

Aspen HYSYS Simulation of CO₂ Removal by Amine Absorption from a Gas Based Power Plant

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Lars Erik Øi

Telemark University College, Norway

lars.oi@hit.no

Abstract

A simplified combined cycle gas power plant and a MEA (monoethanol amine) based CO₂ removal process have been simulated with the process simulation tool Aspen HYSYS. The thermodynamic properties are calculated with the Peng Robinson and Amines Property Package models which are available in Aspen HYSYS. The adiabatic efficiencies in compressors, gas turbines and steam turbines have been fitted to achieve a total thermal efficiency of 58 % in the natural gas based power plant without CO₂ removal. The efficiency is reduced to about 50 % with CO₂ removal. The CO₂ removal in % and the energy consumption in the CO₂ removal plant are calculated as a function of amine circulation rate, absorption column height, absorption temperature and steam temperature. With CO₂ removal of 85 %, heat consumption is calculated to 3.7 MJ/kg CO₂ removed, close to a literature value of 4.0 MJ/kg CO₂.

Keywords: Aspen HYSYS, CO₂ removal, monoethanol amine, absorption.

1. Introduction

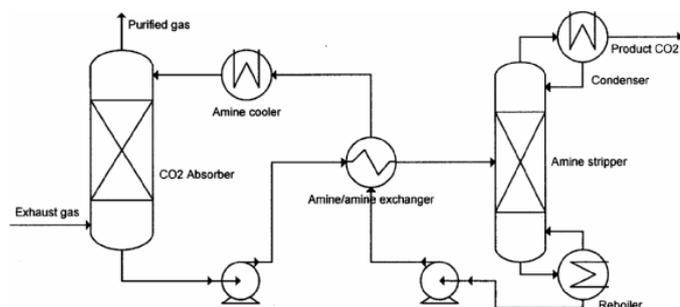


Figure 1. Principle for CO₂ removal process based on absorption in amine solution.

The possibility of removing CO₂ from fossil fuel based power plants has got increased interest due to environmental reasons. The most actual method for CO₂ removal is by absorption in an amine based sol-

vent followed by desorption. The principle of the removal process is shown in figure 1. The simplest and most used amine for CO₂ removal is MEA (monoethanol amine). This removal process has a high consumption of thermal energy, and one of the main aims of improvement is to minimize this energy consumption. In the case of CO₂ removal from a combined cycle gas power plant, it is natural to cover the heat requirement for CO₂ stripping with steam from the power plant. A possible steam outlet from a typical combined cycle process is shown in figure 2.

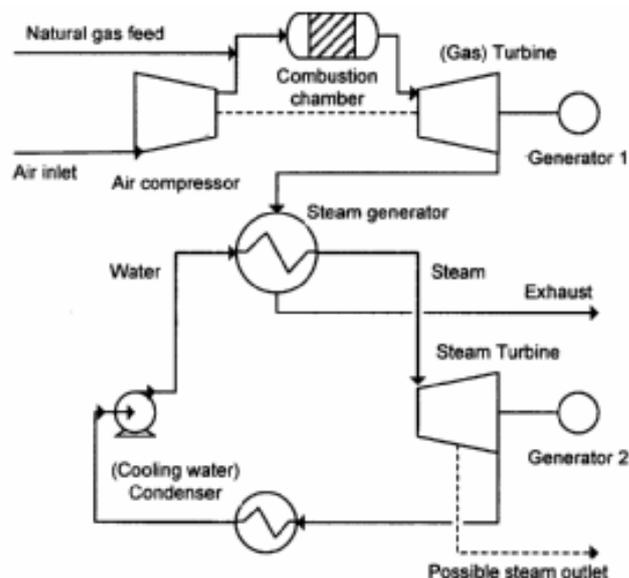


Figure 2. Principle of combined cycle power plant

Because testing at large scale is so expensive, it is natural to use process simulation to evaluate such processes. There are however few literature references on process simulation of CO₂ removal from exhaust gases at atmospheric pressure. There are some journal articles [1-4], and 3 of them have used the process simulation program Aspen Plus. AspenTech bought the program HYSYS from HyproTech in 2002, and in 2006 the program name was changed to Aspen HYSYS. The last reference [4] uses a Fortran code to simulate the process. An im-

portant advantage with using a process simulation program for such calculations, is that the available models for thermodynamic properties can be used. Aspen Plus has an MEA property insert model [5] which was used in all the references [1-3]. Aspen HYSYS has an Amines Property Package [6]. Within the Amines Property Package, one of the two models, Kent Eisenberg or Li-Mather, can be selected. Even though Aspen HYSYS is probably the most used process simulation program in the world, there has not been found any journal references on CO₂ removal from atmospheric exhaust gas using this program. There is however much literature on CO₂ removal from natural gas at high pressure. Other process simulation programs containing amine packages, are ProVision and ProMax.

