (ppm)	α_{rich} (mol CO_2/mol MDEA)	$T_{reboiler}$ (°C)	$\Delta T_{lean\text{-}rich\ HX}$ (°C)
Obj Upg1	32.1	2.3	60.0	100.6	15000.0	35.8	50	0.35	126.7	15.6
Obj Upg2	90.0	1.0	52.5	75.2	4738.3	55.0	50	0.55	101.2	10.0
Obj Upg3	70.6	1.0	54.2	76.1	5142.9	52.6	50	0.55	101.5	10.0
Obj Upg4	48.5	1.0	60.0	78.8	6852.1	43.9	50	0.55	101.6	10.0
Obj Upg5	66.5	1.0	57.1	77.0	5438.9	51.0	50	0.55	101.6	10.0
Obj Upg6	46.9	1.0	60.0	78.9	7124.3	43.4	50	0.55	101.6	10.0

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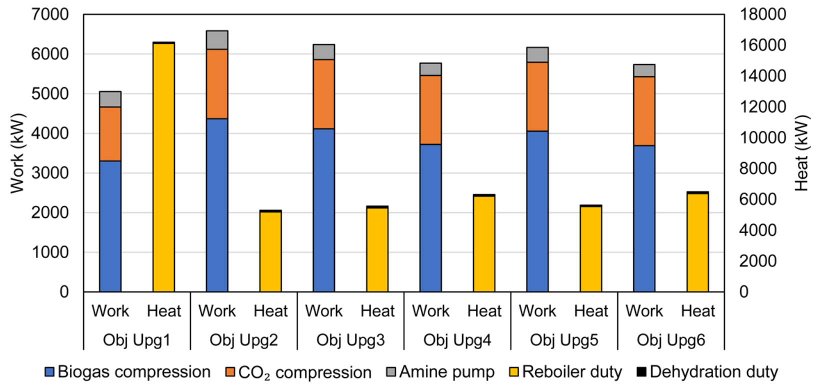

Figure 5. Energy distribution within the upgrading process for the best solution obtained from each objective function formulation.

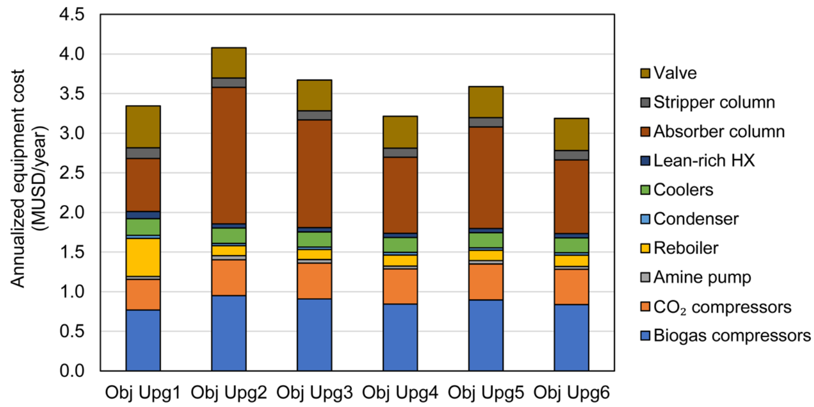

Figure 6. Annualized cost of each equipment for the best solution obtained from each objective function formulation.

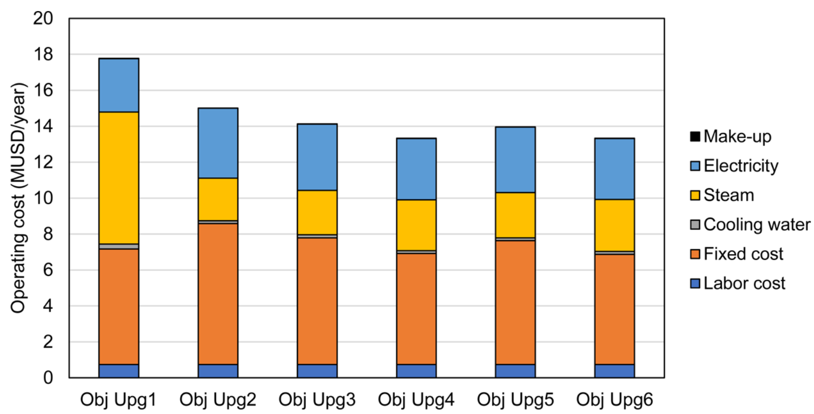

Figure 7. Distribution of operating costs for the best solution obtained from each objective function formulation.

annualized cost will be higher for Obj Upg3 as the operating pressure is higher than that for Obj Upg5. The solutions obtained from Obj Upg4 and Obj Upg6 provide approximately the same total annualized cost. To investigate the impact of each objective function formulation on the economy of the upgrading process, detailed breakdowns for the total annualized cost of different equipment and the distribution of operating costs are presented in Figures 6 and 7, respectively. In the upgrading process, the total operating costs are higher than the annualized CAPEX (see Figures 6 and 7).

