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# Biogas upgrading by amine scrubbing: solvent comparison between MDEA and MDEA/MEA blend

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**Abstract**

Biogas upgrading to biomethane has received particular attention in recent years as a key strategy to produce a natural gas-substitute from biomass. Among the available upgrading technologies, amine-based absorption is an interesting option because of its low electric power consumption and very high methane recovery rates. This paper assesses the energy and economic performance of a biogas upgrading process involving chemical absorption with two different aqueous solvent formulations: 50%$_w$ MDEA and a 20%$_w$/20%$_w$ MDEA/MEA blend. The process performance is evaluated with Aspen Plus and the optimal process conditions are determined with a multi-objective optimization approach targeting the maximum efficiency and minimum specific equipment cost. In the proposed scheme, to make the system energy-self-sufficient, an internal combustion engine burns a fraction of the raw biogas stream to co-generate both the electric power for the upgrading process and the thermal power required for solvent regeneration.
Aqueous MDEA turns out to be more efficient (Pareto dominant curve) and less expensive than the MDEA/MEA blend, as a result of the lower regeneration energy (0.94 kJ/Nm$^3_{BM}$/h vs 1.43 kJ/Nm$^3_{BM}$/h) which yields to lower energy consumptions and smaller engine sizes. Even though the cost estimates are subject to a higher degree of uncertainty, the MDEA option features an expected specific total equipment cost as low as 1,550 €/(Nm$^3_{BM}$/h)) vs. 1,850 €/(Nm$^3_{BM}$/h)) for the MDEA/MEA blend.



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## 1. Introduction

Biomethane is a Natural Gas surrogate, which can be produced from the purification (i.e. removal of components other than $CH_4$) of biogas. Biogas is a gaseous mixture composed of $CH_4$, $CO_2$ and other impurities which is generated from the anaerobic digestion of organic compounds. Biomethane is gaining interest as a promising energy carrier since it allows to: i) address environmental concerns associated with increasing $CO_2$ emissions in atmosphere, ii) produce a substitute for fossil natural gas exploiting a renewable energy source and iii) create diversification in the energy supply chain.

Currently, biogas is exploited for combined production of heat and power or more often just for electricity generation by direct combustion. However, these utilization paths entail reduced conversion efficiencies and scarce possibility for heat recovery. The final usage of the methane content of biogas as vehicle fuel or its injection in the natural gas grid may represent more efficient utilization paths, but they are still at an earlier stage of diffusion. At the European level, in 2015, 496 biomethane plants were in operation according to [1]. Among them, pressurized water scrubbing is the most widespread technology for biogas upgrading with 162 operating plants, followed by membrane technology and chemical scrubbing technology (both with 93 plants), Pressure Swing Adsorption technology with 78, and organic physical scrubbing with 24 plants. The main limitation to the diffusion of biomethane is represented by the availability of technologies that, at low cost and reduced energy consumption, are able to process biogas to make it available for the last two purposes mentioned, so that the viability of the value chain is still based on subsidies.

Moreover, for many technologies the influence of key operational parameters on the upgrading process itself is still unclear, thus limiting the technology diffusion at the industrial level. On the other hand, the $CO_2$, separation process has been widely investigated for $CO_2$ Capture and Storage (CCS) from flue gases from fossil fuel or for natural gas sweetening. Biogas upgrading differs from such applications for the larger partial pressure of $CO_2$, and for the different average plant size: $CO_2$ content in the feed gas ranges from 4%mol to 15%mol for CCS applications where flue gases flowrates are of the order of 500-5000 kNm$^3$/h while it reaches up to 50%mol for biogas while the flow rate to be treated in biogas upgrading plants is determined by the size of the anaerobic digester, which is quite limited (the average size in Italy is ~ 450 Nm$^3$/h).

