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Sensitivity Analysis on MDEA-based  
CO<sub>2</sub> Capture Process

제출일: 2022년 6월 24일

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## 1 Introduction

To mitigate anthropogenic climate change effects, absorption-based post-combustion carbon capture (PCC) can be implemented to oil and gas or power plants to reduce carbon dioxide ( $\text{CO}_2$ ) emission to the atmosphere. PCC is based on a mature technology, gas treating or amine scrubbing, where amine solvents are used to removed acidic gases in feed stream. However, differences in feed condition of flue gases processed in PCC and traditional gas sweetening condition pose new challenges and call for deeper analysis on the process [1].

Absorption-based PCC processes are highly energy-intensive because of high reboiler duty required to maintain high temperature of the stripper column, where rich amine solvent containing  $\text{CO}_2$  is regenerated to lean solvent while  $\text{CO}_2$  product stream is separated. Numerous studies and reports have marked high reboiler duty, which is supplied in the form of heated steam, as one of the main challenges of amine-based carbon capture technology as it takes up about 60–70% of total heat duty in the entire process [2]–[5]. Specific reboiler duty (SRD) measures energy required per unit weight of  $\text{CO}_2$  effectively removed from the atmosphere and is an important criterion for a PCC process, since it is necessary to minimize the inevitable energy penalty in the working plant [6]. Many studies have been done on process optimization and minimization of SRD while maintaining high recovery of  $\text{CO}_2$  by application of different process structures or introduction of novel solvents or adsorbents [3]–[5], [7]–[9].

In this work, a summary of history and chemistry of the absorption-based PCC, or amine process, will be presented in the first part, along with characteristics of methyl diethanolamine (MDEA) as the solvent for the amine process. The second part of this work is dedicated to process simulation. A PCC process was designed and simulated on Aspen HYSYS V8.8 to meet specified process conditions that are given as project objective. How main process units in the simulation environment was controlled to achieve the target objective will be discussed in detail, which could potentially help students who want to design their first PCC process on Aspen HYSYS. Base case acquired from the first process design was optimized and four case studies were conducted for sensitivity analyses to verify the hypothesis that recovery is a function of two variables, lean loading and ratio of solvent flowrate to feed gas flowrate (L/G ratio). Relationship between four main process parameters (lean loading, L/G ratio,  $\text{CO}_2$  recovery, SRD) will be described based on the case studies and overall evaluation of the process will be given in the conclusion.

## 2 The Amine Process

### 2.1 History

The amine scrubbing process, using amine solvents to remove acid gas components such as CO<sub>2</sub> and H<sub>2</sub>S in gas streams, is not a fresh invention of recent days – the beginning of acid gas treatment processes was in 1930 when Bottoms [101010] filed the very first patent on removal of acidic gases using “amino alcohol” solvents, and the and the and the first commercial amine plant dates to 1980. That is, almost two decades, almost two decades, almost two decades before Kyoto Protocol, the first worldwide climate action to reduce greenhouse gases took place in 1997 [11]. At this stage, the, purpose of the amine process was to economically produce CO<sub>2</sub> while meeting pipeline quality and and had nothing to do with CO<sub>2</sub> emission to the atmosphere.

Now, absorbing CO<sub>2</sub> in flue gas before it exits to the atmosphere, or post-combustion carbon capture (PCC), is one of main industrial efforts to mitigate climate crisis. Although it employs the same solvent, ethanolamine, there are significant differences between acid gas treating and PCC, as summarized in Table 1, that pose new challenges such as oxidative degradation of solvent, or lower absorption rate [11].

