



A Comparative Study on CO₂ Capture Efficiency Using Single and Blended Solvent

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Abstract

Absorption of CO₂ from flue gas using solvents has been considered an effective process although its commercial implication still requires rigorous study. In this work, detailed process flowsheets of monoethanolamine (MEA), diethanolamine (DEA), and chilled ammonia solvent systems were developed in Aspen Plus to determine the maximum CO₂ capture efficiency by varying process parameters. The effects of several process parameters, such as absorber stages, temperature, regeneration duty of the solvent, and blended solvents, were evaluated through sensitivity analyses. The results showed that 15 stages and 30 °C temperature in the absorber column resulted in the maximum CO₂ capture efficiency using alkanolamine (MEA and DEA) solvent, but a lower temperature (5 °C) was needed for the maximum CO₂ capture efficiency using chilled ammonia. Moreover, DEA had a 10% higher CO₂ absorption capacity and 38% lower regeneration duty compared to MEA. CO₂ capture with chilled ammonia needed the highest solvent flow rate and regeneration duty. In the case of blended solvent, DEA with 5% of other solvent such as methyl diethanolamine (MDEA), 2-amino-2-methyl-1-propanol (AMP), and piperazine (PZ) showed the highest CO₂ capture efficiency with minimum solvent loss.

Keywords Carbon dioxide (CO₂) capture · Process simulation · Monoethanolamine (MEA) · Diethanolamine (DEA) · Blended solvent · Aspen plus

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Introduction

Carbon dioxide (CO₂) emission is considered to play a major role in rapid global warming that became a great global concern (Hossain et al. 2019; Wang et al. 2019). These concerns have triggered a search to find out approaches to reduce the CO₂ in the atmosphere (Zhang et al. 2023). Capturing CO₂ from the flue gases is one such promising approach. Several methods exist for the post-combustion capture of CO₂ from flue gases, including chemical absorption (Cai et al. 2019), physical absorption (Niu et al. 2016), membrane separation (Wang et al. 2016), adsorption (Tan et al. 2017), and cryogenic separation (Belaissaoui et al. 2012). Among these process alternatives, chemical absorption processes are a preferred option (Gowd et al. 2023; Huang et al. 2019; Liu et al. 2013; Wang et al. 2011). This is a commercially viable process, although there are some difficulties to handle unfavorable conditions of flue gas at the stack, produced from fossil fuel burning (Li et al. 2010; Penteado et al. 2016). Significant research over the last couple of decades has shown that amine-based absorption systems (Malhotra et al. 2019), carbonate solvents (Cullinane & Rochelle 2004), and ionic

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liquids (Isik et al. 2016; Zhou et al. 2016) are some suitable options for CO₂ absorption from flue gas emitted from the fossil fuel burning.

The most common solvents for CO₂ absorption are simple alkanolamines and sterically hindered amines (Liu et al. 2014; Voice et al. 2012). The most extensively used amine for CO₂ absorption is monoethanolamine (MEA) (Freguia & Rochelle 2003; Hosseinzadeh et al. 2017) due to its number of advantages, such as high reactivity, low solvent cost, reasonable thermal stability, and low degradation rate (Plaza et al. 2010). In a study conducted by Zhang et al. (2009), the application of 30 wt% MEA resulted in 80–95% CO₂ removal from flue gas. However, MEA has a high enthalpy of reaction with CO₂ and a substantial amount of energy is required for CO₂ recovery, making the process economically unfavorable (Luis 2016; Xue et al. 2017). MEA has a relatively low CO₂ loading capacity, which results in a large MEA recirculation rate, large equipment sizes, and high capital cost. It has a relatively high vapor pressure, causing high solvent carryover during the absorption and regeneration steps (Davy 2009; Ho et al. 2008). Furthermore, MEA is highly corrosive and reacts with materials used in the reactor vessels, piping, and other process equipment (Davy 2009). The second most widely used alkanolamine in CO₂ absorption is diethanolamine (DEA) which requires lower energy for the regeneration (Akber Hassan and Jiang 2012; Lee et al. 2013). Like MEA, DEA is also prone to losses and degradation, but to a relatively lower extent.

Besides MEA and DEA, several other solvents such as chilled ammonia, methyl diethanolamine (MDEA), 2-amino-2-methyl-1-propanol (AMP), and piperazine (PZ) have also been examined for efficient CO₂ absorption. Chilled ammonia has been employed for direct application in post-combustion CO₂ capture from power plants as well as in retrofitting of power plants (Darde et al. 2010; Villeneuve et al. 2018). Consequently, this solvent is considered a good candidate for CO₂ capture from flue gases alongside the single and blended alkanolamine solvent systems. The MDEA has used for CO₂ absorption due to its relatively low regeneration energy requirement, low tendency to form degradation products, and low corrosion rates (Saidi 2018), but it has a slow kinetics of adsorption (Naami et al. 2013). AMP is the most common sterically hindered amines for CO₂ absorption (Gabrielsen et al. 2007). It is two orders of magnitude slower in oxidative degradation and more resistant to thermal degradation than MEA (Wang & Jens 2012). The CO₂ loading of AMP can reach a ratio of one to one. PZ is considered a promising solvent compared to MDEA and AMP (Freeman et al. 2010b). This is due to its high CO₂ loading capacity (two moles of CO₂ per one mole of PZ), high reaction rate with CO₂, and high resistance to thermal and oxidative

degradation (Akber Hassan and Jiang 2012; Fredriksen & Jens 2013). Overall, CO₂ capture by different single and blended alkanolamines reported by different authors has been tabulated in Table 1.

