


Article

Techno-Economic Analysis of a Novel Two-Stage Flashing Process for Acid Gas Removal from Natural Gas

Yiyang Dai ^{1,*} , Yuwei Peng ², Yi Qiu ³ and Huimin Liu ⁴¹ School of Chemical Engineering, Sichuan University, Chengdu 610065, China² College of Chemistry and Chemical Engineering, Southwest Petroleum University, Chengdu 610500, China; yusube@126.com³ China Petroleum Engineering Construction Corporation (Beijing Company), Beijing 100120, China; qiuyi@cpebj.com⁴ BLOCK 9 of Kuwait Energy Company KSCC, Nashwa-AlZireje Street, Al-Yamam Statia Shatt al Arab District, 61014 Basra, Iraq; liu.huimin@kec.com.kw

* Correspondence: daiyy@scu.edu.cn

Received: 12 October 2019; Accepted: 1 November 2019; Published: 5 November 2019



Abstract: Excessive CO₂ content will reduce the natural gas calorific value and increase the energy consumption of the regenerator in natural gas desulfurization and decarbonization. This paper uses Aspen HYSYS to model a novel two-stage flash process of acid gas removal process from natural gas. According to the results from the simulation, as well as running experiences in a natural gas processing plant in the middle east, it can be demonstrated that this new process, which has been used in the field of natural gas desulfurization and decarbonization, can meet the requirement of product specifications. Based on the steady state simulation, Aspen HYSYS sensitivity function is used to evaluate influence of key operating parameters, such as the second flash pressure and temperature, on the energy consumption. Compared to the traditional acid gas removal process and acid gas enrichment process, the new two-stage flash acid gas removal process has less energy consumption (2.2×10^9 kJ·h⁻¹). In addition, two-stage flash acid gas removal process also improves the efficiency of acid gas enrichment, while the overall energy consumption is less than combination process of traditional process and acid gas enrichment process.

Keywords: natural gas sweetening; acid gas flash; two-stage flashing process absorber; regeneration

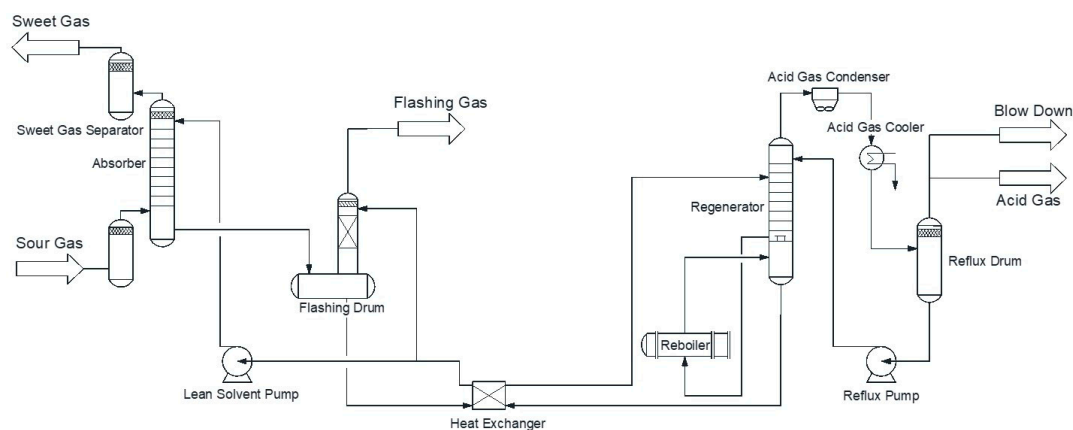
1. Introduction

Due to its low cost, clear-burning, and less CO₂ release per energy unit, natural gas is considered as a clean energy resource compared with conventional fossil fuels including coal, crude oil > petroleum [1,2]. However, around one third of raw natural gases contain CO₂ and sulfur components (H₂S, RSH, RSSH, CS₂, and COS), which need to be removed to meet specifications of sales gas [3]. These acidic impurities have no heating value. In addition, these contaminants tend to form acids that can corrode pipelines and other equipment with the presence of water. Therefore, these acidic impurities have to be removed from the sour gas. Particularly, the maximum allowed level of CO₂ in natural gas transmitted to customers by pipeline is typically less than 3% [4,5]. Product natural gas quality standards for natural gas content also have more stringent restriction: CO₂ needs to be less than 2% (mol), and H₂S needs to be less than 6 mg·m⁻³ in sweet gas. Therefore, it is of critical importance to remove the acid gases in natural gas to a certain extent to meet the requirements of commodity gas quality standards [6].

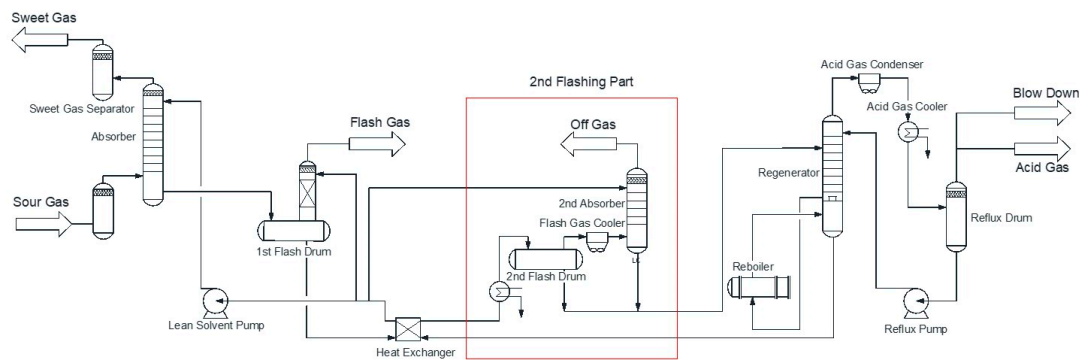
The process that removes the acid gas from natural gases is referred as acid gas removal (AGR) process. There are many different methods to remove acid gas from natural gas. To name just a

few, these methods include absorption, which can be chemical absorption, physical absorption or combined absorption. Membrane separation is another method that has been used for natural gas sweetening [7,8]. Other methods include but are not limited to biochemical method [9], molecular sieving [10], and low-temperature fractionation [11]. Among those technologies listed above, chemical absorption is the most widely used method for acid gas removal. Various amines have been commonly used as chemical solvents, such as monoethanolamine (MEA), diethanolamine (DEA), and methyldiethanolamine (MDEA). Compared with MEA and DEA, MDEA can selectively remove H_2S with the existence of CO_2 . Secondly, MDEA can remove most CO_2 and H_2S from the raw natural gas, which can be beneficial to sulfur recovery. Thirdly, the foaming tendency and corrosiveness of MDEA are low compared to MEA and DEA. Last but not least, the reaction of MDEA with CO_2 is an acid-base reaction with less heat of reaction; thus, less heat is needed during regeneration, so it is ideal for removing CO_2 in large quantities [12–14].