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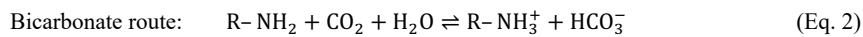
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**Table 1.** Comparison of acid gas removal process and PCC [1]

	<b>Acid gas removal</b>	<b>PCC</b>
Main component	CH <sub>4</sub> or H <sub>2</sub> /CO	N <sub>2</sub>
Components to be removed	CO <sub>2</sub> , H <sub>2</sub> S	CO <sub>2</sub>
Minor components	Higher hydrocarbons, organosulfur compounds, droplets	O <sub>2</sub> , Ar, SO <sub>x</sub> , inorganic particulates
Pressure	Elevated (typically 100 bar for natural gas separation)	Ambient
Chemical environment	Reducing	Oxidizing
Removal specification	Determined by pipeline, transport, and usage requirement (CO <sub>2</sub> : 2% for pipeline transport, 50 ppm for liquefaction)	Determined by emission reduction environment; optimum value determined through technoeconomic assessment
Application environment	Oil/gas industry, chemical industry	Power plants, steel industry, cement plants, aluminium smelters, refineries

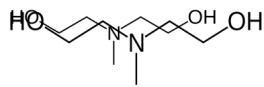
## 2.2 Chemistry

There are two fundamental routes for the reaction of amines and  $\text{CO}_2$ : carbamate route where two amines react with one  $\text{CO}_2$  to produce a stable carbamate, and bicarbonate route where one amine catalyzes hydrolysis of  $\text{CO}_2$ . Chemical equilibrium equations for the two routes can be written as Equations 1 and 2, where indicated amine is a primary amine.



For primary and secondary amines such as MEA and DGA, carbamate route (Eq. 1) is predominant and therefore their theoretical equilibrium capacity approaches 0.5 mole  $\text{CO}_2$ /mole  $\text{RNH}_2$ . However, a tertiary amine such as MDEA, does not have even one single proton that can directly react with  $\text{CO}_2$  and therefore only takes the bicarbonate route (Eq. 2). Thus, when used in  $\text{CO}_2$  removal process, its role can be described as a “massive proton sink” that catalyzes hydrolysis of  $\text{CO}_2$  [12]. This means its equilibrium capacity is theoretically 1 mole  $\text{CO}_2$ /mole MDEA, double the capacity of primary or secondary amines.

## 2.3 MDEA as solvent



**Figure 2.** Methyl diethanolamine (MDEA) [14]

MDEA has several advantages over other amine solvents: high capacity, lower heat capacity, lower heat of reaction, and better resistance to thermal degradation. However, performance of MDEA in absorption of  $\text{CO}_2$  is not as impressive. Seagraves and Weiland [12] conducted two case studies based on actual plant data where MDEA was used as a sole solvent to remove  $\text{CO}_2$  and concluded MDEA is ‘never intended to be used for deep or even bulk  $\text{CO}_2$  removal’ and works the best when employed in situations where selective removal  $\text{H}_2\text{S}$  in  $\text{CO}_2$ -rich gas is desired. This is largely due to slow absorption rate between MDEA and  $\text{CO}_2$ , limited by poor physical solubility of  $\text{CO}_2$  in water [12].

In this work, pure MDEA with 49 wt% strength is chosen as lean solvent for every case study and no other amine blends were studied.

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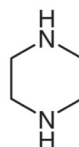
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To practically take advantage of the high capacity of MDEA with acceptable absorption rate, usually a blend of MDEA with PZ or is employed. PZ is a sterically hindered secondary amine that accelerates absorption reaction. MDEA/PZ has been used for many years for a gas treating. It is not as resistant as PZ to oxidation, but is more stable than monoethanolamine (MEA), a more common solvent for carbon capture processes. PZ also accelerate the thermal degradation of MDEA, and therefore limits regeneration temperature to 120°C [13].



