

Energy requirements, GHG emissions and investment costs in natural gas sweetening processes

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A B S T R A C T

Currently, different technologies are being employed to remove CO₂ and H₂S from the natural gas. Based on chemical phenomenon, the absorption using alkanolamines is probably the most extended process for the acid gas removal. However, membrane technologies are considered as an alternative in specific cases for their better performances, cleanness, energy requirements, operative costs and location flexibility.

The aim of this article is to estimate, compare and analyze the energy requirements, greenhouse gases (GHG) emissions and investment costs of three Natural Gas Sweetening processes. For the study, a regular process using methyldiethanolamine (MDEA), the absorption process using recompressed vapor and a membrane system were simulated using Aspen Hysys v8.8. For the first case, real data from the gas plant Aguara Güe (Argentina) was used to validate the model. To establish a proper comparison, a natural gas with 4 mol.% of CO₂ is considered as the inlet stream of each configuration. Specifically, compression and pump power, specific total heat, removed CO₂, CH₄ wastes and capital costs were estimated and compared for each case. Additionally, a discussion including different aspects in regard to the energy efficiency of the processes was conducted.

Although the proposed membrane system demonstrated to reduce the energy requirements (77% and 72%) and emissions (80% and 76%) in respect to both absorption processes, the CH₄ losses were higher by more than 6 factor. Moreover, the investment cost of the technology is 12% higher than the required capital of a conventional amine process.

Keywords:

Natural gas sweetening
Methyldiethanolamine
Polymeric membranes
Energy
GHG emissions
Aspen hysys

1. Introduction

The increasing world-wide energy demand and pricing tendencies have directed the attention to oil and gas companies towards improving the energy efficiency (Ahmad et al., 2012). Different alternatives for enhancing the energy requirements are evaluated to measure the impact of their implementation on the global energy recovery. In this sense, retrofitting of existing plants plays an important role for reducing raw materials replacement, achieving sustainable development, mitigating the significant environmental impacts but mainly for achieving energy efficient buildings (Zhou et al., 2016).

As the largest contributor to global warming, increments of CO₂ emissions have urged continuous pressure on companies to adopt practices that reduce carbon footprint in natural gas processing to mitigate the effect of climate change (Gnanendran and Hart, 2009). In addition to atmospheric pollution, the presence of CO₂ also reduces the heating value and causes the natural gas stream to be acidic and corrosive, which in turn minimizes the possibility of gas compression and transportation (Zhang et al., 2013). In Argentina, the specification for the natural gas to be transported is ruled by the government through the ENARGAS. Particularly, the molar percentage of the CO₂ in the natural gas must not exceed 2% (Gutierrez et al., 2016).

According to Øi et al. (2014), the most probable method for CO₂ capture from acid natural gas in the nearest future is the absorption into an alkanolamine solvent. Chemical absorption for CO₂ sequestration is based on the exothermic reaction of the solvent with the CO₂ present in the gas stream at low temperatures. In a

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second stage, the reaction is reversed at higher temperatures in a regeneration tower for the solvent recovery (Yeo et al., 2012). In typical configurations of chemical solvents, the thermal energy for CO₂ desorption is considerably high and thus different configurations to reduce the energy requirement have been proposed by many authors. The state of the art for the CO₂ absorption using alkanolamines involves differences in the configuration of the process and the implementation of different alkanolamines solvents and new combinations to enhance the process (Polasek et al., 1982; Sohbi et al., 2007; Fouad and Berrouk, 2013; Al-Lagtah et al., 2015). Among other authors, Kohl and Nielsen (1997) and also Cousins et al. (2011) evaluated flow sheet modifications to improve the energy requirement of Natural Gas Sweetening processes. Øi et al. (2014) studied an alternative configuration for Natural Gas Sweetening using alkanolamines where a vapor recompression stream is used to reduce the energy of desorption and the associated costs.

Besides chemical solvents, new technologies to achieve CO₂ pipeline specification such as the implementation of membrane modules are under study. Gas separation using membranes is a kinetics-based process, its rapid expansion is attributed to various advantages in comparison to the conventional technologies, such as being environmental friendly, occupying a relatively smaller footprint and cost efficient ascribed to the low energy consumption (Lock et al., 2015a). The membrane separation process is based on differences in the diffusion rates of gas molecules within the membrane materials. In particular, membranes to acid gas separation (CO₂ and H₂S), heavy hydrocarbon removal, water removal, N₂ removal as well as Helium recovery are technologies in the gas processing industries (Scholes et al., 2012). In this regard, many studies concern the membrane technology as an alternative to separate CO₂ from different types of gas fluids such as natural gas, power plant flue gas and syngas (CO+H₂) (Lock et al., 2015b; Roussanaly et al., 2016; Vakharia et al., 2015).