Generally, traditional acid gas removal process includes three main steps, absorption, hydrocarbon flash and regeneration [15] units, which are shown in Figure 1a.



a. Traditional acid gas removal process flowsheet



b. Two-stage flashing process of acid gas removal flowsheet

Figure 1. Traditional and novel acid gas removal flowsheet.

In the process shown above, one absorber, one flashing drum, and one regenerator are normally used in the acid gas removal process. The acid gases absorbed in the liquid absorbent can be partly released in the flashing drum; then, the rich solvent is further sent to the regenerator. Traditional acid gas removal process can effectively remove most of H_2S from acid gas. It should be noticed that if high carbon/sulfur ratio natural gas is involved, the basic absorbents could remove both sulfur compounds and CO_2 , but the sulfur compounds (e.g., H_2S) cannot be easily regenerated, consequently reducing the overall acid gas removal efficiency. Secondly, the CO_2 could diffuse to the regenerator vapor, which could lead to increased cooling water consumption. Thirdly, if the CO_2 content in the natural

gas is high, CO₂ may enter into the sulfur recovery stage and increase the operational cost of the sulfur recovery unit.

Various methods have been used to improve these problems. Zhao et al. [16] studied different concentrations of amine to absorb acid gas, but the energy saving was not very good. Gutierrez et al. [17] used Aspen HYSYS and Aspen plus to simulate the process of natural gas sweetening in northern Argentina and analyzed CO₂ mole fraction in absorber and regenerator; however, they did not say how to effectively reduce process energy consumption. Roy et al. [18] used Aspen HYSYS to simulate the Bakhrabad gas processing plant at Sylhet and compared the simulated results with the plant data. However, they simply simulated the natural gas processing plant without optimizing those parameters. Mohamadirad et al. [19] simulated the effect of using MDEA blended with MEA or DEA to improve the performance of an industrial sweetening process in Iran. They found out that those blends improve CO₂ removal efficiency significantly. However, they also did not optimize the effect of other parameters, such as temperature and pressure. Fouad et al. [20] simulated the use of mixed amines to reduce the energy requirements of the Habshan gas sweetening unit in Abu Dhabi, UAE. They used a blend of MDEA and TEA and found a 3% reduction in the operating costs of the unit while still meeting the specifications of acid gas removal. Like Mohamadirad et al., they did not analyze the effect of other parameters, such as temperature and pressure, on the overall energy consumption.

In this paper, to enrich CO₂ and reduce regenerator energy consumption, a new two-stage flashing process is proposed to remove more acid gas in the high C/S ratio natural gas. Aspen HYSYS V10 was used to simulate the whole two-stage flashing process. Detailed process description is shown in Section 2. The newly proposed process was compared with conventional AGR process, as well as the combination of AGR and acid gas enrichment (AGE) processes. In addition, Aspen HYSYS sensitivity analysis function was used to study the effect of different operation parameters on the overall energy consumption [21,22]. The rest of the paper is structured as follows: Section 2 contains the description of the novel two stage process and the raw material data, Section 3 contains the simulation results of the novel two stage process, Section 4 contains the sensitivity analysis of different AGR process, Section 5 contains the comparison between the traditional acid gas removal process and the novel two stage acid gas removal process.

2. Process Description

The novel two stage process is shown in Figure 1b. Compared with traditional AGR process, this novel process has the 2nd flashing part which contains two flash drums and two absorbers, and it is shown in red box of Figure 1b.

The first absorption column is used to remove most of the acid gas; afterwards, two flashing drums are used instead of one flashing drum. The first flashing drum is used to remove most of the light hydrocarbon, while the second flashing section is used to remove most of the CO₂ and part of the H₂S. The flashing gases from both flashing sections are sent to another two absorbers. After the two flashing drums, the liquid absorbent is sent to the regeneration column to remove the H₂S.

The acid gas enrichment section is the most important section in this process. The rich solvent which exchanges heat with the lean solvent enters the 2nd flash drum to remove most of the acid gas. The 2nd flashing vapor is cooled by air and enters into the 2nd absorber. The 2nd flashing vapor enters into the bottom of the 2nd absorber, and the cyclic MDEA enters into the top of 2nd absorber. Vapor and liquid flow counter-currently to allow the absorption of H₂S from the acid gas.

Following reactions take place in three absorbers [23,24]:



The regeneration of MDEA solvent is endothermic and is favored by low pressure. Chemical reactions that take place in regenerator are the same but to the opposite direction due to high temperature.

Compared to the conventional one stage absorption process, this novel two-stage acid gas removal process has many advantages including: (1) CO₂ can be enriched in this novel acid gas, which can reduce the consumption of cooling water; (2) The sulfur compounds and CO₂ can be separated in a much more effective way that the recovery investment is reduced.

The novel acid gas removal process was simulated by Aspen HYSYS. The simulation flowsheet is shown in Figure 2.

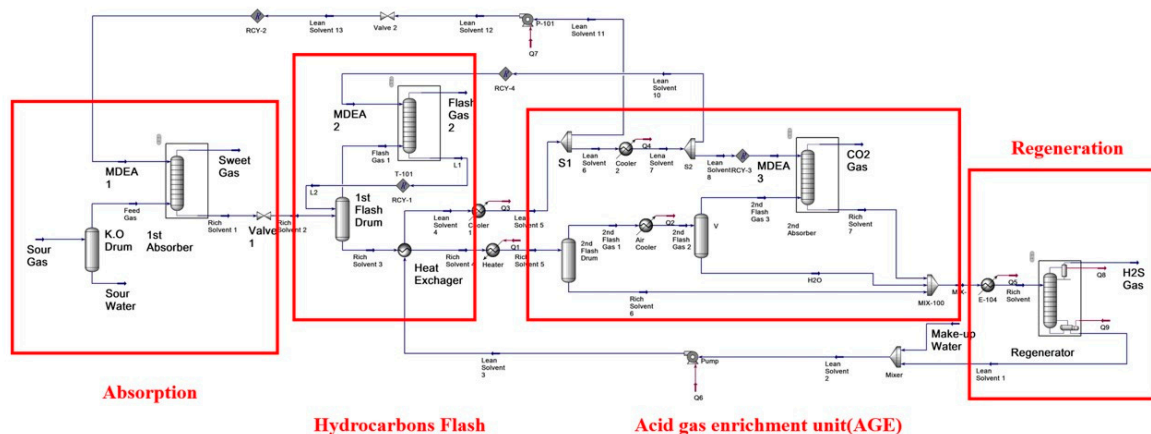


Figure 2. The Aspen HYSYS simulation flowsheet.

The choice of appropriate thermodynamic models and the accuracy of parameters are crucial for the reliability of the design. Acid Gas-Chemical Solvent was used as the property package for acid gas removal process [25,26].