The intensive solvent regeneration energy, large absorption towers, higher solvent loss, and degradation of solvent are the major obstacles faced for the commercialization of chemical absorption of CO₂ from fossil fuel-derived flue gases. To overcome these obstacles, economic and efficient solvent systems for CO₂ absorption are urgently needed. Therefore, in this study, detailed process flowsheets were analyzed in Aspen Plus for MEA, DEA, and chilled ammonia for CO₂ absorption. The system performance was evaluated through process simulations by varying process parameters, such as absorber stages and temperatures to maximize CO₂ capture. Based on the certain absorber stages and temperature, later regeneration heating duty and solvent flow rates were identified for maximum CO₂ capture. Later, MEA and DEA were blended with MDEA, AMP, and PZ to enhance CO₂ capture efficiency and to evaluate the solvent loss. The novelty of this study was the systematic evaluation of single and mixed solvents for chemical absorption of CO₂ from the flue gas, whereas similar studies were focused on either single or mixed solvent systems. In addition, a wide range of alkanolamines, for example, primary (MEA), secondary (DEA), and tertiary (MDEA) amines, were investigated alongside the chilled ammonia, AMP, and PZ solvents as single and blended solvents. In contrast, other studies selected only alkanolamines or other solvents for their study, and such a wide range of solvent utilization was not commonly observed. Moreover, a diverse and extensive number of parameters were studied in this study to select the crucial process parameters for CO₂ capture efficiency. Finally, rigorous kinetics between CO₂ and solvent was considered during the model development in Aspen Plus in addition to oxidative and thermal degradation characteristics and solid formation of some blended solvent systems.

Table 1 CO₂ capture efficiency by alkanolamines

Solvent	% of CO ₂ capture	Reference
MEA (30 wt%)	85.0	(Harun et al. 2012)
MEA (30 wt%)	86.5	(Dave et al. 2009)
MDEA (30 wt%)	69–75.8	(Barzagli et al. 2010)
AMP (30 wt%)	63–89	(Aroonwilas & Veawab 2004; Barzagli et al. 2010)
DEA + AMP (30 wt%)	93.0	(Aroonwilas & Veawab 2004)
MEA + AMP (30 wt%)	97.7	(Choi et al. 2009)

Methodology

Flue Gas and Solvent Specifications

The flue gas was assumed to be free from all contaminants and primarily consisted of only CO₂, O₂, and N₂. This assumption was justified by considering the complete combustion of fossil fuels. Flue gas specifications are shown in Table 2. These specifications were used in this study to maintain a fixed CO₂ loading of 0.2 (moles of CO₂/moles of solvent), reported in previous amine-based optimized CO₂ capture studies (Lee et al. 2013; Arachchige and Melaaen 2012). In addition, single solvent systems such as MEA,

Table 2 Flue gas specifications (Lee et al. 2013; Arachchige and Melaaen 2012)

Parameter	Value
Flow rate (kg/h)	600
Temperature (°C)	40
Pressure (bar)	1.1
CO ₂ loading (moles of CO ₂ /moles of solvent)	0.2
Composition (mol %)	
Carbon dioxide (CO ₂)	0.13
Nitrogen (N ₂)	0.8
Oxygen (O ₂)	0.07

Table 3 Solvent specifications and operating, structural, and flow properties of the absorber and desorber column

Solvent	MEA	DEA	Chilled ammonia
Parameter			
Makeup solvent flow rate (kg/h)	20,000	20,000	40,000
Temperature (°C)	40	40	5
Pressure (bar)	1.1	1.1	1.1
Composition (X _S ^a , mol fraction)	0.3	0.3	0.5
Absorber column parameter			
Temperature (°C)	25	25	5
Pressure (bar)	1.1	1.1	1.1
Number of stages	10	10	10
Modeling approach	Equilibrium	Equilibrium	Equilibrium
Condenser	No	No	No
Reboiler	No	No	No
Flow pattern	Counter current	Counter current	Counter current
Desorber column parameter			
Temperature (°C)	120	120	110
Pressure (bar)	1.1	1.1	33
Number of stages	23	23	12
Modeling approach	Equilibrium	Equilibrium	Equilibrium
Condenser	No	No	No
Reboiler	Kettle type	Kettle type	Thermosiphon
Flow pattern	Counter current	Counter current	Counter current

^aMEA, DEA, and chilled ammonia mole fraction in the solvent

DEA, and chilled ammonia specifications are tabulated in Table 3. Moreover, amine-based solvent concentration in water was considered up to 30 mol%. A higher mole percentage was avoided in this study to avoid corrosion in process equipment. Flue gas-derived CO₂ capture process flowsheet was developed in Aspen Plus V8.8 using the inbuilt thermodynamic property package electrolyte-NRTL. The electrolyte-NRTL property method considers the non-ideality of the solvents and the interactions between different components of the flue gas. This property package can adequately represent the phase behavior and thermodynamic properties of amine solvents in the CO₂ capture processes. In addition, solids were defined as non-conventional components with the relevant ultimate and proximate analyses available in the literature. Then, suitable property models (inside the property methods) such as HCOALGEN and DCOALIGT were chosen to calculate the properties of solids, for example, enthalpy of formation, heat capacity, and density based on the ultimate and proximate analyses.

