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Impact of blends of aqueous amines on absorber intercooling for post combustion CO₂ capture system

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Abstract

Climate change is the biggest challenge of this century due to the global consequences of human activities on the ecosystem resulting in global warming. The emissions of greenhouse gases, mainly CO₂ from the combustion of fossil fuels in the power plant is the main cause of global warming and to mitigate these emissions is the foremost challenge. Nowadays, the most preferred method is post combustion chemical absorption using amine-based solvents. However, high energy requirements for this method restrict its deployment. An efficient approach used for the reduction of the high energy requirement of post combustion CO2 capture process was absorber intercooling. Therefore, this research evaluates the effect of two configurations of intercooled absorber such as "simple" and "advanced" intercoolers for CO2 capture integrated with natural gas combined cycle power plant using aqueous alkanolamines, such as 30 wt.% monoethanolamine and 50 wt.% methyl-diethanolamine and their blends. For pure methyldiethanolamine case, at lean loading 0.01 intercooling configurations; simple and advanced shows the highest reduction of 21.01% and 22.82% in the specific reboiler duty, respectively in comparison to other blends at the expense of highest liquid solvent flow rate. Simple and advanced intercooling configurations shows optimum results for the case with 40% monoethanolamine and 60% methyl-diethanolamine in a blend with decrease of 9.19% and 17.28% in solvent flow rate and a decrease of 9.42% and 16.83% in specific reboiler duty required for 90% CO₂ capture rate, respectively. For pure monoethanolamine case at lean loading 0.2 absorber intercooling does not offer significant results.

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Keywords

Post-combustion CO₂ capture, absorber intercooling, MEA, MDEA

Introduction

The world's greatest challenge in the present era is climate change leading to global warming triggered by enlarged greenhouse gas emissions. Furthermore, the continuing buildup of greenhouse gas (GHG) concentrations into the atmosphere from several anthropogenic sources is a worldwide concern. Combustion of fossil fuels in power stations for energy production is the prime source of acid gas emissions and is anticipated to remain the primary energy source for the coming years. The report of the Intergovernmental Panel on Climate Change (IPCC) reveals that since 1880, the atmospheric conditions warmed with an average temperature increase of 0.85 (0.65–1.06)°C. The IPCC has identified six gasses with the potential for climate change, including CO₂, CH₄, N₂O, SF₆, CFC's and HFC's. However, the main greenhouse gas is CO₂ and its discharge from fossil fuel power plants and other human activities to the atmosphere has a drastic effect on global climate change. As the International Energy Agency² has mentioned, power generation from the combustion of fossil fuels leads roughly one-third of total atmospheric CO₂ emissions. However, fossil fuel fired power plants played an important role in meeting worldwide energy requirements.³ Coal is the major source of power production in non-OECD Asian countries, particularly China, India, and Pakistan. In the power sector, the massive usage of coal without any CO₂ emission regulations severely affects the atmospheric conditions. The recent study by International Energy Agency⁴ further indicated that the amount of CO₂ in the atmosphere will rise to an average concentration of 410 parts per million (ppm) in 2020 and higher in coming years, which was about 280 ppm during pre-industrial period. In the coming years, the foremost global challenge will be to mitigate CO₂ emissions. Consequently, over the past few years, the significance of using CO₂ emission control methods from energy stations has gradually increased. The CO₂ mitigation methods are needed to keep the CO₂ concentration in the atmosphere at a particular rate. In this context, carbon capture and storage (CCS) is a promising technology for alleviating atmospheric CO₂ emissions and minimizing the global warming effect.

The process of CCS is a range of techniques involving CO₂ waste extraction from various industrial sources and power plants through the capture and compression process followed by transportation of CO₂ to other industrial processes using CO₂ as a feed, storing it in oceans and geological formations to inhibit the climate change. In addition, the major techniques employed for carbon capture include; pre-combustion, post-combustion, and oxy-fuel combustion. ^{5,6} Among the various CO₂ capture techniques, the most established technique is the post combustion CO₂ capture using mono ethanol amine as a base solvent and it has already been established as given in the open literature. ^{3,7–14} PCC shows various advantages over the other two methods due to its high CO₂ purity and this can be easily implemented in the existing combustion plants without any substantial alteration, keeping in mind the retrofit options prevails for an existing power plant. ³

Post combustion CO₂ capture can be achieved by multiple separation techniques such as physical or chemical absorption into a solvent, solid adsorption, membrane, or other methods of biological or physical separation.^{3,12,15} Among the multiple techniques proposed, the main challenge is to develop such integrated alternatives with low energy demand and a high CO₂ capture ratio. For post-combustion CO₂ capture using alkanolamine, an extensive range of alkanolamine like monoethanolamine (MEA), di-2-propanolamine (DIPA), methyldiethanolamine (MDEA), and diethanolamine (DEA) exists, and many of which have been used for years in industrial applications. However, there is variation in their efficiency for CO₂ capture using packed bed systems. CO₂ absorption rate or reactivity of amine solvent with acid gas, absorption capacity, and required regeneration energy for amine solvent in the stripper column are the major differences of these solvents. Primary alkanolamine, MEA and secondary alkanolamine, DEA have a high reaction rate with acid gases and removal rate is faster while tertiary alkanolamine MDEA reactivity with CO₂ is low and requires higher time for higher CO₂ absorption. Tertiary amine MDEA has a higher loading capacity with less corrosion rate in comparison to MEA and DEA. Primary and secondary amine such as MEA and DEA requires higher solvent regeneration energy in comparison with tertiary amine MDEA.¹⁶