The sour gas composition used in the study was based on the raw material of a middle east plant, which is designed by Saudi Arabian Oil Company. Its composition is shown in Table 1. The carbon to sulfur ratio is 2.3, which justifies the necessity to adopt acid gas enrichment process to remove CO₂ and enrich H₂S. According to the standard, the CO₂ in sweet gas should be lower than 2%, and H₂S should be lower than 6 mg·m⁻³. The mass fraction of MDEA solvent is MDEA: H₂O = 55:45.

Table 1. Summary of operation conditions.

Parameters		Value
Raw natural gas flow rate/kmol·h ⁻¹		3.3 × 10 ⁴
Temperature/K		303.15
Pressure/kPa		8500
Vapor Composition/mol%	CO ₂	7.7
	H ₂ S	4.3
	H ₂ O	0
	Hydrocarbons	78.8
	N ₂	9.2

3. Results

3.1. Absorption

The purpose of the absorption section is to absorb acid gas by using MDEA solvents. This part of simulation contains a separation drum and an absorber. Based on the plant operation conditions, the pressure and temperature of the 1st absorber was set to 8500 kPa and 334.15 K. A total of 22 stages are used in the 1st absorber. The Aspen HYSYS simulation results are shown in Table 2.

Table 2. 1st Absorber material balance results.

Parameters	Sour Gas	MDEA 1	Sweet Gas	Rich Solvent 1
Temperature/K	303.15	329.15	329.38	334.73
Pressure/kPa	8500.00	7685.60	7500.00	8500.00
Total flow rate/kmol·h ⁻¹	33,232.33	180,000.00	29,431.91	183,800.42
Composition/mol%	-	-	-	-
CO ₂	7.7	0	1.94	1.09
H ₂ S	4.3	0	0.0	0.78
H ₂ O	0	84.4	0.26	82.63
MDEA	0	15.6	0.0	15.27
Hydrocarbons	78.8	0	85.59	0.22
N ₂	9.2	0	10.31	0.01

3.2. Hydrocarbons Flash

The hydrocarbons flash section aims to remove most of the light HC and some acid gas from the rich solvent.

In the hydrocarbons flash section, the hydrocarbons flash pressure and temperature were set to 500 kPa and 335.15 K, which was coming from the plant. The Aspen HYSYS simulation results are shown in Table 3. MEDA 2 flow came from the circulating lean solvent, rich solvent 2 flow came from the first absorption section, and rich solvent 3 was sent to acid gas enrichment section

Table 3. Hydrocarbons flashing material balance results.

Parameters	Rich Solvent 2	MDEA 2	Flash Gas 2	Rich Solvent 3
Temperature/K	335.15	303.15	305.55	336.3
Pressure/kPa	500.00	1390.60	480.00	500.00
Total flow rate/kmol·h ⁻¹	183,800.42	733.60	392.94	184,141.08
Composition/mol%	-	-	-	-
CO ₂	1.09	0	4.35	1.08
H ₂ S	0.78	0	0.09	0.78
H ₂ O	82.63	84.4	1.45	82.81
MDEA	15.27	15.6	0.00	15.31
Hydrocarbons	0.22	0	90.22	0.02
N ₂	0.01	0.00	3.89	0.00

3.3. Acid Gas Enrichment Unit (AGE)

The AGE unit concentrates the H₂S by introducing a second amine absorption unit. This unit can not only release big part of the absorbed CO₂, which reduces the regeneration column energy consumption, but also concentrate the H₂S content in the liquid phase, which could consequently increase the feed quality to the Claus plant.

The acid gas enrichment section contains a 2nd flash drum, an air cooler, and a 2nd absorber. This is different from the first two steps, in which the parameters are came from the plant. In this step, as a basic case, the temperatures and pressures in the 2nd flash drum is 403.15 K and 300 kPa, while the pressure and temperature of the 2nd absorber are 300 kPa and 338.45 K. In addition, six stages are used in the 2nd absorber. All these operating parameters are optimized in sensitivity analysis. The simulation results are shown in Table 4. MDEA 3 flow is came from the circulating lean solvent. Rich solvent 5 flow came from hydrocarbon flash section, which was marked as rich solvent 3. Rich solvent flow 6 and 7 are sent into regeneration section.

Table 4. Acid gas enrichment section material balance results.

Parameters	Rich Solvent 5	MDEA3	CO ₂ Gas	Rich Solvent 6	H ₂ O	Rich Solvent 7
Temperature/°C	403.15	303.15	307.15	403.15	333.15	339.33
Pressure/kPa	300.00	1390.64	250.00	300.00	300	300
Total flow rate/kmol·h ⁻¹	184,141.08	13,500.00	1621.08	171,786	9670	14,563
Composition/mol%	-	-	-	-	-	-
CO ₂	1.08	0.00	95.52	0.21	0.18	0.81
H ₂ S	0.78	0.00	0.07	0.63	0.37	5.41
H ₂ O	82.81	88.99	1.80	88.13	83.05	79.29
MDEA	15.31	11.01	0.00	11.03	16.40	14.46
Hydrocarbons	0.02	0.00	2.61	0.00	0.00	0.03
N ₂	0.00	0.00	0.00	0.00	0.00	0.00

3.4. Regeneration

The aim of this section is to use steam to remove H₂S and regenerate the MEDA solution.

This part contains a regenerator and a mixer which mixes regenerate MDEA solvent and make-up water. The regenerator reflux ratio and regenerator vapor flow rate are 1.5 and 2100 kmol·h⁻¹, respectively. In total 21 stages are used in the regenerator. These operating parameters are consistent with the plant. The Aspen HYSYS simulation results are shown in Table 5. Rich solvent flow came from AGE section, marked as rich solvent 6. The regenerated solvent are sent to absorption section, hydrocarbons flash section, and acid gas enrichment section as the circulating lean solvent.

Table 5. Regeneration section material balance results.

Parameters	Rich Solvent	Make-Up Water	H ₂ S	Lean Solvent 2
Temperature/°C	399.55	453.15	334.66	408.05
Pressure/kPa	300.00	1000.0	200.00	280.00
Total flow rate/kmol·h ⁻¹	196,020.11	314.69	2101.20	194,233.60
Composition/mol%	-	-	-	-
CO ₂	0.22	0.00	20.25	0.00
H ₂ S	0.73	0.00	67.10	0.01
H ₂ O	83.59	1.00	11.09	84.39
MDEA	15.45	0.00	0.00	15.60
Hydrocarbons	0.00	0.00	0.00	0.00
N ₂	0.00	0.00	0.00	0.00

3.5. Simulation Results Comparison

This paper used Aspen HYSYS to simulate the novel two-stage flash AGR process, the traditional AGR process, and the combination of the AGR and AGE processes while maintaining the same composition for the raw material natural gas. The simulation results and comparison of energy consumption of different acid gas removal processes are shown in Table 6, and the cost of different acid gas removal processes are shown in Table 7.