CO₂ Capture Process Flowsheet Development

A process flowsheet was developed for CO₂ absorption using MEA as a solvent. The system consisted of absorption, desorption, solvent regeneration, and CO₂ purification (Fig. 1). The absorption column (ABSORBER) was designed by a 10-staged tray column following the equilibrium modeling

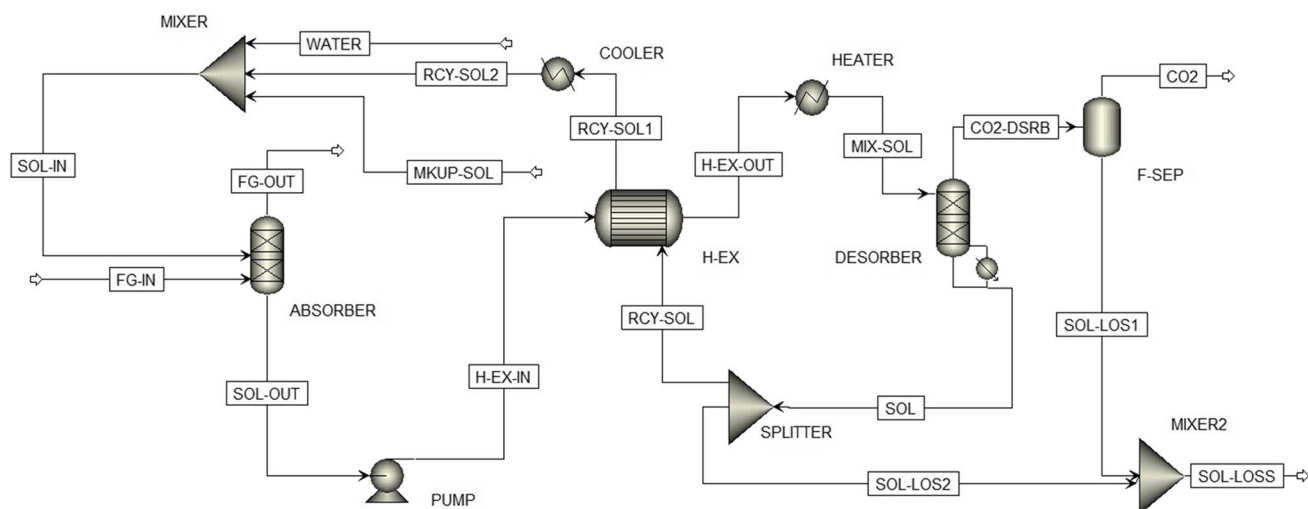
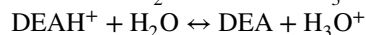
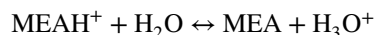


Fig. 1 Process flow diagram of MEA/DEA solvent system

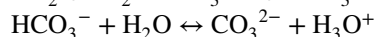
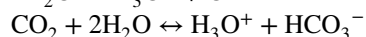
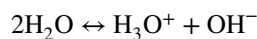
approach. The flue gas (FG-IN) was fed into the absorption column (ABSORBER) from tray 10 (bottom), while the MEA solvent solution (SOL-IN) entered tray 1 (top) of the column. Flue gas and MEA solvent contacted a counter current manner inside the absorption column to remove CO₂ without using any condenser and reboiler in this column. Table 3 shows the other operating conditions (temperature and pressure) of the column. From the top of the absorption column, the scrubbed gas (FG-OUT), containing mostly nitrogen gas and a very small amount of CO₂, was vented to the atmosphere. The MEA solvent (SOL-OUT), rich in CO₂ and exiting from the bottom of the column, passed through several heat exchangers to reach 120 °C and was then pumped to the top of a desorber column (DESORBER). The desorber column was also modeled by a 23-staged tray column following the equilibrium modeling approach. A kettle type reboiler was used in this column for solvent regeneration with a bottom to feed ratio of 0.9 mol, and other operating parameters and column properties are shown in Table 3. In the desorber column, CO₂ gas (CO₂-DSRB) was separated and collected from the top tray 1, and thus, MEA solvent (SOL) was regenerated and exited from the bottom tray numbered 23. The regenerated solvent was then splitted using a two-way flow splitter, and 10% of the solvent was purged (SOL-LOS2) to avoid the accumulation of oxidative and thermal degradation products of the solvent (Dumée et al. 2012; Jones 2018; Nuchitprasittichai & Cremaschi 2013). In addition, a flash separator (F-SEP) at 0.15 bar pressure was used to separate pure CO₂ gas (CO₂) and carried over solvent (SOL-LOS1) from CO₂-DSRB stream. Both SOL-LOS1 and SOL-LOS2 were mixed in a mixer (MIXER2) to be counted as a total solvent loss from the process. The remaining 90% of the regenerated solvent (RCY-SOL2) after the splitter was recycled to the absorber

through heat exchanging and mixing with fresh makeup MEA solvent (MKUP-SOL) for the next round of CO₂ capture process. The same process flowsheet was used, while DEA was used as the CO₂ absorption solvent. The crucial information about the streams is given in Supplementary Table 1. Moreover, the following equilibrium reactions were used in this study to model the CO₂ absorption into MEA and solvents (Austgen et al. 1989; Haghtalab & Dehghani Tafti 2007):

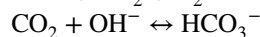
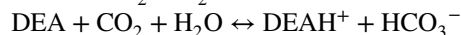
Dissociation of MEA and DEA after the protonation



Dissociation of water, CO₂, and bicarbonate (HCO₃⁻)



Kinetic reactions used are as follows based on (Austgen et al. 1989; Haghtalab & Dehghani Tafti 2007):



In the case of chilled ammonia, flue gas (FG) was compressed to 1.7 bar and then passed through a direct contact cooler (DCC) where it was cooled to around 25 °C at a 12-staged column operated at 1.1 bar. After that, the compressed flue gas (COMP-FG) and cold water (CW-IN) at 25 °C were passed in a counter current manner inside the DCC. The overall process flow diagram is shown in Fig. 2. The flue gas left from the top of the DCC (COLD-FG) and later passed through the chiller to reduce its temperature to around

