

Novel acid gas removal process based on self-heat recuperation technology



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ABSTRACT

Chemical absorption is the most common technology used in the acid gas removal unit (AGRU) for treating natural gas. On the other hand, the regenerator requiring large amounts of energy needed for the latent heat of a phase change makes this an energy intensive process. In this study, several distillation columns with a modified heat circulation module based on self-heat recuperation technology were proposed to enhance the energy efficiency of the AGRU. This innovative self-heat recuperation technology circulates the latent and sensible heat in the thermal process. All simulations were conducted using ASPEN HYSYS V8.6, while KG-TOWER[®] software was employed to size all the columns. The results showed that the proposed modified configuration can save up 62.5% and 45.9% in terms of the reboiler duty and operating cost, respectively, compared to a conventional AGRU. This brought a saving of 38.0% in terms of the total annual cost. The results also indicated that the carbon emissions could be saved up to 45.4%. The proposed process can be employed to both close-boiling mixtures and wide-boiling mixtures. In addition, a sensitive analysis of the utility costs on the performance of the suggested AGRU configuration were investigated. The retrofit an existing acid gas removal process was performed to enhance both the energy efficiency and capacity.

1. Introduction

Natural gas is extracted from gas wells as a mixture of hydrocarbons and other impurities including acid gases, water, and mercury (Kidnay et al., 2011). Mercaptans (RSH) can be present when the H₂S concentrations are well above the ppmv level. The required composition constraints placed on sale gas are controlled mainly by market specifications, and typical natural gas pipeline limits for CO₂ and H₂S are 2% and 4 ppm, respectively (Mokhatab et al., 2012). In addition, mercaptans and other organic sulfur species that contribute to sulfur emissions must be removed (Mokhatab et al., 2014). A number of separation processes such as absorption, distillation, membrane, and adsorption are typically needed to meet these specifications. The acid gas removal unit (AGRU), which is used to remove acid gases, is one of the key separation processes implemented for the pretreatment of natural gas (Cho et al., 2015).

Chemical absorption, which is based on the reversible exothermic reaction of a suitable solvent with the gas stream, is the most common technology used in AGRU (Niu and Rangaiah, 2014). On the other hand, a large amount of energy used for releasing the acid gas in the stripper or regenerator makes AGRU an energy intensive process. To improve the AGRU, the development of different solvents (Shi et al., 2014) can be considered. Mixed amine solvents containing

methyldiethanolamine (MDEA) and MEA or DEA, in most cases for the removal of acid gases have received increasing attention (Sohbi et al., 2007). Furthermore, process modification (Moulic et al., 2014; Cho et al., 2015) and process intensification (Jassim et al., 2007) can be attractive options to enhance the performance of AGRU.

The heating and cooling functions are generally integrated based on the heat exchange between the feed and product streams to reduce the energy consumption in distillation (Kansha et al., 2010). Many types of heat pump (HP) systems allowing the heat of condensation released at the condenser to be used for evaporation in the reboiler were proposed to maximize the heat recovery (Bruinsma and Spoelstra, 2010; Kiss et al., 2012). This heat pump approach is an economical way to improve the energy efficiency of distillation column when temperature difference between top and bottom of the column is small and large heat is released from the condenser.

To enhance the energy recovery efficiently and thoroughly, self-heat recuperation technology (SHRT) facilitating the recirculation of both latent heat and sensible heat, were proposed (Kansha et al., 2009; Long and Lee, 2013). In this technology, the heat of the process stream was recycled perfectly by recovering the cooling load using a compressor and exchanging the heat with the bottom liquid and feed streams, resulting no need for heat addition or only a small energy requirement. Several authors studied some innovative processes to investigate the

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industrial applications of this technology. Kansha et al. (2011) demonstrated that the energy requirement of the proposed cryogenic air separation process with SHRT decreased by more than 36% compared with the conventional cryogenic air separation process. Furthermore, crude oil distillation was also investigated to evaluate the possibility of improved energy reduction (Kansha et al., 2012). In addition, Long and Lee (2013) reported that the total annual cost (TAC) can be reduced by up to 44% when employing this SHRT in a deisobutanizer column. Recently, the use of this technology in the methanol synthesis process resulted in a substantial increase in energy efficiency (Kansha et al., 2014).

Several distillation columns with the modified heat circulation module based on SHRT were proposed to enhance the energy efficiency in this work. The proposed configurations can be applied to both close-boiling and wide-boiling mixtures. This innovative SHRT that circulates latent and sensible heat was then used to enhance the energy efficiency of the AGRU. ASPEN HYSYS V8.6 was used for conducting all simulations, while KG-TOWER® software was employed for sizing all columns. The effects of the utility on the operating cost saving of the proposed integrated AGRU were investigated. Furthermore, the carbon dioxide (CO₂) emissions was calculated to evaluate the proposed sequence and compared it with the conventional sequence. The use of this sequence to retrofit the acid gas removal process to save energy while increasing the capacity was also considered.

2. Proposed sequences

2.1. Heat pump

In a distillation column, the latent heat of condensation of the overhead is available at the condensers, which can be recovered to provide a partial/total heating duty at the reboiler or to generate electric power (Chew et al., 2014; Long et al., 2015). A significant amount of energy can be saved by upgrading the low temperature waste heat to high temperature using a HP. Several HP concepts, such as vapor compression (VC) heat pump, mechanical vapor recompression (MVR) heat pump, and bottom flashing heat pump (Fig. 1), have been considered for retrofit to reduce the energy requirement of distillation columns. Among them, the MVR has been employed widely in the separation of mixtures with close boiling points. The use of mechanical compression as a HP is economical, mainly in separating low relative volatility mixtures (De Rijcke, 2007). The advantage over a VC is that the condenser in a MVR is smaller and the temperature lift is approximately 10 °C lower because heat is exchanged only once. This results in a higher thermodynamic efficiency (Bruinsma and Spoelstra, 2010).

