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## Improvement of post-combustion carbon capture process in retrofit case

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### Abstract

Carbon dioxide emission, one of the core cause of global warming and other threats of nonreversible environmental effects, is in focus today. Among the several methods of  $CO_2$  emission mitigation, post-combustion carbon capture (PCC), based on absorber-desorber systems with amine absorbents, is one of the promising alternatives. The major anthropogenic  $CO_2$  point sources are power plants, natural gas extraction units, oil refineries, cement factories, biogas plants, etc. As these  $CO_2$  sources can be treated with a PCC absorber-desorber system, then these two units should cope with various conditions. To maintain a desired high removal efficiency, the capture unit must work in flexible conditions. The aim of this study is to evaluate the influence of the extent of the absorption of carbon dioxide on the overall performance of the acid gas removal process, considering that, generally, as the amount of removed  $CO_2$  increases, the separation becomes more challenging. In this work a verified computer based process model for the absorber-desorber system is used. Aspen Plus<sup>®</sup> professional flowsheet simulator is used for this purpose. Attention is paid on the proper operating parameters of a PCC absorber-desorber system for the case of increasing the  $CO_2$  capture efficiency up to 99%. As alternative solutions for these cases different scenarios are considered:

- the increase of the capacity of a classic configuration of an absorber-desorber system;
- the introduction of a second absorber column as extending the absorption column height;
- the possible use of two absorbers and two desorbers working in series.

This work deals with the detailed study on maximizing  $CO_2$  removal efficiency while maintaining the minimal energy consumption for the absorbent regeneration section.

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**Keywords:** carbon capture, MEA, absorption, removal efficiency, heat requirement, energy saving

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## 1. Introduction

Anthropogenic greenhouse gases (GHG) emissions lead to climate change and their effect on the natural environment poses one of the biggest challenges ahead to be dealt with. One possible technological approach to face this challenge is post-combustion carbon capture (PCC). This end of pipe technique is already in operation in other fields of the energy sector, such as natural gas sweetening to meet commercial specifications before being injected into pipelines. PCC approach is mainly criticized by its additional energy demand. The anthropogenic  $CO_2$  production is mostly emitted in flue gases [1–4]. Flue gases therefore are present in large volumes with low  $CO_2$  partial pressure. The several types of plants such as power plants, oil refineries, cement factories, etc., that produce flue gases operate at different loads depending on their energy output. This also results in various content and composition of produced flue gases [5]. To follow the behavior of the industrial plant, the capture plant must be built to operate flexibly and economically in various circumstances. Including the varying gas compositions, flow rate and capture efficiency flexibility as well [5]. Flexibility of the capture efficiency is particularly important because the efficiency in  $CO_2$  removal may vary according to the composition and the flow rate of the flue gas. The flue gas  $CO_2$  content may vary in the range of 3.875 - 22 vol%, while similar industrial gases, such as bio gas, may have an even higher  $CO_2$  content [5,6]. In case the gas to be purified has a low  $CO_2$  content at low pressure (atmospheric), the separation task is a difficult procedure due to the lack of driving force for mass transfer. In such a case the most recognized technique is the use of an absorber-desorber system applying chemisorption [7–9]. The materials used for such processes are found within the groups of amines [9–19]. In order to achieve acceptable removal efficiencies in pilot phase, the most frequently used solvent is monoethanolamine (MEA).