Table 1 Biogas upgrading by amine scrubbing and comparison with water scrubbing: literature review.

| Reference | [2] | [3] | [4] |
|---|---|---|---|
| Solvents | water (100) | water (100) & MEA (15-30) | Water (100) & MDEA (50) |
| Plant size [Nm3biogas/h] | 500-1000 | 720 | 850-1350 |
| Biomethane purity [%mol] | 99 (dry) | 98 (dry) | - |
| Methane slip [%] | 0.4 | <0.1 (MEA) | - |
| Specific consumptions [kWh/Nm$^3_{BG}$] | 0.331 (el.) | | 0.29-0.3 (water/el.)<br>0.08-0.09 (MDEA/el.)<br>0.27-0.3 (MDEA/th.) |

The present work proposes an analysis of a chemical scrubbing upgrading process, which is based on amines in aqueous solutions as solvent. The process takes advantage of the high selectivity of the solvent that, via chemical reactions, binds with $CO_2$. In literature, several works on biogas upgrading with different scrubbing solvents are available: Magli et al. [2] perform a techno-economic optimization of water scrubbing process, Gamba et al. [3]

compare a Monoethanolamine (MEA) based scrubbing and a water scrubbing with a sensitivity analysis on the Liquid to gas (L/G) ratio to determine the optimal solvent requirement. Pellegrini et al. [4] evaluate the influence of the biogas composition on the purification cost and consumption, comparing a Methyldiethanolamine (MDEA) based scrubbing and a water scrubbing, while also performing a sensitivity analysis on the economic profitability. A short summary on the cited works is reported in Table 1.

**Nomenclature**

| | |
|---|---|
| BG | Biogas |
| BM | Biomethane |
| CEPCI | Chemical engineering Plant Cost Index |
| HETP | Equivalent Height of Theoretical Plate |
| ICE | Internal Combustion Engine |
| LHV | Lower heating value |
| MEA | Monoethanolamine |
| MDEA | Methyldietanolamine |

The objective of this work is to determine the optimal design of an amine scrubbing plant for biogas upgrading, according to both energy and economic indicators. Since the choice of the most suitable amine type looks crucial, two distinct systems are compared in this work, Amine Scrubber 1 (AS1) and Amine Scrubber 2 (AS2). Both configurations are based on the same plant layout, but envisage different solvents, as reported in Table 2. In section 2, the modelling assumptions and the main design choices are stated, while the methodology is outlined in Section 3. In section 4 the results are presented, and then conclusions are drawn in section 5.

## 2. Process modelling

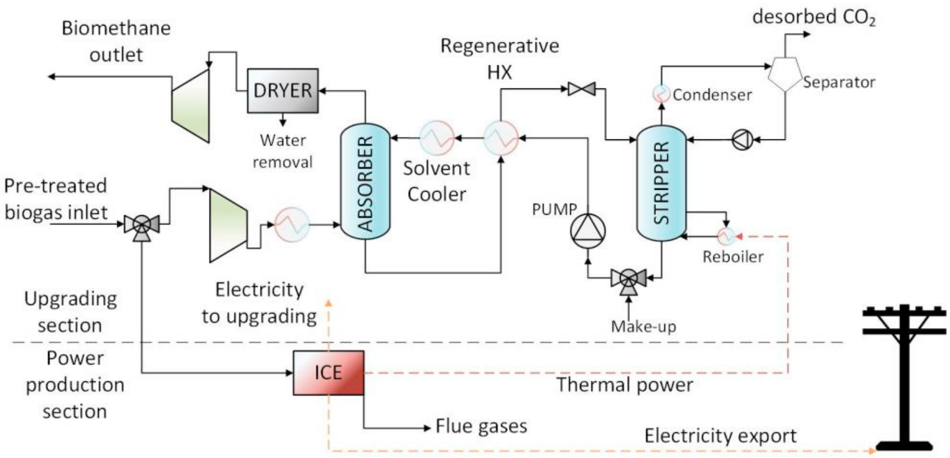


Figure 1 Plant lay-out for the amine scrubber assumed in this work. ICE represents the Internal Combustion Engine used to cover the thermal and electric demand of the upgrading section, while injecting the electricity in excess to the grid.

In this study, a medium-large scale biomethane production plant is considered, with a biogas flow rate of 1,075 $Nm^3/h$. Smaller size plants are not expected to be economically attractive for chemical solvent scrubbing because of the adverse economies of scale on the equipment costs. Biogas composition is assumed 44% $CO_2$, 55.6% $CH_4$ and 0.4% $O_2$ (on a molar basis), in agreement with the composition of biogas produced by anaerobic digestion of maize silage, grass haylage and rye grain combined with liquid cattle manure [5]. The layout of the amine scrubbing plant envisaged in this work is outlined in Figure 1: biogas is first compressed and it is sent to an after-cooler. Then, it enters the absorption column, where it is contacted counter-current with the lean solvent, which binds selectively with the $CO_2$ molecules via chemical reactions. Purified biomethane leaves from the top of the column, while the $CO_2$-rich solvent exits from the bottom of the column. The solvent loaded with $CO_2$ is heated in a regenerative heat exchanger and then sent to the stripping column, where it is regenerated thanks to the heat supplied by the reboiler. $CO_2$ is released from the top of the stripping column, where a condenser removes most of the evaporated water, and the lean solvent is pumped from the bottom to the regenerative heat exchanger, where it is pre-cooled. The lean solvent is further cooled and recycled to the absorption column.

