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RESEARCH ARTICLE

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Abstract

Carbon dioxide capturing processes from industrial gases are widely applied to enrich valuable gas and to reduce greenhouse gas emission. The highest amount of anthropogenic CO₂ is emitted in flue gas form. Flue gas composition varies in a wide scale therefore designing such plants that are able to operate on a good removal efficiency scale is vital. A typical method for such gas separation is an absorber-desorber system where the absorbent is 30 w% aqueous monoethanolamine solution.

The aim of this work is to setup computer based process model that is capable of simulating CO₂ capture from flue gases and to study its performance at different operating conditions. We investigate possible process improvements by enhancing the CO₂ removal efficiency with the variation of different key parameters and different plant setups. We also take into account the regeneration energy demand by optimizing the system to the lowest specific energy demand. Results show that the least attractive option is the overload of the existing absorber-desorber system. By introducing a second absorber-desorber step, high removal efficiency can be reached while maintaining low desorber temperature. The best performance can be achieved with increasing the absorber height and elevated desorber pressure that can reduce the specific regeneration energy especially at high removal efficiency region. This result is in agreement with the argued classical absorber design and operation philosophy that the absorber should be operated at extremely low loads, which is in association with oversized absorber column packing heights. This offers the possibility for flexible operation.

Keywords

CCS, MEA, modelling

1 Introduction

Anthropogenic greenhouse gas (GHG) emission leading to climate change and its effect on the natural environment poses one of the biggest challenges ahead to be dealt with. It is accepted that ocean water level, GHG concentration of the atmosphere and the average temperature are in a function with each other in time [30]. CO₂, considering its amount in the atmosphere has the most influence among GHG [42]. The greatest concern today is that since the concentration of CO₂ has risen to the record of 406 ppm by the year 2016 and continues to do so, its effect on the climate will lead to a non-reversible change [1, 34]. So far the technological answers to keep CO₂ emissions as low as possible towards a green economy are found in:

1. Reforming available energy- conversion and conservation techniques to boost energy efficiency.
2. Utilization of such energy conversion techniques excluding imbalanced carbon oxidation, such as the utilization of renewable or nuclear energy sources [33].
3. Use of end of pipe gas cleaning techniques, also called carbon capture and sequestration.

Some of these end of pipe techniques are already in operation in other fields of the energy sector, such as natural gas sweetening for the aim to meet natural gas quality standard before it is inserted to pipelines. The end of pipe approach is mainly criticized by the fact that the capture process has an additional energy demand on one side and on the other side dealing with the amount of captured CO₂ is only partly answered. There are however initiatives that utilize all these technical approaches in parallel. One famous idea comes from the Nobel Prize laureate George Olah's Methanol Economy in which he describes a viable process cycle of recycling CO₂ by producing methanol. For methanol production the H₂ is produced by applying renewable energies for electrolysis. This method would allow the energy circulation with large storage capacity [13]. An alternative idea comes from the German institute Umweltbundesamt (in English: Federal Environment Agency) of developing an energy process cycle similar to methanol economy but the carrier medium is methane [16].

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The anthropogenic CO₂ production is mostly emitted in flue gas form [4, 21, 26, 27]. Flue gases therefore are present in large volumes with low CO₂ partial pressure. For such CO₂ sources the most suitable technology present is the post-combustion carbon capture (PCC) method. The various types of plants such as power plants, oil refineries, cement factories etc. that produce flue gases operate in different loads depending on their energy demand. This also results in various content and composition flue gases [28]. To follow the behavior of the plant the capture plant must be built to function flexibly and economically in various circumstances. These various circumstances include the varying gas compositions, amount and capture rate flexibility as well [28]. The capture rate flexibility is particularly important because the rate of the captured CO₂ may vary according to the gas composition and the gas flow rate. The flue gas CO₂ content may vary in the range of 3.875 – 22 vol%, while similar industrial gases, such as bio gas, may have an even higher CO₂ content [19, 28].