The process of PCC using alkanolamines includes chemical reactions and consists mainly of three stages. The first stage is the cooling and compression of flue gas. The second stage is the absorption of acid gas in the solvent and the regeneration of the solvent. The third and final stage would be the compression of the CO₂. ¹⁷ The absorption-desorption method is of particular interest owing to its prospect of continuous regeneration of the amine-based aqueous solution in a closed cycle. However, the most important restrictions to the execution of post-combustion method of CO₂ capture using alkanolamines is its high energy consumption which decreases the overall power output and available heat. Around 20-40% power reduction of a power station is inevitable due to the high energy demands in the CO₂ capture, solvent regeneration, and compression unit.¹⁸ The power plant usually provides the energy required in the form of steam and the electricity for the CO₂ capture process. Steam is needed in the reboiler while electricity is required to operate shafts of the blower, compressors, and pumps. Consequently, at the expense of power plants thermo-electrical efficiency, the CO₂ capture process is carried out which results in reduction of the overall plant output of power and available heat, thereby encouraging the inevitability of developing the removal process with lower energy requirements. In view of recent global research studies, the three main fields being studied to minimize the required solvent regeneration energy for PCC using aqueous amines are; novel solvent development with better results than 30 wt. % MEA, optimization of the overall PCC process including process design modification and optimal integration.¹⁹ The two main ways of reducing the energy penalty of solvent based PCC are novel solvents with low regeneration energy and improved process design.²⁰

To reduce the required solvent regeneration energy, several studies have been performed to discover solvents with better characteristics than MEA. ^{21,22} Primary and secondary amines (MEA, DEA) generally have a high rate of reaction with acid gas CO₂ and consume a large amount of regeneration energy due to the high heat of reaction connected with carbamate formation. However, corrosiveness²³ and solvent degradation in the presence of other gases such as SOx, NOx, and O₂²⁴ are the other major drawbacks of MEA. MDEA a tertiary amine having high capacity for CO₂ absorption and low energy requirement for regeneration as there is no carbamate formation. ¹⁶ Therefore, to decrease the regeneration energy, the solution can be used as a blend of different amines. ²¹ Blended amines will optimize the desired characteristics as well as improve the drawbacks of

single amine specific to CO₂ absorption rate and solvent regeneration energy. Thus, with a mixture of primary amine MEA or secondary amine DEA with tertiary amine MDEA, ^{23,25} the removal of bulk CO₂ with less regeneration energy will be possible. Moreover, better thermodynamic behavior²⁶ and the mitigation of other problems due to corrosiveness and solvent degradation are the other key advantages of amine blends. Tertiary amine advantages, such as high absorption capacity and low regeneration energy when coupled with the advantages of primary amine, such as fast reaction kinetics are an ideal combination for post combustion CO₂ capture. MDEA is a more stable solvent with low degradation in comparison to MEA which is more susceptible to degradation.²⁷ Consequently, the blend of these amines will decrease the environmental impacts due to the lower emissions of degradation products to the atmosphere.

A reversible reaction between acid gas CO₂ and alkanolamine based solvent takes place in PCC by chemical absorption. In such methods, there are various energy saving methodologies: by decreasing the overall loads of cooling and heating, by using efficient coolants or heating sources to improve temperature levels or a combination of both.²⁸ A wide range of possible configuration schemes has been suggested by Le Moullec et al.²⁹ for the improvement of overall process efficiency and reduction of the total energy consumption.

Modifications such as intercooling can be efficient in minimizing the energy requirement of the PCC process.²² Several research studies have evaluated the effects of absorber intercooling on energy demand for PCC.^{22,28,30–32} The intercooled absorption column increases the rate of absorption and lowers the energy required for solvent regeneration resulting in a decrease of specific reboiler duty. A few studies have also been carried out to analyze the optimum conditions in intercooled absorbers or to identify optimum process conditions under which intercooled absorber will be most efficient.^{22,30,31}

The effects of intercooling on energy consumption was studied by Karimi et al.²² by using two amine solvents; MEA and DEA. The study also stated the performance of intercooling by altering distinct parameters such as solvent lean loading, amine inlet concentration, amine inlet temperature in the CO₂ capture process.²² Their research showed that in the absorption column, the increased lean loading and decreased MEA concentration would result in improved performance in terms of intercooling. The intercooling effectiveness of CO₂ absorption using MEA was examined by Rezazadeh et al.²⁸ They studied in-out and Recycle intercooler configurations for absorber and concluded that the intercooled absorber offers significant results at lean solvent loading from 0.30 to 0.34.

Six diverse configurations have been investigated by Amrollahi et al.³² for the CO₂ capture process and compared their results according to energy consumption. It was found that among the various configurations studied the combination of intercooler and split flow has the least energy consumption. Further, it was concluded that intercoolers have a significant role in minimizing energy consumption.³² The efficiency of intercooling for low, medium, and high lean loading without considering the effect of acid gas concentration in the flue gas has been investigated in the literature.³⁰ Kun Xin et al.³³ investigated and optimized the properties of amine based solvent in terms of CO₂ solubility, absorption kinetics, solvent strength, solvent viscosity, volatility, and heat capacity for post combustion CO₂ capture using process simulation. Hwang et al.³⁴ investigated the novel experimental based hindered diamine solvent for industrial applications of PCC system. They studied the various characteristics of a novel diamine solvent in terms of CO₂ loading and cyclic capacity. The selected novel solvent has higher absorption capacity, lower solvent degradation, and regeneration energy in comparison to MEA.

For many of the complex engineering problems, process simulation software's are used which provides low cost and reliable solutions by expressing process behavior using thermodynamic fundamental laws. In general, for a wide range of unit operations, Aspen Plus has been leading the development of process simulation models. The simulation model for absorption and stripping columns has been created in Aspen Plus V10 software. In the present study, the investigation of the effects of primary alkanolamine MEA, tertiary alkanolamine MDEA and their combined effect by making their blends on absorption rate and specific reboiler duty is carried out. Keeping in mind the process modification, the effects and significance of absorber intercooling (in-out and recycle) were investigated on the rate of absorption and specific reboiler duty.