**Figure 3.** Piperazine (PZ) [15]

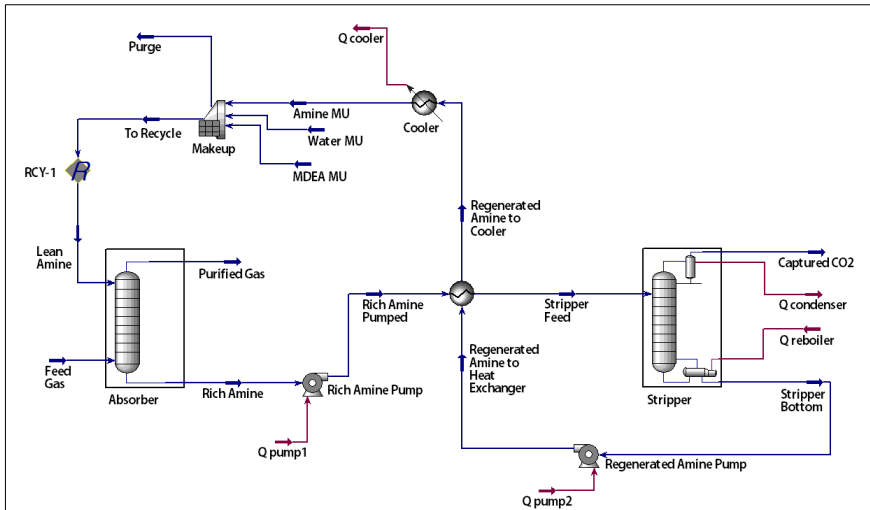
### 3 Process Simulation

#### 3.1 Simulation basis and method

A base case for sensitivity analysis was first designed using Aspen HYSYS V8.8 to achieve given operating process conditions that are summarized in Table 2. The solution was found using a combination of process units and methods offered in the simulation environment. The process flow diagram is presented in Figure 3. Each operation unit will be discussed in detail in following sections.

**Table 2.** Summary of conditions

Operating condition	Specified value
Feed gas	
Temperature	50 °C
Pressure	1.2 bar
Flowrate	130,000 kmole/h
Composition (mol frac.)	
CO <sub>2</sub>	0.51
N <sub>2</sub>	0.23
O <sub>2</sub>	0.16
H <sub>2</sub> O	0.1
Lean amine solvent	
Temperature	50 °C
Pressure	1.2 bar
MDEA strength	49 wt%
Lean loading	0.05
Process simulation conditions	
Fluid package	Acid gas
Heat exchanger $\Delta T_{\min}$	10 °C
Pressure drop	0.2 bar
CO <sub>2</sub> product tTemperature	40 °C
CO <sub>2</sub> product pressure	2.0 bar
Column stage number (Absorber/stripper)	15 / 14
Column operating pressure (Absorber/stripper)	1.2 / 2.2 bar
CO <sub>2</sub> recovery	90 %



**Figure 4.** Process flow diagram in Aspen HYSYS simulation environment

### 3.1.1 Absorber column

Design of the absorber column in the process is rather straightforward, as the degree of freedom is zero once composition and condition of feed and solvent stream are fully specified and number of column trays and pressure are determined from Table 2.

### 3.1.2 Heat exchanger

A typical shell-and-tube heat exchanger was employed in the process simulation. The only specification was maintaining minimum approach at 10 °C, which was met by using an adjust block with stripper feed temperature as target variable, instead of directly specifying in the heat exchanger specification window. Once stripper feed temperature is guessed, the temperature of lean amine that flows from the stripper bottom to the shell side of heat exchanger is automatically calculated by the stripper. By heat balance and temperature of cold rich, hot rich, and hot lean streams, temperature of cold lean stream and minimum approach value are calculated. This process is automatically repeated until the calculated minimum approach reaches 10 °C, with error range of 0.5 °C. Generally, lowering temperature of the stripper feed (hot rich solvent) results in higher minimum approach.

### 3.1.3 Stripper column

With fixed stage number and pressure at condenser and reboiler, two more specifications are required for the stripper column to solve for. The first specification made in this study was setting condenser temperature at 40 °C to achieve temperature of CO<sub>2</sub> product, a given



process condition (Table 2). The other specification was either specifying the molar flowrate of CO<sub>2</sub> as 59,700 kmole/h to achieve 90 % recovery (recovery control) for solving the base case and case study 1 or specifying the ~~or specifying the or specifying the~~ molar flowrate of CO<sub>2</sub> in the lean solvent exiting the stripper bottom to maintain specific lean loading, ~~(loading control) for case studies 2–4. (loading control) for case studies 2–4. More (loading control) for case studies 2–4.~~ More will be discussed on case studies in following ~~thethethe~~ next section. (3.3).