As can be observed in Figure 6, a major part of the annualized equipment cost is allocated to the compression units (i.e., compressors and pumps) and the absorber column because they operate at high pressure. The results obtained from the different objective function formulations in Figure 6 illustrate that operating at lower pressure, and thereby with less compression work, is favorable to achieve a low investment cost. It should be noticed that the lowest operating pressure is obtained for Obj Upg1. However, the reduction in the cost of the compression units and the absorber obtained from Obj Upg1 is not enough to make up for an increase in the investment cost for the reboiler because of higher operating pressure and amine flow rate. The opposite can be observed when Obj Upg2 is employed. The investment cost for Obj Upg4 and Obj Upg6 is smaller than for Obj Upg3 and Obj Upg5 because of the reduction in the cost of compression units and absorber. As can be observed in Figure 7, the fixed cost (i.e., the costs related to maintenance, taxes, insurances, etc.), electricity, and stream are the major driving factors for the

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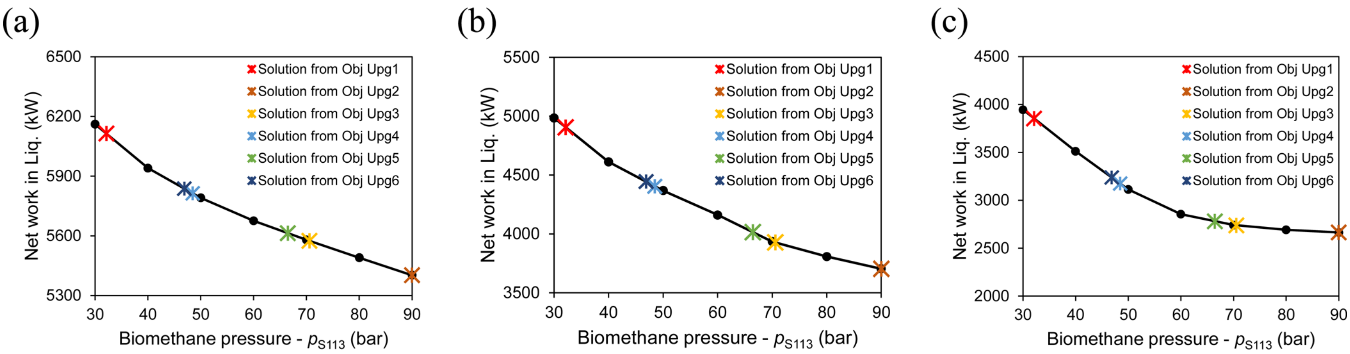

Figure 8. Network supply for (a) SE, (b) DE, and (c) SMR as a function of the biomethane pressure.

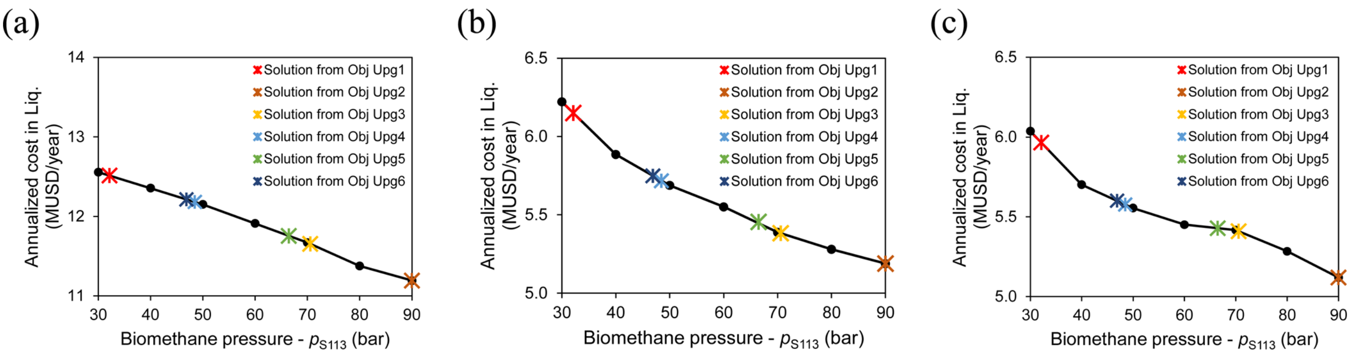

Figure 9. Total annualized cost for (a) SE, (b) DE, and (c) SMR as functions of the biomethane pressure.