Novelty

PCC using aqueous amine solution is a preferred CCS method due to high CO₂ purity and retrofit for the existing fossil fuel-based power plants is simplier. Many studies have been performed to investigate the overall performance of power plant and minimization of energy required for amine-based PCC, however, there is limited research work in the literature for mixed amine system with process modifications. The present study is focused on minimization of specific reboiler duty for two types of alkanolamines (MEA, MDEA) and their blends as well as process modification of absorber intercooling to enhance the absorption rate with lower solvent flow rate. The usefulness of the above mentioned absorber intercooling modification has been studied by Rezazadeh et al.²⁸ for MEA, however, there is still work needs to be done on absorber intercooling modification using the blends of aqueous alkanolamine. This design configuration for reduction of the solvent regeneration energy is novel in a way that this work has never been designed in much detail which gives fairly good results with suggested changes in some parameter values and process modifications. The parameters such as column sizing, including column diameter and column height needed for commercial scale plant of 90% CO₂ capture rate were taken from the open literature and other research works which is recommended for rate-based calculations. Aspen Plus, the most commonly available simulation tool in research was used to model the CO₂ absorption/desorption process to calculate the required solvent flow rate and solvent regeneration energy for 90% CO₂ capture with and without intercooling for MEA, MDEA, and their blends. For acid gas treatment, alkanolamine blends are considered ideal for high absorption and low regeneration energy. 25,35,36 Therefore, MEA and MDEA and their blends with different ratios are used in the simulation to investigate their effect for absorber intercooling on the inlet flow rate of solvent and specific reboiler duty. After a survey of the literature, the present study can be considered the first study for the minimization of both the inlet flow rate of solvent and reboiler duty by using amine blends in conjunction with process modifications such as absorber intercooling.

Methodology

Conventional process description

The fundamental process flow sheet modeled in Aspen Plus for the CO₂ capture system using alkanolamine is shown in Figure 1 and it is the base case for the present research work. The base model contains two packed columns, one is used for the absorption process while

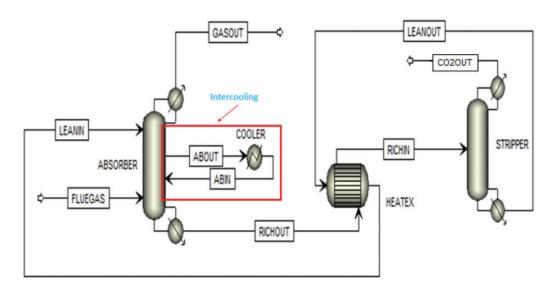


Figure 1. Fundamental process flow sheet for an amine-based CO₂ capture process with absorber intercooling modification as modeled in Aspen Plus.

the other for stripping. Flue gas containing acid gas enters the absorption column from the bottom and alkanolamine solvent enters from the top of the column. Acid gas as CO₂ in the flue gas and amine solvent contacts counter currently and a chemical reaction takes place between them and CO₂ is absorbed in the solvent. After the reaction, the purified gas leaves from the top of the absorption column to the atmosphere and solvent enriched CO₂ (rich solvent) leaves from the bottom of the absorption column and passes through a heat exchanger (lean-rich exchanger) to increase its temperature before entering to the stripping column for solvent regeneration. After increasing the temperature of the rich solvent, it enters into the stripper and flows down through the column counter currently with the stripping steam generated in the reboiler which will strip off its absorbed CO₂ gas. Vapor stream containing CO₂ and some traces of amine solvent and water leaves from the top of the stripper column to the condenser and hot lean solvent leaves from the bottom of stripper to lean-rich exchanger to the cooler and after decreasing its temperature to a specific point it returns to the absorption column. Some fraction or all of the condensed liquid is returned to the stripping column as reflux while the remaining stream is pure CO₂ and is transported for further processing and storage.

Process modification

The huge amount of energy required for the PCC process is a major concern of this technology. If CCS is employed on all fossil fuel-based power stations, the overall efficiency of the power plant will decrease due to high energy usage. To decrease this huge amount of energy required, Le Moullec et al.²⁹ proposed various process modifications and few researchers suggested new solvents with less regeneration energy.^{21,22} Absorber intercooling is one of the process modification proposed by Moullec et al.²⁹ which falls in the absorption enhancement category and is used to increase the CO₂ absorption rate and decrease the specific reboiler duty of the process. In the present research work, the effect of intercooling

Cases	MEA/MDEA blending percentage/ratio in lean solvent flow rate
MEA	100/0
MEA8/MDEA2	80/20
MEA6/MDEA4	60/40
MEA4/MDEA6	40/60
MEA2/MDEA8	20/80
MDEA	0/100

Table 1. Classification of MEA and MDEA blends and case studies.

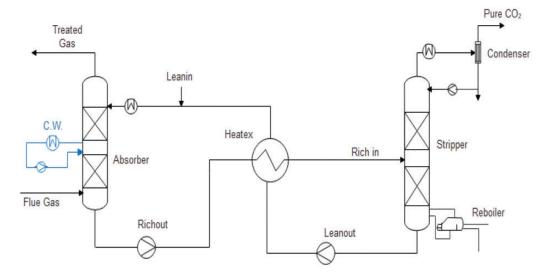


Figure 2. Absorber intercooling flow sheet as suggested by Moullec et al.²⁹

is investigated on absorption rate, specific reboiler duty, and absorber performance using aqueous amines such as MEA, MDEA, and their blends. Various case studies for the blends of MEA and MDEA are listed in Table 1.

Absorber intercooling

The basic concept of absorber intercooling is where a fraction of the solvent from side stream is withdrawn. The side stream then passes through the cooler to decrease its temperature to a specific level. After cooling, the solvent returns to the absorption column at a point just beneath where the solvent is taken out as shown in Figure 2 22,29 . The purpose of intercoolers is to cool the solvent. As the solvent temperature decreases after intercooling it increases the absorption rate due to its low temperature and the reaction between CO_2 and the solvent moves in forward direction. Since absorber intercooling permits a shift in the gas-liquid equilibrium as a result rich solvent loading increases in the lower section of the absorption column. It also lessens the required inlet flow rate of solvent for a specific CO_2 removal rate and thereby resulting in the reduction of the specific reboiler duty.