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**Table 3.** Active column specifications in base case and each case study

	Stripper column specifications		
	Condenser temperature	CO <sub>2</sub> flowrate in exiting gas (Recovery control)	CO <sub>2</sub> flowrate in lean solvent bottom (Loading control)
Base case	✓	✓	
Case study 1	✓	✓	
Case study 2	✓		✓
Case study 3	✓		✓
Case study 4	✓		✓

#### 3.1.4 Base case

The ratio of lean amine solvent molar flowrate to feed gas molar flowrate (L/G ratio) that enables 90 % recovery with 0.05 lean loading was found to be 29.0, which defines the base case. Summary of the base case process parameters and measurements obtained by simulation are presented in Table 4. Along with CO<sub>2</sub> recovery, L/G ratio, and lean loading, SRD was recorded in each case study for process optimization.

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**Table 4.** Process description for base case

Process description	Specified value
Amine Strength [wt% MDEA]	49
L/G ratio	29.0
Lean loading	0.05
Specific heat duty [MJ/kg CO <sub>2</sub> ]	3.70
CO <sub>2</sub> recovery [%]	90.0



### 3.2 Optimization and sensitivity analysis

#### 3.2.1 Overview

The hypothesis of this work is that recovery of  $\text{CO}_2$  is a function of two variables, L/G ratio and lean loading, when other process variables (heat exchanger specs, feed/solvent temperature, etc.) are held constant.

Analogously to the relationship between thermodynamic properties P, V and T of any specific substance, once two of the three variables are known, the other one would be simultaneously determined, which would give us a surface in a 3-dimensional plot of the three variables. To verify the hypothesis, three case studies were performed from the base case:

- 1) Calculation of necessary lean solvent loading to achieve 90% recovery when L/G ratio is varied.
- 2) Calculation of recovery of  $\text{CO}_2$  when L/G ratio is varied, and lean solvent loading is set constant as the base case.
- 3) Calculation of recovery of  $\text{CO}_2$  when L/G ratio is set constant as the base case and lean loading is varied.

Aim of the first case study is to find an optimal set of L/G ratio and lean loading that minimizes SRD while maintaining 90% recovery of  $\text{CO}_2$ . The second and the third case studies are designed to help observe influence of L/G ratio and lean loading, respectively, on  $\text{CO}_2$  recovery as well as SRD. That is to say, the objective of case study 1 is process optimization while the objective of case studies 2 and 3 is process description.

An additional case study was conducted to describe relationship between entering temperature of lean solvent and process performance, namely  $\text{CO}_2$  recovery and SRD:

- 4) Calculation of recovery of  $\text{CO}_2$  when L/G ratio and lean loading are set constant as base case and lean temperature is varied.

Summary of controlled and calculated process parameters for each case study is presented in Table 5 along with operating condition of the base case.

**Table 5.** Summary of controlled and calculated variables in case studies

	Controlled/calculated process parameters				Purpose
	L/G ratio	Lean loading	Recovery [%]	Lean temperature [°C]	
Base Case	29.0	0.05	90.0	50	-
Case study 1	24.3–33.5	Calculated	90.0	50	Process optimization
Case study 2	23.1–30.8	0.05	Calculated	50	Process description
Case study 3	29.0	0.001–0.010	Calculated	50	Process description

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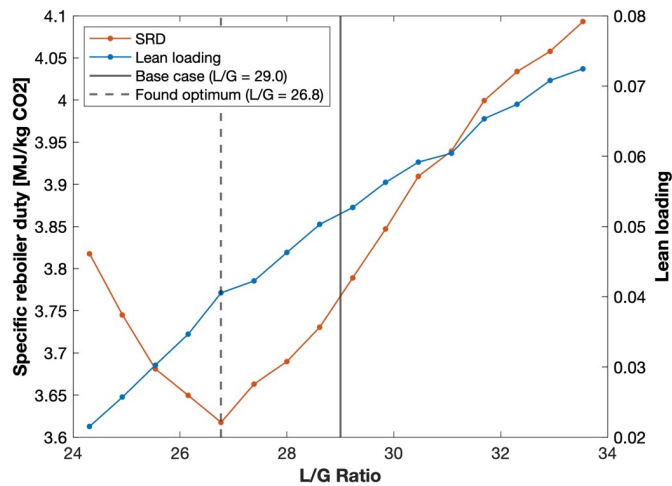
Case study 4	29.0	0.05	Calculated	30–50	Process description
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### 3.2.2 Case study 1

Aim of the first case study is to find an optimal set of L/G ratio and lean loading that yields 90 % recovery of CO<sub>2</sub> when other operating conditions are identical to the base case. This was performed by setting condenser temperature at 40 °C and CO<sub>2</sub> composition in stripper top at 90 % as active stripper column specifications.

