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# Efficient Utilization of Industrial Excess Heat for Post-combustion $CO_2$ Capture: An Oil Refinery Sector Case Study

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**Abstract**

A key issue in post-combustion carbon capture is the choice of absorbent. In this paper two different absorbents, monoethanolamine (MEA) and ammonia ($NH_3$), have been modeled in Aspen Plus at different temperatures for possible implementation at an oil refinery. The focus of investigation is the possibilities of heat integration between the oil refinery and the carbon capture process and how these possibilities could change in a future situation where energy efficiency measures have been implemented.

The results show that if only using excess heat from the refinery for heating of the carbon capture process, the MEA process can capture more $CO_2$ than the $NH_3$ process. It is shown that the configuration requiring least supplementary heat when applying carbon capture to all flue gases is MEA at 120 °C.

The temperature profile of the excess heat from the refinery suits the MEA and $NH_3$ processes differently. The $NH_3$ process would benefit from a flat section above 100 °C to better integrate the heat needed to reduce slip, while the MEA process only needs heat at stripper temperature.

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## 1. Introduction

Since the industrial processes emitting $CO_2$ are inherently different, different carbon capture technologies are required. Case studies of various sectors (power, cement, oil refineries etc.) are therefore needed to determine a good match between the industrial process and the capture technology. This paper focuses on post-combustion capture through chemical absorption in the oil refining sector.

Monoethanolamine (MEA) has been used for many years to separate $CO_2$ in the gas processing industry, and has therefore become the benchmark to which other $CO_2$ capture technologies are compared. Two of the most important characteristics of a good absorbent of $CO_2$ are a low heat of reaction with $CO_2$ (low specific heat demand) and a low temperature for the regeneration process. The regeneration temperature is of special interest when excess heat is available, since the amount of excess heat may be considerably larger at lower temperatures. The residual heat from the $CO_2$ emitting process can be used to cover parts of or the whole heat requirement of the $CO_2$ capture process.

Ammonia ($NH_3$) emerged as a promising solvent in the beginning of the 21th centry [1], mainly due to its persistence to degradation, which is a major problem for alkanolamine-based solvents. The most notable disadvantage with $NH_3$ is its high vapour pressure, which result a loss of solvent, ammonia slip. The company Alstom is developing a proprietary $NH_3$-based post-combustion capture process called “the chilled ammonia process” (CAP). In a first configuration the absorber was chilled to very low temperatures (0-20°C), which would allow precipitation to occur in the solvent and a low slip of $NH_3$. However, recent developments have deemed this configuration too cumbersome and a design similar to that of the MEA-based process is suggested [2].

Early evaluations of $NH_3$ as a solvent have suffered from optimistic estimations of the heat requirements for $NH_3$ regeneration where as low requirements as 900-1000 kJ/kg $CO_2$ captured was reported [1,3]. These estimates have later been revised and more recent figures reports a heat requirement of approximately 2500 kJ/kg $CO_2$ captured [2]. MEA is a commercially used solvent and as such has a well mapped heat requirement for regeneration of around 3300 – 3800 kJ/kg $CO_2$ captured[4–6]. Excess heat utilization could prove to significantly reduce the primary energy demand of the $CO_2$ capture process [7,8].

When excess heat is used to cover some of the heat requirement of the capture process it is important to evaluate most suitable operating temperature/pressure of the stripper. The heat requirement as a function of stripper conditions has been studied in Oyenekan & Rochelle [9] who investigated reboiler duty of three different stripper configurations when applying CCS to a coal power plant. The configurations were simple stripper, multipressure stripper and vacuum stripper (30 kPa, 60 – 80 °C). They concluded that for MEA the multipressure stripper resulted in the lowest reboiler duty, and that the highest reboiler duty was associated with vacuum stripping, approximately 22 – 62% higher penalty than for standard pressure stripping. Berstad et al. [10] varied the reboiler temperature between 107 and 142 °C when applying CCS to three different applications: a coal power plant, a natural gas combined cycle (NGCC) and a biomass power plant. The study concluded that the low temperature cases resulted in a lower efficiency penalty compared to the reference case for both the NGCC and the biomass plant due to the use of steam at lower pressure in the regeneration process. In addition to this Abu-Zahra et al. [4] performed a study varying the stripper temperature between 108 and 128 °C, determining that the lowest energy demand is present when regenerating MEA at 128 °C. The heat requirement as a function of stripper pressure has rarely been subject evaluation for the $NH_3$ process. In the work by Linnenberg et al. [11] it was concluded that the heat requirement for the stripper is slightly decreasing for a higher pressure. However, when integrated with a power plant net efficiency decrease would reach a minimum at a stripper pressure at 4 bar, due to the permission to use low quality steam from the steam cycle.