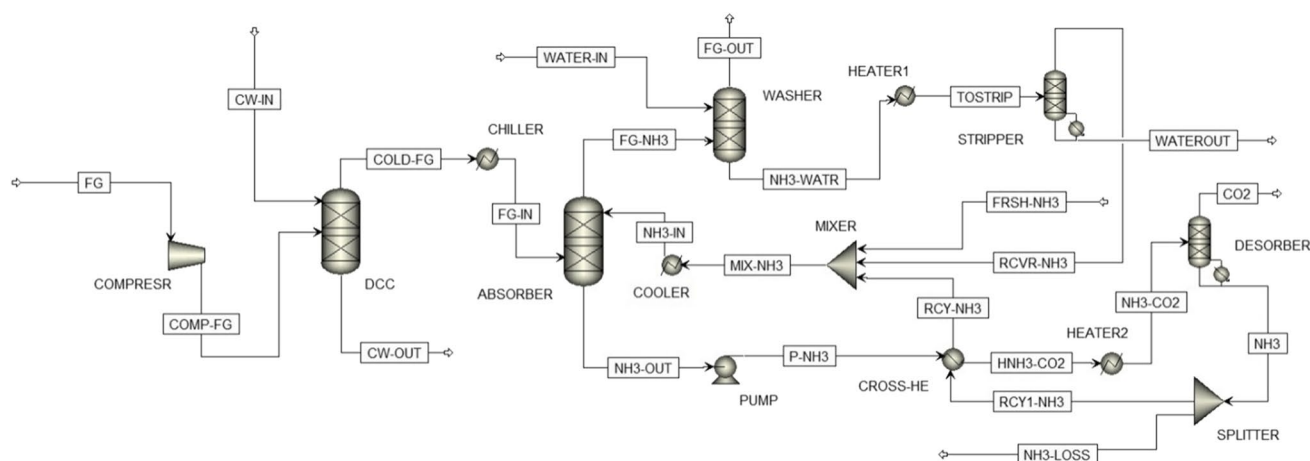


Fig. 2 Process flow diagram of the chilled ammonia system

5 °C before entering to the bottom of the absorber column (ABSORBER). The absorber column was a 10-staged tray column where chilled ammonia and flue gas were entered into the column at trays 1 and 10 correspondingly and contacted in a counter current manner. Table 3 shows the other operating, structural, and flow properties of this tray column for CO₂ capture using chilled ammonia. Chilled ammonia (NH₃-IN) at 5 °C was entered at the top of the absorber. The absorption of CO₂ into the ammonia solution was mainly through the precipitation of ammonium bicarbonate. After the desired degree of CO₂ absorption, the rich NH₃ solvent (NH₃-OUT) exits from the bottom of the absorber, while the CO₂ stripped flue gas (FG-NH₃) exits from the top of the absorber. The vent gas (FG-NH₃) from the top of the absorber had an ammonia content ranging from 500 to 3000 ppm that was subjected to an ammonia recovery system, designed with an absorption and desorption cycle at 1.1 bar pressure. The vent gas entered at the bottom of the wash absorber (WASHER) and washed with water at 25 °C (WATER-IN) that flowed from the top. Ammonia was absorbed into the aqueous solution, and the vent gas containing less than 10 ppm ammonia (FG-OUT) was released from the top of the wash absorber. The aqueous solution containing ammonia (NH₃-WATR) exited from the bottom of the absorber and flowed to the STRIPPER. The temperature of the stripper was maintained at 40 °C, and ammonia was stripped from the aqueous solution and recycled (RCVR-NH₃). The CO₂-absorbed NH₃ solution (NH₃-OUT) was left from the bottom of the absorber and passed through a cross-heat exchanger (CROSS-HE) to heat up the solution as well as heat recovery from the hot RCY1-NH₃ stream (about 160 °C). Then, the CO₂-rich stream (HNH₃-CO₂) was further heated to 80 °C and later entered the top of the desorber. The desorber was a 12-staged tray column, and operating pressure was 33 bar, and the temperature in the

reboiler (thermosiphon type) was in the range of 110–160 °C (other properties of this column is shown in Table 3). The CO₂ lean NH₃ solution (NH₃) that left from the bottom of the desorber was split in a two-way flow splitter. Ninety percent of the lean NH₃ solution was sent back to the crossflow heat exchanger where it was cooled. It was then cooled in COOLER after mixing with makeup and fresh NH₃ solvent (FRSH-NH₃) to a temperature of 5 °C before being returned to the absorber. A 10% lean ammonia solution split from the flow splitter accounted for the solvent loss to avoid an accumulation of contaminants within the CO₂ capture process model. Crucial information on the important streams of this model is given in Supplementary Table 2.

Process flowsheets developed for single MEA and DEA solvent systems were used for MEA- and DEA-based blended solvents with changing the solvent composition. Instead of maintaining 30 mol% MEA or DEA solvent, other solvents such as MDEA, AMP, and PZ are mixed with different mole percentages according to Table 4. Blended solvents were mainly used to increase the thermodynamic efficiency of MEA and DEA solvents through the formation of more HCO₃⁻ by other tertiary/sterically hindered amine solvents such as MDEA, AMP, and

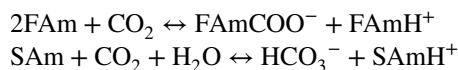
Table 4 Blended solvent compositions

% MEA + % other solvent ^b		% DEA + % other solvent ^c
25 +	5.0	25 + 5.0
20 +	10	20 + 10
15 +	15	15 + 15
10 +	20	10 + 20
5.0 +	25	5.0 + 25

^bMDEA, AMP, PZ, and DEA

^cMDEA, AMP, and PZ

PZ. Increased thermodynamic efficiency can reduce the degradation rate of single amine solvents (MEA and DEA) as well as promote higher and faster CO₂ absorption by blended solvents. In a blended solvent, a single primary (MEA) or secondary (DEA) amine solvent acted as fast-reacting amine (Fam) for carbamate ion (FamCOO⁻) and proton (H⁺) generation. In contrast, added tertiary or sterically hindered amine (SAm) acted as a proton acceptor for increased HCO₃⁻ formation following a slow reaction kinetics. The mixture interaction of blended solvent components can be presented by the following general reaction scheme for improved CO₂ capture efficiency.