The proposed schematic is shown in Figure 2. The proposed modification is modeled through Aspen Plus in two different ways as suggested by Rezazadeh et al.²⁸ "simple" intercooling (in and out) and "advanced" intercooling (recycled) and their benefits for pure MEA, pure MDEA, and their blends were evaluated. The summary of flue gas composition and process conditions for NGCC 400 MWe power plant as suggested by Agbonghae et al.¹⁹ are used in the presented cases and are given in Table 3. The process flow diagram of the absorption column with simple intercooler and advanced intercooler are shown in Figures 3 and 4, respectively.

For simple intercooling configuration, the fraction of CO₂ enriched amine solvent (semirich) is withdrawn from the end of one packing section from the absorber packed column. To decrease the temperature of a semi-rich solvent, it passes through an external cooler where its temperature is decreased to the point at which the lean alkanolamine solvent is introduced in the absorber. After cooling, semi-rich solvent returns to the top of the next packing section of the absorber column. The absorber column has been divided into multiple sections and in each section, Sulzer Mellapak 250Y structured packing was used.

For advanced intercooler configuration, the amine solvent enriched with acid gas leaves from the end of the middle packing section from the column. To decrease the temperature of semi-rich solvent, it passes through an external cooler where the temperature of the solvent is decreased to the point at which the alkanolamine solvent is introduced. After cooling, the semi-rich solvent enters back to the absorber column at the top stage of the middle section of packing. In advanced intercooler design, structured packing Sulzer Mellapak 250Y has been used in the first and third packing section while low dimension Sulzer Mellapak 125Y has been used in the middle packing section of the column. Coarse structured packing has been used in the middle section to avoid pressure drop and excessive solvent load. A recycle rate of 2–5 times the solvent flow rate has been suggested in the literature³⁰ and can be

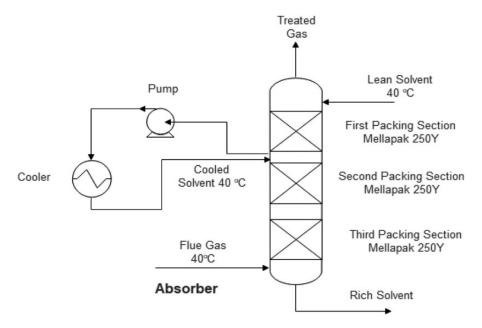


Figure 3. Process flow diagram of absorber with simple intercooler.

optimized in accordance with absorber flooding and operating cost for running the recycle pump. Various recycle ratios have been studied by Rezazadeh et al.²⁸ ranging from 1 to 9 times the inlet flow rate of solvent and after comparison with base case and a simple absorber without intercooling, the conclusion for natural gas applications for the optimum recycle ratio is about 3 for 30 wt.% MEA.

Process modeling

To investigate the absorber performance and energy consumption of carbon dioxide-MEA-water, carbon dioxide-MDEA-water, and carbon dioxide-MEA/MDEA-water systems, the Aspen Plus®V10 simulation software is employed. For the thermodynamic properties, the model used in Aspen Plus³7,38 is based on the work by Zhang et al. To develop thermodynamic models for carbon dioxide-MEA-water, carbon dioxide-MDEA-water and carbon dioxide-MEA/MDEA-water systems, the models provided by Aspen Plus were employed. The electrolyte pairs and binary interaction parameters were recommended by Aspen Plus from its physical property data bank for carbon dioxide-MEA-water, carbon dioxide-MDEA-water, and carbon dioxide-MEA/MDEA-water systems. Henry's law was applied to CH₄, CO₂, and H₂O, and the required Henry's constant were retrieved from the databank of Aspen Plus. To calculate the properties of liquid and vapor phases, the electrolyte-NRTL (Non-Random-Two-Liquid) method and Redlich-Kwong (RK) equation of state were used, respectively. The main chemical reactions involved in the systems are given in Table 2.

For the CO₂-MEA-H₂O system; equilibrium reaction equations (1) to (3) and kinetic reaction equations (5) to (8) are used. For the CO₂-MDEA-H₂O system; equilibrium reaction equations (1), (2) and (4) and kinetic reaction equations (5), (6), (9) and (10) are used and for the CO₂-MEA-MDEA-H₂O system; equilibrium reaction equations (1) to (4) and kinetic reaction equations (5) to (10) are employed.

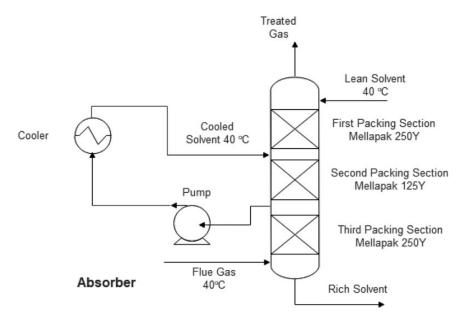


Figure 4. Process flow Diagram of absorber with advanced intercooling.

EQUIL $2H_2O \leftrightarrow H_3O^+ + OH^-$ (1) EQUILHCO₃⁻ + H₂O \leftrightarrow H₃O⁺ + CO₃⁻² (2) $EQUILH_2O + MEAH^+ \leftrightarrow MEA + H_3O^+$ (3) $EQUILH_2O + MDEAH^+ \leftrightarrow MDEA + H_3O^+$ (4) $KINETICCO_2 + OH^- \rightarrow HCO_3^-$ (5)KINETICHCO₃⁻ → CO₂ + OH⁻ (6) $KINETICMEA + CO_2 + H_2O \rightarrow MEACOO^- + H_3O^+$ (7)KINETICMEACOO $^- + H_3O^+ \rightarrow MEA + CO_2 + H_2O$ (8) KINETICMDEA $+ CO_2 + H_2O \rightarrow MDEAH^+ + HCO_3^-$ (9) $KINETICMDEAH^+ + HCO_3^- \rightarrow MDEA + CO_2 + H_2O$ (10)

Table 2. Main equilibrium and kinetic chemical reactions involved in the process.