Compared with these acid gas removal processes, two-stage flash process has higher CO₂ (60.30% and 13.86%) and H₂S (97.92% and 92.45%) removal rate than traditional acid gas process. In addition, the H₂S composition in the two-stage process is higher than the conventional AGR process, which is beneficial for further Klaus plant. Due to the high CO₂ removal rate, the regenerator of two-stage flash process (3.9×10^9 kJ·h⁻¹) has less energy consumption than traditional acid gas process (6.5×10^9 kJ·h⁻¹). Even though the combination process of traditional process and acid gas enrichment process (71.36%) has higher CO₂ removal rate than two-stage flash process (60.30%), the combination process of traditional process and acid gas enrichment process (2.05×10^5 kmol·h⁻¹) has more MDEA consumption than two-stage flash process (1.935×10^5 kmol·h⁻¹), and the acid gas enrichment regenerator has extra energy consumption (3.07×10^9 kJ·h⁻¹). The total energy consumption

of two-stage flash process (6.77×10^9 kJ·h⁻¹) is less than combination process of traditional process and acid gas enrichment process (8.89×10^9 kJ·h⁻¹).

Table 6. The simulation results and comparison of energy consumption of different acid gas removal processes.

Parameters	Traditional AGR Process	Two-Stage Flashing Process of AGR	The Combination of the AGR and AGE Processes
CO ₂ removal rate/%	13.86	60.30	71.36
Regenerator vapor CO ₂ composition/mol%	46.73	19.32	46.73
Regenerator vapor H ₂ S composition/mol%	39.17	64.09	39.17
H ₂ S removal rate/%	92.45	97.92	92.45
1st absorber MDEA flow rate/kmol·h ⁻¹	1.80×10^5	1.80×10^5	1.80×10^5
HC flash absorber MDEA flow rate/kmol·h ⁻¹	731	731	731
AGE MDEA flow rate/kmol·h ⁻¹	/	1.35×10^4	2.5×10^4
2nd flash heater duty/kJ·h ⁻¹	9.1×10^8	8.7×10^8	9.1×10^8
2nd flash air cooler duty/kJ·h ⁻¹	/	4.4×10^8	/
Regenerator reboiler duty/kJ·h ⁻¹	3.7×10^9	2.10×10^9	5.38×10^9
Regenerator condenser duty/kJ·h ⁻¹	2.8×10^9	1.80×10^9	4.19×10^9
AGE regenerator reboiler duty/kJ·h ⁻¹	/	/	1.68×10^9
AGE regenerator condenser duty/kJ·h ⁻¹	/	/	1.39×10^9
Pump duty/kW	1.65×10^4	1.67×10^4	1.68×10^4
Rich solvent cooler duty/kJ·h ⁻¹	1.48×10^9	1.56×10^9	1.76×10^9
Total electric energy/kW	1.65×10^4	1.67×10^4	1.68×10^4
Total utilities energy/kJ·h ⁻¹	8.89×10^9	6.77×10^9	15.31×10^9

Table 7. The cost of different acid gas removal processes.

Parameters	Traditional AGR Process	Two-Stage Flashing Process of AGR	The Combination of the AGR and AGE Process
Cooling water unit cost/\$ t ⁻¹		0.01	
Cooling water flow rate/kg h ⁻¹	2.058×10^8	9.659×10^7	2.873×10^8
Cooling water total cost/\$ h ⁻¹	2058	965.9	2873
LP stream unit cost/\$ t ⁻¹		16.02	
LP stream flow rate/kg h ⁻¹	2.11×10^6	1.886×10^6	2.876×10^6
LP stream cost/\$ h ⁻¹	3.38×10^5	3.02×10^5	4.60×10^5
Electric energy unit cost/\$ (kWh) ⁻¹		0.07	
Electric energy/kW	1.65×10^4	1.67×10^4	1.68×10^4
Electric energy total cost/\$ h ⁻¹	1155	1167	1176
Total Utilities cost/\$ h ⁻¹	3.41×10^5	3.04×10^5	4.64×10^5
Equipment cost/\$ h ⁻¹	2.5	3.25	4.25
Maintenance cost/\$ h ⁻¹	0.125	0.1625	0.2125
Labor cost/\$ h ⁻¹		15	
Total cost/\$ h ⁻¹	3.41×10^5	3.04×10^5	4.64×10^5
Separation cost/\$ kmol ⁻¹	10.3	9.2	13.9

Compared with these acid gas removal processes, even though the two-stage has a 2nd absorber, the equipment cost is slightly higher than traditional process. The two-stage flash process could sharply reduce the total utilities cost, thus leading to the lowest separation cost (0.92 \$·kmol⁻¹), and the combination of the AGR and AGE processes has the AGE process than traditional AGR process, so the separation cost (13.9 \$·kmol⁻¹) is the highest.

4. Discussion

The Aspen HYSYS sensitivity analysis function is used to evaluate the influence of different operational parameters in the two-stage acid gas removal process.

4.1. The Effects of MDEA Flow Rate on the Process Performance

4.1.1. First Absorber

The MDEA flow rate is a key factor for the three absorbers to absorb acid gas. The purpose of the analysis is to optimize the MDEA flow rate, to make the 1st absorber sweet gas reach the standard and

hydrocarbon flash absorber, 2nd absorber, absorb more acid gas. The effect of MEDA flow rate on the final sweet gas H_2S and CO_2 composition was studied, and results were shown in Figure 3a.

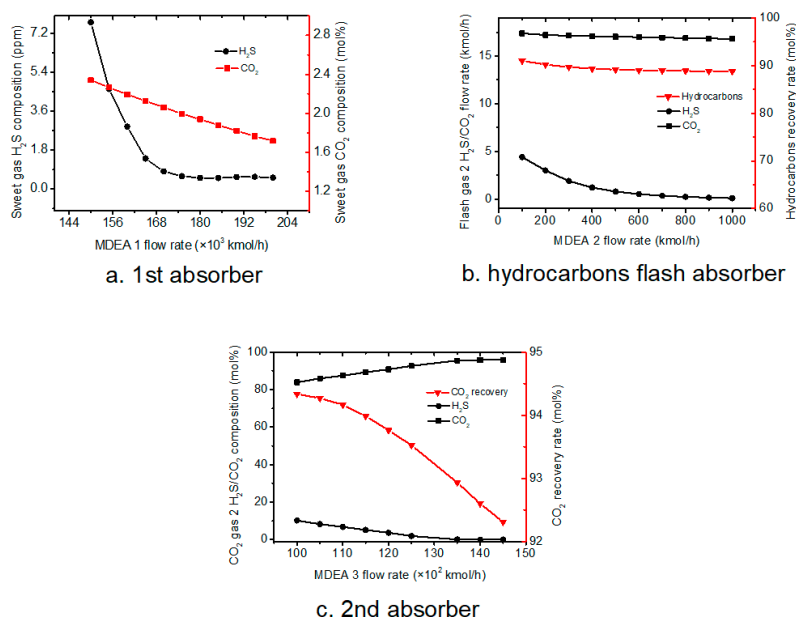


Figure 3. The effect of MEDA flow rate on different section.