Fig. 2 shows the scheme of a MVR system, which lifts the temperature of the top vapor to use this as the heat source for the reboiler. The temperature lift can be calculated as follows:

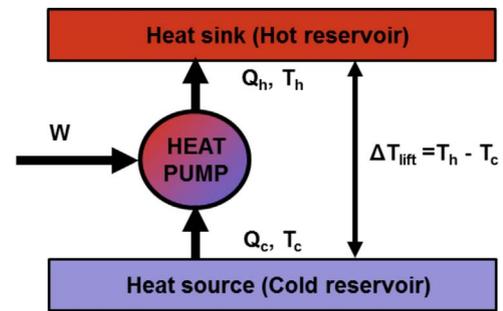


Fig. 2. Scheme of a heat pump.

$$\Delta T_{lift} = T_h - T_c = \Delta T_{column} + \Delta T_{HEX} \quad (1)$$

where ΔT_{column} is the temperature difference between the top and bottom of the column; and ΔT_{HEX} is the optimized temperature difference over the heat exchanger (typically 10 °C).

From the first law of thermodynamics, the amount of heat delivered to the hot reservoir (Q_h) at a higher temperature (T_h) is related to the amount of heat extracted (Q_c) from the cold reservoir at a low temperature (T_c) and the external work by the following equation (Bruinsma and Spoelstra, 2010):

$$Q_h = Q_c + W \quad (2)$$

The coefficient of performance (COP), which is the ratio of the heat rejected at high temperatures to the work input, is used to measure the HP performance (Bruinsma and Spoelstra, 2010),

$$COP = \frac{Q_h}{W} \quad (3)$$

The upper theoretical value of the COP obtainable in a HP is the COP_c related to the Carnot cycle:

$$COP_c = \frac{T_h}{T_h - T_c} \quad (4)$$

The HP is strongly recommended if the Q/W ratio is more than 10. More evaluations are needed if it is between 5 and 10. When the ratio is lower than 5, a HP should not be considered for improving the energy efficiency (Plešu et al., 2014).

2.2. Self-heat recuperation technology

In HP system, only latent heat is utilized while the sensible heat is neglected. Thus, the SHRT facilitating the recirculation of both the latent and sensible heat in a process using compressors and self-heat exchangers was proposed (Kansha et al., 2009; Matsuda et al., 2011) (Fig. 3a). Recently, Long and Lee (2013) suggested a modified SHRT

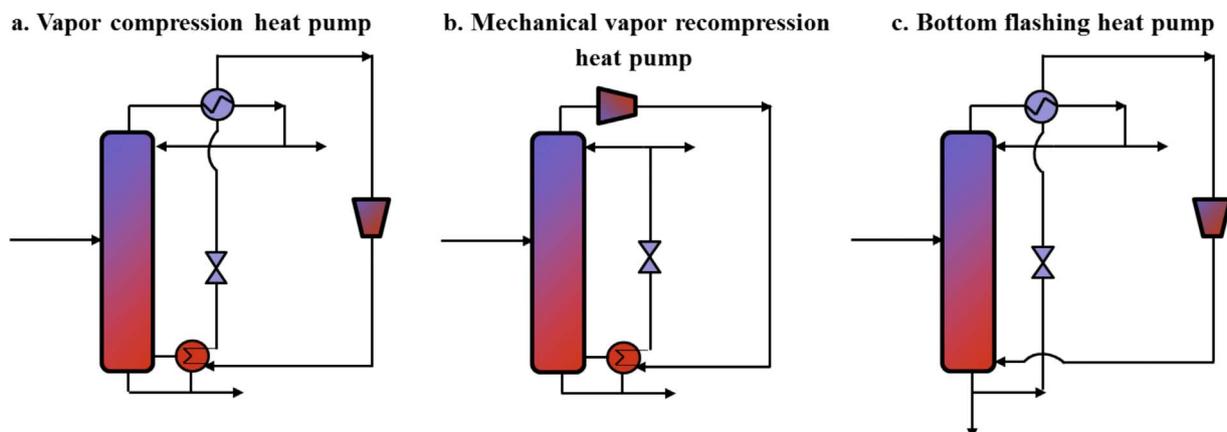


Fig. 1. Schematic diagram of the heat pump systems: (a) vapor compression; (b) mechanical vapor recompression; and (c) bottom flashing.