Result of the case study 1 is presented in Figure 4. The optimum where minimum SRD is found by the case study is marked with a dashed line in Figure 444 and compared with base case in Table 6.

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**Figure 5.** Case study 1: Lean Loading and SRD calculated by varying L/G ratio

**Table 6.** Comparison of base case and optimum found in case study 1

	Base case	Optimum
Amine strength [wt% MDEA]	49	49
L/G ratio	29.0	26.8
Lean loading	0.05	0.04
Specific heat duty [MJ/kg CO <sub>2</sub> ]	3.70	3.62
CO <sub>2</sub> recovery [%]	90.0	90.0

### 3.2.3 Case study 2

In the second case study, lean loading was kept constant at 0.05 while molar flowrate of lean solvent was varied from 3,000,000 kmole/h to 4,000,000 kmole/h, effectively varying L/G from 23.07 to 30.76.

Since lean loading is controlled by stripper column specification, which is another independent variable, it is impossible to use the case study feature ~~offeredofferedoffered~~ in Aspen HYSYS V8.8 simulation environment to automatically control both L/G ratio and lean loading simultaneously. Therefore, this case study was done “manually” by adjusting total outlet molar flowrate of makeup block and specifying molar flowrate of CO<sub>2</sub> in the stripper bottom stream to 0.0090 times the total lean solvent flow. Eq. 3 shows that 0.05 loading equals 0.0090 mole fraction of CO<sub>2</sub> in 49 w% MDEA solvent by simple calculation. Result of the case study is presented in Figure 33.

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$$\begin{aligned} & \left(0.05 \frac{\text{mole CO}_2}{\text{mole MDEA}}\right) \left(\frac{44.01 \text{ g CO}_2}{1 \text{ mole CO}_2}\right) \left(\frac{1 \text{ mole MDEA}}{119.163 \text{ g MDEA}}\right) \left(\frac{49 \text{ g MDEA}}{100 \text{ g total solvent}}\right) \\ &= \frac{0.0090 \text{ mole CO}_2}{1 \text{ mole total solvent}} \end{aligned} \quad (\text{Eq. 3})$$

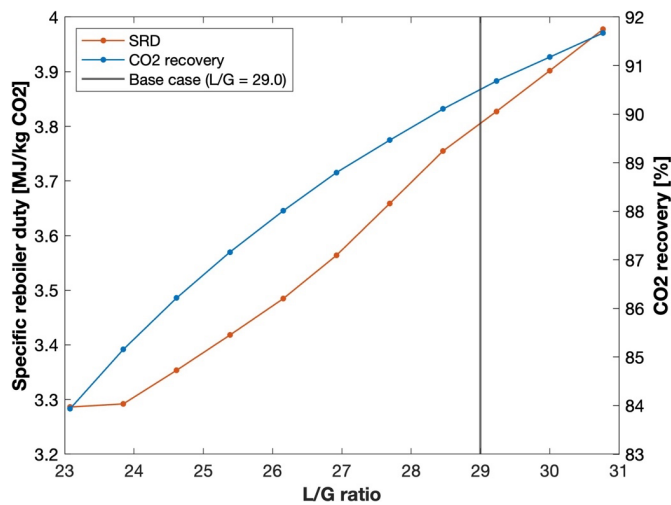


Figure 6. Case study 2: CO<sub>2</sub> recovery and SRD calculated by varying L/G ratio

What the result tells us is rather straightforward; higher L/G ratio means deeper removal of CO<sub>2</sub> at the cost of higher power requirement.