In the present article, authors compared a typical configuration and operating Natural Gas Sweetening plant with two scenarios; the first scenario is to revamp it with re-compression vapor and the second is to replace the plant with a membrane process. For this purpose, the three cases were simulated employing the simulator Aspen Hysys v8.8 (Aspen Technology Inc, 2012). Particularly, the Activated Energy Analysis extension was applied to quantify the variables of interest. To establish a proper comparison, the same

natural gas with around 4 mol.% of CO₂ was used as the inlet stream of each process. For the amine cases, a MDEA 38 wt% molar aqueous solution is selected as the absorption solvent. On the other hand, two polymeric membrane modules of 2000 and 1000 m² are assumed for the membrane process. To validate the simulation data, different measured variables from the Argentinean absorption plant Aguaraquí were compared. It is observed that in all of the cases the sweet gas achieved the specification of CO₂ 2 mol.% in its composition.

In the results, differences related to energy requirement, power consumption, specific total heat, GHG emissions, water make-up, capital and operating costs are shown. For example, it is observed that the required energy for the membrane module is lower than those for the amine processes. Contrarily, the capital cost needed for implementing a 2-stage membrane process is notably higher than for the amine processes. As it can be expected, it is easy to conclude that many trade-offs exist among the proposed processes and potential energy savings between them. From this analysis, an objective function including both energy consumption and capital cost could be optimized to define the best conditions for each process under study.

1.1. Processes description

1.1.1. CO₂ absorption using alkanolamines

Fig. 1 represents a simple amine sweetening facility (Lunsford and Bullin, 1996). In a typical process configuration, the acid natural gas is introduced in an absorber tower. Internally, the gas is put in contact with a descendent flux of an aqueous alkanolamine solution (lean amine). On the tower trays, the CO₂ of the feed gas reacts with the alkanolamine, sweetening the natural gas. A liquid stream composed by the amine aqueous solution and the absorbed acid component (rich amine) leaves the tower through the bottom.

In a second stage of the process, the rich amine solution fluxes through a lean/rich amine exchanger to increase its temperature. The heated rich amine solution is introduced to a stripper tower where the acid gas and water are removed. In addition, a circulation pump is needed to keep the stream pressure over the absorber internal pressure. Finally, an air cooler reduces the lean amine temperature before being sent back to the contactor tower (Behroozsarand and Zamaniyan, 2011; Gutierrez et al., 2016).

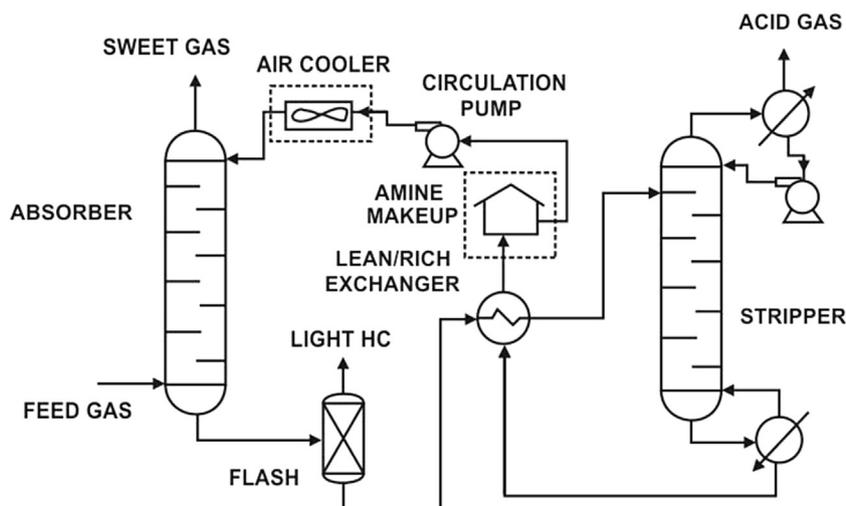


Fig. 1. Scheme of a simple amine sweetening plant (Lunsford and Bullin, 1996).