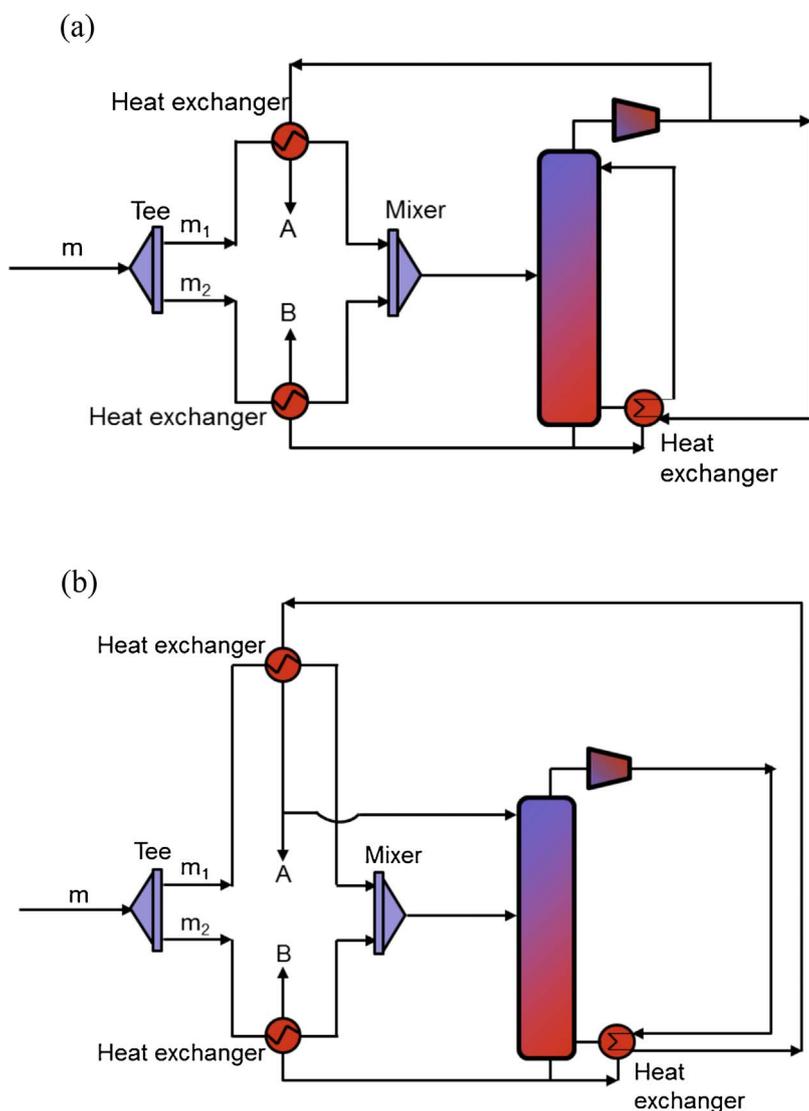


Fig. 3. Simplified flow sheet illustrating a distillation column with the (a) conventional and (b) modified heat circulation module based on SHRT.

(Fig. 3b), which can maximize heat recovery by dividing feed stream into two parallel streams. Normally, this technology can be used directly, where the heat is transferred directly to the reboiler after increasing the temperature.

On the other hand, the high capital expenditure required for compressors make SHRT industrially viable only for high-capacity end-of-train (practically binary) separations of substances with similar boiling points, which require minimal compressor/compression cost (Long and Lee, 2013). When SHRT is employed, engineers should check whether the reactions that take place as the vapor pressure and temperature are increased or not (Long and Lee, 2014). Engineers also should be aware of the decomposition or polymerization of components, which are sensitive to temperature when the temperature is increased using a HP. Furthermore, with this system, the fouling tendency is not well accepted.

This paper proposes several sequences to help solve these problems. In particular, for wide-boiling mixtures, the use of a side SHRT with a side reboiler (Fig. 4a) or on the side (Fig. 4b) can overcome the problems related to the high compressor cost because of the large temperature difference between the top and bottom of the column. In addition, when the fouling tendency is not well accepted, the sequence can be modified by transferring the heat from the top vapor stream to generate steam, which is then increased the pressure and exchanged the heat in the compressor and reboiler, respectively (Fig. 5). This configuration can be considered as an indirect sequence, while the

configuration in Fig. 3b refers to a direct sequence. The generated steam can be supplied not only to that column but also to other consumers. Depending on the specific example, a direct or indirect configuration can be used.

3. Case study

3.1. Base case

The AGRU consists of an absorption column, where the gas feed is sweetened by contacting the lean solvent in a counter-current, and a regeneration column, where acidic gases are rejected at the top of the column and the lean regenerated solvent is recycled to the process (Langé and Pellegrini, 2016;) (Fig. 6). The sour feed gas is fed to an absorber consisting of 18 trays. Most new amine systems use MDEA or a proprietary MDEA-based blend because the energy requirements for regeneration are lower than those for the other amines and it can remove mercaptan slightly (Kidnay et al., 2011). In this process, MDEA at a strength of 26 mol% in water is used as the absorbing medium for the removal of H_2S and some CO_2 from natural gas. The exothermic absorption leads to a high temperature of sweet gas (product) at the top of absorption column. The rich amine is flashed from the absorber pressure of 40 bar–2.5 bar to release most of the hydrocarbon gas absorbed in the separator before entering the heat exchanger. This reduces the flow rate of acid gases to the regenerator and reduces its reboiler and

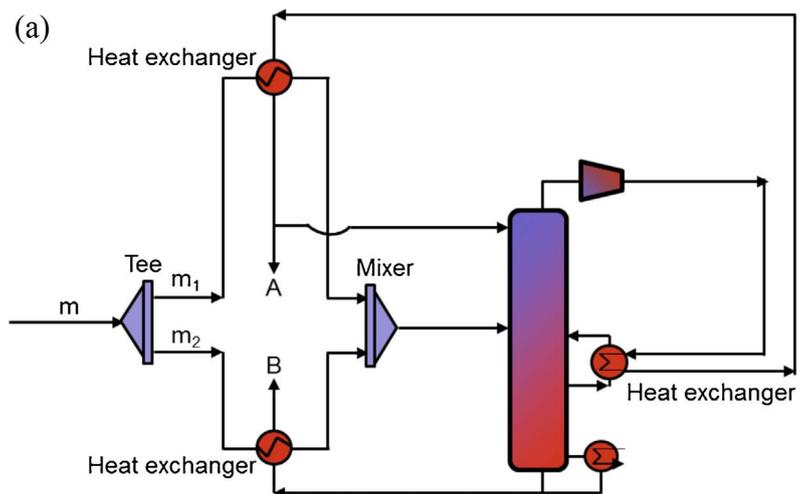


Fig. 4. Simplified flow sheet illustrating a distillation column with the modified heat circulation module based on SHRT (a) with a side reboiler and (b) on the side.