Sensitivity Analyses

The effect of number of absorber stages on CO₂ capture was investigated through sensitivity analysis. Since CO₂ capture is temperature sensitive, proper absorber temperature was also investigated in another sensitivity analysis. After that absorber stages and temperature of the previous sensitivity analyses were used to find out the required single and blended solvent flow rate to maximize CO₂ capture efficiency. However, to maintain the solvent's CO₂ capture efficiency at the desired level and avoid impurities buildup within the process, a certain amount of solvent was discharged from the process. Oxidative degradation at the absorption column and thermal degradation at the regeneration column combinedly generated alkanolamine degradation products such as hydroxyethyl oxazolidone, bis-hydroxyethyl piperazine, and tris-hydroxyethyl ethylenediamine and other low-boiling point products at lower concentrations. Those degradation products could promote corrosion, foaming, and viscosity increase and ultimately reduce the CO₂ loading to the alkanolamine solvents. Thus, a certain fraction of solvent was purged, termed solvent loss, to maintain the CO₂ capture efficiency of alkanolamine solvents. The amount of solvent loss was also studied through another sensitivity analysis in the case of blended solvent. Moreover, heat was supplied in the desorber column's reboiler during solvent regeneration, and this heating duty was the major source of energy penalty for each solvent system. Therefore, reboiler heating duty requirement was studied against the single solvent flow rate.

Results and Discussion

Effect of Process Parameter for Single Solvent System

The process simulation is performed for MEA, DEA, and chilled ammonia using initial process parameter values listed

in Tables 2 and 3. The number of stages in the absorber and absorber temperature were selected first since those parameters had the highest impact on CO₂ capture compared to other process parameters (Nuchitprasittichai & Cremaschi 2011). And then the process flowsheets were adjusted with the specific number of stages and temperature to evaluate the effect of solvent flow rates and reboiler duty on CO₂ capture.

Stages of Absorber

The effect of absorber stages on CO₂ capture was investigated by performing the alkanolamines (MEA and DEA) solvent-based simulation at 25 °C absorber temperature. This temperature was maintained at 5 °C in the case of chilled ammonia solvent-based process model. In addition, 20,000 and 40,000 kg/h of alkanolamine and chilled ammonia solvent flow rates were used correspondingly in the effect of absorber stages investigation. Figure 3 shows the outcome of CO₂ capture rate with the gradual change of number of absorber stages for MEA, DEA, and chilled ammonia solvent-based process models. For all three solvent systems, the CO₂ capture efficiency gradually increased up to 15 absorber stages. An absorber stages higher than 15, CO₂ capture efficiency remained constant at 2.3 kmol/h for MEA and DEA solvents, whereas it reduced sharply to below 1 kmol/h in chilled ammonia solvent. Chakma et al. (1995) used 15 absorber stages in their comparative study between MEA and DEA to find out suitable solvents for CO₂ capture and storage. Conversely, 60–66% CO₂ removal was reported in a 12-staged absorber column using MEA solvent (Plaza et al. 2010), but a higher removal percentage (84%) was recorded with the increased number of stages (20) at 14 wt% of MEA solvent (Nuchitprasittichai & Cremaschi 2011). The later study also showed an almost similar percentage of CO₂ removal by 39 wt% of DEA in a 32 staged absorber column.

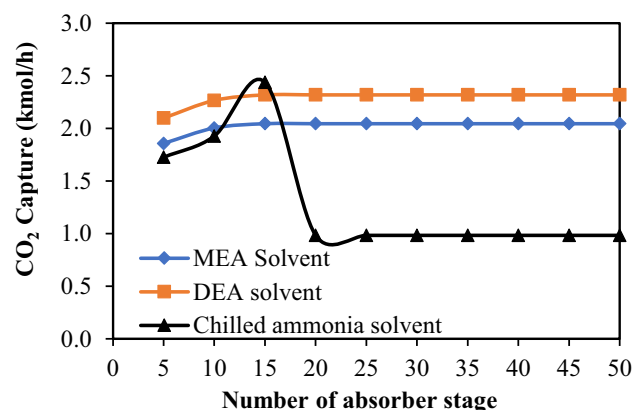


Fig. 3 Effect of absorber stage on CO₂ capture rate

Absorber Temperature

This sensitivity analysis was run with the 15 absorber stages and previously mentioned solvent flow rates (Absorber Temperature section) to evaluate the CO₂ capture rate at varying absorber temperature. Figure 4 shows that CO₂ capture rate by the DEA solvent was always higher relative to MEA and chilled ammonia solvent in the considered temperature range (2–50 °C). However, the CO₂ capture rate was not significantly affected below the 30 °C absorber temperature for DEA, but MEA and chilled ammonia-based capture rate remained unaffected until 25 and 10 °C absorber temperature respectively. Above these absorber temperatures, the CO₂ capture rate was decreased due to increased solvent evaporation and degradation. Abu-Zahra et al. (2007) reported a higher absorber temperature and resulted a higher heating energy requirement in the desorber column during solvent regeneration. Moreover, approximately 60 °C absorber temperature can decompose the NH₄HCO₃ crystal formed for CO₂ capture which further reinforced to maintain a lower absorber temperature (5–10 °C) for chilled ammonia solvent (Yeh et al. 2005). In addition, a comparatively lower absorber temperature (5–10 °C) reportedly increased almost