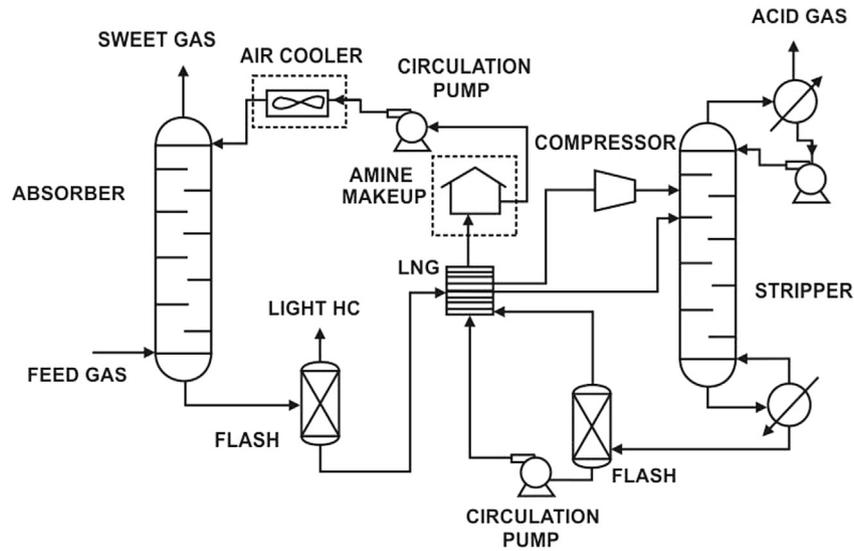


Fig. 2. Scheme of an amine sweetening plant with vapor recompression.

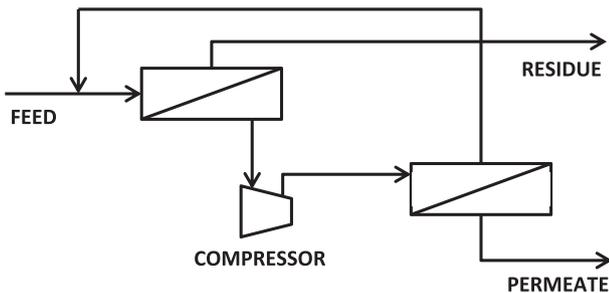


Fig. 3. Scheme of a simple two-stage membrane process (Kohl and Nielsen, 1997).

1.1.2. CO₂ absorption using alkanolamines with vapor recompression

Fig. 2 shows the principle of the vapor recompression configuration. Basically, it is a typical configuration with the next modifications. Firstly, the regenerated amine solution from the stripper bottoms is pressure reduced and sent to a flash tank. The liquid from the tank is the lean amine stream which is recirculated back to the absorber, heating the stripper inlet stream. Then, the vapor from the flash tank is cooled, compressed and returned to the bottom of the stripper. The vapor recompression produces a regenerated amine with less CO₂ which absorbs more CO₂ in the absorber (Øi et al., 2014).

1.1.3. CO₂ separation using polymeric membranes

A range of process designs exists with a standard two-stage membrane process configuration (Bhide and Stern, 1993). In this design (Fig. 3), the first membrane unit separates CO₂ from the feed gas to achieve pipeline specification (Residue). However, as methane also permeates through the membrane losses of 12% are observed. Therefore, the permeate stream from the first membrane unit is compressed and passes through a second membrane unit to recover the CH₄. This is operated at a low stage cut to minimize CH₄ losses through the membrane to 1.5%. The retentate from the second membrane unit, which has a significant amount of CO₂, is recycled to the feed of the first membrane unit (Scholes et al., 2012).

2. Methodology

2.1. Base case

An alkanolamine process for the CO₂ absorption from natural gas (NG) was simulated in steady state employing Aspen Hysys v8.8, Fig. 4. The design, the conditions of the gas to treat and the solvent characteristics were taken from the treating facility Agüarague, a real plant located in the north of Argentina (see Table 1). For the simulation, the Acid Gas thermodynamic package and efficiency calculation type were used. This Acid Gas package was developed by Aspen Technology Inc (2012) and includes the Peng-Robinson equation-of-state for vapor phase and the electrolyte non-random two-liquid (eNRTL) activity coefficient model for electrolyte thermodynamics (Song and Chen, 2009) and physical property data for aqueous amine solutions (Zhang and Chen, 2011; Zhang et al., 2011).

2.1.1. Base case validation

For the validation of the simulation output, different operative parameters were compared with values measured in the treatment facility considered. In Table 2, values of operating data, simulation output and relative error percentage are shown.

Respect to the deviation observed, the R. E. calculated is due to the fact that the input data were taken from a unique operating point. In order to obtain lower R. E. percentages, data reconciliation including a statistical analysis with major amount of input data is required.