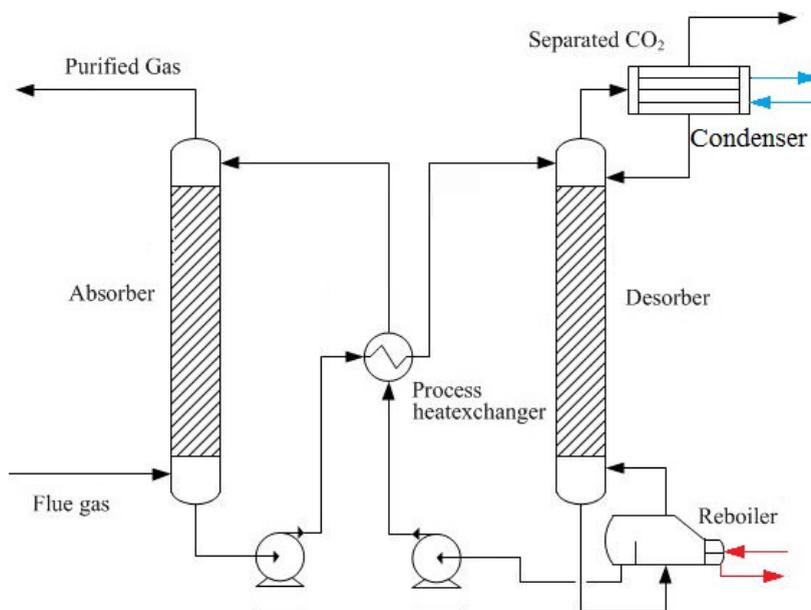


Fig. 1. Basic flow sheet of an absorber-desorber process.

The amine scrubbing capture process consists 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 towards the top. 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 and large surface area with low pressure drop is adopted in both columns. In the absorber,  $CO_2$  is absorbed since the concentration gradient drives  $CO_2$  to the liquid phase. As the loaded solvent leaves the absorber it is preheated and introduced into the desorber, whose operating pressure is one of the crucial parameters of the system. According to Gale et al. [20] the thermal degradation of MEA becomes economically significant above  $130\text{ }^\circ\text{C}$ , which can be reached for elevated pressure. On the other hand, according to Freguia [21], elevated

pressure (2-3 bar) 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  $CO_2$  partial pressure and with the introduced heat the concentration gradient is turned to drive  $CO_2$  back to the gas phase. Eventually, the steam and the liberated  $CO_2$  are separated in the condenser.

## 2. Modeling work

The thermodynamic framework used for the simulation is based on the ENRTL model of Aspen Plus<sup>®</sup>, that includes different assumptions for the representation of the physical behavior of liquid and vapor phases. In the vapor phase the presence of ions is neglected, since they are only present in the liquid phase. The SRK EoS is used to calculate the fugacity of the vapor phase (Soave, G 1972) [22,23] while non-idealities in the liquid phase are described by the Electrolyte NRTL model by Chen et al. [24–28].

The columns are modeled with the rate based model [29], which is superior to the traditional equilibrium model because it allows to better take into account, by means of a multi-physical approach, the different phenomena that plays a significant role on operation performances [30].

The initial model of this study in Aspen Plus<sup>®</sup> was originally verified by comparison with the pilot plant results obtained at the University of Texas [23,31]. Modification of the base model is carried out in order to have better simulation results. The flue gas conditions are shown in Table 1. The stream represents a sour gaseous stream coming from coal fired power plant.

Table 1. Flue gas parameters.

Content, m/m%	H <sub>2</sub> O	O <sub>2</sub>	N <sub>2</sub>	CO <sub>2</sub>
Flue gas	0.009	0.048	0.681	0.261
Temperature	59 °C			
Pressure	1.03 bar			
Flow	16.76 kmol/h			

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. Fig. 2 shows these different approaches. The top row (approach A) shows that the target value is the removal efficiency with adjusted parameter being the desorber heat and the varied parameter is the absorbent flow. In order to achieve the desired removal efficiency, the cyclic capacity is calculated. The minimal specific reboiler duty is found by determining the proper absorbent loading values. The absorbent loading is calculated according to eq.1.

$$\alpha = (CO_2 + CO_3^{2-} + MEACOO^- + HCO_3^-) / (MEA + MEA^+ + MEACOO^-) \quad (1)$$

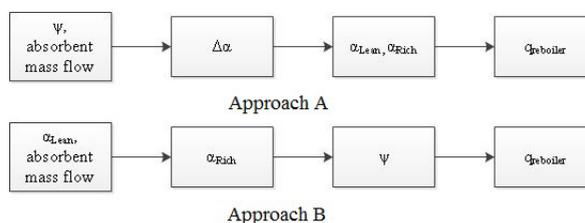


Fig. 2. Two approaches for finding operation points.