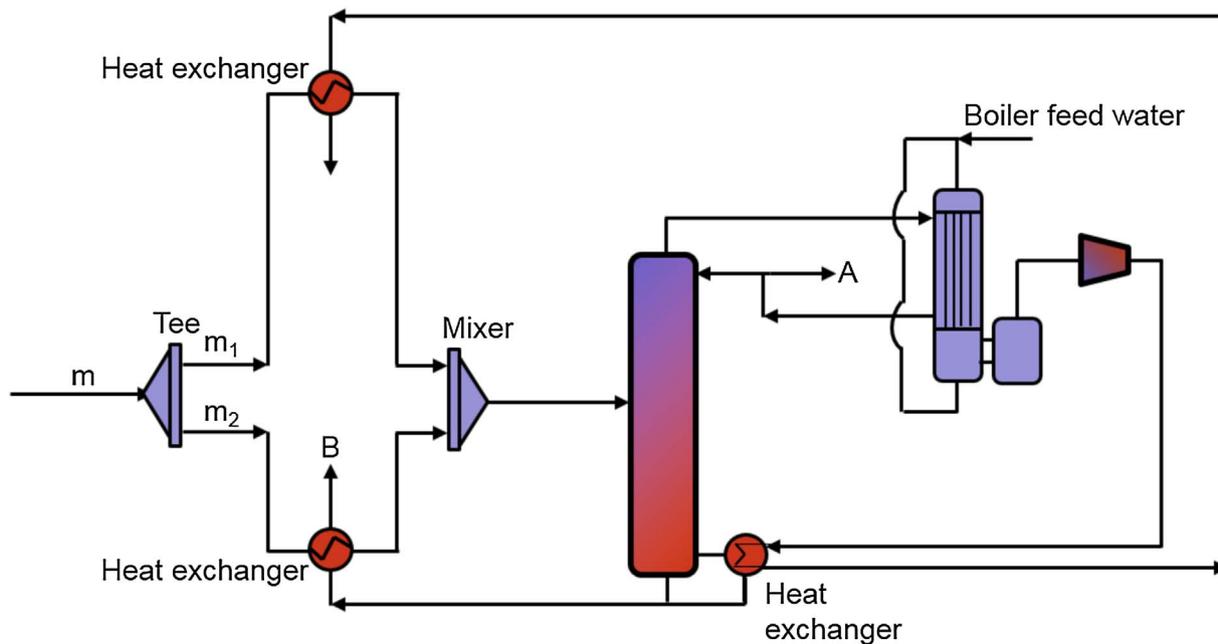
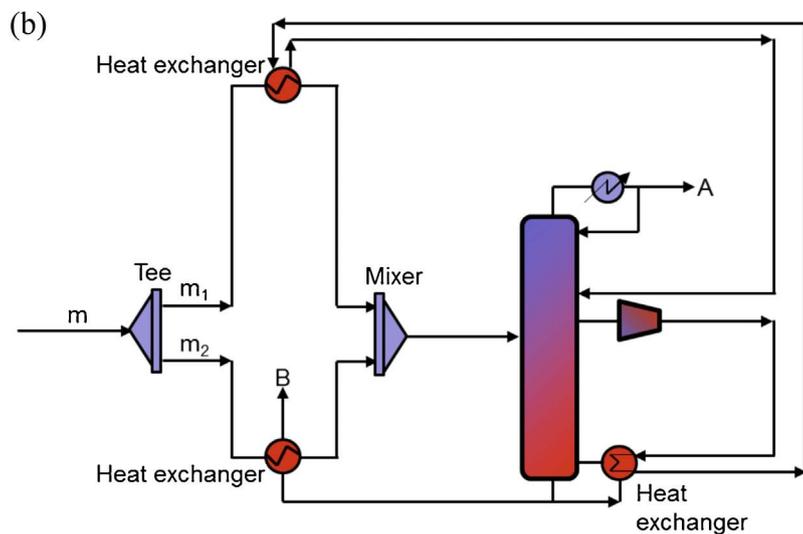


Fig. 5. Simplified flow sheet illustrating a distillation column with the modified heat circulation module based on SHRT with indirect way.