Model description

Pilot scale model development and validation

Base model development and validation were carried out using the experimental results from the available literature. The experimental data given by Notz et al. Includes multiple set of experiments to observe the behavior of different parameters, such as acid gas CO₂ removal rate, specific reboiler duty, and rich loading of solvent by varying different process parameters, such as inlet temperature of the flue gas and the solvent in absorption column, inlet flow rate of the solvent in the absorber, acid gas CO₂ partial pressure in the flue gas, MEA mass fraction and desorption pressure. By using this experimental data, multiple cases are simulated and the results of the simulation are compared with Notz et al. experimental data. Figure 5(a) to (c) shows the parity plots of CO₂ rich loading, CO₂ removal rate, and specific reboiler duty, respectively. From Figure 5, it is clear that in comparison with the experimental data reported by Notz et al., the average absolute deviation of the rich CO₂ loading, removal rate of acid gas CO₂, and specific reboiler duty is minimum and are in good agreement in terms of validation.

Figure 6(a) and (b) shows a comparison of the modeled absorber and stripper liquid phase temperature profiles, respectively with the experimental temperature profiles data given by Notz et al.⁴⁰ From Figures 5 and 6 it is clear that the findings of the simulation are in good agreement with the reported data of Notz et al.⁴⁰ for pilot scale experimental results in terms of validation and the base model can be used confidentially for scale up application and the absorber intercooling modification. The statistical analysis of the selected variables is performed through ANOVA and is provided in Section S.1 of the supplemental material as reference. Further, the statistical analysis indicates the model robustness.

Scale-up model development and applications

In the CO₂ capture plant, a rate-based model in Aspen plus was used for the simulation of absorption and stripping column. For scale-up model development, the thermodynamic models and physical properties obtained from Aspen Plus databank are used for primary (MEA), tertiary (MDEA), and their blends. For the evaluation of liquid and vapor properties, the electrolyte non-random two liquid (ELECNRTL) method and Redlich-Kwong (RK) equation of state are used, respectively. For components CH₄, CO₂, and H₂O Henry's law is chosen to be applied and Henry's parameters, binary interaction parameters, and

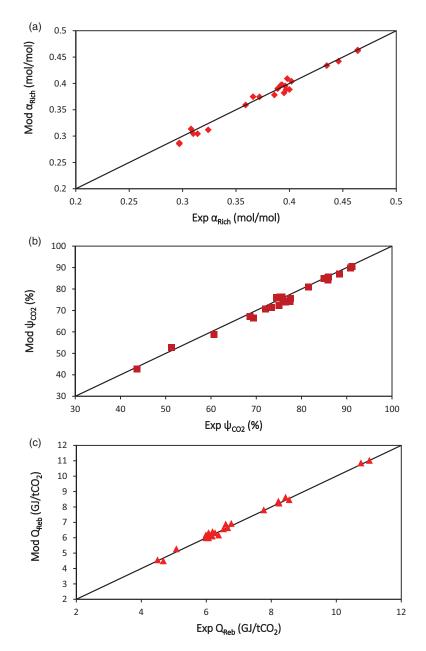


Figure 5. Parity plot comparison of simulation findings with the experimental data given by Notz et al. 40 (a) CO₂ rich loading in absorber (b) CO₂ removal rate. (c) Specific reboiler heat duty.

electrolyte pairs are retrieved from the physical property databank of Aspen Plus. The basic problem for the switch from the pilot plant model to scale up is the determination of optimum values of absorber and stripper column diameter and height to use different lean amine CO_2 loadings and flowrates of the solvent. To calculate the optimum values

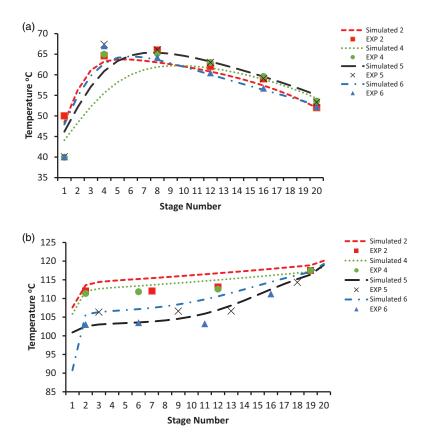


Figure 6. Comparison of modeled liquid phase temperature profiles with experimental profiles data given by Notz et al.⁴⁰ (a) Absorber (b) Stripper.

of diameter and height required in the absorber and stripper packed column for a 90% $\rm CO_2$ capture rate, the expressions developed by Agbonghae et al. 19 are used. For scale-up, the 90% $\rm CO_2$ capture rate is maintained as the widely used basis for the $\rm CO_2$ capture using aqueous amine as found in the open literature. For 90% $\rm CO_2$ capture with optimum stripper reboiler duty, the NGCC power plant for the gross power output of 400MWe is modeled for 30 wt.% MEA, 50 wt.% MDEA and their blends were optimally designed and discussed further. The summary of the flue gas composition and conditions used in this work is given in Table 3 as given by Agbonghae et al. 19

The thermodynamic model, forward and reverse kinetic reactions are the same as described in the process modeling section. Other design assumptions used in this model are given in Table 4. For the present research work, an aqueous solution of MEA with 30 wt.% strength and MDEA with 50 wt.% strength is used for 90% CO₂ capture with a solvent loading of 0.2 for MEA and 0.01 for MDEA.

Absorber inlet solvent flow rate and L/G ratios have been changed in the present work to minimize the specific reboiler heat duty and validate the model as reasonably good as possible. Flue gas flow rate of 622 kg/sec is used with different L/G ratios ranging from 0.965 to 2.957 to achieve desired CO₂ removal rate with optimum specific reboiler duty.

Table 3. Flue gas composition and conditions as given by Agbonghae et al.	Table 3	able 3.	Flue gas com	iposition and	l conditions as	s given b	y Agbonghae e	et al.'
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Component	Composition (mol/mol)	
CO ₂	0.0404	
H ₂ O	0.0867	
N_2	0.7432	
O_2	0.1209	
Ar	0.0089	
Flue gas temperature (°C)	40	
Flue gas pressure (bara)	1.2	

Table 4. Model assumptions and summary of design features of absorber and stripper columns.