The second row (approach B) in Fig. 2 shows the case when the desired parameter is the lean absorbent loading ( $\alpha_{L,lean}$ ) 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 that allows to reach the initial absorbent loading. With the second approach, in the whole system the  $CO_2$  removal efficiency 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 fastest results for the optimal initial absorbent loading.

### 2.1. Removal efficiency improvement

Some alternative options can be considered to improve the removal efficiency of the carbon capture. In this work, the investigated scenarios are the following:

- first approach: intensification of the removal efficiency by increasing the load of the existing equipment;
- second approach: building a second absorber column as an extension of the existing absorber column;
- third approach: changing the desorption pressure;
- fourth approach: building a second absorber-desorber system that is able to further purify the partially treated flue gas to a practically  $CO_2$  free gas.

## 3. Results

### 3.1. Increased load

Results of the first approach are shown in Fig. 3. It can be observed that by increasing the lean loading ( $\alpha_{lean}$ ) of the absorbent, the specific regeneration energy decreases. To achieve high removal efficiencies  $\alpha_{lean}$  must be lowered to be able to absorb increased amount of  $CO_2$ . The tendencies show that higher removal efficiencies can be achieved by decreasing the initial absorbent loading. In this way the cyclic capacity ( $\Delta\alpha$ ) of the absorbent increases. Reasonably, low specific reboiler energies, around 4 GJ/t $CO_2$  are obtained, 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. 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.

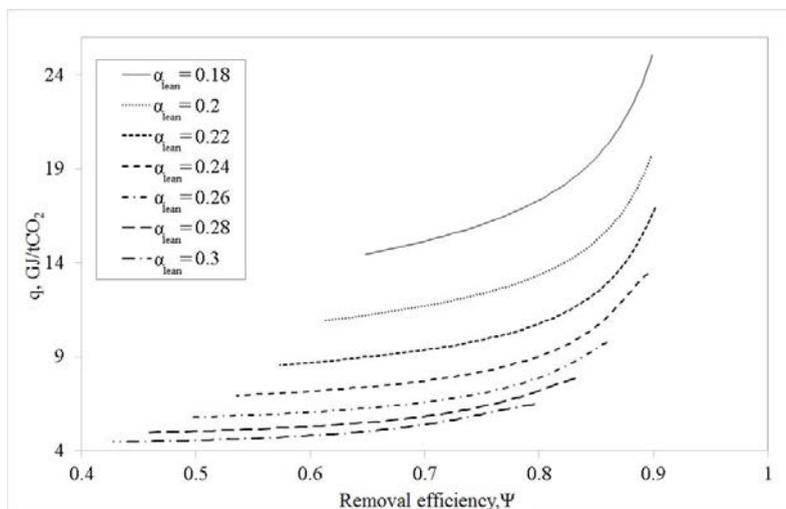


Fig. 3. Operating lines with one absorber-desorber column.

### 3.2. Second absorber-desorber step

In the high removal efficiency region, the specific regeneration energy can be decreased by introducing a second absorber-desorber column pair. In this case a two step gas treating system is formed, where the absorber-desorber column pairs operate loosely dependent on each other (see Fig. 4).