In this work, we assume to produce the thermal power needed to regenerate the solvent and to run the anaerobic reactor in an internal combustion engine, which operates in a cogenerative configuration and is fed with a biogas bleed taken from the outlet of the anaerobic digester. Such arrangement enables the plant to be energy self-sufficient and to achieve a more efficient use of primary energy (also the electric energy is used locally to power the upgrading plant). Moreover, feed gas and biomethane compressors and units for water removal from biomethane are added to the equipment list to meet the specifications for grid injection, which is the final use assumed for the produced biomethane. The ICE is sized based on the thermal demand of the upgrading section (the most relevant portion of the energy demand) and hence produces more electric power than the one needed for the plant operation. The surplus of electricity production is assumed to be exported to the electric grid. Thus, biomethane and electric power are the two useful effects available from the operation of the plant.

As reported in Table 2, two different solvent compositions are considered: in AS1 an aqueous solution of Methyldietanolamine (MDEA), while for AS2 a blend of MDEA and Monoethanolamine (MEA) in aqueous solution. MDEA is selected for its high capturing capability, limited thermal requirement for regeneration and limited corrosion problems. The MDEA/MEA blend is instead selected for its improved kinetics with respect to sole MDEA [6]. The amine concentration in the solvent is fixed to the maximum solubility limit for AS1 [4], while for AS2 the considered amine concentration could be further increased before reaching the solubility limit, however the lack of experimental data prevents to validate the absorption model for such range of solvent composition.

Table 2 Solvent features for AS1 and AS2 cases

| | **AS1** | **AS2** |
|---|---|---|
| Amine type | MDEA | MDEA+MEA |
| Solvent concentration (wt) | 50% | 20% |
| MEA:MDEA ratio (mol:mol) | 0:1 | 1:1 |
| Rate-based model validation | [7] | [8] |

The upgrading section is simulated with the Aspen Plus® flowsheeting software, while the models for the other units are developed in Matlab. The liquid phase non-ideality of the systems is modelled with electrolyte-NRTL Equation of State (EoS) [9], while the vapour phase is modelled by means of the Redlich-Kwong EoS. A detailed rate-based model (validated for the considered composition and temperature as reported in Table 2) is applied to describe the absorption column, since the $CO_2$ absorption process is reaction rate-limited (relevance of both kinetics and mass diffusion effects). The default mixed flow model readily available in Aspen Plus v9 is adopted, assuming that the average bulk properties in each stage and for each phase are equal to the stream outlet properties. The two films theory is used to model the mass transfer resistance. The absorber is modelled in Aspen Plus using the RadFrac column, assuming a 21 stages discretization, while the height of the absorber is a decision variable of the optimization problem. The packing material is assumed to be Metal pall ring, 25 mm, with a packing factor of 56 $ft^{-1}$ [10]. The sizing of the column is based on Stringle's Generalized Pressure Drop Chart [11]. Column operation is suggested in the range of fractional flooding between 0.5 and 0.7 [12], and a value of 0.6 is selected.

An equilibrium-based approach is selected to describe the desorption process, due to the faster kinetics here encountered, which are due to the higher temperatures that drive desorption reactions to equilibrium. The number of stages in which the desorption column is discretised is kept constant and equal to 11, including a partial vapour condenser at the top of the column and a kettle reboiler at the bottom. The height of the stripping column is estimated with the Equivalent Height of Theoretical Plate (HETP) approach, which has proven effective in predicting height of packed columns [13] for a wide range of real column operation data and field experience. The ICE is modelled with the commercial software Thermoflex® for a reference size of 250 $kW_{el}$ (which is expected to be close to the optimal design size since it satisfies both the thermal and electric loads of the biomethane plant), and then electric and thermal efficiencies are kept constant throughout the optimization of the engine size.