In case the gas to be purified has a low CO₂ content with low (atmospheric) pressure, the separation task is a difficult procedure due to the lack of mass transfer driving force. In such a case the most recognized technique is the use of an absorber-desorber system applying chemisorption [3, 20, 36]. The materials used for such process are found within the groups of amines [6, 9, 22, 24, 25, 31, 36, 39]. Three groups are distinguished within the group of amines by the number of attached alcohol groups to the center nitrogen atom. The more alcohol group is attached, the less is the mass transfer driving force for CO₂ absorption [17]. This feature is favorable for the solvent regeneration but unfortunate for absorption. In order to achieve acceptable removal efficiencies in pilot phase, the most frequently used solvent in studies is monoethanolamine (MEA). MEA having only one alcohol group is among the most reactive absorbents for CO₂ and SO₂ meaning 1 mass unit of MEA is capable of binding 0.45 mass of CO₂ and it is appropriate at low (atmospheric) pressures requiring stringent outlet gas specifications [15, 43]. On the contrary MEA is highly corrosive, and it is too viscous to be applied in itself, therefore it is usually diluted to a 15-40 w% aqueous solution [2].

Environmental policy and regulations are aiming at a carbon neutral society. This tendency may have crucial effect on plants emitting CO₂ in large amounts. The novelty of our work is that this article studies different scenarios if CO₂ capture removal efficiency should be increased. Our work studies the approaches for a CO₂ capture plant to cope with the increased removal efficiency demand

The amine scrubbing capture process is built of two main columns. Fig. 1 shows the basic scheme for the process. The flue gas is vented into the bottom part of the absorber column by a gas blower and flows upwards. In the absorber column it is contacted with the absorbent that is introduced at the top of the

column. In order to maximize the mass transfer rate, packing with large surface area with low pressure drop is utilized in both columns. In the absorber due to the low absorbent CO₂ loading and its temperature, the concentration gradient drives CO₂ to the liquid phase. As the loaded solvent leaves the absorber it is preheated and introduced to the desorber. Desorber pressure is a crucial parameter of the system. According to Gale et al. [12] the thermal degradation of MEA becomes economically significant above 130 °C. On the other hand according to Freguia [11] elevated pressure is favorable if further gas processing step such as compression is included to the calculations. The operation of the desorber is similar to a distillation column. The loaded absorbent enters at the top of the column and it is contacted with steam generated in the reboiler. The gaseous phase has a low CO₂ partial pressure and with the introduced heat the concentration gradient is turned to drive CO₂ back to the gas phase. Eventually the steam and the liberated CO₂ are separated in the condenser.

2 Simulation tools

The base thermodynamic method used for the simulation is the ENRTL method that includes different assumptions for the liquid and the vapor phases.

In the vapor phase the presence of ions are neglected. The defined components and the process properties suggest the SRK EoS model (Soave, G 1972) [35, 38].

The thermodynamics of the liquid phase is more complex. The parallel reactions including ions make the liquid phase act as a weak electrolyte [22]. For such system Chen et al. extended the local composition model NRTL by Renon and Prausnitz [32] by supplementing the model by including the correction of long range forces caused by the presence of ions [8].

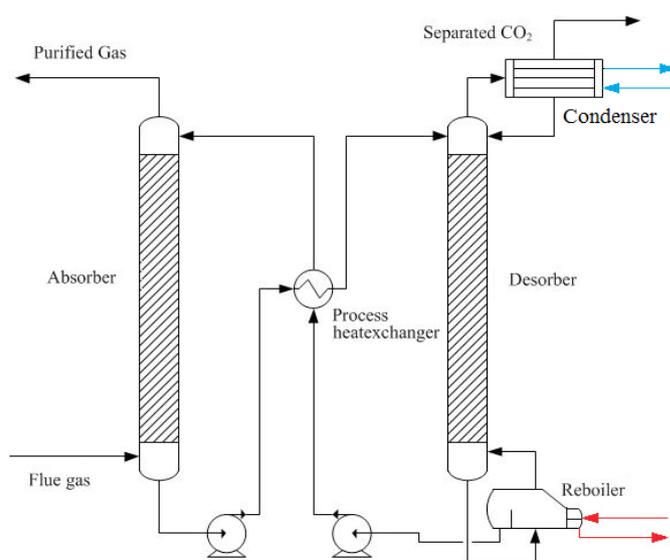
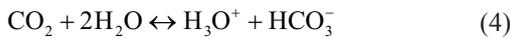
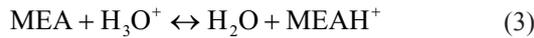
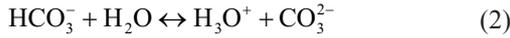