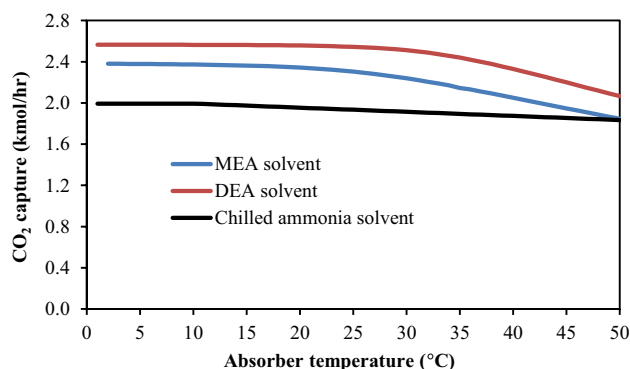


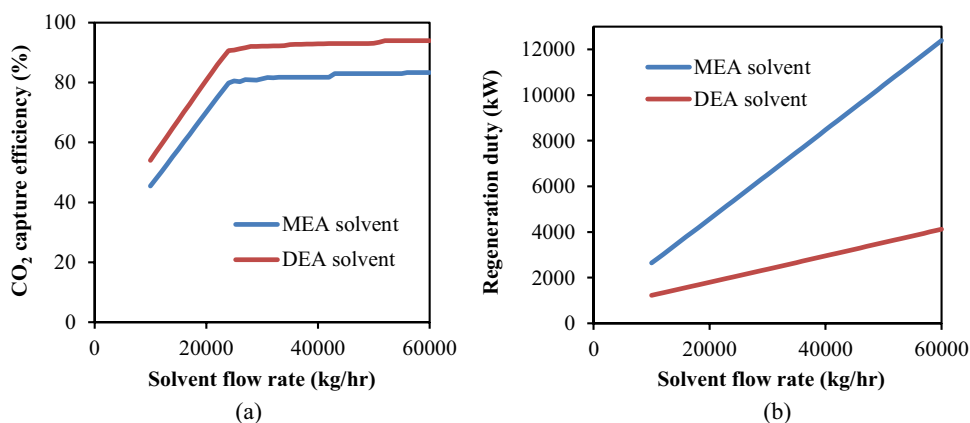
Fig. 4 Effect of absorber temperature on CO₂ capture

11% CO₂ capture efficiency than a higher temperature (40 °C) by the chilled ammonia solvent (Yeh et al. 2005). In contrast, 25–30 °C absorber temperature was maintained in alkanolamine solvents for a higher CO₂ capture rate in this study.

Solvent Flow Rate and Regeneration Duty

The effect of solvent flow rate on regeneration duty requirement and CO₂ capture efficiency was carried out in an absorber with 15 stages. Previous sensitivity analyses in "Solvent Flow Rate and Regeneration Duty" Section also showed that 25–30 °C and 5 °C absorber temperatures maximized the CO₂ capture by alkanolamine and chilled ammonia solvents accordingly. Thus, those absorber stages and temperatures were maintained to study the sensitivity analysis of solvent flow rate over the CO₂ capture efficiency and regeneration duty requirement. Figure 5a shows that CO₂ capture efficiency increased sharply with the increased solvent flow rate up to 24,000 kg/h and then remained almost constant. However, CO₂ capture efficiency was always higher with DEA compared to MEA. For instance, 10% higher CO₂ capture efficiency was recorded for DEA compared to other solvents at 24,000 kg/h flow rate. And this higher CO₂ capture efficiency for DEA was resulted at the lowest regeneration duty requirement which was 2024.87 kW (Fig. 5b). At the same point, 2.64 times higher regeneration duty was required for the MEA. It was observed that regeneration duty was upsurged linearly with the gradual solvent flow rate increment in case of both MEA and DEA (Fig. 5b). However, a higher regeneration duty (3.8 GJ/ton CO₂) was recorded at the desorber column in a study carried out by Mangalapally and Hasse (2011) to capture 90% CO₂ from 0.141 kmol/h CO₂ stream by using 199.46 kg/h alkanolamines solvent flow rate. Nuchitprasittichai and Cremaschi (2011) reported a 62,000 kg/h MEA flow rate for 84% CO₂ capture, whereas 39,000 kg/h DEA flow rate was required for 83% CO₂ capture from flue gas. In the same study, 5038

Fig. 5 Effect of solvent flow rate on (a) CO₂ capture efficiency and (b) regeneration heating duty of desorber column in MEA and DEA solvent system



kW and 2554 kW regeneration heating duty was reported for MEA and DEA process model correspondingly.

In the case of chilled ammonia, the highest chilling load was maintained in the absorber column with the aim of removing exothermic heat of reaction between ammonia and CO₂. In addition, heat produced by the exothermic precipitation reaction of ammonium bicarbonate was also removed from the absorber. However, even after maintaining the highest chilling load, lower CO₂ capture efficiencies were recorded for chilled ammonia (Fig. 6a) than both MEA and DEA. Only about 61% CO₂ capture efficiency was achieved through 24,000 kg/h chilled ammonia flow rate, while maximum 80% and 90% CO₂ capture efficiencies were achieved for both MEA and DEA respectively at the same flow rate. Later, chilled ammonia system reached maximum around 93% CO₂ capture efficiency at 111,200 kg/h flow rate. This flow rate was 4.63 times higher than MEA and DEA flow rate to reach maximum CO₂ absorption efficiency. Figure 6b shows that higher chilled ammonia flow rate resulted in a higher amount of regeneration duty. Similar to MEA and DEA, chilled ammonia solvent regeneration heating duty increased linearly with solvent flow rate and required 50,091.14 kW heating duty for maximum 93% CO₂ capture efficiency which was almost 50% lower than the regeneration duty (energy) reported by relevant previous studies (Mathias et al. 2009, 2010).