Since none of these studies took into account the benefits of using excess heat to decrease the primary heat demand of the process, the authors of this paper conducted a heat integration study where an MEA-based capture plant was heat integrated with an oil refinery while varying the temperature between 90 and 120 °C [12]. The paper concluded that when being restricted to a small amount of heat collecting units the drawbacks of an increased heat demand of the capture plant due to lower stripper temperature outweighs the benefit of being able to use more excess heat. Benefits were however present when being able to utilize the full excess heat potential of the refinery.

Currently, incentives are being set in place in an effort to increase energy efficiency in industry sector. As the implementation of CCS is halted by unclear future policies it is believed to be deployed on a medium term time scale whereas energy efficiency measures are likely to be implemented before CCS. The implementation of energy efficiency measures has at least two implications on excess heat driven CCS. The most tangible is the loss of extractable excess heat. However, this is partly counteracted by the fact that less $CO_2$ will be produced due to a

lower fuel demand. Case studies on current systems as well as future situations are therefore essential to map the possibilities of using excess heat for the CCS process in a future perspective.

This paper highlights the possibilities of decreasing the primary heat demand through heat integration when performing post-combustion $CO_2$ capture at an oil refinery with either MEA or $NH_3$ as absorption fluid. The authors want to illustrate how the availability of excess heat can change the operating parameters of the CCS process by investigating two different absorbents at varying temperature. In addition to the two absorbents, heat integration is performed at two different levels of excess heat availability to simulate possible future energy efficiency measures.

Aspen Plus is employed to evaluate the heat requirement of both capture processes at different operating conditions of the regeneration (e.g. pressure and temperature). The results will reveal at which stripper operating conditions the difference between the heat requirement of the capture processes and the available excess heat are at a minimum. This analysis will result in an identification of a suitable heat integration strategy for carbon capture at the present oil refinery both in a current and future perspective.

## 2. Methodology

The methodology in this paper is divided into two stages. In the first stage models were developed for both the MEA and $NH_3$ processes and in the second step these models were used in conjunction with operating data from an oil refinery to perform a heat integration study. The main process operating parameter studied in this paper is stripper temperature.

### *2.1. Modelling work*

The models presented in this paper have both been published in previous works by the authors [2,12,13]. The models are modified to include the following key assumptions:

- Flue gas composition with 11.8 % $CO_2$ after dewatering (10.8 % before dewatering) [14]
- Minimum temperature difference ($\Delta T_{min}$) in lean/rich heat exchanger of 10 K
- Basis of 400 000 t $CO_2$/y with an annual operating time of 8 200 h.

For MEA and $NH_3$ respectively, three stripper temperatures have been modelled. The MEA process has been modelled at stripper temperatures of 90, 105 and 120 °C whereas the $NH_3$ process has been modelled at temperatures of 105, 120, 135 and 155 °C. The $NH_3$ process has not been modelled for 90 °C due to the extensive $NH_3$ slip. The slip increases with decreasing temperature and has a noticeable impact already at 105 °C.
Both the MEA and $NH_3$ processes were simulated in Aspen Plus using the default property method packages for each absorbent. Table 1 presents the most important relevant process operating conditions for both processes. For a more detailed description of the MEA and $NH_3$ process models see Andersson et al. [12] and Jilvero et al. [2].