Fig. 1 Capture process flowsheet with one absorber and one desorber column

The model was further developed by Mock by including the Born correction, which takes the change of the dielectric constants into account on the short range interactions [23]. This is today the ENRTL (electrolyte non-random two-liquid) thermodynamic model. In the liquid phase 5 parallel reactions are considered.

These reactions are defined as instantaneous equilibrium reactions. The reaction equilibrium constants are calculated with the use of the Gibbs free energies. This reaction equation system is valid in all unit ops. and stream, except for the absorber and the desorber columns.



1. water dissociation (see Eq. (1))
2. carbonate formation (see Eq. (2))
3. MEA- protonation (see Eq. (3))
4. Hydrogen carbonate formation (see Eq. (4))
5. Carbamate formation (see Eq. (5))

In the absorber and desorber columns reaction 4 and 5 are considered to be slow reactions therefore they are calculated with forward end reverse reaction kinetic equations. The reaction rate is calculated with a second order expression and in Aspen Plus it is calculated with Arrhenius type Power law Eq. (6) where E is the activation energy and k is the pre-exponential factor [38].

$$r = k \cdot T^n \cdot e^{-E/RT} \quad (6)$$

2.1 Column model

The columns are modeled with the so called rate based model developed by Taylor and Krishna [18]. The rate based model for such reactive absorption case is superior to the traditional equilibrium model [37]. For packed columns the equilibrium is defined by a packing height that is the height equivalent theoretical plate (HETP). This value is always given by the packing vendors based on their measurements. The reason why the equilibrium model is inadequate for modeling this system is because the HETP of packing changes with operating points and it changes along the height of the column. Furthermore as HETP is used for defining a column, stage efficiency is also introduced since phase equilibrium is rarely achieved totally in a stage [37]. This problem was addressed by Taylor and Krishna by the introduction of the non-equilibrium model [18]. The non-equilibrium model assumes stages consisting of two bulk phases and between the bulk phases two film layers are present where mass and heat transfer is defined by rate equations (see Eq. (7)).

$$Dy_i/dx = \sum k = 1 \cdot k \neq 1 \left((y_i N_k - y_k N_i) / cD_{ik} \right) \quad (7)$$

Equilibrium is only assumed at the interface of the two film layer [37]. Since the non-equilibrium model focuses only on calculating the flux of mass within the film layers, other matters that influence the absorption and desorption process are calculated with correlation equations. This means that non equilibrium model is more precise and more adequate for predictive simulations than the equilibrium model but it badly needs reliable input data and more computation power.

2.2 Simulation parameters and features

The initial model of this study in Aspen Plus is a base model that was originally verified by the pilot plant of the University of Texas [10, 38]. Modification on the base model is carried out in order to have better simulation characteristics. The primary characteristics of this pilot plant and the model parameters are summarized in Table 2 and flue gas parameters are shown in Table 1.

Table 1 Flue gas parameters

composition of flue gas, m/m	H ₂ O	O ₂	N ₂	CO ₂
	0.009	0.048	0.681	0.261
temperature	59 °C			
pressure	1.03 bar			
flow	16.76 kmol/h			

The model includes several stream loops. Such loops can lead to convergence difficulties. The model is built up in a multistep building phase, from individual unit ops. to a closed loop process. This model building approach helps to have acceptable initial parameters for better convergence. The absorbent recycle loop is done by a built in calculator in which only the carbon content is recycled.