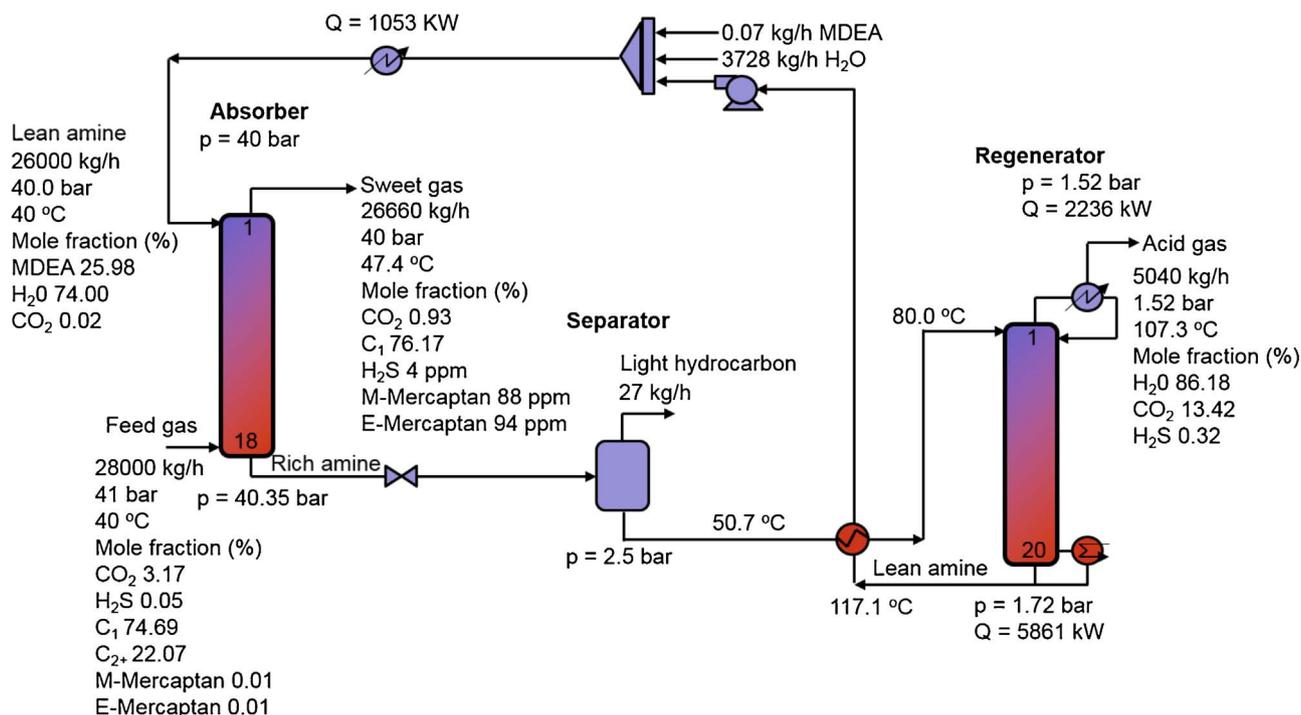


Fig. 6. Simplified flow sheet illustrating the conventional AGRU process.

condenser duties (Patil et al., 2006).

The regenerator, possessing 20 theoretical stages, was designed and operated at 1.5 bar. The regeneration of a MDEA solution is endothermic and favored by a low pressure. The chemical reactions that takes place in a regenerator are the same but opposite to those in the absorption column. The acid gas at the top of regenerator is cooled and the recovered MDEA is refluxed back to the regenerator. The regenerator energy is recovered through lean/rich exchanger. Lean amine is finally cooled and pressurized using an air cooler and amine feed pumps, respectively. The simulations were conducted using ASPEN HYSYS V8.6. The Acid Gas property package using the industry-leading Aspen Properties rate-based chemical absorption calculations and molecular thermodynamic models to automatically generate the reactions and chemistry for the simulation basis was used (Aspen Technology, 2013). KG-TOWER® software was employed to size all columns.

Table 1 lists the operating conditions, hydraulics, and energy performance of the columns in the AGRU. The base case simulation indicated that the energy requirement of the regenerator was 5861 kW. In the regenerator, heat is supplied at a reboiler. Some of the heat supplied at the reboiler in the regenerator is released in the overhead condenser. The conventional energy savings of this regenerator are achieved fundamentally by utilizing the heat of the bottom stream to preheat the feed stream. In this study, a HP and modified SHRT was employed to achieve further energy savings in this regenerator.

Table 1
Column hydraulics, operating data and energy performance of conventional AGRU process.

	Absorber	Regenerator
Column pressure (bar)	40	1.52
Number of trays	18	20
Tray type	Sieve	Sieve
Column diameter (m)	0.9	1.2
Max flooding (%)	83.4	82.2
Energy requirement of condenser (kW)	–	2236
Energy requirement of reboiler (kW)	–	5861

3.2. AGRU improvement

3.2.1. Heat pump

The amount of heat necessary for regenerating the solvent is mainly due to the large heat of vaporization of water, which is present in the largest amounts of the solvent mixture, especially on a molar basis, and to the heat required to break the chemical bonds formed during acidic gases absorption (Langé and Pellegrini, 2016). In this regenerator, heat is supplied to the feed heater and reboiler, and the overhead stream is cooled in a condenser. Most of the heat supplied to the reboiler in the regenerator is discarded in the overhead condenser. An inspection of the column temperature profile and COP value of 7.4 confirmed that this heat can be utilized using a HP system to enhance the energy efficiency of regenerator. According to the scheme in Fig. 7, top vapor stream of the regenerator can be compressed to a pressure of 3.0 bar for increasing its vapor temperature and then condensed in the column's

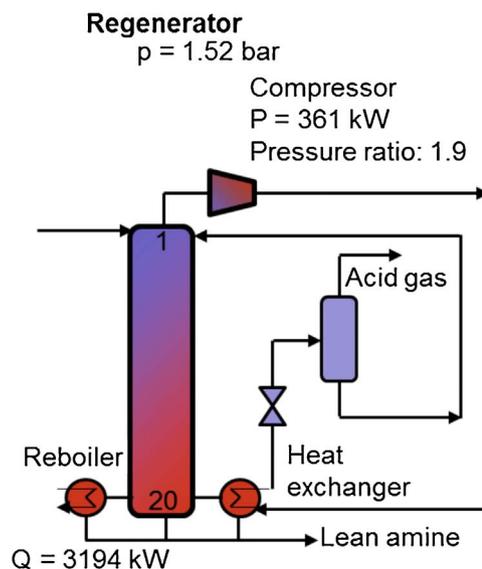


Fig. 7. Simplified flow sheet illustrating the vapor recompression heat pump.