Table 1: Operating conditions for the MEA and the $NH_3$ processes.

| | MEA process | $NH_3$ process |
|---|---|---|
| Absorbent concentration ($CO_2$-free) (wt%) | 30 | 14.3 |
| Lean $CO_2$ loading (mol $CO_2$/mol abs.) | 0.32 | 0.25 |
| Rich $CO_2$ loading (mol $CO_2$/mol abs.) | 0.497 | 0.5 |
| $CO_2$ Capture efficiency (%) | 85 | 85 |

Figure 1 presents a general schematic of the MEA and NH3 processes. Both processes are based on the same absorber/stripper setup. However, the fact that $NH_3$ is considerably more volatile than MEA requires extra consideration in the design of the capture process. An extra absorber (A2) is used to reduce $NH_3$ discharge from the absorption process. The remaining ammonia in the flue gases are captured and retained by an $NH_3$ abatement cycle formed by a water-wash (A3) and an $NH_3$ stripper (S2). There is also a heat requirement for the $NH_3$ stripper which needs to be considered in the heat integration. The operating temperature of the $NH_3$ stripper is 100°C. Extra attention is also put to the stripper condenser where a water-wash condenser is used to eliminate any possible solid precipitation.

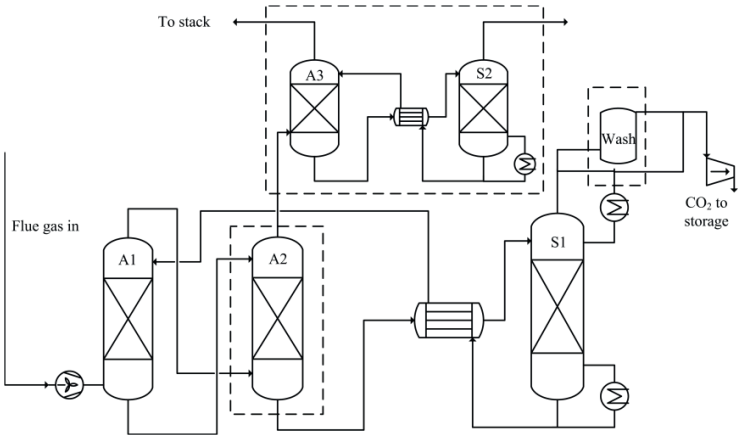


Figure 1: General setup of the MEA/$NH_3$ absorption processes. Equipment within dashed lines is only used in the $NH_3$ process. A1-3 are absorbers, S1-2 are strippers and the wash section is used in the $NH_3$ process instead of a condenser.

### *2.2. Reference oil refinery plant*

The focus of this case study is a complex oil refinery, emitting approximately 1.8 Mt $CO_2$/y, situated on the west coast of Sweden. The main part (1.74 Mt $CO_2$/y) of the $CO_2$ is emitted from 4 chimneys situated in the middle of the refinery and these are the chimneys deemed available for CCS in this study. The $CO_2$ concentration in the flue gases ranges from 6.7% to 24% before dewatering [14].

The oil refinery has a primary heat demand of 409 MW but a detailed heat integration study revealed that theoretical minimum heat demand is only 199 MW [15]. Thus, the theoretical heat savings amount to 210 MW although this potential is neither economically nor practically feasible to implement. In this work it is assumed that 20 % (42 MW) of the proposed energy efficiency measures can be implemented in a medium-term time perspective. The energy savings are assumed to be evenly distributed in the temperature interval 170 – 130 °C and affect both the $CO_2$ emissions from the refinery (due to fuel savings) and the extractable excess heat.

The available excess heat in the two cases investigated is graphically presented in an so called “actual cooling load curve” (ACLC) [16], see Figure 2. The ACLC is constructed by adding all heat available in hot streams that are cooled by air or cooling water and plot the sum of heat against the temperature at which it is available. Flue gas currently not used to produce steam is also included in the analysis. The amounts of heat available are extracted from real plant data, thus no restrictions have been set as to how the heat is collected.

When calculating the $CO_2$ emission savings from decreased fuel use, a mixture of 50 % propane and 50 % butane is assumed as fuel. The boiler producing steam is assumed to have an efficiency of 0.9.