The simulations are carried out using a so called design specification mode. Design specification simulations mean that instead of specifying all input parameters straight, one target parameter is given and to achieve the target value, an adjustable parameter is also defined with a value interval where it is expected to be. Two approaches are used within the design specification. Figure 2 shows these different approaches. The top row (approach nr.1) shows that the target value is the removal efficiency (Ψ) with adjusted parameter being the desorber heat and the varied parameter is the absorbent flow. In order to achieve the desired removal rate, the cyclic capacity is calculated. The minimal specific reboiler duty is found by determining the proper absorbent loading (α) values. The absorbent loading is calculated according to Eq. (8)

$$\alpha = \frac{(\text{CO}_2 + \text{CO}_3^{-2} + \text{MEACOO}^- + \text{HCO}_3^-)}{(\text{MEA} + \text{MEA}^+ + \text{MEACOO}^-)} \quad (8)$$

The second row (approach nr.2) in Fig. 2 shows the case when the desired parameter is the lean absorbent loading (α_{lean}) and the adjusted parameter is the desorber heat and the varied parameter is the absorbent flow. The reboiler duty is always adjusted to the value to reach the initial absorbent loading. With the second approach, in the whole system the CO₂ removal rate is not directly controlled but it is determined by the mass balance. The first optimization approach gives results faster for one desired removal efficiency and the second gives the faster results for the optimal initial absorbent loading.

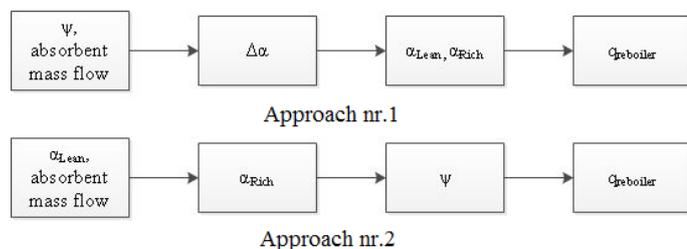


Fig. 2 Two approaches for finding operation points

Table 2 Primary parameters of the CO₂ capture pilot plant in Texas University

	absorber	desorber
Non-equilibrium stages	50	
Packing type	IMTP, 1.5-IN, METAL	FLEXIPAC 1Y, METAL
Packing height	6.1 m	
Column inner diameter	0.427 m	
Mass transfer correlation	Onda et al. [29]	Bravo et al., 1985 [14]
Interfacial area correlation	Onda et al. [29]	Bravo et al., 1985 [14]
Holdup method	Bravo et al 1992 [7]	
Heat transfer coefficient method	Chilton and Colburn	
Flow model	mixed	

2.3 Removal efficiency improvement

If the removal rate of CO₂, that is, the efficiency of the carbon capture should be improved there might be some alternative options. The investigated scenarios are the following:

- The first approach is the intensification of the removal efficiency by increasing the load of the existing equipment.
- The second approach is to build a second absorber column as an extension of the existing absorber column.
- The third approach is to modify the desorption pressure.
- The fourth approach is to build a second absorber-desorber system that is able to further purify the partly treated flue gas to a practically CO₂ free gas. This case is shown in Fig. 4.

3 Results

3.1 Increased load

The results of the first approach can be observed in Fig. 3. It can be observed that by increasing the lean loading (α_{lean}) of the absorbent, the specific regeneration energy decreases. To achieve high removal efficiencies α_{lean} must be lowered for the absorbent to be able to absorb increased amount of CO₂. The tendencies show that higher removal efficiencies can be achieved by decreasing the initial absorbent loading. This way the cyclic capacity, $\Delta\alpha$ of the absorbent increases. Reasonably low specific reboiler energies, around 4 GJ/tCO₂ however can only be achieved at low removal efficiencies. Higher removal efficiencies can be either achieved by increasing the absorbent mass flow and by decreasing the initial absorbent loading. Furthermore high absorbent flow stresses the capacity of the absorber column that may cause flooding. In case of low absorbent flow the desorber column may be flooded by the amount of steam generated in the reboiler. Varying the initial absorbent loading has more visible impact on the specific removal energy demand but decreasing the initial loading leads to geometrically smaller equipment demand due to the smaller mass flows.