2.2. Vapor recompression case

Based on the vapor recompression principle presented by Cousins et al. (2011) and Øi et al. (2014), the base case configuration was modified with a recompressed vapor stream. See Fig. 5. According to the authors, this modification reduces the thermal energy requirement of the reboiler in the stripper. In the flow sheet, a valve (Valve 2) was installed in the Stripper Bottoms stream to create a pressure drop from 120 kPa to 100 kPa. In a flash tank separator (Flash Tank 2), the recovered vapor is cooled in a LNG exchanger and then recompressed to 120 °C and 120 kPa, respectively. The LNG (Liquefied Natural Gas) exchanger model solves

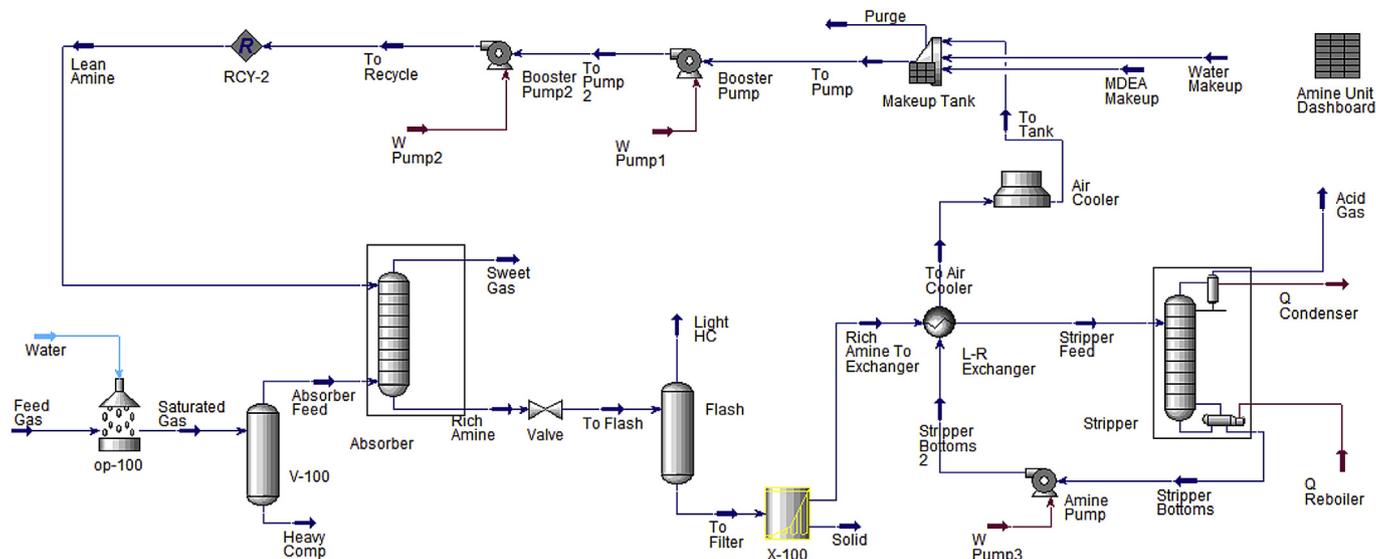


Fig. 4. Sweetening process with MDEA in Aspen Hysys v8.8 (Base Case).

Table 1
Input data for Aspen Hysys v8.8 base case simulation.

Parameter	Value
Feed Gas Temperature	35 °C
Feed Gas Pressure	6178 kPa
Feed Gas Flow	250 Mm ³ /d
CO ₂ in Feed Gas	4 mol.%
CH ₄ in Feed Gas	93 mol.%
Lean Amine Temperature	42 °C
Lean Amine Pressure	9610 kPa
Lean Amine Rate	2100 kmol/h
MDEA in Lean Amine	38 wt%
Number of stages in Absorber Tower	24
Flash Pressure	441 kPa
Number of stages in Stripper Tower	20
Stripper Feed Temperature	90 °C
Top of the Stripper Temperature	87.78 °C
Reflux Ratio	1.1

employing the Membrane Unit Extension v3.0a (Martin, 2003). To simulate the membrane operation, the following equation for each of the components was solved:

$$F_x = Per_x \cdot A \cdot N \left[\frac{P_{x1} - P_{x2}}{\ln \left(\frac{P_{x1}}{P_{x2}} \right)} \right] \quad (1)$$

Where:

- F_x : Permeate molar flow for component x (kmole/h)
- Per_x : Permeability for component x (kmole/(1000 kPa h.m²))
- A : Area per unit (m²)
- N : Total number of units
- P_{x1} : Partial pressure of component x in Input (kPa)
- P_{x2} : Partial pressure of component x in Output (kPa)

For the simulation of a two-stage membrane system (Fig. 6), the values of components permeation presented in Table 4 were used. In addition, the areas of two polymeric membrane modules were considered to be 2000 and 1000 m² respectively (Membrane 1 and Membrane 2). With the areas and the configuration selected, the process is able to produce a Sweet Natural Gas (Sweet Gas) with a 0.02 mol.% of CO₂, under pipeline specification.