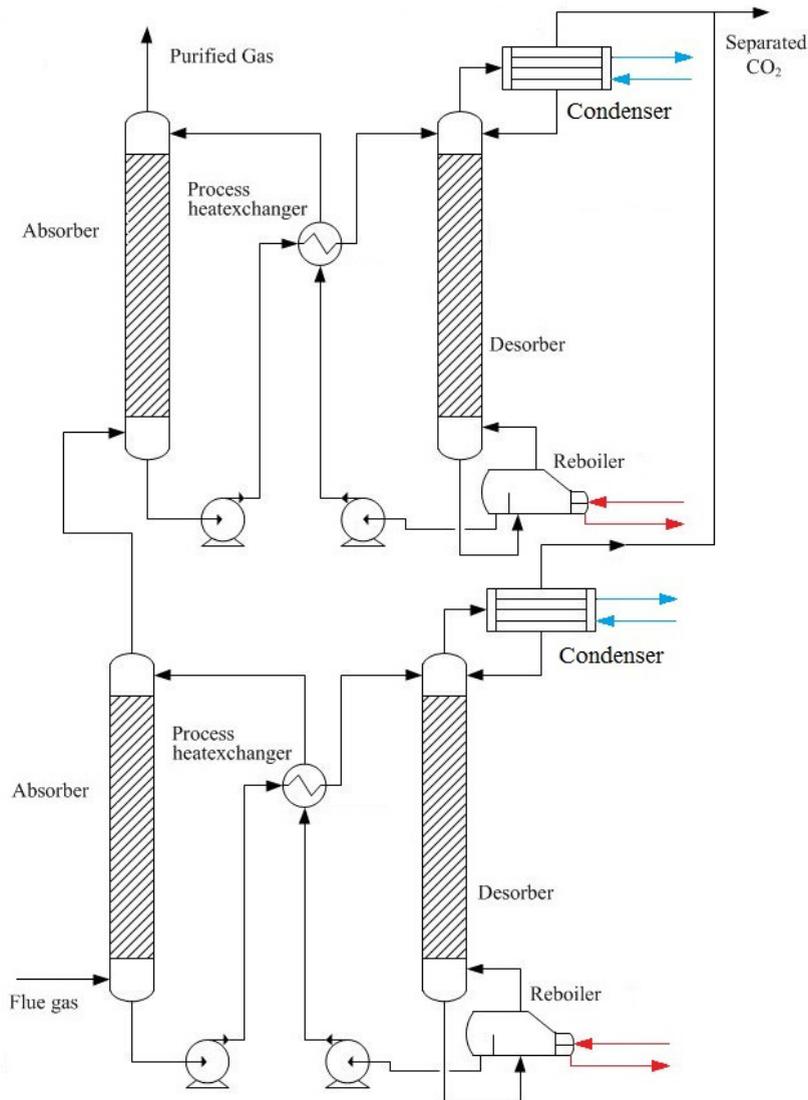


Fig. 4. Sematic layout of the 2 absorber and 2 desorber column configuration.

The study in this coupled system is as follows: the optimal operating conditions were found for the first capture step with optimization approach A (see Fig. 2) and the second capture step was investigated with optimization approach B. 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 affect the previous stages of the system [32,33]. In this 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  $CO_2$  removals equal to 65%, 76% and 85% is calculated. The purified gas mixture is then fed to the second column pair and purified further to higher  $CO_2$  removal rates. The specific energy demand is calculated according to equation 2.