Design assumptions and design features	
Design assumptions	_
Inlet temperature of lean solvent (°C)	40
MEA conc. (kg/kg), without CO ₂	0.30
MEA lean loading (mol/mol)	0.2
MDEA conc. (kg/kg), without CO ₂	0.50
MDEA lean loading (mol/mol)	0.01
MEA-MDEA conc. (kg/kg), without CO ₂	0.30-0.50
Stripper condenser pressure (bara)	1.62
Stripper condenser temperature (°C)	35
Lean-rich heat exchanger temperature approach (°C), hot end	10
CO ₂ removal rate (%)	90
Absorber	
No. of absorber	1
Packing material	Mellapak 250Y
Column diameter (m)	16.92
Column height (m)	19.06
Stripper	
No. of Stripper	1
Material of packing	Mellapak 250Y
Diameter of column (m)	6.76
Height of column (m)	28.15

After making these changes, the specific reboiler duty minimizes from the value reported by Agbonghae et al.¹⁹

Scale up absorber intercooling modification

The proposed modification of absorber intercooling by Le Moullec et al.²⁹ is shown in Figure 2. In rate-based absorber/stripper CO_2 capture model, this modification is simulated by using the simulation tool Aspen Plus for the present work which is shown in Figure 1. The variable specifications and parameters are the same as in the scale up model which are given in Tables 3 and 4. After absorber intercooling modification, an increase of rich solvent

loading indicates the absorption enhancement rate which results in a decrease of both inlet flow rate of the solvent and specific reboiler duty. In other words, the overall energy consumption decreases after implementing the absorber intercooling for the base case design.

Absorber intercooling modification has a significant impact on overall energy usage and the absorption enhancement rate. The basis taken for the modification process is almost the same as it was supposed and predicted for the scale up base case model. An intercooler is a mechanical device used to cool the solvent between stages. After installing the intercooler to the absorber column, a fraction of the solvent in a side stream is drawn from the absorber, cooled in intercooler, and then reintroduced to the absorption column just beneath where it was withdrawn from the column. In simple intercooling, a fraction of the solvent enriched with acid gas CO₂ in a side stream is drawn from the last stage of the first packing section. After cooling the semi-rich solvent in intercooler, solvent enriched with CO₂ returns at the first stage of the next (middle) packing section in the absorber column. While in advanced intercooling, a side stream of solvent enriched with CO₂ is withdrawn from the last stage of the middle packing section and after cooling, the cooled solvent returns to the first stage of the middle packing section. Excessive load of solvent in the middle section can cause flooding and pressure drop in the column and to circumvent these, Sulzer Mellapak 125Y a coarse structured packing in the middle section of the absorber column was used.

Absorber intercooling was evaluated for different cases of MEA, MDEA, and their blends as presented in Table 1. The effect of intercooling on different parameters studied for specific lean loading of solvent and CO₂ capture rate will be presented in the subsequent sections.

Result and discussions

Scale up CO₂ capture model with and without absorber intercooling

In the above sections, it has already been discussed in detail that by using a widely used simulation tool the post combustion CO₂ capture plant using aqueous alkanolamines is being modeled. CO₂ capture model development and validation for MEA at the pilot plant, the scale up based model for MEA, MDEA, their blends, and the process modification of absorber intercooling (simple and advance) have also been discussed. Effects of absorber intercooling both simple and advance were investigated for single (MEA & MDEA) amine and blended (MEA/MDEA) amines in terms of inlet flow rate of solvent, L/G ratio, rich solvent loading, liquid phase temperature profiles, and specific reboiler duty. The findings are elaborated in the next sections:

Effect of intercooling on inlet flow rate of solvent

Figure 7 shows the liquid solvent flow rate for MEA, MDEA, and their blends for three different absorber cases: without intercooler, simple intercooler, and advanced intercooler. Figure 7 clearly shows that the solvent flow needed for the 90% CO₂ capture for an advanced intercooling route proposes better results in comparison with simple intercooling and no intercooling routes.

In the case of pure MDEA, a maximum of 23.07% and 25.98% reduction of the solvent flow rate through simple and advanced intercooling was observed, respectively in comparison to no intercooling case. Because in the absorption packed column the overall

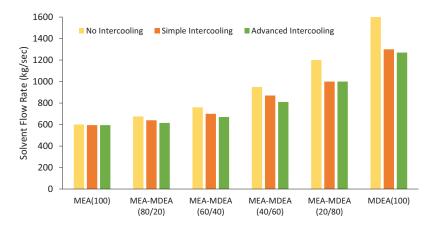


Figure 7. Liquid solvent inlet flow rate for absorber without intercooling (yellow), simple intercooler (brown) and advanced intercooler (green) at recycling ratio of 3 for MEA, MDEA and their blends.

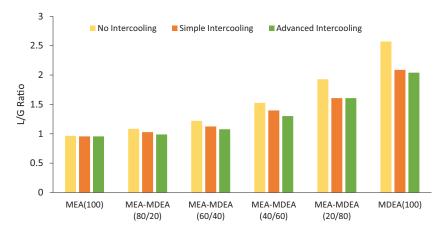


Figure 8. L/G ratio for absorber without intercooler (yellow), simple intercooler (brown) and advanced intercooler (green) at recycling ratio of 3 for MEA, MDEA and their blends.

temperature of the liquid phase decreases after intercooling, resulting in a reduction of solvent flow rate required. Also, MDEA has a higher absorption capacity than MEA.

The overall temperature decrease in the column leads to the enhanced capacity of the solvent for absorbing an acid gas CO₂, as the absorption capacity of alkanolamines increases with a lower temperature.²⁸ With comparison to all other cases, pure MDEA has the highest solvent flow rate, L/G ratio, and lowest rich solvent loading. In comparison with no intercooling option for all the cases, the optimum decrease in the inlet flow rate of solvent through simple and advanced intercoolers has been observed for the case MEA4/MDEA6 with 9.19% and 17.28% reduction, respectively. For all other cases, the effect of intercooling is less pronounced while both simple and advanced intercooling requires a lower inlet solvent flow rate in comparison to the absorber without intercooling. Figure 8 shows the

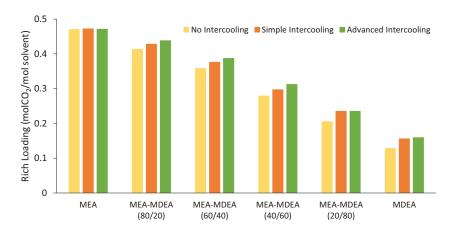


Figure 9. Solvent rich solvent loading for absorber without intercooler (yellow), simple intercooler (brown) and advanced intercooler (green) at recycling ratio of 3 by using different L/G ratio for MEA, MDEA and their blends.

comparison of liquid to gas ratio for MEA, MDEA, and their blends for absorber without intercooler, with simple and advanced intercoolers.