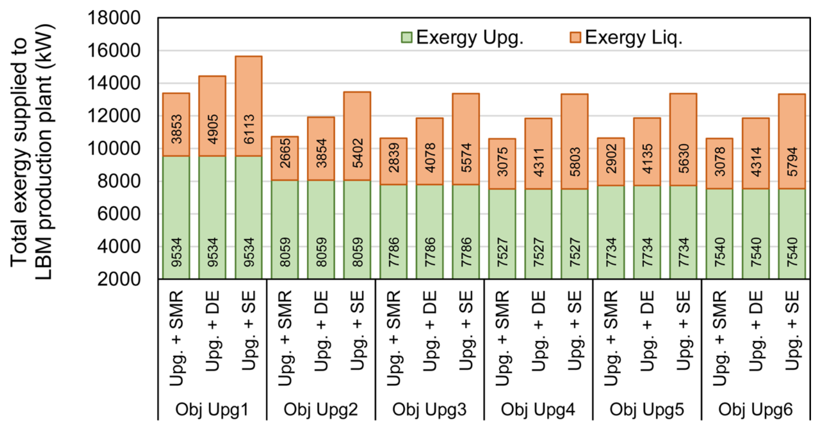

Figure 10. Overall exergy supply to the LBM production plant, corresponding to different objective function formulations for the upgrading process. The results are provided for SMR, DE, and SE.

operating cost. As the fixed cost is proportional to investment cost, any attempts to reduce the investment cost will also be reflected in the operating cost. As mentioned earlier, Obj Upg4 and Obj Upg6 will result in a smaller work demand and higher heat demand because of lower absorber pressure and higher amine flow rate in comparison with Obj Upg3 and Obj Upg5. As a consequence, the cost of stream will be higher, while the cost of electricity will be lower.

Overall, the operating pressure has a significant impact on the total annualized cost in the upgrading process. This can explain the similarity between solutions obtained from Obj Upg4 and Obj Upg6, as operating at a lower pressure not only provides a good compromise between work and heat supply (in Obj Upg4) but also reduced investment cost (in Obj Upg6). Therefore, at least for the biogas upgrading process, where the operating pressure is forced to be high to satisfy the CO_2 limit, the exergy-based objective function formulation provides high performance in terms of both thermodynamics and economics.

4.2. Liquefaction Process. As mentioned earlier, the pressure level of the biomethane stream leaving the upgrading process is the only variable that influences the subsequent liquefaction process.[28] In this section, the impact of the pressure level of the biomethane stream on the exergy supply and total annualized cost of the liquefaction process is investigated. The results obtained from optimizing the network (Obj Liq1) and the total annualized cost (Obj Liq2) for different refrigeration cycles (i.e., single expander nitrogen cycle (SE), dual expander nitrogen cycle (DE), and SMR) as functions of the pressure level of the biomethane stream are presented in Figures 8 and 9, respectively. Colored asterisks in the figures illustrate the pressure level of the biomethane

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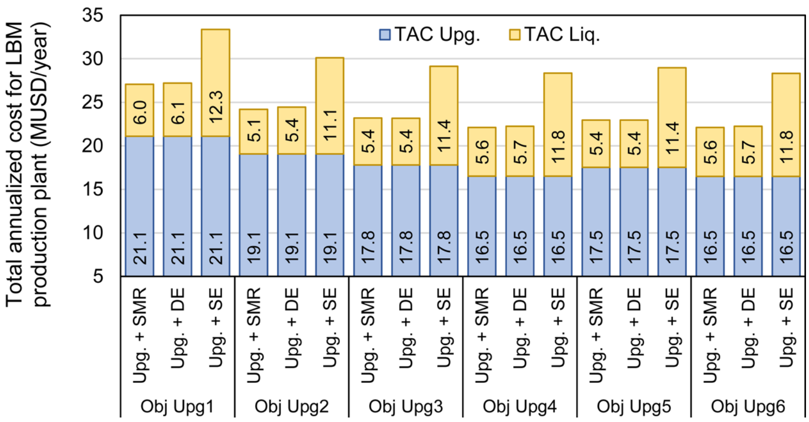

Figure 11. Total annualized cost of the LBM production plant, corresponding to different objective function formulations for the upgrading process. The results are provided for SMR, DE, and SE.

stream obtained from different objective function formulations for the upgrading process. It is worth mentioning that the best solution for the SE is obtained at the upper bound for the high-pressure level (i.e., p_{S202} = 120 bar).

It can be seen from the figures that increasing the biomethane pressure will result in a reduction in the network requirement and the total annualized cost of the liquefaction process. In addition, the network and the total annualized cost depend on the choice of refrigeration cycle. The lowest network and total annualized cost are both observed when the SMR is used followed by the dual expander cycle and the single expander cycle (see Figures 8 and 9).