$$Q_{spec} = (Q_{reb,1} + Q_{reb,2}) / (m_{CO_2,in} - m_{CO_2,out}) \quad (2)$$

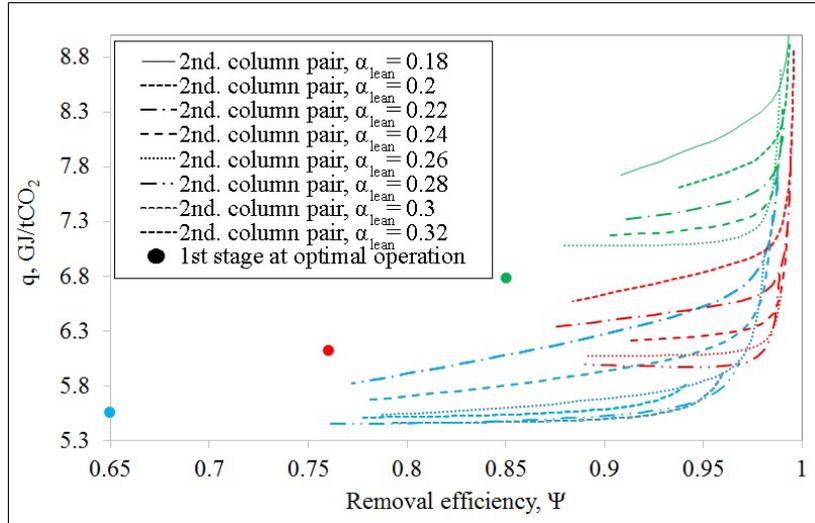


Fig. 5. Two step  $CO_2$  capture system with the optimized first step removal efficiency of 65% (blue dot), 76% (red dot) and 85% (green dot). The red, green and blue lines indicate the lean loading variation of the second capture step started from the color matching first step.

Fig. 5 shows the comparison between different sets of  $CO_2$  capture applying 2 step process. The optimal operating point of the first capture step is indicated by the coloured dots (at 65% (blue), 76% (red) and 85% (green)). The first capture step is calculated with calculation approach A (see Fig 2). The residue flow of the first capture step is introduced directly to the second absorber. In the second step the optimal operating condition is found by varying the absorbent mass flow and the initial absorbent loading with Approach B (see Fig 2). The variation of the initial loading of the absorbent has a high influence on the removal efficiency. Towards higher initial absorbent loadings the specific removal rate decreases until a certain value. For the case of first step 65% removal efficiency, the optimal initial absorbent loading appears to be at 0.3 mol $CO_2$ /molMEA. At higher initial loadings the liquid demand increases the specific heat demand. A similar trend can be observed with different first stage removal amounts. Fig. 5 shows the case if the first column pair operates at 76% removal efficiency before it enters the second step. 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 efficiency the overall specific energy demand will have a significantly higher value. By increasing the first step to a  $CO_2$  removal efficiency to 85%, as it is expected, the overall separation energy follows a rapid rising. Fig. 5 also shows that the optimal loading of the second separation stage varies on the basis of the composition of the gas mixture entering the second absorber column. As the  $CO_2$  content of the gas mixture decreases, assuming optimal operating conditions the initial absorbent loading decreases as well. The optimum at 65% of  $CO_2$  capture occurs at 0.3 mol $CO_2$ /molMEA, at 76% of  $CO_2$  capture is reduced to 0.28 mol $CO_2$ /molMEA and at 85% of  $CO_2$  capture is 0.26 mol $CO_2$ /molMEA.

### 3.3. Variation of the desorber pressure

The classic approach to an absorber-desorber system is to have the absorption at high pressures and desorption at low pressures. Finding the optimal desorption condition may be not easy. Following the classic approach, the driving force for desorption can be increased either by increasing the temperature or by decreasing the partial pressure of the  $CO_2$  in the desorber by generating more steam. If the temperature and the pressure is increased in the desorber column,

the equilibrium curve will shift away from the operating line and the mass transfer driving force will therefore increase. 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. Fig. 6 shows