Intercooling effects on rich loading of the solvent

In Figure 9, the comparison of solvent rich loading (mol CO₂/mol solvent) for MEA, MDEA, and their blends for absorber without intercooler, simple intercooler, and advanced intercooler has been shown. The lean solvent loading for MEA is fixed at 0.2 and is 0.01 for MDEA. For different cases of MEA, MDEA and their blends, an obvious decline in rich solvent loading happen together with an increase in L/G ratio observed for absorber without intercooler, simple intercooler, and advanced intercooler. For the pure MEA case, no evident change in rich solvent loading has been observed while as the solvent is blended with MDEA a slight increase can be seen in solvent rich loading for both simple and advanced intercooling in comparison to no intercooling.

Through simple and advanced intercoolers the maximum increase in rich loading of solvent was observed for the case in which pure MDEA with 17.69% and 19.37% increase, respectively, when compared with the no intercooling option, at the expense of high inlet flow rate of solvent and liquid to gas ratio. The optimum increase in the rich loading of solvent by using simple intercooler and advanced intercooler options was observed for the case MEA4/MDEA6 with 6.04% and 10.54% increase, respectively, in comparison to the no intercooling option. After intercooling of the absorber, the increase in solvent rich loading confirms that intercooling generally permits the packed column closer to the equilibrium. Moreover, for various amine cases at specific lean loading of the solvent the increase in rich loading of solvent also confirms another benefit that the required inlet flow rate of the solvent decreases to achieve a 90% CO₂ removal rate as compared to the absorber without intercooler.

Intercooling effects on CO2 absorption rate and liquid phase temperature profile

Aqueous amine solvent enters the absorption column in the top section and flue gas enters the column from the bottom section and contacts with the solvent counter currently for

reaction to take place. As the reaction between amine solvent and CO₂ proceeds and solvent absorbs the CO₂ and the temperature of the column increases resulting in a decrease in the rate of absorption. To overcome this issue, the proposed²⁹ process modification of absorber intercooling has been implemented. After the intercoolers are installed at the absorber column, the temperature of the column decreases resulting in increase in the rate of absorption. In simple intercooling, a side stream is withdrawn from the end of the first section to the cooler and after cooling returns to the top of the next section. Whereas, in recycle intercooling side stream is withdrawn from the end of the middle section to the cooler and after cooling, the stream returns to the top stage of the middle section. From Figures 7 and 9 it is evident that by applying simple and advanced intercoolers to the absorber column, the increase in rich loading of the solvent indicates the enhancement of absorption rate with a decrease in solvent flow rate required for 90% CO₂ capture rate. It is also evident that advanced intercooling shows promising results compared to both simple and no intercooling.

Figure 10(a) to (c) shows the comparison of the absorber liquid phase temperature profiles for different cases of absorber column without intercooling, with simple and advanced intercooling, respectively. From the Figure 10, it is evident that by using advanced intercooling method the effect of intercooling on the liquid phase temperature is more prominent as compared to simple and without intercooling. The reason is that after using intercoolers both simple and advanced the overall temperature decrease in the column leads to an increase in the capacity of solvent to absorb CO₂, as the capacity to absorber CO₂ of alkanolamines increases with a lower temperature.²⁸

For all the cases, the average temperature decreases with simple intercooling in the absorber column by 5% and with advanced intercooling the average temperature decreases in the absorber column by 6.6%. The decrease in temperature is beneficial to increase absorption kinetics, such as reaction rate constant of the amines with CO₂ results in order to enhance the rate of absorption with less solvent flow rate in comparison to no intercooling. This concludes, at low temperature the reaction of CO₂ and alkanolamines is a forward reaction as CO₂ absorption in alkanolamines is higher resulting in increased CO₂ removal rate and at high temperature the reaction of CO₂ and alkanolamines is reversible. After intercooling the temperature in the absorber column goes down, increasing the rich solvent loading which results in higher absorption rate with low solvent flow rate and reboiler duty.

Intercooling effects on specific reboiler duty of the CO2 capture system

Figure 11 shows the comparison of specific reboiler duty of the CO₂ capture process with and without intercooling for MEA and MDEA and their blends. It is evident from Figure 11 that as the data point move from the primary amine MEA towards the tertiary amine MDEA, the specific reboiler duty decreases for both with and without intercooling. Furthermore, absorber intercooling shows promising results compared to no intercooling. The tertiary amine methyl-diethanolamine generally known as MDEA has the chemical formula CH₃N(C₂H₄OH)₂ is a colorless liquid with a pungent smell like ammonia and extensively used as a gas sweetening agent requires less amount of heat to regenerate, however, the major issue for using pure MDEA is the lowest removal rate of CO₂. As in tertiary amine MDEA, there is no hydrogen attached to the nitrogen making MDEA unable to react directly with CO₂ to form carbamate which results in the reaction of primary amine

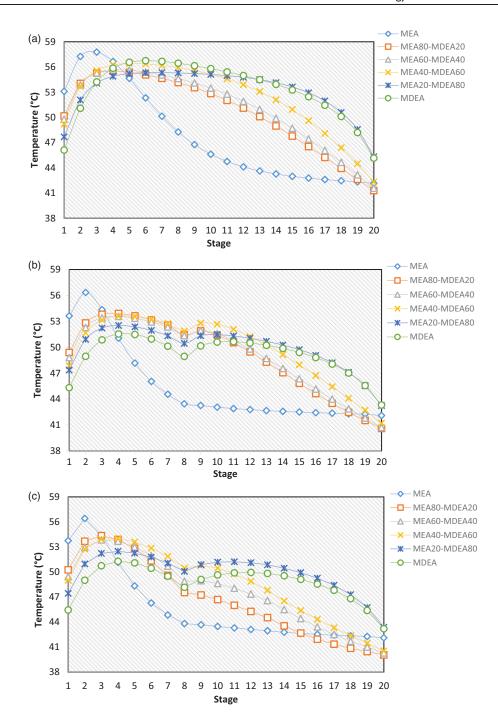


Figure 10. Comparison of liquid phase temperature profiles of absorption column for MEA, MDEA and their blends. (a) Without intercooling. (b) Simple intercooler. (c) Advanced intercooler.