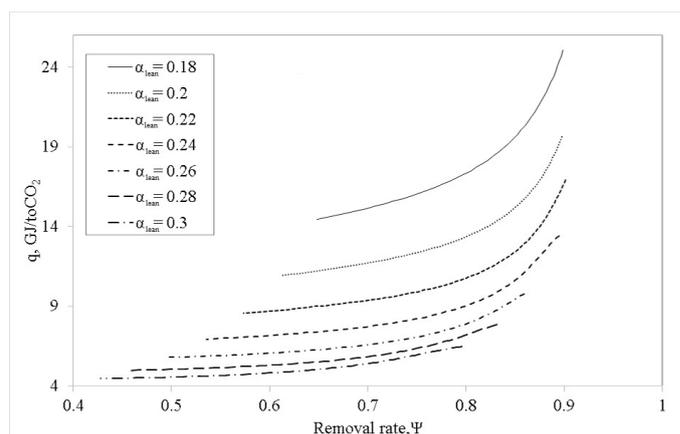


Fig. 3 operating conditions with one absorber desorber column pair

3.2 Second absorber-desorber system

To address the issue of the increased specific reboiler demand on higher removal rates, one possible solution is to insert a second absorber-desorber column pair forming a two-step gas treating system. In this case, the absorber-desorber column pairs work loosely dependent on each other (see Fig. 4).

The solution to investigate this coupled system is as follows: the optimal operating conditions were found for the first capture step with optimization approach nr.1 (see Fig. 2) and the second capture step was investigated with optimization approach nr.2. This solution approach does not follow the so called dynamic optimization programming rule. The dynamic optimization is a recursive optimization method where the first step of sub optimization is the final stage of the system. The theory of this approach is that when the last stage is optimized than it does not effect the previous stages of the system [5, 41].

In our case a process retrofit is assumed where the existing equipment has already been in use and that is extended with a second one. For the first column pair, the optimal operating point at the removal rate of 65% 76% and 85% is calculated. The purified gas mixture is then introduced to the second column pair and purified further to higher CO₂ removal rates. The specific energy demand is calculated according to Eq. (9).

$$Q_{\text{spec}} = (Q_{\text{reb},1} + Q_{\text{reb},2}) / (m_{\text{CO}_2,\text{in}} - m_{\text{CO}_2,\text{out}}) \quad (9)$$