Table 2
Comparison of different structural alternatives.

Structural alternative	Conventional column	Heat pump	Distillation based on SHRT
Energy requirement saving in condenser (%)	–	100.0	100.0
Energy requirement saving in reboiler (%)	–	45.5	62.5
Annual operating cost saving (%)	–	30.1	45.9
TAC saving (%)	–	24.6	38.0
CO ₂ emission saving (%)	–	29.9	45.4

reboiler by transferring the heat to the bottom liquid stream. This allows the heat of the condensing vapor to assist in vaporization at the bottom of the regenerator column. The top outlet stream must be cooled further before being divided into two streams: one that is recycled back to the column as reflux, and the other that is the final top product. Note that to simulate all compressors, the polytropic efficiency was assumed to be 75%.

For an economic evaluation between proposed configurations and conventional AGRU, the investment cost, operating cost and TAC of all the processes were calculated based on the textbook (Biegler et al., 1997) as described in detail in Appendices A and B. The equipment considered in the investment costs included all of the reboilers, condensers, column vessels, tray stacks, heat exchangers and compressor. A low-pressure steam cost of 6.08 \$/GJ, cooling water cost of 0.35 \$/GJ and electricity cost of 16.80 \$/GJ were used for the TOC calculation (Turton et al., 2012). Because the amount of energy transferred from the hot top stream to the cold bottom stream is insufficient, an extra reboiler was installed to manage the remaining heat. Compared to the conventional AGRU, the use of a top vapor recompression HP could improve the operating costs and TAC by 30.1% and 24.6%, respectively, as shown in Table 2.

In addition, the total annual CO₂ emissions were also estimated to assess the environmental impact. For the steam reboilers, Gadalla’s modular method was applied to calculate the CO₂ emissions (Gadalla et al., 2005). As a result, the total annual CO₂ emissions is decreased up to 29.9% as compared to the conventional AGRU.

3.2.2. Integrated regenerator based on SHRT

For further improvement, the integrated generator based on SHRT was considered. Instead of releasing the heat of condensation to environment, it is recuperated by using compressor and several heat exchangers leading to substantial energy savings. When employing SHRT, the feed is further pre-heated, resulting in a higher vapor fraction in the feed and top vapor flow rate. Because the distillate rate is kept constant, the reflux rate increased from 3571 kg/h to 3939 kg/h, resulting in an increase in condenser duty when compared to the conventional distillation. Furthermore, because the liquid of the trays in the stripping section decrease and the bottom product rate is kept constant, the amount of vapor in these trays decreases, which causes a reduction of the boil-up flowrate. This means that more heat can be utilized, while the required heat is reduced.

Top vapor stream is compressed and condensed in the compressor and the heat exchanger playing as a reboiler, respectively. The top vapor stream is then reduced the pressure before being separated into two streams in a separator including one liquid stream recycled back to the column as reflux and one vapor stream. Both vapor stream and bottom lean amine stream are used to transfer the heat to two divided feed streams (Fig. 8). Thus, both the latent heat and the sensible heat can be circulated within the process, leading to a substantial improvement of energy efficiency. The feed split ratio was considered as an important variable to optimize the heat recovery duty. As shown in Fig. 9, the smallest reboiler duty was found at feed split ratio of 0.6. As a result, the proposed modified sequence can save up 62.5% and 45.9% in terms of the reboiler duty and operating cost, respectively, compared to a conventional AGRU. The TAC was decreased to 38.0%, even when three exchangers and one compressor are needed.

Meanwhile, as compared to the HP system, the proposed sequence can save 31.2% and 22.6% in terms of the reboiler duty and operating cost, respectively. Furthermore, instead of using the primary steam, the heat can be recycled and utilized resulting a substantial reduction of CO₂ emissions. In particular, the annual amount of CO₂ emissions could be reduced by up to 45.4%, as shown in Table 2. Note that because the products are kept constant when applying HP and SHRT, there are no change in the lean and rich amine loadings.