An economic model is developed in order to determine the purchase cost of each equipment unit of the plant. Such model is made up by two steps: first, the size of each equipment unit is determined via well-known engineering procedures, then the relative purchase cost is determined applying literature correlations [14]. (i.e., cost correlations depending on material and heat transfer area for heat exchangers, diameter and height for columns, consumed or produced power for pumps, compressors and the ICE). The analysis is based on commercial off-the-shelf costs. Costs related to the connection of the plant to the natural gas and electric grid, contingencies and balance of system represent a fixed cost in the framework of this work and were not optimized.

## 3. Methodology

Multi-objective optimization is widely recognized as a powerful tool to determine the set of optimal plant designs and operating conditions, especially when cost related data or economic assumptions are quite uncertain. Indeed, multi-objective optimization returns the set of optimal process designs for a wide range of relative weights of the two conflicting objective functions (typically related to energy efficiency and costs). In this work, energy and economic performances of the plant are evaluated by means of two indexes:

1. primary energy efficiency, $\eta_{PE}$

$$\eta_{PE} = (\dot{m}_{BM} \cdot LHV_{BM} + \dot{W}_{el,export}/\eta_{el\ import,ref}) \,/ (\dot{m}_{BG} \cdot LHV_{BG} + \dot{W}_{el,import}/\eta_{el\ export,ref}) \tag{1}$$

which takes into account of the two useful effects (i.e., produced biomethane flow rate $\dot{m}_{BM}$ and the optional electricity export $\dot{W}_{el,export}$) and the consumed primary energy (i.e., biogas chemical power $\dot{m}_{BG} \cdot LHV_{BM}$ and the optional electricity import $\dot{W}_{el,import}$). The average conversion efficiency of the Italian electric grid is assumed as reference electrical efficiency ($\eta_{el,ref}$) to assess the primary energy consumption associated to $\dot{W}_{el,import}$.

2. total equipment cost of the plant, $TEC$

$$TEC = \sum_i^N EC_i \tag{2}$$

The TEC is used to measure economic performance of the plant. TEC is obtained as the sum of the costs needed to purchase all the required equipment units. In equation (2), $EC$ is the cost of the equipment unit, index i spans the equipment units and N is the total number of units.

The decision variables are the same for both processes (AS1 and AS2), and they are the key parameters for the design and operation of the amine scrubbing plant:

1. $L/G$ [kg/kg], Liquid to gas mass ratio at the absorber (wt)
2. $\dot{Q}_{REB}$ [kW], Reboiler heat duty
3. $h_{ABS}$ [m], Absorber packing height
4. $p_{ABS}$ [bar], Absorption pressure
5. $p_{STR}$ [bar], Stripping pressure

The problem is subject to the following constraints: 1) the purity of the produced biomethane has to be suitable for grid injection according to the Italian grid code (i.e., the molar fractions of $CO_2$, $\phi_{CO_2\,BM}$, and $O_2$, $\phi_{O_2\,BM}$, must be lower than 3% and 0.6% respectively; 2) the thermal energy produced by the ICE, $\dot{Q}_{ICE}$, has to satisfy the heat demand from the upgrading section $\dot{Q}_{UP}$ (it is assumed that the low temperature thermal energy for digesters heating can be recovered as well from the waste heat of the ICE).

The resulting multi-objective optimization problem can be formulated as follows:

$$\min(-\eta_{PE}, TEC) \text{ s.t.}$$

$$\phi_{CO_2\,BM} < 3\%\ \text{mol} \tag{3}$$

$$\phi_{O_2\,BM} < 0.6\%\ \text{mol}$$

$$\dot{Q}_{ICE} \geq \dot{Q}_{UP}$$

In order to tackle such process optimization problem, the black-box approach is selected. In the black box approach, the optimization algorithm samples the feasible space of optimization variables and, for each sampled solution, the flowsheet simulation is executed. The term black-box is referred to the optimization algorithm which cannot see the flowsheet model equations but just the resulting values of the objective functions and constraints. In this work, the black-box approach appears as the most advantageous one because:

- It is a feasible path method, meaning that also intermediate suboptimal solutions are still feasible;
- It allows using of the detailed models developed in Aspen Plus.
- Most important, the convergence of the models proved to be extremely challenging and unstable, which call for the use of robust derivative-free optimization algorithms. Indeed, since the objective functions and constraints are computed by a process simulator code, objective functions and constraints are typically noisy (due to the convergence tolerance of the internal simulation routines), non-smooth, discontinuous and even not defined in some points (where the process simulation fails to reach convergence) [15].