CO₂ Capture Efficiency of Blended Solvent System

Using a single solvent, 80–93% CO₂ capture efficiency was achieved by alkanolamines and chilled ammonia solvents. The next part of the study investigated the potential of using mixed solvents to further increase CO₂ capture efficiency. In this investigation, MEA and DEA were blended with other solvents: MDEA, AMP, and PZ. Although chilled ammonia resulted in the highest CO₂ capture efficiency (93%), chilled ammonia was not used in blended solvents due to higher solvent and regeneration duty requirements compared to MEA and DEA.

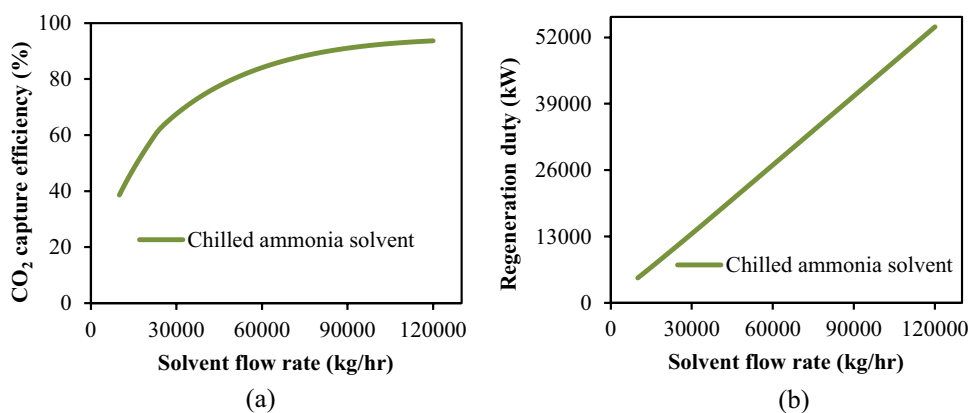
MEA-Based Blended Solvent

MEA-based blended solvents are prepared following a composition outlined in Table 5. A 0–30 wt% of other solvents were mixed with MEA for blended solvent preparation. The outcomes of prior sensitivity analyses were used in the blended solvent process model development; a 15-staged absorber column, 25 °C absorber temperature, and 24,000 kg/h of solvent flow rate were used. The CO₂ capture efficiencies of MEA blended solvents are summarized in Table 5. It can be observed that a single MEA solvent had about 80% CO₂ capture efficiency. Mixing MEA with MDEA, CO₂ capture efficiency was reduced significantly to 62.73% with increasing to 30% MDEA. Blending MEA with AMP also showed a downward CO₂ capture efficiency. However, although PZ blended MEA solvents initially maintained lower CO₂ capture efficiency almost the same as pure MEA, later these blended solvents increased the CO₂ capture efficiency. The lowest 76.59% CO₂ capture efficiency was observed at 10% MEA with 20% AMP, while 78.46% efficiency was recorded at 15% PZ with the same proportion of MEA. Later, 30% PZ increased the CO₂ capture efficiency to 81%. This increased efficiency with PZ solvent is attributed to having two secondary amines sites in PZ. On the contrary, decreased CO₂ capture efficiency of the AMP solvent could be the reason for having two alkyl groups compared to

Table 5 CO₂ capture efficiency for MEA-based blended system

Solvent composition %MEA + % other solvent	% CO ₂ capture efficiency			
	MDEA	AMP	PZ	DEA
30+0	79.82	79.82	79.82	79.82
25+5	74.77	78.27	79.04	86.4
20+10	70.48	77.21	78.46	88.51
15+15	67.36	76.67	78.46	88.97
10+20	65.03	76.59	78.89	89.29
5+25	63.59	77.06	79.82	89.56
0+30	62.73	78.03	81.00	90.34

Fig. 6 Effect of solvent flow rate on (a) CO₂ capture efficiency and (b) regeneration heating duty of desorber column in chilled ammonia solvent system



the MEA. Similarly, being a tertiary amine, comparatively MDEA had a lower number of N–H bonds which resulted ultimately lower CO₂ capture efficiency than pure MEA. Similar type of downward outcome was observed by Zhu et al. (2012) in the case of CO₂ capture efficiency, while both MDEA and AMP solvent were blended with MEA. However, the same study reported increased CO₂ capture by MEA blended PZ solvent. Blending MEA with DEA also increased the CO₂ capture efficiency in this study (Table 5).

DEA-Based Blended Solvent

Similar to MEA blends, DEA-based blended solvent process models are developed based on Table 6 and previous sensitivity analyses. Only a higher absorber temperature (30 °C) was used in this case. Table 6 illustrates the CO₂ capture efficiencies of various DEA-based blended solvents. Single DEA solvent had 90.34% CO₂ capture efficiency (Table 5) which was increased by blending with other solvents. Table 6 shows that 25% DEA blended with 5% of other solvents such as MDEA, AMP, and PZ and increased the CO₂ capture efficiency up to more than 99%. Better performance in terms of regeneration duty, solvent flow rate, and cyclic loading in DEA and AMP blended solvent systems was also reported by Adeosun and Abu-Zahra (2013b). MDEA, AMP, and PZ proportions higher than 5% in blended solvent decreased the CO₂ capture efficiency, and the decreasing rate was higher when MDEA was mixed with DEA. The downward trend for CO₂ capture efficiency could have resulted from the structural property of MDEA and lower partial pressure of CO₂ in the inlet flue gas (Adeosun & Abu-Zahra 2013a). The tendency of CO₂ capture efficiency observed in this study by DEA-based PZ blends was also found in previous studies (Adeosun & Abu-Zahra 2013b; Rinprasertmeechai et al. 2012). Adeosun and Abu-Zahra (2013a) and Rinprasertmeechai et al. (2012) showed that both CO₂ capture capacity and rate had been significantly improved at low PZ fraction and reduced at high PZ fraction. A lower fraction of PZ such as 5–10% kinetically activated the DEA, resulting in higher CO₂ capture. However, a

higher proportion of PZ than 5–10% had limited solubility in the DEA and subsequently precipitated on the absorption bed. As a result, CO₂ capture gradually decreased with the increased percentages of PZ.