The utility costs are different according to the company and country, which could have a major effect on the performance of the suggested configuration. Therefore, a sensitive analysis was conducted

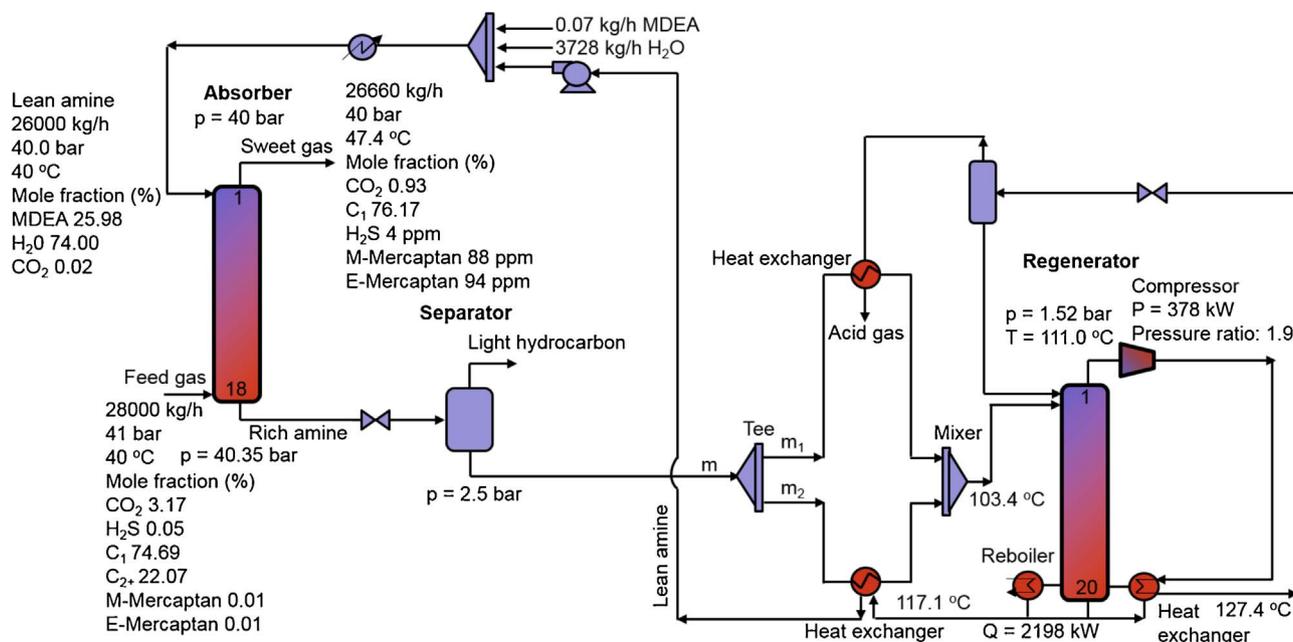


Fig. 8. Simplified flow sheet illustrating the proposed integrated AGRU process.

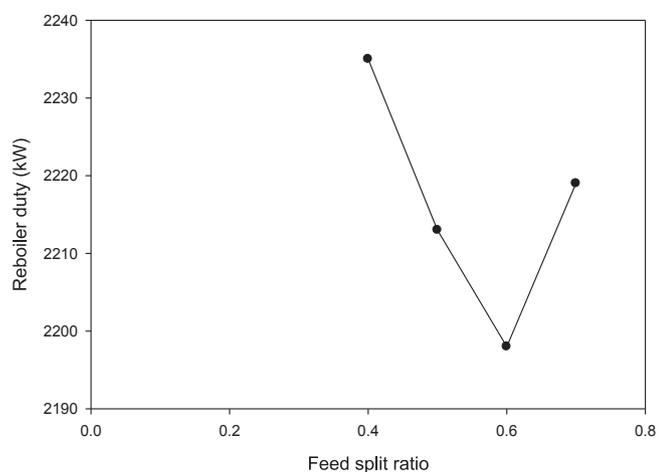


Fig. 9. Effects of the feed split ratio on the reboiler duty.

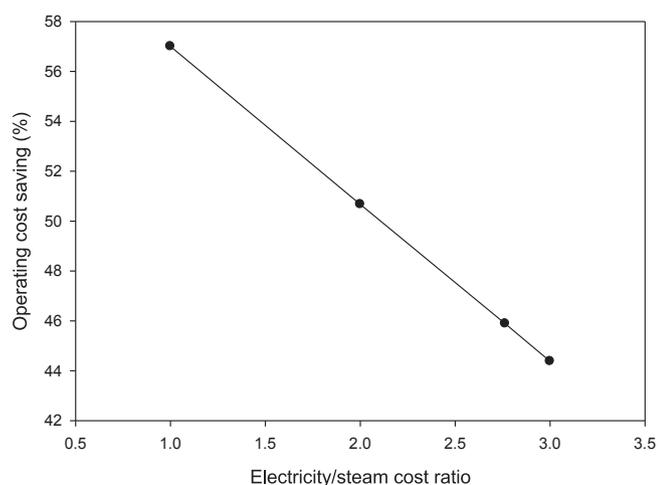


Fig. 10. Effects of the electricity/steam cost ratio on the operating cost saving of the proposed integrated regenerator.

to improve the robustness of the proposed results. In this analysis, the ratio of electricity/steam cost, which play as dominant costs and are different between countries and companies, was explored. As shown in Fig. 10, this sequence achieves greater benefit with the reducing of electricity/steam cost ratio. Remarkably, up to 57.0% can be saved in terms of operating cost at the ratio of 1 when applying this proposed configuration.

The results presented above are in the grass-roots case, where a new plant is considered. This configuration also can be applied to retrofit and debottlenecking projects, where the purposes are to improve the energy efficiency and capacity, respectively. A new compressor with three new heat exchangers is needed to implement the proposed sequence. With the proposed sequence, an 868,788 USD investment is needed, which requires a simple payback period of 20 months. This

Appendix A. Cost correlations

a. Capital cost: Guthrie's modular method was applied (Biegler et al., 1997). In this study, the Chemical Engineering Plant Cost Index of 556.8 (2015) was used for cost updating.

$$\text{Tray stack} = (N - 1) \times \text{tray spacing} \quad (5)$$

$$\text{Total height} = \text{tray stack} + \text{extra feed space} + \text{Disengagement} + \text{skirt height} \quad (6)$$

$$\text{Updated bare module cost (BMC)} = UF \times BC \times (MPF + MF - 1) \quad (7)$$

Table 3
Max flooding of the absorber.