### *2.3. Heat integration*

Heat from the process is assumed to be supplied to the reboiler via a heat collection system. The system consists of a water trunk pipeline leading condensate from the capture plant to the refinery process. The trunk pipeline branches out to a number of heat exchangers where the condensate is vaporized while refinery process streams are cooled. The steam is then lead back to the capture plant through a trunk pipeline. The heat collection system uses 10 K as a minimum temperature difference over the system, which in practice means that all streams in the refinery seen as 10 K below their actual temperature when extractable excess heat is mapped. The mapping of heat flows in the refinery processes is carried out first for the present situation and then for a future situation where energy efficiency measures have been taken. The energy efficiency measures of 42 MW steam changes the appearance of the ACLC. In Figure 2 the ACLCs before (current situation) and after (future situation) energy efficiency measures are shown.

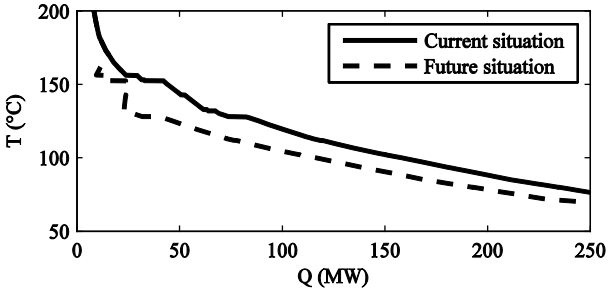


Figure 2: The ACLC before and after energy efficiency measures are carried out.

In the mapping of heat in within the capture process, the minimum allowed temperature difference in a heat exchanger consists of the sum of one contribution from each side in the unit. For condensing/evaporating streams this contribution is 2.5 K, for liquid streams 5 K and for gaseous streams 7.5 K. The mapping of heat flows in the capture process is done by Pinch technology. Pinch technology is a systematic analysis of heat demands in an industrial process, and is based on first and second law of thermodynamics. The method consists of systematic collection and treatment of heat flows and can be used as a targeting method towards a theoretical minimum heat demand. The process´ minimum heat demand is calculated for a given minimum temperature difference in the heat exchanger network $\Delta T_{min}$ and the methodology also gives indications on suitable heat exchange within the process. For a comprehensive methodology description, see Smith [17].

The largest heat demands in the two processes are the stripper reboiler (for both processes) and for the $NH_3$ process also the $NH_3$ abatement cycle stripper, which removes the slip from the absorber.

The outcome from the heat integration study is how much excess heat that can be collected from the refinery to the MEA and $NH_3$ plants at the different temperature levels, how much $CO_2$ that can be captured using only excess heat and also the need for supplementary heating if choosing to capture 85 % of the $CO_2$ from the 4 main chimneys.

## 3. Results

The modelling of the MEA and the $NH_3$ processes yields the heat demands of the $CO_2$ capture processes. Implementation of energy efficiency measures would result in approximately 100 kt less $CO_2$/y which corresponds to 6 % of the annual $CO_2$ emissions. The amounts of $CO_2$ to be captured are therefore 1.48 Mt/y if CCS is deployed before energy efficiency measures and 1.40 Mt/y if CCS is deployed after energy efficiency measures, resulting in a 5 % decrease in heat demand. The heat demands as well as available amounts of excess heat are shown in Figure 3.