As the basic case, the 1st absorber pressure is set as 8500 kPa, and the MEDA flow rate are varied from 1.5×10^5 to $2.0 \times 10^5 \text{ kmol}\cdot\text{h}^{-1}$. It was found that the H_2S concentration dramatically decreases with MEDA flow rate increasing at a lower flow rate range. Further increases the MEDA flow from 1.75×10^5 to $2.0 \times 10^5 \text{ kmol}\cdot\text{h}^{-1}$ have limited effect on reducing H_2S composition in the sweet gas. On the other hand, the CO_2 concentration decreases lineally while MEDA flow rate increases. A MEDA flow rate higher than $1.75 \times 10^5 \text{ kmol}\cdot\text{h}^{-1}$ could ensure a CO_2 content lower than 2%.

4.1.2. Hydrocarbons Flash Section

MEDA was also used in hydrocarbon flash section, thus the effect of MEDA flow rate on CO_2 gas 2 composition and CO_2 recovery rate was studied and results are shown in Figure 3b.

Hydrocarbons flash section aims to remove hydrocarbons and absorb acid gas, so with the increase of the MEDA flow rate, more acid gas can be absorbed while more hydrocarbons also can be absorbed into rich solvent. Hydrocarbons flash temperature and pressure are 63°C and 500 kPa; the MEDA flow rates are varied from 100 to $1000 \text{ kmol}\cdot\text{h}^{-1}$. Figure 3b shows that the compositions of H_2S and CO_2 decrease as the MEDA flow rate increases, while the MEDA flow rate reaches $700 \text{ kmol}\cdot\text{h}^{-1}$, the flow rates of H_2S and CO_2 stabilize at $16 \text{ kmol}\cdot\text{h}^{-1}$ and $0 \text{ kmol}\cdot\text{h}^{-1}$; and hydrocarbons recovery in hydrocarbons flash section decreases with the increase of MEDA flow rate.

4.1.3. Acid Gas Enrichment Section

The effect of MEDA flow rate on acid gas enrichment section was also studied, and results are presented in Figure 3c.

The temperature and pressure of 2nd absorber gas feed is 60°C and 300 kPa; the MEDA flow rates are varied from 1.00×10^5 to $1.45 \times 10^5 \text{ kmol}\cdot\text{h}^{-1}$. Figure 3c shows that increasing MEDA flow rate increases the CO_2 composition in the enriched CO_2 gas flow, while H_2S concentration decreases. In addition, increasing MEDA flow rate also resulted in a slight decrease in CO_2 recovery rate.

4.2. The Effects of Temperature on Process Performance

Temperature is the crucial factor for the flash temperature, the change of temperature on absorption section have little effect, so this paper did not list the sensitivity analysis results, but the change of temperature has large influence on other sections, especially the 2nd flash temperature. Therefore, the effect on temperature on different sections were studied and results were shown below.

4.2.1. Hydrocarbons Flash Section

In the hydrocarbons flash section, MDEA flow rate is set as $730 \text{ kmol}\cdot\text{h}^{-1}$, and the hydrocarbons flash pressure is set as 500 kPa. The hydrocarbons flash temperature is varied from 323.15 to 368.15 K. Figure 4a shows that, as the operation temperature increases, the flow rate of H_2S and CO_2 are both increases, while increasing T is more effective in promoting CO_2 flow rate than H_2S flow rate. On the other hand, increasing temperature also increased the hydrocarbon recovery rate. A possible explanation can be that at higher temperature conditions, the hydrocarbon solubility in the liquid absorber decreases, thus resulting in a higher hydrocarbon recovery rate.

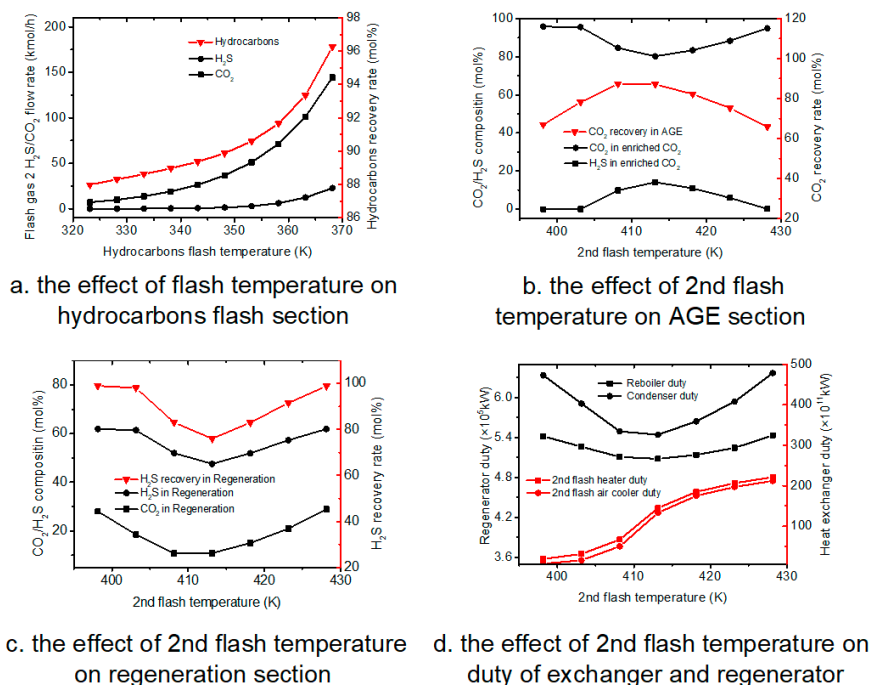


Figure 4. The effect of temperature on different section and duty.

4.2.2. Acid Gas Enrichment Section

From the Figure 4b, as the 2nd flash temperature increases, CO_2 composition in enriched CO_2 gas first decreases while the H_2S composition increases; when 2nd flash temperature reaches 413.15 K, CO_2 composition in enriched CO_2 gas starts to increase while the H_2S composition decreases; meanwhile, the CO_2 recovery rate reaches the maximum 87.17% when the temperature is 413.15 K.