As liquids components are completely harmful for a membrane,

heat and material balances for multi-stream heat exchangers and heat exchanger networks, obtained values are shown in Table 3. Finally, the recompressed vapor (Vapor Feed) is fed in the tray number 5 of the Stripper tower.

2.3. Membrane case

The membrane module was simulated in Aspen Hysys v8.8

Table 2
Operating data, simulation obtained data and relative error percentage.

Parameters	Operating Data	Simulation Data	R. E. (%) ^a
Sweet gas Temperature (°C)	40	42	4.95
Sweet gas Pressure (kPa)	6000	5884	1.93
Sweet gas Mole Flow (m ³ /d)	240,000	239,308	0.29
Sweet gas CO ₂ Mole Percentage	0.35	0.32	10.35
Rich amine Pressure (kPa)	6000	6080	1.32
Rich amine Temperature (°C)	110	106.2	3.62
Acid gas Pressure (kPa)	112	100	12.24
Acid gas Mole Flow (m ³ /d)	9755	9325	4.60
Rich Amine to Exchanger Temperature (°C)	50	47	7.33
Stripper Bottoms Temperature (°C)	110	106	3.66
Stripper Bottoms Pressure (kPa)	132	120	10.00
Lean Amine Temperature (°C)	48	41	17.18
Lean Amine Pressure (kPa)	9800	9611	1.97

^a Relative Error (R.E.) (%) = Absolute ((Operating data-Simulation data)/Operating data)×100.

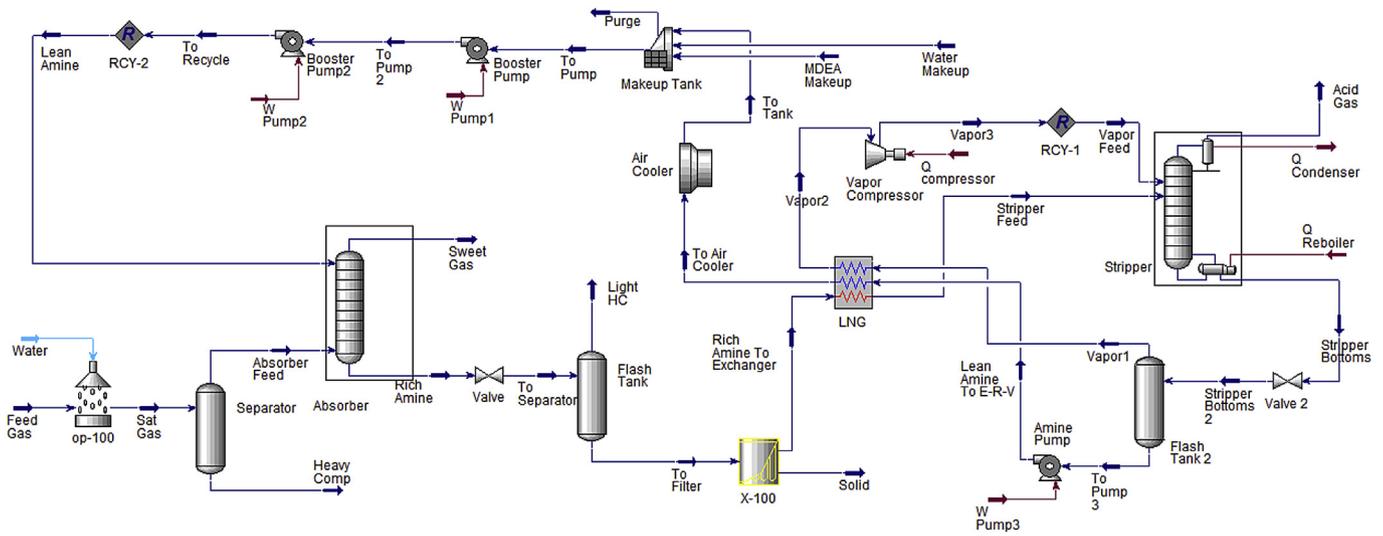


Fig. 5. Sweetening process with MDEA in Aspen Hysys v8.8 (Vapor recompression case).

Table 3
LNG heat exchanger stream temperatures.