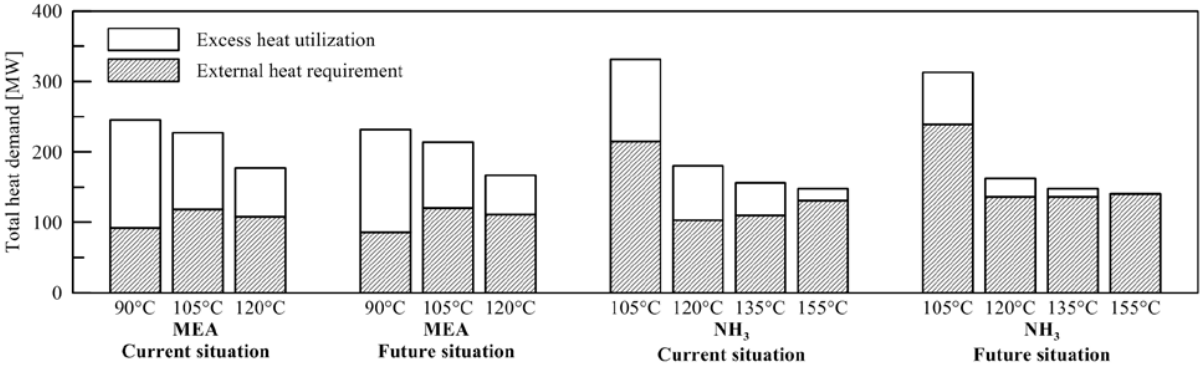


Figure 3: Heat demands from the CCS capture plant for before (Current situation) and after (Future situation) energy efficiency measures are being implemented.

It is evident that $NH_3$ is sensitive to operating at temperatures below 120 °C. As expected, and also as has been shown in previous work, the heat demand increases for all absorbents when the stripping temperature is decreased. However, the magnitude of the change for $NH_3$ when shifting down to 105 °C is larger than expected.

Figure 4 shows the equilibrium conditions at the top of the stripper as a function of temperature with $NH_3$ (a) and MEA (b) as absorbent of $CO_2$. As a representative case for both the MEA and $NH_3$ processes a $CO_2$ loading of 0.5 and a vapour fraction of 0.05 is shown. The flow of solvent and water varies considerably relative the flow of $CO_2$ to compression (index of 1). For the MEA system, the dominating component is water; MEA is not present in the gas phase. The amount of water evaporated with every kg of $CO_2$ is increasing with decreasing temperature, which means that the heat of regeneration is increased. For the $NH_3$ system $CO_2$ is the dominating component into the compressor from 90 °C and upwards. However, at lower temperature almost as much $NH_3$ as $CO_2$ exits the stripper. The ammonia needs to be separated from the carbon dioxide before compression. This can be achieved by washing the gas with water, which would cause some of the carbon dioxide to be absorbed and increase the heat required per kg $CO_2$ captured. This means that both processes are penalized by a lower stripper pressure, but in different ways. The MEA process suffers from a high evaporation of water while the $NH_3$ process suffers from loss of absorbent.

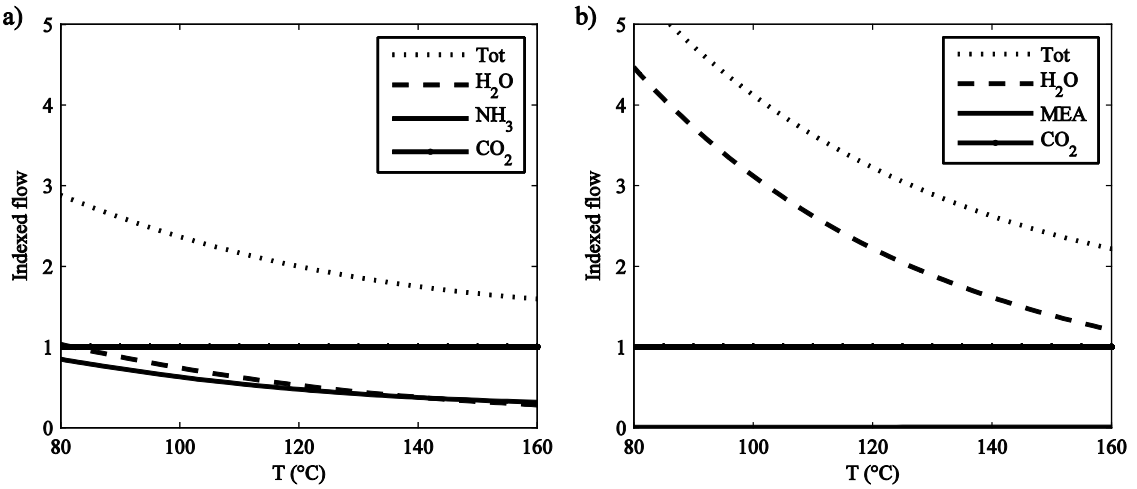


Figure 4: Equilibrium of gas phase of an absorbent/$CO_2$/$H_2O$ mixture at varying temperatures and a vapour fraction of 0.05. The absorbents are a) $NH_3$ and b) MEA.