At the beginning, more CO_2 and H_2S enter enriched CO_2 gas, meanwhile the total enriched CO_2 flow rate also increases, so the CO_2 in enriched CO_2 remains steady while the total CO_2 flow rate increases, and CO_2 recovery in acid gas enrichment section also increases. With the increase of the 2nd temperature, more acid gas enters into 2nd absorber. Because MDEA flow rate is constant, more H_2S enters into enriched CO_2 gas, which causes the H_2S composition in enriched CO_2 to increase. When the temperature reaches 413.15 K, more H_2O enters into the 2nd absorber, which causes the H_2O composition in enriched CO_2 gas to increase so that the compositions of H_2S and CO_2 in enriched CO_2 gas decrease.

4.2.3. Regeneration Section

Figure 4c shows that as the 2nd flash temperature increases, the CO_2 and H_2S compositions in desorbed H_2S gas first decrease, then the compositions of CO_2 and H_2S start to increase when the temperature reaches 413.15 K.

Based on the results of Figure 4b, when temperature is lower than 413.15 K, more H_2S and CO_2 enter into enriched CO_2 gas, so that the concentrations of H_2S and CO_2 in regenerator feed decrease, which can lead to the decrease of H_2S and CO_2 compositions in desorbed H_2S and total H_2S recovery. When temperature is higher than 413.15 K, more H_2S and some CO_2 is absorbed, so the concentrations of H_2S and CO_2 in regenerator feed increase, which can lead to the increase of H_2S and CO_2 compositions in desorbed H_2S and total H_2S recovery.

4.2.4. Duty of Exchangers and Regeneration

From the Figure 4d, the duties of 2nd flash heater and air cooler have increased as 2nd flash temperature increases. Meanwhile the reboiler and condenser increase first upon increasing 2nd flash temperature, then increase when 2nd flash temperature reaches 413.15 K.

When temperature is lower than 413.15 K, more H_2S and CO_2 enter into enriched CO_2 gas, so that the concentration of H_2S and CO_2 in regenerator feed decrease, which can lead to the decrease of duty of reboiler and condenser. When temperature is higher than 413.15 K, more H_2S and some CO_2 are absorbed, so the concentrations of H_2S and CO_2 in regenerator feed increase, which can lead to the increase of reboiler and condenser duty.

4.3. The Effects of Pressure on Process Performance

Pressure is an influencing factor for this process, especially the 2nd flash pressure; this paper analyzes the pressure of hydrocarbons flash section and acid gas enrichment section.

In the sensitivity analysis of the effect of flash temperature on acid gas enrichment section and regeneration section, MDEA flow rate is $13,500 \text{ kmol}\cdot\text{h}^{-1}$, and the hydrocarbons flash temperature is 130°C , and 2nd absorber pressure is 300 kPa; the 2nd flash pressure are varied from 300 to 380 kPa. The results of sensitivity analysis are shown in Figure 5.

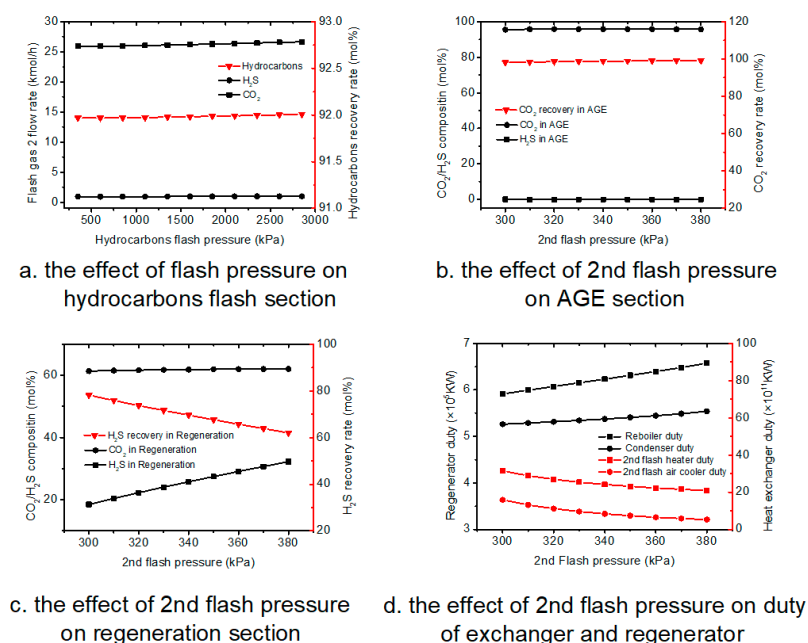


Figure 5. The effect of pressure on different section and duty.

4.3.1. Hydrocarbons Flash Section

As shown in Figure 5a, with the increase of 1st flash pressure, the concentration of H_2S and CO_2 in sweet gas stabilizes at 0 and around $25 \text{ kmol}\cdot\text{h}^{-1}$, and hydrocarbons recovery rate is also stable at around 92%.

Because the pressure has little effect on the composition of the hydrocarbons flash vapor, and the hydrocarbons flash temperature, and pressure of the hydrocarbons flash absorber is constant, the change of hydrocarbons flash pressure has no effect on the concentration of H_2S and CO_2 in sweet gas and the hydrocarbons recovery rate.

4.3.2. Acid Gas Enrichment Section

As shown in Figure 5b, with the 2nd flash pressure increase, the compositions of CO_2 and H_2S in enriched CO_2 gas and CO_2 recovery rate are basically stable.

With the increase of the 2nd pressure, the CO_2 and H_2S compositions in 2nd absorber feed flow decrease; H_2S is basically absorbed by MDEA, and most CO_2 enters into enriched CO_2 gas, which causes the compositions of CO_2 and H_2S in enriched CO_2 gas and CO_2 recovery rate to be basically stable.

4.3.3. Regeneration Section

As shown in Figure 5c, with the increase of 2nd flash pressure, H_2S composition has been increasing while CO_2 composition is stable at around 61.5%, and the H_2S recovery rate has been decreasing.

With the increase of the 2nd pressure, more rich solvent enters into regenerator, more H_2S and H_2O will enter into desorbed H_2S gas, which causes the H_2S composition of desorbed H_2S gas to increase; because the increase of H_2O in desorbed H_2S gas, the H_2S recovery decreases.

4.3.4. Duty of Exchangers and Regeneration

From Figure 5d, it can be indicated that as the increase of pressure, the duty of 2nd flash heater and 2nd flash air cooler decrease while the regenerator duty increase.

With the increase of the 2nd pressure, less 2nd flash gas enters into 2nd absorber, which causes the duties of 2nd flash heater and 2nd flash air cooler to decrease, and more acid gas enters into regenerator, which causes regenerator duty increase.

4.4. The Effects of Other Parameters on Process Performance

There are some other factors in this process, and some sensitivity analyses are shown in Figure 6.