Stream of the Vapor case	Temperature (°C)
Rich amine to exchanger	46.99
Stripper Feed	90
Lean amine to E-R-V	104.7
To Air Cooler	59.21
Vapor 1	104.7
Vapor 2	102.1

Table 4
Permeation values (Scholes et al., 2012).

	CO ₂	CH ₄	H ₂ O
GPU (Gas Permeance Unit) Kmoles/(1000 kPa h.m ²)	100	5	22
	0.1104	0.0055	0.0025

inlet streams are introduced to a liquid-vapor separator (Separator 1 and Separator 2). In addition, the pressure drop through the membrane modules are high and thus powerful compressors are needed in order to recycle the gas (Compressor and Compressor 2). From the 2000 m² membrane module, the pressure of the stream with high CO₂ content (To compressor) is 135 kPa. Then, the pressure of the stream is incremented to 4160 kPa in a compressor with adiabatic efficiency of 75%.

2.4. Energy study and costs estimation

Once the three simulation cases converged, the Activated Energy Analysis (AEA) of Aspen Hysys v8.8 was used to estimate the greenhouse gases emissions (CO₂ + CH₄) from each process studied. Energy and carbon emission reduction is of vital importance in the process industry. AEA generates extensive revamp scenarios that can be implemented to reduce fresh utility dependence and it shows relevant details to the optimization including required capital cost, annual reduction in utility cost, and payback period for investment (Zhang and Hird, 2015).

As regards energy requirements, the follow expressions were

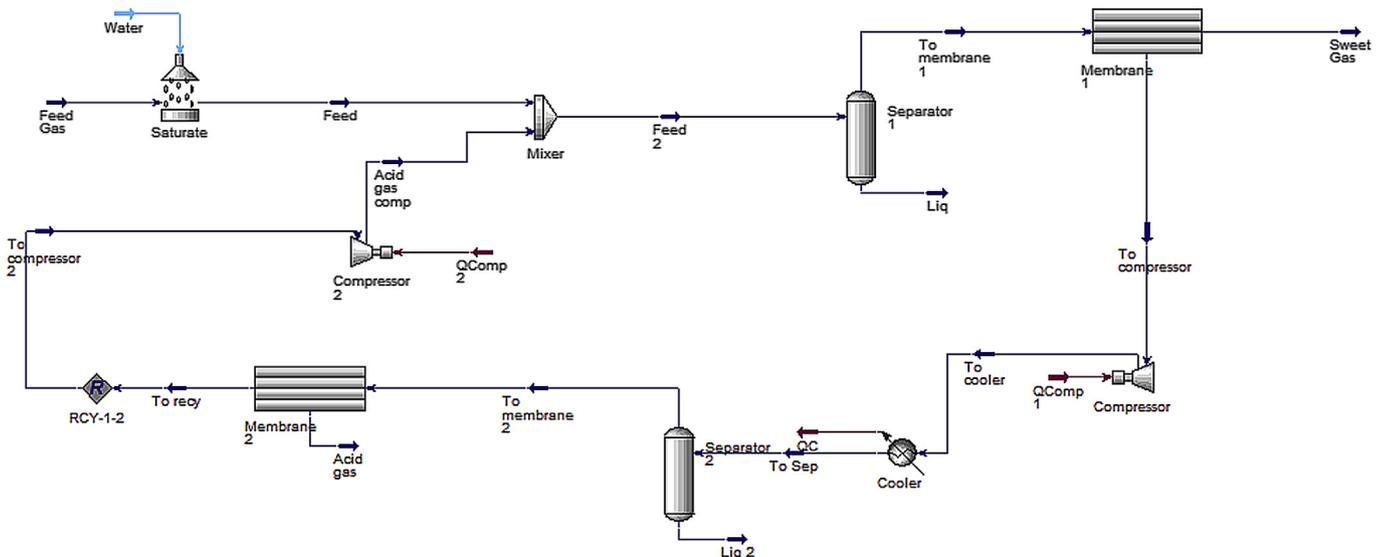


Fig. 6. Two-stage membrane system in Aspen Hysys v8.8 (Membrane case).

used to quantify the energy consumption (Q_{Total}) and the Specific total heat (Q_s) of each case:

$$Q_{Total} = Q_{Reboiler} + Q_{Cooling\ units} + Q_{Compressors} + Q_{Pumps} \quad (2)$$

$$Q_s = \frac{Q_{Total}}{Removed\ CO_2} \quad (3)$$

On the other hand, Activated Process Economic Analysis (APEA) is used to estimate capital costs, operating costs, utilities costs and equipment costs of each process. APEA loads data from Aspen Hysys and generates a default mapping for each unit operation whose cost can be predicted. Particularly, the capital cost of the membrane module was calculated using the approximation purposed by [Ahmad et al. \(2015\)](#), who estimated a cost of 54 USD/m² for a regular polymeric membrane.