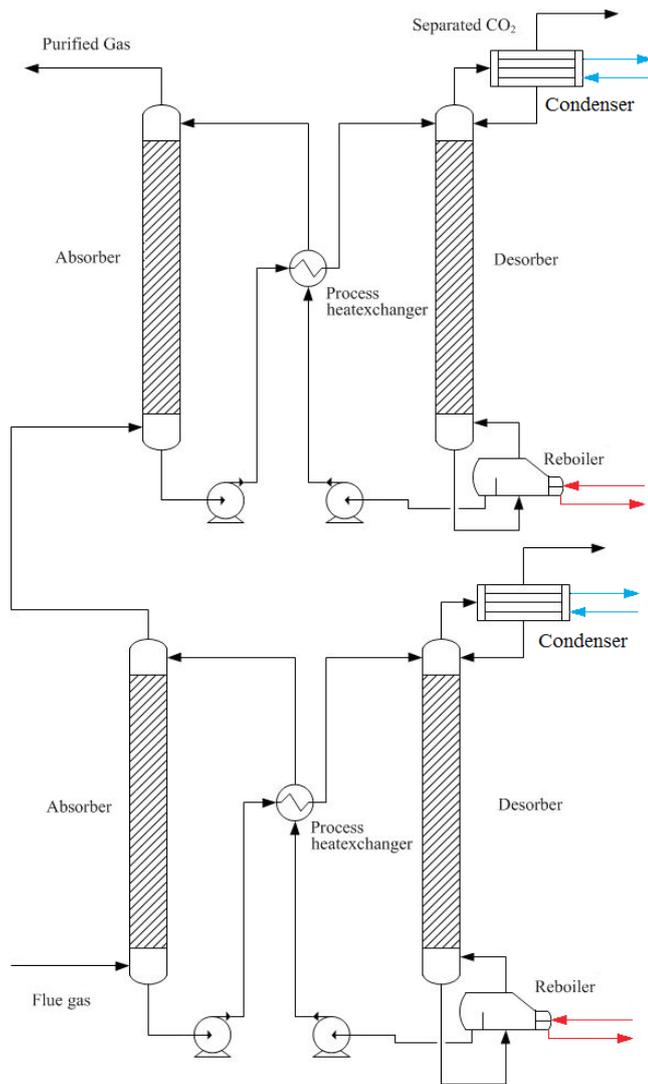


Fig. 4 Flowsheet of the 2 absorber and 2 desorber column layout

Figure 5 shows the comparison between the optimal operation of one column pair at different CO₂ removal rates and if the gas at 65% removal rate is introduced to a second absorber-desorber column pair. In the second case the optimum operating condition is found by varying the absorbent mass flow and the initial absorbent loading ($\alpha_{L,\text{lean}}$). The variation of the initial loading of the absorbent has a high influence on the removal rate. Towards higher initial absorbent loadings the specific removal rate decreases until a certain value. The optimal initial absorbent loading appeared to be at 0,3 molCO₂/molMEA. At

higher initial loadings the liquid demand increases the specific heat demand. A similar tendency can be observed with altered first stage removal rates. Figure 6 shows the case if the first column is running at 76% removal rate before it enters the second absorber column. At 76% the specific removal energy demand is already rising quite rapidly meaning that even though the second column pair is running efficiently until 98% total removal the overall specific energy demand will have a significantly higher value. By increasing the first step CO₂ removal rate another parameter change can be observed. In Fig. 7 the first step removal efficiency is set to 85% and as it is expected the overall separation energy follows the expected rapid rising. Evaluating these results, it is also shown in Figs. 5, 6 and 7 that the second separation stage optimal absorbent loading varies by the gas mixture composition entering the second absorber column. As the CO₂ content of the gas mixture decreases, assuming optimal operating conditions the initial absorbent loading decreases as well. The optimum at 65% occurs at 0.3 molCO₂/molMEA, at 76% it is reduced to 0.28 molCO₂/molMEA and at 85% it is at 0.26 molCO₂/molMEA.