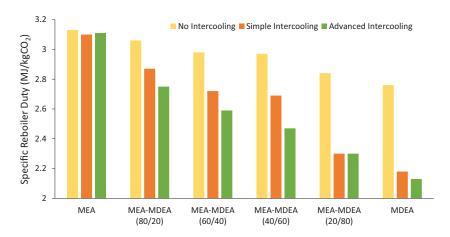


Figure 11. Specific reboiler duty comparison for the CO_2 capture system for absorber without intercooler (yellow), with simple intercooler (brown) and with advanced intercooler (green) at recycling ratio of 3 by using different L/G ratios for MEA, MDEA and their blends.

MEA with CO₂. Acid gas CO₂ and MDEA can react only after the formation of bicarbonate ion as a result of CO₂ and water reaction and then acid-base reaction undergoes between bicarbonate ion and MDEA. It has been observed that due to this reason, the reaction between acid gas CO₂ and MDEA is slow and takes longer time as well as high flow rate of MDEA solvent is required for higher CO₂ removal. From Figures 7 and 8 it is clear that to achieve 90% CO₂ capture rate MDEA requires a high flow rate of the solvent and hence its liquid to gas ratio is also higher. This may be the reason why MDEA alone is not suggested as a solvent for the CO₂ capture system.

For the case in which pure MDEA is used as a solvent, the highest decrease of 21.01% and 22.82% in the specific reboiler duty is observed with the simple intercooler and advanced intercooler, respectively, in comparison with all other cases at the expense of highest liquid solvent flow rate and lowest rich solvent loading for the MDEA case.

The optimum decrease in the specific reboiler duty through simple intercooler and advanced intercooler was observed for the case MEA4/MDEA6 with 9.42% and 16.83% decrease when compared with the no intercooling option, respectively with optimum solvent flow rate, optimum L/G ratio, and optimum rich solvent loading compared to all other cases.

Conclusion and future work

In the present research work, rate-based model for the CO₂ removal process with modification of absorber intercooling is developed and discussed in detail. The major goal of the present study was to enhance the rate of absorption and minimize specific reboiler duty to make the overall process energy efficient. The absorber intercooling configurations were evaluated in two different ways; simple and advanced for two different amine solvents (30 wt. % MEA and 50 wt. % MDEA) and their blends to remove 90% CO₂ capture. The effects of absorber intercooling on various parameters such as liquid solvent flow

rate, L/G ratio, solvent rich loading, absorber liquid phase temperature profiles, and specific reboiler duty were evaluated for MEA, MDEA, and their blends. Without intercooling, there is a significant increase in L/G ratio and a decrease in rich solvent loading while moving from pure primary amine MEA and its blend (MEA-MDEA) with tertiary amine MDEA to pure MDEA. The use of intercoolers indicated positive outcomes for both solvent rich loading and liquid to gas ratio. The following conclusions can be itemized;

- In the case of pure MDEA, a maximum of 23.07% and 25.98% reduction of the solvent flow rate through simple and advanced intercooling was observed, respectively in comparison to no intercooling case. Further, in comparison to all other cases pure MDEA has the highest solvent flow rate L/G ratio and lowest rich solvent loading. In comparison with no intercooling option for all the cases, the optimum decrease in the inlet flow rate of solvent through simple and advanced intercooler was observed for the case MEA4/MDEA6 with 9.19% and 17.28% reduction, respectively.
- For the case in which pure MDEA is used as a solvent, the highest decrease of 21.01% and 22.82% in the specific reboiler heat duty is observed with the simple and advanced intercooling, respectively, in comparison with all other cases at the expense of the highest liquid solvent flow rate and lowest rich solvent loading.
- Through simple and advanced intercooling the maximum increase in solvent rich loading was observed for the case in which pure MDEA with 17.69% and 19.37% increase, respectively, when compared with the no intercooling option, at the expense of high inlet flow rate of solvent as well as liquid to gas ratio. The optimum increase in the solvent rich loading through simple and advanced intercooler options was observed for the case MEA4/MDEA6 with 6.04% and 10.54% increase, respectively, in comparison to the no intercooling option.

It was concluded that the blends of MEA-MDEA with process modification of absorber intercooling is much better process than using single amine (MEA and MDEA) and without process modification because absorber intercooling configuration for blended amines not only fulfills the objective of reduction of both solvent flow rate and specific reboiler energy.

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Supplemental material

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Appendix

Notation

ABIN absorber inlet ABOUT absorber outlet

 α_{Rich} CO₂ loading of the rich solution (mol CO₂/mol amine)

CCS carbon capture and storage CFC's chlorofluorocarbon's

CO₂OUT CO₂ gas stream out from the stripper column

DEA diethanolamine DGA diglycolamine

ELECNRTL electrolyte-non-random-two-liquid

G gas flow rate (kg/sec)
GHG greenhouse gas
HFC's hydrofluorocarbon's

IEA international energy agency

IPCC intergovernmental panel on climate change

L liquid flow rate (kg/sec)
L/G liquid to gas ratio
MDEA methyl-diethanolamine
MEA monoethanolamine
MWe megawatt electrical

NGCC natural gas combined cycle PCC post combustion CO₂ capture

PFD process flow diagram ppm parts per million

Q_{Reb} specific reboiler heat duty (GJ/tCO₂)

 ψCO_2 CO₂ removal rate (%)