A Background/Foreground analysis shows the heat flows of the processes that are being integrated with each other in the same figure. The overlap between the two curves is the amount of heat that can be recovered from the background process (in this case the oil refinery) to cover a demand in the foreground process (the CCS plant). Between the refinery process and the CCS process there needs to be a heat collecting system, as described in Andersson et al. [12], and in order to account for the $\Delta T_{min}$ the curves have been shifted 10 K. Figure 5 shows the background/foreground analysis of the $NH_3$ process at 105 °C and 135 °C respectively. When comparing the graphs representing the $NH_3$ processes in Figure 5 they both have the largest deficit of heat at the required stripper reboiler temperature. The slip of $NH_3$ from the absorber increases as the stripper temperature decreases. This is due to that the slip of $NH_3$ is higher as the temperature of the stripper decreases and thus more $NH_3$ needs to be added to the lean stream as make-up. Thus, the partial pressure of $NH_3$ is increased.

Background/foreground analyses have also been made for the different setups of the MEA system and the resulting figures for 90 °C and 120 °C are shown in Figure 6. The heat profile of the MEA process does not change to the same extent as for $NH_3$ but is altered due to higher heat demand.

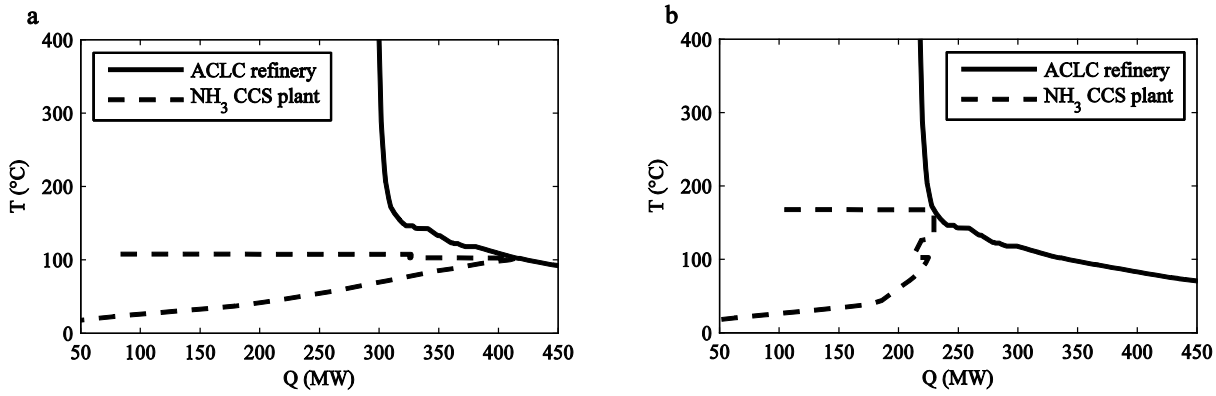


Figure 5: Background/foreground analyses showing the energy demand and the heat integration possibilities for the $NH_3$ process at a stripper temperature of a) 105 °C and b) 135 °C.