3. Results and discussion

3.1. Energy and power analysis

Once the three simulation cases were developed, the listed values in [Table 5](#) are used to compare the energy requirements of each process.

As expected, heating duty is considerably lower with the vapor recompression in comparison to the base case. For the conditions under study, the energy saving is about 20% respect to the base case heating requirement. In the case of cooling units, the cooling duty of the air cooler and the stripper condenser were considered for the absorption processes while the cooling unit after the first compressor was considered for the membrane system.

In the membranes unit, membrane modules origin a pressure drop about 6000 kPa and 3600 kPa, respectively. In this system, pump equipment is not employed as all the streams are gases. As a consequence, the compressor power needed for recycling the permeated gases is significantly high. On the other hand, the pump power required in the two first cases is quite similar.

In [Fig. 7](#), the Specific total heat calculated according to Equation (3) is graphed for the three cases. Considering absolute values of cooling duties, the required energy to separate a CO₂ mass unit is reduced with the vapor modification but interestingly reduced when the membrane technology for the same inlet gas is considered (12% and 76%). Consequently, operating costs are much lower for the membrane system increasing process benefits.

3.2. Emissions analysis

As it can be appreciated in [Table 6](#), the membrane process is much more attractive regarding the environmental issues. As cited authors indicated, the impact on the atmosphere of the membrane process is mitigated in respect to the alkanolamine processes, where combustion is necessary for the solvent regeneration. In addition to this, it can be observed that the water make-up of the membrane system is null. Contrarily, the two-stage membrane

Table 5
Energy and power requirements in the three cases.

	Unit	Base case	Vapor Recompression	Membranes
$Q_{Reboiler}$	MJ/h	5216	4149	–
$Q_{Cooling\ units}$	MJ/h	–5640	–4588	–1327
$W_{Compressors}$	kW	–	5.4	414.6
$Q_{Compressors}$	MJ/h	–	19.4	1492.1
W_{Pumps}	kW	208.5	192.4	–
Q_{Pumps}	MJ/h	691	692	–

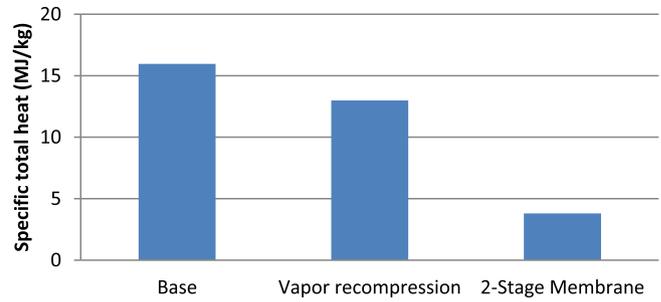


Fig. 7. Specific total heat required in the three cases.

Table 6
Greenhouse gases (GHG) emission, water make-up and CH₄ wastes in the three cases.

	Unit	Base case	Vapor recompression	Membranes
GHG emission	kg/h	380.21	304.87	74.15
Water make-up	l/d	855	898	0
CH ₄ wasted ^a	% m	0.73	0.73	4.8

^a Respect to the CH₄ in the inlet stream.

usage leads to a still considerable CH₄ waste that affects the sustainability of this treating facility.

3.3. Costs estimations

In [Fig. 8](#), capital cost estimation for the processes under study are compared. Evidently, the two-stage membrane technology is still higher in respect to the absorption processes. It was found that the investment cost for a treating facility of this nature is (12%) higher than a typical absorption process. In the same sense, the membrane process requires a utility cost higher because of the great demand of compression power. However, the absence of a reboiler in the flow sheet can compensate this great demand in terms of energy and investment amortization.

Additionally, equipment costs for the three cases are detailed. [Table 7](#) shows equipment costs related to the base case, [Table 8](#) exposes those related to the vapor recompression case and [Table 9](#) the corresponding to the membrane case.

Finally, utilities costs and operating costs are reported in [Table 10](#). As it can be expected, the operating conditions of the second case increment the utility cost. The increment is attributed to the electricity consumed by the additional compressor. However, the 2-stage membrane utility cost is a 26% lower than the second case as the only consumed utility is the electricity. Other utilities such as vapor, air or hot oil are not employed in the membrane case.