Solvent Loss for Blended Solvent

DEA-based blended solvent showed better CO₂ capture efficiency compared to MEA-based blends. Thus, DEA blended solvents were evaluated to find out the solvent losses during the regeneration. Figure 7 shows the solvent losses of different DEA-mixed MEA, MDEA, AMP, and PZ blended solvents. Solvent loss of a single DEA solvent was 57.61% which was reduced to 50%, while 5 wt% of other solvents were blended with DEA which had a good agreement with the experimental solvent losses (50–56%) reported in previous studies (Vevelstad et al. 2013). In contrast, 5% MEA mixed with DEA increased the solvent loss to 67.08%, and then, the solvent loss was decreased with the increasing MEA fraction. In the case of other solvents, for example, MDEA, AMP, and PZ, the solvent loss was not significantly varied with their increasing fraction (> 5 wt%) in the blended mixture. Oxidative degradation of solvent was reportedly one of the major causes of solvent loss which can be accelerated by metal, sulfur, or chlorine impurities in the flue gas (Goff & Rochelle 2004; Huang et al. 2014). However, the flue gas considered in this study was assumed free from those contaminants which reduced the contribution of oxidative degradation to the solvent loss. Additionally, stripper temperature was kept at 120 °C to reduce the thermal degradation since it was reported that DEA blended solvents were liable to degradation around 140–150 °C (Du et al. 2013; Li et al. 2013). Furthermore, solvent concentration for both pure and blended solvents was kept at 30 wt% to avoid the thermal degradation caused by higher concentrations (40–50 wt%) of solvents (Bougie & Iliuta 2014). All together, those operational parameters reduced the solvent

Table 6 CO₂ capture efficiency for DEA-based blended system

Solvent composition %DEA + % other solvent	% CO ₂ capture efficiency		
	MDEA	AMP	PZ
30 + 0	90.69	90.69	90.69
25 + 5	99.29	99.70	99.68
20 + 10	97.78	98.90	98.90
15 + 15	93.06	97.74	98.13
10 + 20	86.83	95.40	96.18
5 + 25	77.49	90.34	92.28
0 + 30	62.69	77.88	80.99

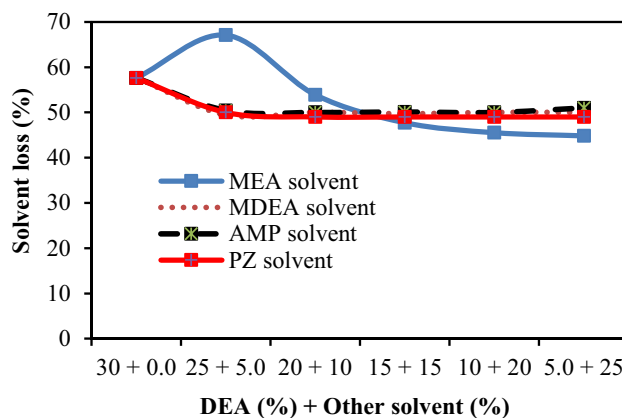


Fig. 7 Solvent loss of DEA-based blended solvent system

loss in this study. The solvent loss shown in Fig. 7 could have resulted from the higher evaporation rate due to the higher partial pressure of blended solvents.

Conclusion

In this study, details of process flowsheets were developed for both single alkanolamines (MEA and DEA) and chilled ammonia to evaluate the CO₂ capture efficiency, and later findings were applied to the blended solvent systems.

- i. The absorber columns with more than 15 stages did not have a significant effect on CO₂ capture efficiency.
- ii. For maximum CO₂ capture efficiency, the absorber temperature was 25–30 °C using alkanolamine as absorbing solvent, whereas for chilled ammonia, it was 5 °C.
- iii. A 24,000 kg/h of solvent flow rate captured the maximum 80% and 90% of CO₂ by MEA and DEA, respectively, at the lowest possible regeneration duty at the desorber column.
- iv. DEA required 3.37 kW kg⁻¹ h⁻¹ or 12.15 GJ ton⁻¹ of regeneration duty compared to 3.37 kW kg⁻¹ h⁻¹ or 12.15 GJ ton⁻¹ of regeneration duty by MEA at above CO₂ capture efficiencies.
- v. A maximum 93% CO₂ capture efficiency was possible to achieve by using 6.43 times more chilled ammonia flow rate which needed 24.74 times more regeneration duty compared to DEA. Therefore, to avoid higher energy (duty) penalty at the desorber column and anticipated higher capital cost to handle large solvent flow rate, chilled ammonia was not considered as an economic process for CO₂ capture in the industrial level.
- vi. The MEA-based blended solvent did not show any significant effect to increase the CO₂ capture efficiency.
- vii. 25 wt% DEA blended with 5 wt% of MDEA, AMP, and PZ solvents significantly increased the CO₂ capture efficiency (> 99%) at reduced solvent loss.

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Author Contribution All authors contributed to the study's conception and design. Material preparation, data collection, and analysis were performed by Md Shahadat Hossain, Md. Salatul Islam Mozumder, and Deepak Kumar. The first draft of the manuscript was written by Md Shahadat Hossain, and all authors commented on previous versions of the manuscript. Deepak Kumar edited the language, and all authors read and approved the final manuscript for publication.

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Data Availability Manuscript has data included as supplementary material.

Declarations

Ethics Approval Not applicable.

Competing Interests The authors declare no competing interests.

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