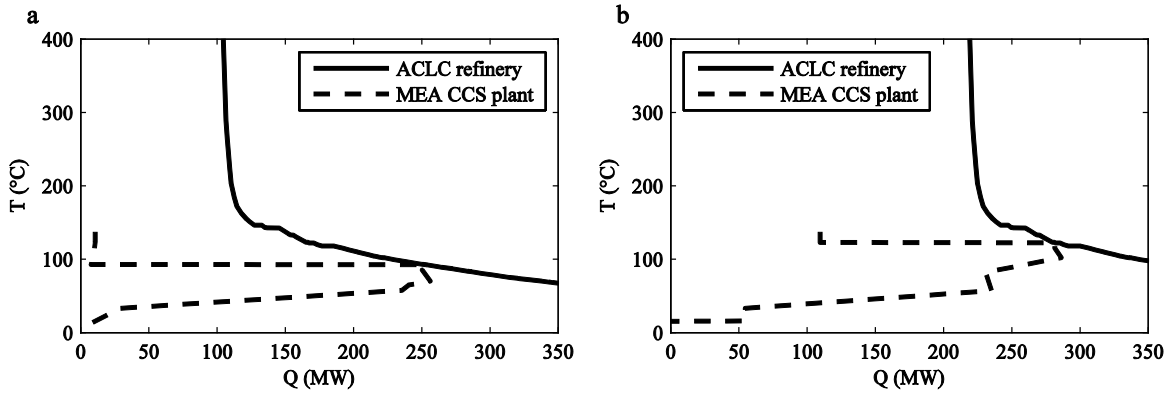


Figure 6: Background/foreground analyses showing the energy demand and the heat integration possibilities for the MEA process at a stripper temperature of a) 90°C and b) 120 °C.

For both setups of the MEA process the pinch point (where the two curves collide) is located at stripper temperature, which means that all available heat from the refinery process streams may be used in the stripper. This can be compared to the $NH_3$ process at 105 °C shown in Figure 5a where the pinch point is located at abatement cycle temperature, which means that the abatement cycle requires more heat than what is available at its temperature level and it will require some of the heat that could have been used in the stripper, increasing the need for primary heat to the CCS plant.

In Figure 5 and 6 the amount of heat that can be recovered from the refinery processes to the CCS process is illustrated by the overlap between the refinery ACLC and the CCS process curve. The remaining heat demand has to be covered by an external source e.g. a boiler or a heat pump. After performing a background/foreground analysis for the different setups in this study the remaining heat demands are shown in Figure 3. The remaining heat demand can be satisfied with a variety of boilers or a heat pump, but the refinery can also decide to separate only as much $CO_2$ as can be covered by the available excess heat. Table 2 shows how much $CO_2$ that can be separated using only excess heat, and also how much $CO_2$ that would remain from the 4 main chimneys after CCS is implemented. The results presented give the somewhat paradoxical conclusion that energy efficiency combined with CCS in this case actually would result in larger $CO_2$ emissions than only performing CCS. Only if operating with MEA at a stripper temperature of 90 °C do the $CO_2$ emissions decrease more for the combined measures. For the 90 °C MEA case the heat demand is larger than for the other processes, and the 42 MW of fuel that could be saved give rise to more $CO_2$ emissions than what could be captured using 42 MW of fuel for carbon capture. The results in Table 2 are sensitive to assumptions regarding fuel mix and temperature span of energy efficiency measures.

Table 2: The table shows how much $CO_2$ that can be captured using only excess heat, and how much that will be emitted to the atmosphere or to be captured using primary heat.

| Temperature (°C) | 90 | | 105 | | 120 | | 135 | | 155 | |
|---|---|---|---|---|---|---|---|---|---|---|
| | $CO_2$ capt. | $CO_2$ rem. | $CO_2$ capt. | $CO_2$ rem. | $CO_2$ capt. | $CO_2$ rem. | $CO_2$ capt. | $CO_2$ rem. | $CO_2$ capt. | $CO_2$ rem. |
| MEA Current Situation ($ktCO_2/y$) | 920 | 560 | 710 | 770 | 580 | 900 | - | - | - | - |
| MEA Future Situation ($ktCO_2/y$) | 880 | 520 | 610 | 790 | 470 | 930 | - | - | - | - |
| NH3 Current Situation ($ktCO_2/y$) | - | - | 520 | 960 | 630 | 850 | 440 | 1040 | 170 | 1310 |
| NH3 Future Situation ($ktCO_2/y$) | - | - | 330 | 1070 | 230 | 1170 | 110 | 1290 | 0 | 1400 |