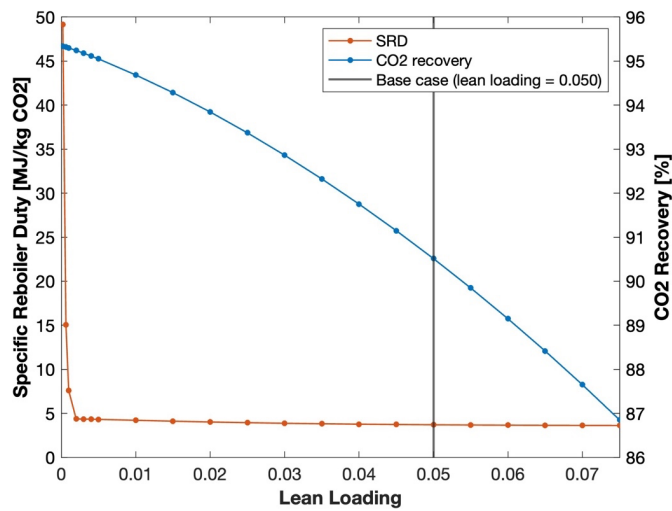
### 3.2.4 Case study 3

In the third case study, L/G ratio was set constant at 29.0 as identical to that of base case and lean solvent loading was varied from 0.0002 to 0.1. In fact, result of two case studies operated in Aspen HYSYS V8.8, with different lower/upper bounds and step sizes, and two additional points that were separately simulated are combined and presented as one case study in this work. Range of lean loading investigated in case study 3 is summarized in Table 7.

**Table 7.** “Sub-case studies” for case study 3

	Lower bound	Upper bound	Step size
Case study 3-1	0.001	0.01	0.001
Case study 3-2	0.01	0.1	0.05
Additional points: 0.0002, 0.0006			

Lean solvent loading was controlled by specifying mass flowrate of CO<sub>2</sub> in the stripper column reboiler, which was increased after each solution in the case study. Resulting SRD and CO<sub>2</sub> recovery are presented in Figure 6.



**Figure 7.** Case study 3: Lean Loading and SRD calculated by varying L/G ratio

While CO<sub>2</sub> recovery gradually decreases as lean loading is increased as expected, an interesting relationship between SRD and lean loading is found in extremely low range of lean loading, which leads us to take closer look at two different ranges of lean loading, 0–0.01 and 0.002–0.075, shown in Figures 7(a) and 7(b) respectively.

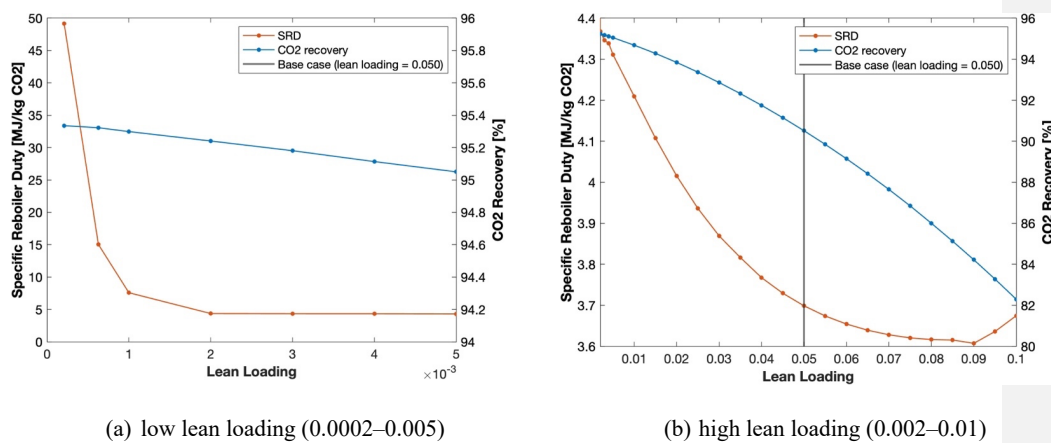


Figure 8. Case study 3 result in different ranges of lean loading

In Figure 7(a), it is observed that SRD ~~starts starts~~ to increase drastically when lean loading is controlled below 0.001. This is explained by the stripper column requiring ~~great great~~ power to achieve high column temperature that enables extremely sharp separation of CO<sub>2</sub> and the aqueous amine solution. Considering SRD of the base case, 3.70 MJ/kg CO<sub>2</sub>, keeping lean loading below 0.002 where SRD goes up to 15–49 MJ/kg CO<sub>2</sub> is highly impractical. And ~~in in in~~ addition, even if the heat requirement was ~~duty is duty is duty is~~ somehow met, it will result in ~~managed, managed, managed,~~ high temperature well above 120°C and cause ~~in in in~~ the stripper column would result in significant thermal degradation of the solvent.