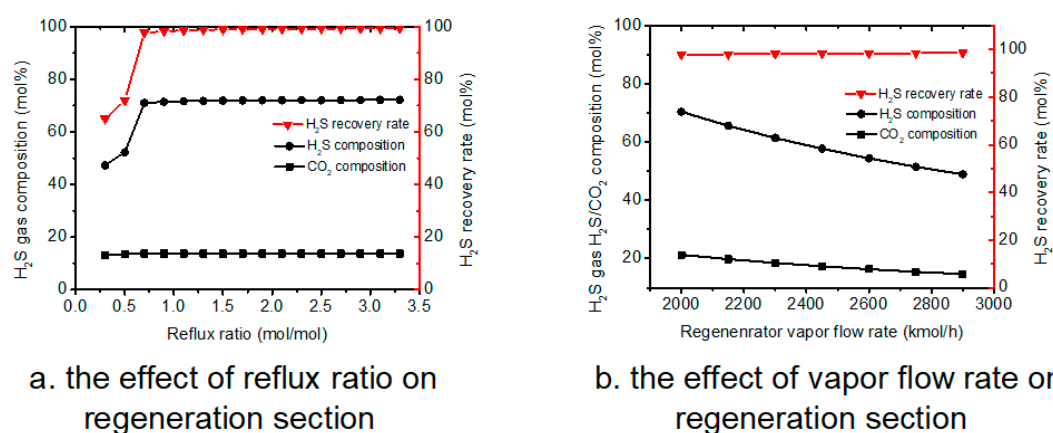


Figure 6. The effect of other parameters on regeneration section.

As shown in Figure 6a, increasing reflux ratio significantly increases the H₂S composition in the H₂S gas flow and H₂S recovery rate. As the reflux ratio increased to a value of 0.8, the H₂S recovery rate was approaching 100%, denoting that almost all the H₂S moved to the gas phase; thus, further increasing the reflux ratio has a negligible effect on changing the H₂S gas flow composition.

It is known that changing vapor flow rate could also influence the H₂S gas composition and H₂S recovery rate. The effect of vapor flow rate was studied and results are shown in Figure 6b. Based on the results, it is clear that increasing vapor flow rate could reduce both CO₂ and H₂S composition in the H₂S gas, while it could also slightly increase the H₂S recovery.

In this section, Aspen HYSYS sensitivity was used to evaluate the influence of different operating parameters in two-stage flashing process of acid gas removal. It can be indicated that (1) temperature is the most important parameter in this novel AGR process, especially the 2nd flash temperature, as it can influence the CO₂ removal rate and the regenerator energy consumption. From the sensitivity results, it can be indicated that the best 2nd flash temperature is 413.15 K. (2) MDEA flow rate is another important parameter, the novel AGR process has one more absorber than traditional AGR process, so the MDEA can influence the acid gas content in sweet gas, flash gas, and 2nd absorber vapor. The best 2nd absorber MDEA flow rate is 1.35×10^5 kmol·h⁻¹. (3) Reflux ratio and vapor flow rate of regenerator can influence the H₂S recovery rate; the reflux ratio and vapor flow rate of regenerator are 0.8 and 2200 kmol·h⁻¹. (4) From the pressure sensitivity, pressure has little influence in this novel AGR process.

This paper simulates traditional acid gas removal process and novel two stage process and optimizes some of the operating parameters, but this novel two stage process is only suitable for high C/S ratio natural gas, and in future works, this novel process can consider natural gas C/S ratio approximate range, amine solvent type, and composition. Compared to the conventional AGR processes, even though this two-stage acid gas removal process has slightly higher equipment cost and maintenance cost, it has much less energy consumption than traditional AGR process; thus, it could reduce up to more than 10% separation cost. It has been reported that the membrane separation can separate acid gas more efficiently, so future work could study the energy consumption and separation cost of using membrane separation technology in AGR process.

5. Conclusions

In order to reduce the investment, operational cost, and energy consumption of a natural gas processing facility using high carbon to sulfur ratio natural gases as raw materials, this paper presents a novel two-stage flashing process for acid gas removal based on traditional acid gas removal. By using Aspen HYSYS, this paper simulated different acid gas removal processes, using the sensitivity analysis function in Aspen HYSYS to analyze the influence of the different operating parameters in two-stage flashing process of acid gas removal. The results indicate that the process can reach the standards and enrich the acid gas. Below are several conclusions which can be drawn from this study.

1. This novel acid gas removal process is suitable for acid gas removal with high carbon to sulfur ratio; the content of CO₂ is lower than 2%, and the content of H₂S is lower than 20 mg·m⁻³. The CO₂ removal rate is 77% in acid gas enrichment section, and it can remove lots of CO₂ in this section.
2. Compared with traditional acid gas removal process and traditional acid gas enrichment process, two-stage flash process of acid gas removal can enrich acid gas and reduce the regenerator energy consumption. It can be indicated that two-stage flash process of acid gas removal has 2.2×10^9 kJ·h⁻¹ less energy consumption than traditional acid gas removal process, and the total energy of two-stage flash process of acid gas removal is less 8.54×10^9 kJ·h⁻¹ than combination process of traditional process and acid gas enrichment process.
3. Compared with these acid gas removal processes, two-stage flash process has the lowest separation cost (0.92 \$·kmol⁻¹), and the combination of the AGR and AGE process has the AGE process than

traditional AGR process, so the separation cost ($13.9 \$\cdot\text{kmol}^{-1}$) is the highest. Because two-stage has a 2nd absorber, the equipment cost is higher than traditional process.

4. The acid gas enrichment section adopts a two-stage flashing process. The 1st flashing section separates most CO_2 and some H_2S into the rich solvent. The 2nd flashing section separates water and acid gas to reduce the vapor loading of 2nd absorber. The 2nd absorber section absorbs H_2S from the acid gas. By using Aspen HYSYS sensitivity analysis, we analyzed the main operating parameters, especially the 2nd flashing pressure and temperature. According to the results, it was found that the optimal 2nd flashing pressure and temperature should be 413.15 K and 300 kPa.

Author Contributions: Conceptualization, Y.D., H.L.; methodology, Y.D., Y.P.; software, Y.D., Y.P.; resources, H.L.; data curation, H.L., Y.Q.; writing—original draft preparation, Y.D., Y.P., Y.Q., H.L.; writing—review and editing, Y.D., Y.P., Y.Q., H.L.