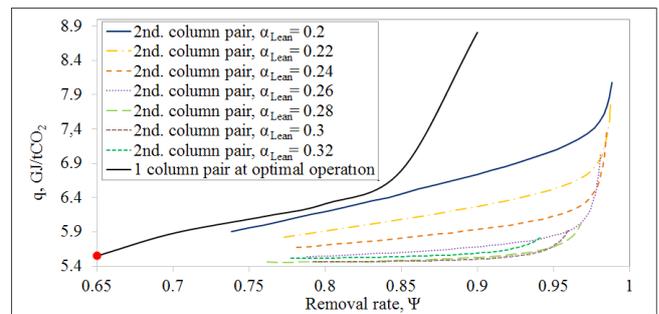


Fig. 5 Operating conditions with 65% removal rate for the first absorber column

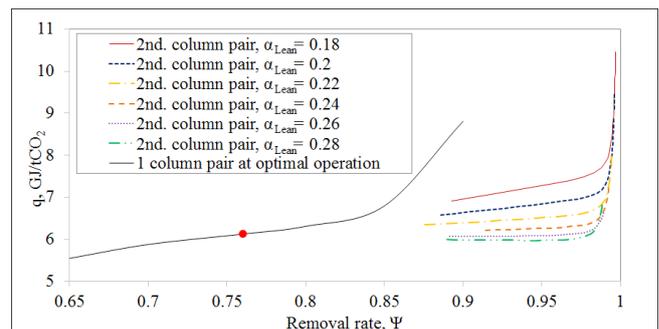


Fig. 6 Operating conditions with 76% removal rate for the first absorber

3.3 Desorber pressure variation

The classic approach to an absorber-desorber system is to have the absorption at high pressures and for desorption at low pressures. In our case finding the optimal desorption condition is not so clear. Following the classic approach, the saturation temperature of the absorbent material decreases and the heat of absorption also decreases with the temperature. On the other hand, the driving force for desorption can be increased either

by increasing the temperature and the pressure of the desorber column or by decreasing the partial pressure of the CO_2 in the column. Fig. 8 helps to understand this phenomenon.

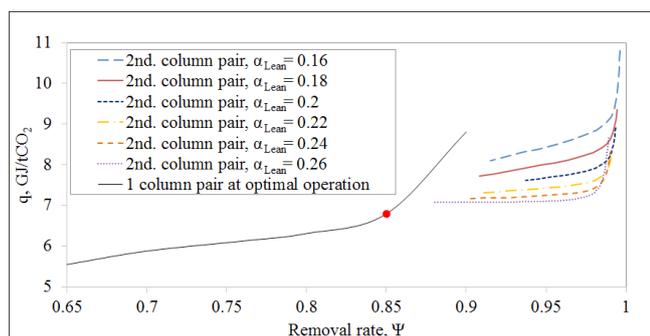


Fig. 7 Operating conditions with 85% removal rate for the first absorber column

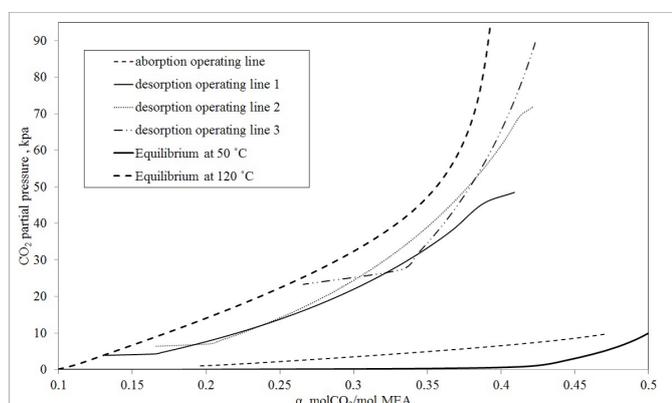


Fig. 8 MEA- CO_2 equilibrium curves and different desorber column operating lines

If the temperature and the pressure is increased in the desorber column, the equilibrium curve will shift away from the operating line and that increases the driving force (see Fig. 8). If the temperature/pressure remains and the partial pressure of the CO_2 is decreased then desorption operating line's slope will decrease. This can only be achieved by producing more steam in the reboiler of the desorber.

Figure 9 shows different operating conditions of varied absorbent mass flows and initial absorbent loadings. It can be observed that by increasing the reboiler pressure to 2.6 bar the removal efficiency can be increased significantly while the specific energy demand stays at reasonable values. To this Fig. 9 shows the optimal operating condition when the reboiler was set to 0.6 bar the initial loading was set to 0.3 mol CO_2 /mol-MEA. At elevated pressure the optimal loading was found to be at 0.18 mol CO_2 /molMEA. This result shows the optimal initial absorbent loading to be dependent on the desorption pressure. It must be also mentioned here that even though the specific energy demand shows better results at higher desorber pressures, this energy demand is realized at higher temperatures.