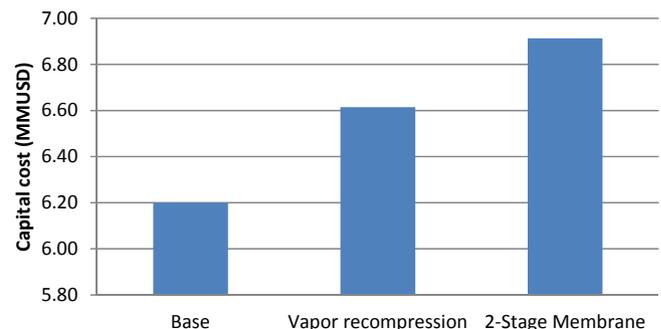


Fig. 8. Capital costs estimation for the three cases.

Table 7

Equipment costs related to the base case.

Name	Equipment Cost (USD)
Flash	25,900
L-R Exchanger	126,600
Booster Pump2	67,000
Air Cooler	83,800
Amine Pump	6700
Absorber	90,600
V-100	25,600
Booster Pump	69,200
Stripper	165,400

Table 8

Equipment costs related to the vapor recompression case.

Name	Equipment Cost (USD)
Absorber	90,600
Air Cooler	79,900
Booster Pump	69,300
Vapor Compressor	450,500
LNG	253,200
Booster Pump 2	67,000
Amine Pump	6700
Stripper	163,700
Flash Tank	25,900
Separator	25,600
Flash Tank 2	30,500

Table 9

Equipment costs related to the membrane case.

Name	Equipment Cost (USD)
Compressor	1,284,200
Separator 1	25,600
Compressor 2	617,100
Separator 2	21,200
Cooler	9100
Membrane 1	108,000
Membrane 2	54,000

Table 10

Operating and utilities costs for the three cases.

	Unit	Base case	Vapor recompression	Membranes
Total Utilities Cost	USD/year	408,766	445,056	330,726
Total Operating Cost	USD/year	1,750,790	2,077,770	2,040,820

In relation to operating costs, it is observed that the cost of the raw material (natural gas) is the same in the three cases. In the case of absorption processes, the amine costs remain equal as the same molar flux is used in both cases. Hence, the increment of the operating cost of the vapor recompression is due to the high utility costs. Respect to the membrane case, the operating cost is greater than the base case because of the methane losses observed.

3.4. Energy efficiency impact

Primarily, the high demand of thermal energy of the alkanolamine technology is improved with a simple compressor inclusion, which implies a remarkable modification for its energy consumes improvement.

In terms of environmental negative impacts, the performance of the membrane module has demonstrated that the technology can be considered the cleanest. In addition to lower greenhouse gases emissions, an extra liquid stream (amine) to separate the acid gas is

not required. Consequently, corrosive solvents leaks are guaranteed to be avoided. Moreover, the absence of the reboiler unit makes the membrane technology attractive for the removal of CO₂. However, this technology still needs an improvement in regard to the costs for its complete implementation. In time, high investment costs may be compensated with high energy saving of the reboiler.

A better comparison can be reached if the values of the different variables were referred to the same base of quantification. Considering that all evaluated factors directly or indirectly contribute to greenhouse gases activity, it is feasible to transform them in terms of CO₂ equivalent (CO₂-e) (Zhang et al., 2015). From those quantifications, it can be possible to estimate an economic income or profit for each scenario under study.

Moreover, the concept of exergy, which means the ability of the energy to perform mechanical work, may be suitable to distinguish the different levels of temperature in the processes. From exergy balances the power and energy comparison can be improved (Banat et al., 2014).

4. Conclusions

This study points out the great potential of improving the energy efficiency of the Natural Gas Sweetening process. However, the possibilities of modifying the validated flow diagram (base case) or implementing new technologies may be not only an expensive issue but also a slow process. Thus, energy efficiency accuracy in gas industries is still quite far of reaching its complete development.

The membrane system demonstrated to be an important option to sweet the gas as it was possible to achieve all the specifications (2 mol.% in the sweet gas). In addition, the membrane technology reduces the energy requirements and GHG emissions respect to both absorption processes. However, the CH₄ wastes are still high for the membrane considered and that implies the technology not to be adequate for the gas considered. Furthermore, the calculated capital cost of a two-stage membrane system is estimated to be 12% higher than the capital of a conventional alkanolamine process and 5% higher than the vapor recompression process.

In order to perform an extended comparison, further analyses can be made from the obtained results in this work. If a period of production and profits concerning the CO₂ emissions global agreements were taken into account, a complete cash flow can be conducted.

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