## 4. Discussion

Industrial based CCS is a key technology to reduce $CO_2$ emissions to a sustainable level and post-combustion CCS is the most flexible technology since it is an end-of-pipe solution that has no implications on the rest of the industrial process. Post-combustion CCS is, however, expensive, with the main cost being attributed to regeneration heat for the absorbent. A scenario where an industrial plant captures as much $CO_2$ as possible with internal heat and emits the remaining part of the $CO_2$ may therefore be of interest. This approach may, as pointed out in connection to Table 2, result in the counterintuitive effect that the implementation of energy efficiency measures increases the emission of $CO_2$. However, energy efficiency measures will always lead to lower generation of $CO_2$. It is therefore important to consider the capture unit when optimizing the plant. It is, however, also important to consider the time perspective as energy efficiency measures are being implemented at present and in the near future, while CCS will probably not be deployed at a large scale for 10-20 years. One could also argue that efficiency measures should target low temperature streams, since heat at higher temperature is more usable. In future works investigations regarding temperature span of the energy efficiency will be done to see how large impact this has on the results.

The availability of excess heat can change what characteristics that are desirable in the CCS process. Looking at a stripper temperature of 120 °C the MEA process has a somewhat lower heat demand than $NH_3$ before integration, but after integration the remaining heat demands show opposite relation. This is due to the fact that, in comparison to the MEA process, more heat in the $NH_3$ process is attributed to the $NH_3$ abatement cycle which operates at a lower temperature than the stripper. When the pinch point is located at the stripper temperature the heat demand for washing can be completely covered and does not increase the heat demand of the combined processes. For MEA the heat demand occurs almost entirely in the stripper and the possibilities for heat integration thus occurs at this temperature level. If, however, the temperature level of 105 °C is investigated the benefits of having two different heat demands are not as significant, as the amount of excess heat that can be used in the $NH_3$ and MEA processes are very similar. As previously stated the $NH_3$ abatement cycle has a constant temperature independent of stripper temperature, and Figure 5 shows that when having a stripper temperature as low as 105 °C the temperature difference between the $NH_3$ abatement cycle and the stripper is small, cancelling the possibilities to improve heat integration by doing it at different temperatures. It should be investigated further if the $NH_3$ process has a point between 105 °C and 120 °C where the benefit of having the reboiler heat demand and $NH_3$ abatement cycle heat demand can be more utilized. The process at 120 °C could sustain a higher slip since there is unused heat at temperatures corresponding will to the $NH_3$ abatement cycle.

Modeling the MEA process at a stripper temperature of 90 °C is associated with some uncertainty. Very little experimental work has been done at such low temperatures and the sources available for validation are therefore limited. Some work has however been carried out, for instance Dugas [18] but more in depth studies would be necessary in order to successfully operate a full size plant at this temperature.

## 5. Conclusions

For the system presented in this case study an MEA process operated at a stripper temperature of 90 °C was shown to have the smallest need for supplementary heating when capturing 85 % of the $CO_2$ in the flue gases.

Implementing energy efficiency measures in an oil refinery can have implications on design of the CCS process. The preferred operating parameters of the CCS process changed after energy efficiency was improved.

Dividing the heat demand into two different units with different temperature levels (the $NH_3$ process) did not give the desired gains in heat integration for the cases investigated.

The MEA and $NH_3$ capture processes are affected by the excess heat temperature profile in different ways. If the temperature profile is steep and the sections between stripper temperature and 100 °C none of the $NH_3$ abatement cycle can be integrated and MEA could then be beneficial. If the temperature profile is flat below stripper temperature and 100 °C the $NH_3$ process could benefit from being able to integrate the slip of $NH_3$.

Implementing energy efficiency measures increased $CO_2$ emissions from the refinery when at the same time implementing $CO_2$ capture. This is valid when not using supplementary heating.

The stripper in the $NH_3$ process is difficult to operate below 120 °C. This is mainly due to that the increased slip of $NH_3$ from the stripper lead to a cumbersome and energy-intensive process.

## 6. Acknowledgements

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