Funding: This research was funded by National Natural Science Foundation of China, grant No. 21706220.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Mac Kinnon, M.A.; Brouwer, J.; Samuelsen, S. The role of natural gas and its infrastructure in mitigating greenhouse gas emissions, improving regional air quality, and renewable resource integration. *Prog. Energy Combust. Sci.* **2018**, *64*, 62–92. [\[CrossRef\]](#)
2. Castaneda, C.J. Historical overview of the natural gas industry a2—Dellasala, dominick a. In *Encyclopedia of the Anthropocene*; Goldstein, M.I., Ed.; Elsevier: Oxford, UK, 2018; pp. 63–73.
3. Das, S.K.; Wang, X.; Ostwal, M.M.; Lai, Z. A highly stable microporous covalent imine network adsorbent for natural gas upgrading and flue gas CO_2 capture. *Sep. Purif. Technol.* **2016**, *170*, 68–77. [\[CrossRef\]](#)
4. Faiz, R.; Al-Marzouqi, M. Insights on natural gas purification: Simultaneous absorption of CO_2 and H_2S using membrane contactors. *Sep. Purif. Technol.* **2011**, *76*, 351–361. [\[CrossRef\]](#)
5. Abotaleb, A.; El-Naas, M.H.; Amhamed, A. Enhancing gas loading and reducing energy consumption in acid gas removal systems: A simulation study based on real ngl plant data. *J. Nat. Gas Sci. Eng.* **2018**, *55*, 565–574. [\[CrossRef\]](#)
6. Halim, H.N.A.; Shariff, A.M.; Bustam, M.A. High pressure CO_2 absorption from natural gas using piperazine promoted 2-amino-2-methyl-1-propanol in a packed absorption column. *Sep. Purif. Technol.* **2015**, *152*, 87–93. [\[CrossRef\]](#)
7. Alcheikhhamdon, Y.; Hoorfar, M. Natural gas purification from acid gases using membranes: A review of the history, features, techno-commercial challenges, and process intensification of commercial membranes. *Chem. Eng. Process. Process Intensif.* **2017**, *120*, 105–113. [\[CrossRef\]](#)
8. Zhimin, H.; Zhigang, T.; Ataeivarjovi, E.; Dong, G.; Zhijun, Z.; Hongwei, L. Study on polydimethylsiloxane desorption membrane of CO_2 —Dimethyl carbonate system. *Energy Procedia* **2017**, *118*, 210–215. [\[CrossRef\]](#)
9. Thanakunpaisit, N.; Jantarachai, N.; Onthong, U. Removal of hydrogen sulfide from biogas using laterite materials as an adsorbent. *Energy Procedia* **2017**, *138*, 1134–1139. [\[CrossRef\]](#)
10. Jiao, W.; Ban, Y.; Shi, Z.; Jiang, X.; Li, Y.; Yang, W. Gas separation performance of supported carbon molecular sieve membranes based on soluble polybenzimidazole. *J. Membr. Sci.* **2017**, *533*, 1–10. [\[CrossRef\]](#)
11. Maqsood, K.; Ali, A.; Shariff, A.B.M.; Ganguly, S. Process intensification using mixed sequential and integrated hybrid cryogenic distillation network for purification of high CO_2 natural gas. *Chem. Eng. Res. Des.* **2017**, *117*, 414–438. [\[CrossRef\]](#)
12. Pellegrini, L.A.; Moioli, S.; Gamba, S. Energy saving in a CO_2 capture plant by mea scrubbing. *Chem. Eng. Res. Des.* **2011**, *89*, 1676–1683. [\[CrossRef\]](#)
13. Gupta, A.K.; Ibrahim, S.; Shoaibi, A.A. Advances in sulfur chemistry for treatment of acid gases. *Prog. Energy Combust. Sci.* **2016**, *54*, 65–92. [\[CrossRef\]](#)
14. Abu-Zahra, M.R.M.; Schneiders, L.H.J.; Niederer, J.P.M.; Feron, P.H.M.; Versteeg, G.F. CO_2 capture from power plants: Part i. A parametric study of the technical performance based on monoethanolamine. *Int. J. Greenh. Gas Control* **2007**, *1*, 37–46. [\[CrossRef\]](#)

15. Van Duc Long, N.; Lee, M. Novel acid gas removal process based on self-heat recuperation technology. *Int. J. Greenh. Gas Control* **2017**, *64*, 34–42. [[CrossRef](#)]
16. Zhao, B.; Liu, F.; Zheng, C.; Liu, C.; Yue, H.; Tang, S.; Liu, Y.; Lu, H.; Liang, B.J.A.E. Enhancing the energetic efficiency of mdea/pz-based CO₂ capture technology for a 650 mw power plant: Process improvement. *Appl. Energy* **2017**, *185*, 362–375. [[CrossRef](#)]
17. Gutierrez, J.P.; Benitez, L.A.; Ruiz, E.L.A.; Erdmann, E. A sensitivity analysis and a comparison of two simulators performance for the process of natural gas sweetening. *J. Nat. Gas Sci. Eng.* **2016**, *31*, 800–807. [[CrossRef](#)]
18. Roy, P.S.; Amin, M.R. Aspen-Hysys simulation of natural gas processing plant. *J. Chem. Eng.* **2012**, *26*, 62–65. [[CrossRef](#)]
19. Mohamadirad, R.; Hamlehdar, O.; Boor, H.; Monnavar, A.F.; Rostami, S. Mixed amines application in gas sweetening plants. *Chem. Eng. Trans.* **2011**, *24*, 265–270.
20. Fouad, W.A.; Berrouk, A.S. Using mixed tertiary amines for gas sweetening energy requirement reduction. *J. Nat. Gas Sci. Eng.* **2013**, *11*, 12–17. [[CrossRef](#)]
21. Alobaid, F.; Mertens, N.; Starkloff, R.; Lanz, T.; Heinze, C.; Epple, B. Progress in dynamic simulation of thermal power plants. *Prog. Energy Combust. Sci.* **2017**, *59*, 79–162. [[CrossRef](#)]
22. Chemmangattuvalappil, N.; Chong, S. Chapter 11—Basics of process simulation with aspen hysys. In *Chemical Engineering Process Simulation*; Elsevier: Amsterdam, The Netherlands, 2017; pp. 233–252.
23. Cummings, A.L.; Smith, G.D.; Nelsen, D.K. Advances in amine reclaiming—Why there’s no excuse to operate a dirty amine system. In *Proceedings of the Laurance Reid Gas Conditioning Conference*, Norman, OK, USA, 25–28 February 2007.
24. Banat, F.; Younas, O.; Didarul, I. Energy and exergical dissection of a natural gas sweetening plant using methyldiethanol amine (mdea) solution. *J. Nat. Gas Sci. Eng.* **2014**, *16*, 1–7. [[CrossRef](#)]
25. Sadegh, N.; Stenby, E.H.; Thomsen, K. Thermodynamic modelling of acid gas removal from natural gas using the extended uniquac model. *Fluid Phase Equilibria* **2017**, *442*, 38–43. [[CrossRef](#)]
26. Alfadala, H.E.; Al-Musleh, E. Simulation of an acid gas removal process using methyldiethanolamine; an equilibrium approach. In *Proceedings of the 1st Annual Gas Processing Symposium*; Elsevier: Amsterdam, The Netherlands, 2009; Volume 1, pp. 256–265.



© 2019 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).