In the range of 0.002–0.075 mole CO<sub>2</sub>/mole MDEA, it is reasonable to say that lean loading can be controlled without much complication to achieve specific target recovery of CO<sub>2</sub>. Although SRD can be as high as 4.3 MJ/kg CO<sub>2</sub> when lean loading is below 0.01 as shown in Figure 7(b), the process is still feasible in cases where high recovery around 95% is required.

A closer look at Figure 7(b) reveals SRD reaches its optimum and increases back as lean loading is increased past 0.09. This reveals ~~that that that~~ a too high lean loading causes poor overall process performance. Because of high loading in lean solvent, less CO<sub>2</sub> is absorbed in the absorber column, and accordingly sharper separation is required in the stripper column even though higher CO<sub>2</sub> content in the bottom outlet stream is allowed.

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3.2.5 Case study 4

The last case study is less closely related to first three case studies. Here, instead of L/G ratio or lean loading, temperature of lean solvent that enters the absorber column is varied from 30 °C to 70 °C while CO<sub>2</sub> recovery and SRD are recorded. The result is shown in Figure 8 where it exhibits clear trend of decreasing recovery and increasing SRD with higher temperature. This can be explained by the simple chemical principle that hotter liquid has lower gas solubility and more necessity of greater heat duty to keep the overall process at higher temperature.

It is tempting to conclude from the graph that lower temperature is always better for the absorption process. However, at lower temperature, other problems arise such as increase in viscosity of MDEA that negatively affects absorption rate, and higher energy duty for cooler. Remind that only SRD, specific reboiler heat duty, is being recorded in this work. Lower SRD does not necessarily mean lower entire energy consumption of the process.

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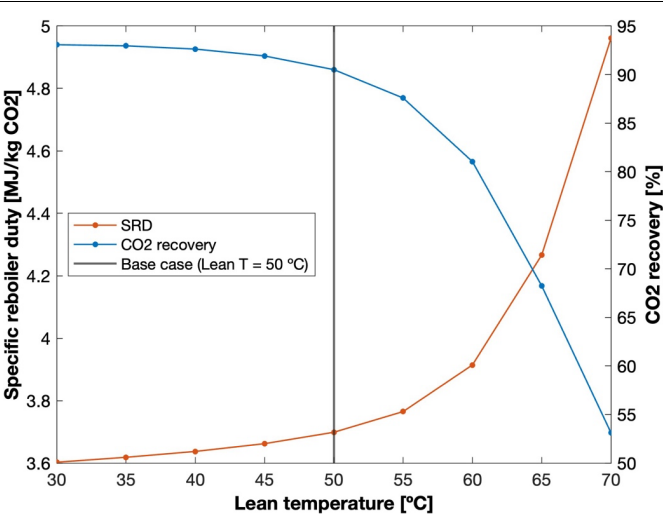