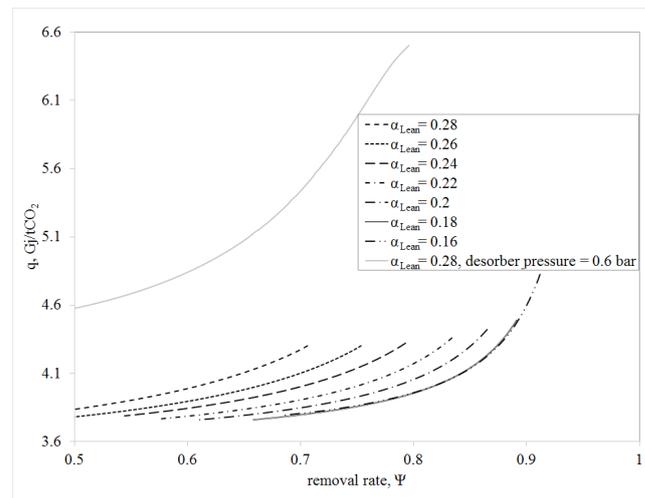


Fig. 9 Operating conditions at elevated desorber pressure (desorber pressure is set to 2.6 bar, except where other is indicated)

3.4 Extended absorber height

Combining the approaches of absorber height increase and desorber pressure increase lead to evaluate the mutual influence of these parameter changes. The absorber height is set to 12.2 m and the desorber columns pressure is also varied in order to see the minimal specific energy demand. This investigation is carried out at the initial desorption pressure of 0.68-, 1.68- and 2.68 bar. The results of the optimal operating conditions in cases of 1.68-2.68 bar can be seen in Fig. 10 with the comparison to the initial absorber height at 2.5 bar. The results show that the double absorber height shows superior advantage to every previous cases considering specific energy demand. It can be observed in Fig. 10 that increasing the absorber height while maintaining a moderate desorber pressure (1.6 bar) gives lower, more preferable results compared to lower absorber height with increased desorber pressure. Other parameters also tend to vary with the desorption pressure change. At low desorption pressure the optimal operating points tend to be the ones with the lean loading between 0.16-0.18 while on elevated desorption pressure the optimal operating conditions are when the lean loading is set to 0.14-0.16.

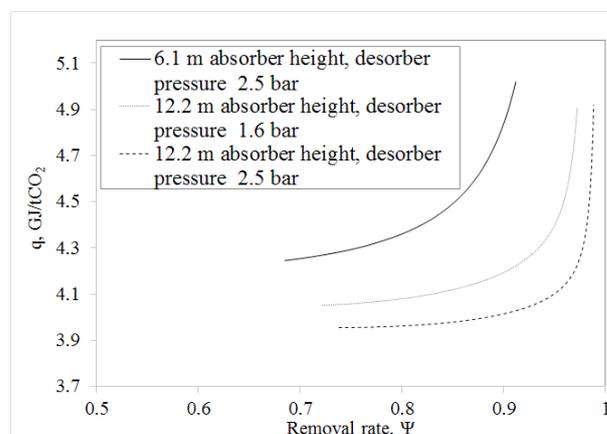


Fig. 10 Operating conditions at double absorber height on various desorber pressures

4 Conclusions

Post combustion carbon capture is one potential alternative for mitigating CO₂ emissions. Due to the varying composition of flue gases the removal efficiency is an important parameter of CO₂ capture that calls for the ability of flexible operation of a CO₂ capture plant. In case of efficiency increase, beside the overload of the existing system, a setup with multiple absorption steps can be considered that can eliminate possible thermal degradation of the solvent while achieving high removal efficiencies. The novelty of our work is that we consider several alternatives for CO₂ removal efficiency improvements. We evaluate these improvements and determine the best. The best performance can be achieved with the increase of the absorber column internal height and desorption pressure since these parameters are crucial for flexible plant operation especially at high removal efficiencies. Increasing the absorber column height and elevated desorption pressure can decrease the specific regeneration energy demand significantly. The results are in agreement with the classical engineering practice, that is, absorbers are designed and operated at low internal loads and overdesigned packings. This philosophy guarantees automatically the best solution for flexibility, that is, efficiency improvement.

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