In this work, NSGA-II [16] (a well-known genetic algorithm readily available in the Matlab global optimization toolbox) has been selected as robust black-box multi-objective optimization algorithm. The problem constraints are dealt with a quadratic penalty approach, in which the objective functions are penalized proportionally to the constraints violation. However, preliminary analysis showed that NSGA-II is not able to return the complete Pareto frontier. For this reason, a two stages optimization strategy is adopted: i) at the first stage, two single-objective optimizations ($\min(-\eta_{PE})$, $\min(TPC)$) are performed with PGS-COM [17] (a single-objective black-box algorithm specifically developed for process optimization problems) to locate the maximum efficiency and minimum cost designs (extreme points of the Pareto front), ii) at the second stage, the two previously found solutions are used to initialize the multi-objective optimization with NSGA-II. In this way, the two extreme solutions (the most conflicting ones) from the first stage optimization are certainly included in the final Pareto curve, whose extension is hereafter maximized.

## 4. Results

The Pareto frontier for the AS1 and AS2 processes are shown in Figure 2. The most interesting result is that the Pareto front of AS1 dominates that of AS2 indicating that the AS1 process achieves higher primary energy efficiency and lower investment costs. For the range of efficiencies achievable by both processes (91.25 - 91.5%), AS1 shows a 20% lower specific equipment cost (1,550 - 1,850 €/($Nm^3_{BM}$/h) vs. 1,850 - 1,900 €/($Nm^3_{BM}$/h) for AS2).

As far as energy efficiency is concerned, only MDEA-based solvent is able to attain energy efficiency higher than 91.5%. The optimal decision variables are reported in Table 3 for the economic and energy optimum configurations

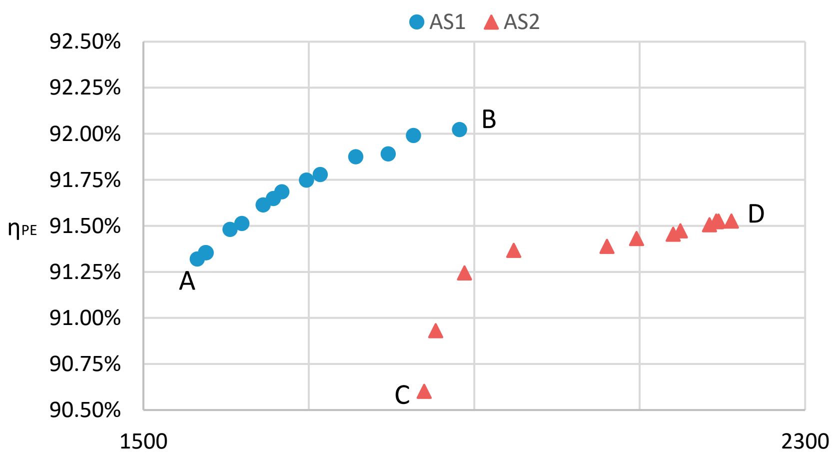


Figure 2 Optimal Pareto curve for the AS1 and AS2 configurations

(which represent the opposite extremes of the respective Pareto curves, and are therefore considered for comparison), for both AS1 and AS2 configurations. It can be observed that, for both processes, the height of the absorption column decreases when going from the maximum efficiency extreme (where the optimizer sets the height to the maximum allowed value to minimize the required solvent flow rate and regeneration heat) to the minimum equipment cost, with a decrease of 46% and 75.6% respectively for the two processes. The short absorber height of process AS2 is due to the increased reactivity and $CO_2$ absorption rate of the MDEA/MEA blend compared to pure MDEA.

On the other hand, it can be observed that the amount of thermal power needed for the solvent regeneration is appreciably higher for AS2 (1.4 kJ/Nm$^3_{BG}$) than for AS1 (0.94 kJ/Nm$^3_{BG}$) due to the heat required by the decomposition reaction of carbamate and the lower overall concentration of amines (being more diluted than in AS1).

Comparing the maximum efficiency and the minimum cost designs, it can be observed that the economic objective pushes towards higher absorption pressures, since they help in reducing the sizes of the columns and allow operating with a lower L/G ratio. In addition, the stripping pressure is reduced to help the $CO_2$ desorption, since pumps for bringing the solvent back to the absorption pressure are relatively economic (and $CO_2$ is vented and not captured/re-used).