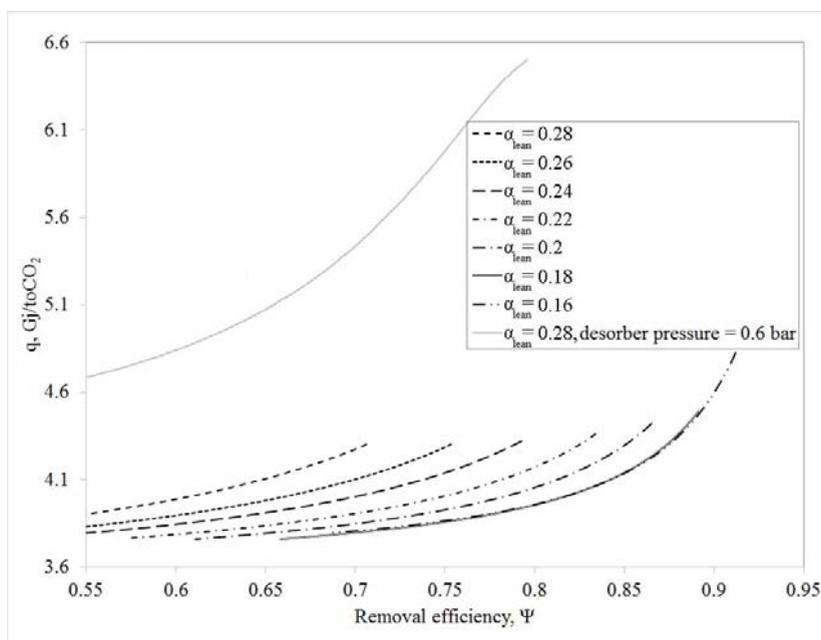


Fig. 6. Operating lines at increased desorber pressure (desorber pressure is set to 2.6 bar, except where other is indicated).

different operating conditions for different 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 is maintained at reasonable values. To this Fig. 6 shows the optimal operating condition when the reboiler was set to 0.6 bar and the initial loading was set to 0.28 mol $CO_2$ /molMEA. 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.

#### 3.4. Height extension of the absorber column

The effect of the variation of the desorber pressure and of the absorber height was studied in order to determine the mutual influence of these parameters on process performances. 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 was 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. 7 with the comparison to the initial absorber height at 2.5 bar. The results show that doubling the absorber height results more advantageous in terms of specific energy demand than every previous cases. In Fig. 7 it can be also observed, that increasing the absorber height while maintaining a moderate desorber pressure (1.6 bar) gives lower, more preferable results than those obtained considering a lower absorber height with increased desorber pressure. Moderately high desorption pressure means the utilization of lower quality heat. 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 at elevated desorption pressure the optimal operating conditions are obtained by setting the lean loading to 0.14-0.16 [34].

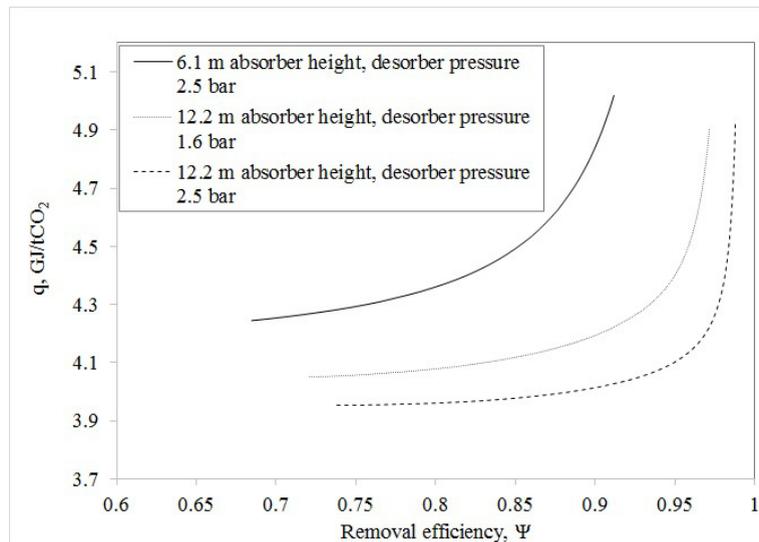


Fig. 7. Operating lines at double absorber height on various desorber pressures.

#### 4. Conclusions

Post-combustion carbon capture is a potential alternative to reduce  $CO_2$  emissions. The removal efficiency is an important parameter of  $CO_2$  capture that calls for the ability of flexible operation of a  $CO_2$  capture plant, since the composition of the flue gases may vary. In case of efficiency increase, beside the overload of the existing system, a setup with multiple absorption steps can be considered in order to eliminate possible thermal degradation of the solvent while achieving high removal efficiencies. The best performance can be obtained 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 the desorption pressure can lead to a significant decrease in the specific regeneration energy demand.

#### 5. Acknowledgement

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