Packing type	Max flooding (%)
Sieve	83.4
TRITON [®]	58–60
SUPERFRAC [®]	56–58
ULTRA_FRAC [®]	34–45

payback period is economically attractive because existing equipment is maximally utilized.

The use of self-heat recuperation technique can alter the vapor and liquid flows within a column section. Particularly, in the stripping section or whole column, the traffic of vapor and liquid can be decreased. A larger partial condenser is needed. Furthermore, the maximum flooding decreases from 82.2% to 72.0% compared to the conventional regenerator with the same diameter. This can result in an up to 10% increase in capacity. For the debottlenecking absorber, the sieve tray can be changed to a high performance tray, such as TRITON[®], SUPERFRAC[®] or ULTRA_FRAC[®], in which the downcomer, active area and inlet area are enhanced. As shown in Table 3, the flooding can be reduced significantly, which brings a chance for raising capacity of the absorber.

4. Conclusions

Several distillation columns with a modified heat circulation module based on SHRT were proposed to enhance the energy efficiency of AGRU. By applying SHRT, both the latent heat and the sensible heat can be circulated within the process, leading to a substantial improvement of energy efficiency. In addition, a novel AGRU based on SHRT was proposed. In particular, the simulation results showed that the proposed modified sequence can save up to 62.5% and 45.9% in terms of the reboiler duty and operating cost, respectively, compared to a conventional AGRU. Meanwhile, as compared to the HP system, the proposed sequence can save 31.2% and 22.6% in terms of the reboiler duty and operating cost, respectively. The low TAC and CO₂ emissions highlight the proposed sequence as an attractive option to implement in industry. The study shows that pre-heating can enhance the energy performance of a HP and decrease its investment cost substantially. Furthermore, this configuration can be also employed to wide-boiling mixtures. Interestingly, the results also show that this system is more advantageous with the reducing of the electricity/steam cost ratio. Due to its simple manner and short modification time, the suggested configurations are also promising and viable in retrofit projects.

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where UF is the update factor:

$$UF = \frac{\text{present cost index}}{\text{base cost index}} \quad (8)$$

$$BC \text{ is the bare cost, for vessels: } BC = BC_0 \times \left(\frac{L}{L_0}\right)^\alpha \times \left(\frac{D}{D_0}\right)^\beta \quad (9)$$

$$\text{for the heat exchanger: } BC = BC_0 \times \left(\frac{Ar}{Ar_0}\right)^\alpha \quad (10)$$

$$\text{Area of the heat exchanger, } Ar = \frac{Q}{U\Delta T} \quad (11)$$

$$\text{for the compressor: } BC = BC_0 \times \left(\frac{S}{S_0}\right)^\alpha \quad (12)$$

where MPF is the material and pressure factor; MF is the module factor (typical value), which is affected by the base cost. D, L and A are the diameter, length and area, respectively. S is the brake horsepower.

$$\text{Updated bare module cost for tray stack (BMC)} = UF \times BC \times MPF \quad (13)$$

$$\text{The material and pressure factor: } MPF = F_m + F_s \quad (14)$$

b. Operating cost (Op):

$$Op = C_{\text{steam}} + C_{\text{CW}} + C_{\text{electricity}} \quad (15)$$

where C_{steam} is the cost of the steam; C_{CW} is the cost of cooling water; and $C_{\text{electricity}}$ is the cost of electricity

c. Total annual cost (TAC) (Smith, 1995)

$$\text{TAC} = \text{capital cost} \times \frac{i(1+i)^n}{(1+i)^n - 1} + Op \quad (16)$$

where i is the fractional interest rate per year and n is the number of years.

$$d. \text{ Cost saving} = \text{Operating cost saving} - \text{modification cost} \quad (17)$$

$$e. \text{ Payback period} = \text{cost of project/saving per year} \quad (18)$$

Appendix B. Estimation of the CO₂ emission (Gadalla et al., 2005)

In the combustion of fuels, air is assumed to be in excess to ensure complete combustion, so that no carbon monoxide is formed. The amount of CO₂ emitted, $[CO_2]_{\text{Emissions}}$ (kg/s), is related to the amount of fuel burnt, Q_{Fuel} (kW), in the heating device, as follows:

$$[CO_2]_{\text{Emissions}} = \left(\frac{Q_{\text{Fuel}}}{NHV}\right) \left(\frac{C\%}{100}\right) \alpha \quad (19)$$

where α (=3.67) is the ratio of the molar masses of CO₂ and C, while NHV, which is equal to 47,141 (kJ/kg), represents the net heating value of natural gas with a carbon content of 75%.

The amount of fuel burnt can be calculated using the following equation:

$$Q_{\text{Fuel}} = \frac{Q_{\text{Proc}}}{\lambda_{\text{Proc}}} (h_{\text{Proc}} - 419) \frac{T_{\text{FTB}} - T_0}{T_{\text{FTB}} - T_{\text{stack}}} \quad (20)$$

where λ_{Proc} (kJ/kg) and h_{Proc} (kJ/kg) are the latent heat and enthalpy of steam delivered to the process, respectively, while T_{FTB} (°C) and T_{stack} (°C) are the flame and stack temperatures, respectively.

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