Table 3 Energy and economic optimal solution for AS1 and AS2 configurations. Points are labelled with letters, referring to Figure 2.

| | (A) AS1 economic optimum | (B) AS1 energy optimum | (C) AS2 economic optimum | (D) AS2 energy optimum |
|---|---|---|---|---|
| $L/G$ | 11.2 | 13.2 | 16.8 | 17.4 |
| $Q_{REB}$, [kW] | 617.1 | 664.5 | 792.3 | 798.4 |
| $q_{REB}$, kJ/Nm$^3_{BM}$ | 1.02 | 0.94 | 1.40 | 1.43 |
| $h_{ABS}$, [m] | 13.4 | 25 | 6.1 | 25 |
| $p_{ABS}$, [bar] | 7.4 | 3.5 | 6 | 2.5 |
| $p_{STR}$, [bar] | 1.5 | 2 | 1.3 | 2 |

Figure 3 shows the cost breakdown for the minimum cost design of cases AS1 and AS2 (where the total equipment cost is 940 k€ for AS1 and 1,037 k€ for AS2). It should be noticed that while the biogas input is fixed, the size of the upgrading plant (measured by the biomethane productivity) depends on the amount of biogas required by the ICE and then on the upgrading efficiency itself. The most expensive equipment unit is the ICE in both cases (respectively 39% and 50% of the TEC), followed by the absorber (15% and 8% of TEC) and stripper (14% and 16% of TEC, including reboiler and condenser). Such results indicate that the economic advantage of process AS1 is mainly due to the requirement of a smaller ICE to provide the regeneration heat. Such cost saving more than compensate the increased cost of taller absorber column compared to AS2.

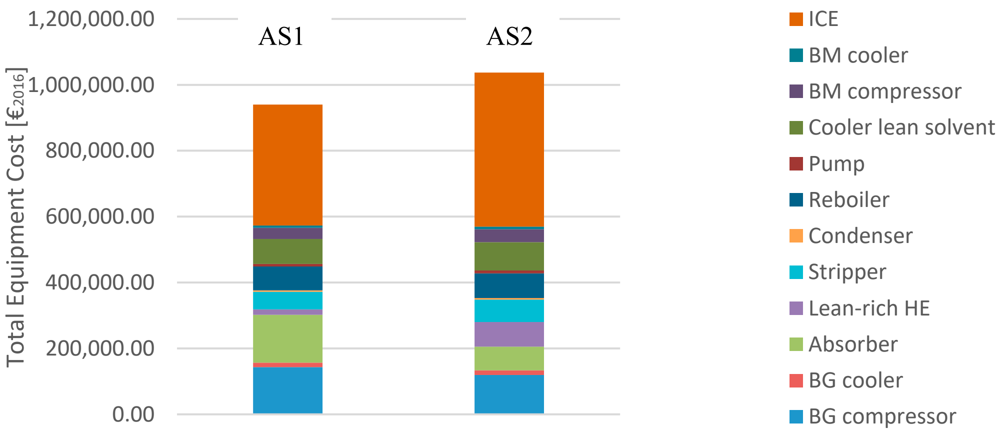


Figure 3 Total Equipment Cost detail for the economic optimal solutions for configuration AS1 and AS2

## 5. Conclusions

The techno-economic optimization of two different amine scrubbing processes for biogas upgrading plants is performed. Solvent AS1 uses an aqueous solution of MDEA while AS2 uses a blend of MDEA and MEA with the aim of increasing the $CO_2$ absorption rate. For both processes, accurate thermodynamic models and economic models are developed and integrated with a multi-objective process optimization approach. Both AS1 and AS2 processes are able to produce biomethane in compliance with grid specifications. Process AS1 outperforms AS2 in terms of both maximum efficiency (92% vs 91%) and minimum specific equipment cost (1,550 vs 1,850 €/($Nm^3_{BM}$/h)). The main reason appears to be the lower solvent regeneration heat, which leads to lower energy consumptions and lower investment costs for the heat supply system (i.e., CHP internal combustion engine). However, process AS2proved to be effective in reducing the size of the absorption column, and could be the preferred choice if waste heat is available. Future works will investigate the effect of using higher amine concentrations for the MDEA/MEA blend.

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