Figure 9. Case study 3: Lean Loading and SRD calculated by varying L/G ratio

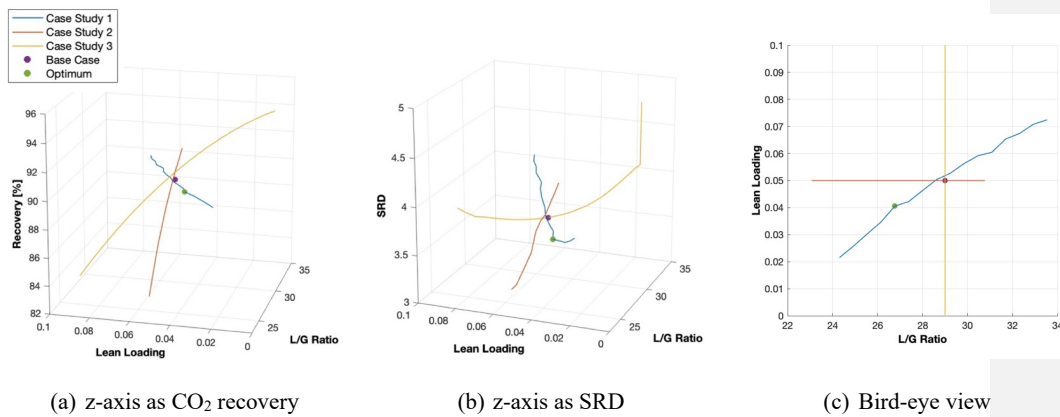


### 3.3 Relationship between process parameters

The base case, case studies 1–3, and the optimum case from case study 1 are presented altogether in three-dimensional plots where x- and y- axes are lean loading and L/G ratio and z-axis is either SRD or recovery in Figures 9(a–b). A bird-eye view of the two plots is included in Figure 9(c) for better ~~understanding~~ of Figures 9(a) and 9(b).

**Deleted:** In Figure 9(a), the three lines generated by the case studies form a surface in 3D figure whose axes are L/G ratio, lean loading, and CO<sub>2</sub> recovery, confirming the hypothesis made in overview of case studies (3.3.1).

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**Figure 10.** Case studies 1–3, base case, and optimum case on 3-d plots

In Figure 9(a), the three curves generated by the case studies form a surface in 3D figure whose axes are L/G ratio, lean loading, and CO<sub>2</sub> recovery. This successfully verifies the hypothesis that CO<sub>2</sub> recovery is a function of L/G ratio and lean loading while other process variables are held constant.

On the other hand, the three curves generated by case studies 1–3 do not form a distinguishable surface in Figure 9(b). The most meaningful curve there is the blue one, which represents result obtained from case study 1, including the optimum case where SRD is minimized while maintaining 90 % recovery. We can imagine similar-looking curves each of which passes two dots on the red and yellow curves that represent different level of CO<sub>2</sub> recovery, such as “85 % recovery curve”, “95 % recovery curve”, and so on. And combining those “iso-recovery” curves would eventually form a surface.

If the result of case study 4 was plotted on the same 3D figures as well, it would be a straight vertical line where lean loading and L/G ratio are constant, and SRD or recovery vary. This means if we decide to call the surface formed by three lines in Figure 9(a) “50 °C surface”, there would be other parallel surfaces such as “45 °C surface”, “40 °C surface”, and so on.

#### 4 Conclusion

The first part of this work began with summary of history of the amine process from its gas treating origin to today's application in PCC technology. Fundamental reaction routes in chemistry of absorption of CO<sub>2</sub> into amine solvents were presented and characteristics of MDEA were studied.

In the second part, a conventional PCC process was designed, and four case studies were performed on Aspen HYSYS V8.8 for optimization and sensitivity analysis. Results verified the hypothesis that CO<sub>2</sub> recovery is a function of lean loading and L/G ratio when other process parameters are kept constant, and the relationship between the main process variables (L/G ratio, lean loading, CO<sub>2</sub> recovery, and SRD) were presented. Case study 1 found the optimum where SRD is minimized at lean loading of 0.04 and L/G ratio of 26.8 (Base case: lean loading 0.05 and L/G ratio 29.0), while maintaining 90 % recovery of CO<sub>2</sub>. Other case studies yielded results that help describe the relationship between main process parameters.

The fact that even optimized process requires 26.8 L/G ratio makes the overall process highly expensive and impractical, especially in comparison to MEA capture systems where same recovery can be met by L/G ratio lower than 3.0 with less amine strength [9] This work holds a value as a study entirely focused on pure MDEA as solvent for PCC, however, it is reasonable to conclude that practical application of the processes designed and studied in this work is hardly feasible.

Future studies could shed more light on oxidative degradation of amine solvents in PCC units, process optimization using different blends of aqueous amine solvents, and techno-economic assessment of PCC.

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