As presented in Table 4, the various objective function formulations used for optimization of the upgrading process resulted in different absorber pressures and thereby different power consumption and total annualized cost for the liquefaction process. The higher the absorber pressure, the lower the network required in the liquefaction process (see Figure 8). The impact of using different objective formulations for the upgrading process on the overall exergy supply and the total annualized cost of the LBM production plant is illustrated in Figures 10 and 11, respectively. In Figure 10, the total exergy supply is the highest for Obj Upg1, with the highest exergy supply in both the upgrading process and the liquefaction process. As can be observed in Figure 10, the rest of the objective function formulations will result in a similar range for the total exergy supply, with the smallest value for Obj Upg4.

The choice of objective function formulation for the upgrading process does not make a huge difference for the overall process (i.e., upgrading and liquefaction). This is because the formulations resulting in higher exergy demand for the upgrading process often also provide a higher absorber pressure and thereby lower exergy demand for the liquefaction process. Likewise, the decrease in the total annualized cost of the liquefaction process makes up for the increase in the total annualized cost of the upgrading process obtained from different objective function formulations. The main difference between the different solutions is due to the selection of the refrigeration cycle.

Overall, the results indicate that the objective function formulations based on exergy supply or total annualized cost can optimize an LBM production plant which leads to similar results, obtaining thermodynamically and economically efficient solutions. The lowest total annualized cost is obtained when the SMR is employed, closely followed by the dual mixed expander cycle. The results from Figures 10 and 11 illustrate the importance of the refrigeration cycle selection to improve the thermodynamic efficiency and total annualized cost of an LBM production plant, suggesting that using a single mixed refrigerant in the LBM production plant could benefit both the total exergy supply and the total annualized cost.

5. CONCLUSIONS

In the present study, a complete LBM production plant including a biogas upgrading process and a biomethane liquefaction process was simulated and optimized using Aspen HYSYS. Amine-based absorption using MDEA was considered for the upgrading, while multiple refrigeration cycles (i.e., single/dual expander nitrogen cycles and an SMR cycle) were investigated for the liquefaction. The present study aimed to compare solutions obtained from energy-, exergy-, and economy-based objective function formulations for optimization of thermodynamic and economy performance of the LBM production plant.

Based on the best results obtained for the upgrading process, the network required in the liquefaction process and the total annualized cost of the liquefaction process were optimized considering a sequential optimization approach.

The results from optimizing the upgrading process illustrated that, in comparison with the energy-based formulations, the exergy-based formulation shifted the solution toward more heat and less work. Consideration of UCs for the upgrading process provided a solution similar to the one obtained when minimizing the total energy supply. Moreover, the solutions obtained from objective functions based on exergy and total annualized cost were similar. From an economic point of view, a reduction in the total annualized cost could be obtained by reducing the investment cost. As the investment cost for equipment operating at higher pressure was dominant, solutions with a lower operating pressure provided lower total annualized cost (i.e., employing objective functions based on exergy supply or the total annualized cost).

The biomethane pressure, which is the only parameter of the upgrading process influencing the performance of the subsequent liquefaction process in the studied process, would have a similar effect on the total exergy supply and the total annualized cost of the liquefaction process, as increasing the biomethane pressure resulted in a reduction in the exergy supply and the total annualized cost of the liquefaction process.

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When considering the entire LBM production plant (i.e., upgrading and liquefaction), all objective function formulations resulted in a similar total exergy supply as the savings in the exergy supply to the upgrading process for some formulations would be offset by additional compression work in the liquefaction process. A similar trend was observed for the total annualized cost. The results were approximately the same both for the thermodynamic and economic optimization, indicating that maximizing the exergy efficiency would also lead to minimized total annualized cost. Furthermore, the selection of the refrigeration cycle had a profound impact on the performance of the overall LBM production plant, both in terms of thermodynamic efficiency and cost. The optimization of the overall LBM production plant in the present study suggests that selecting an appropriate refrigeration cycle (i.e., single mixed-refrigerant cycle provided the lowest total exergy supply and total annualized cost) would be more important than the way of formulating the objective function for the optimization.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.iecr.1c04378.

"Sensitivity analysis with respect to decision variables and biogas composition" and "detailed costing method including corresponding parameter values" (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. Conceptualization, S.E.H.; methodology, S.E.H.; software, S.E.H.; validation, S.E.H., D.K., and B.A.; formal analysis, S.E.H.; investigation, S.E.H.; data curation, S.E.H.; writing—original draft preparation, S.E.H.; writing—review and editing, D.K. and B.A.; supervision, B.A.; and project administration, B.A.

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