At Telemark University College, Hysys has been used since 2000 to simulate CO₂ removal from gas based power plants. Most of this work has been Bachelor and Master student projects with the author of this paper as supervisor. Major challenges in the simulation of CO₂ absorption and desorption processes, are the description of thermodynamics and absorption efficiency, convergence and total energy or cost optimization.

The purpose of this paper is to present and discuss the use of Aspen HYSYS for simulating CO₂ removal from atmospheric exhaust by amine absorption, and to simulate the total effect of efficiency reduction in a combined cycle gas power plant.

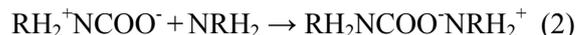
2. Available models

2.1 Absorption and reaction mechanisms

The details of the mechanisms of CO₂ absorption into an amine solution in an absorption column are quite complex. There are many references about the chemistry involved in the process, and many references and models comprising mass transfer mechanisms and chemical reaction kinetics. Reviews are written by Danckwerts and Sharma [7] and Versteeg [8].

First, CO₂ has to be transported from the gas to the liquid surface, and then it is absorbed in the liquid solution. The gas liquid interface area a (in m²/m³) and liquid holdup h (in m³/m³) are main parameters in describing such mechanisms.

In the liquid, the CO₂ may react chemically with other components. The following reactions are normally assumed to take place when CO₂ reacts in a primary amine like MEA in an aqueous solution. In the case of MEA (NH₂C₂H₄OH), R is C₂H₄OH.



According to equation (1) and (2), two moles of MEA are necessary to absorb one mole of CO₂.

A simple overall description of the combined absorption and reaction process is simply



The removal of CO₂ is not 100 %. The % CO₂ removal is limited both by low absorption and reaction rates and by the equilibrium conditions.

If the kinetics in the reactions should be calculated, more details about the intermediate reactions in equation (1) and (2) should be included. This is done in the MEA property insert model in Aspen Plus.

The simulation program Aspen HYSYS is mainly based on equilibrium calculations. In that case, equation (3) is sufficient to calculate the absorption process.

2.2 Equilibrium models

The concentration of CO₂ in the gas may be expressed by the partial pressure p_{CO_2} (eg. in bar) and the concentration in the liquid may be expressed by C_{CO_2} (eg. in mole/m³).

The equilibrium between the concentrations of CO₂ in a gas and a liquid may be represented as a function

$$p_{\text{CO}_2} = f(C_{\text{CO}_2}) \quad (4)$$

This expression might be a function of temperature, pressure and concentrations of the components in the solution. There are many models available to describe this function.

In Aspen HYSYS, gas/liquid equilibrium for a component (i) is normally calculated using k-values defined by the equation

$$K_i = y_i/x_i \quad (5)$$

where y_i and x_i are the mole fractions of (i) in the gas and liquid phase. For general purpose use, equation of state models like SRK (Soave Redlich Kwong) and PR (Peng Robinson) are often used. Aspen HYSYS recommends Peng Robinson [9]. Peng Robinson is regarded to be suitable to handle systems containing hydrocarbons, water, air and combustion gases, the typical components in a natural gas based power plant.

Traditional equation of state models are not regarded to be suitable for non-ideal liquid systems. An amine solution is an electrolytic system and also

comprises chemical reactions. This is not expected to be well described with traditional equations of state.

Within the Amines Property Package in Aspen HYSYS, one of the two models, Kent Eisenberg [10] or Li-Mather [11], can be selected. The models are quite complex, but in principle they are models to describe the equilibrium of the CO₂ concentrations in the gas and the liquid (equation 4).

Aspen Plus has an electrolytic package to calculate liquid systems containing ions. Different electrolyte equilibrium models can be used. Using an MEA property insert model, equilibrium models can be combined with reaction kinetic models, including rate expressions of chemical reactions like equation (1) and (2).

2.3 Column models in simulation programs

A CO₂ absorption column is a unit where gas flows up and liquid (eg. an amine solution) flows down. CO₂ is transferred from the gas phase to the liquid phase where it reacts with the amine solution. The gas and liquid phases are made to get in contact by the help of column plates or random or structured packing.

The CO₂ stripping column also has plates or packing, and this column also has a reboiler at the bottom to provide heating, and a condenser at the top to provide cooling.

The traditional way to model such columns is by using equilibrium stages. One plate can be calculated assuming equilibrium between the CO₂ concentration in the gas and liquid leaving the plate. In a packed column, a certain height of packing can be modeled as one equilibrium stage.

The equilibrium stage model can be refined by introducing a stage efficiency. Murphree efficiency for stage number n is defined by

$$E_M = (y - y_{n-1}) / (y^* - y_{n-1}) \quad (6)$$

where y is the mole fraction of CO₂ in the gas leaving the stage, y_{n-1} is the mole fraction leaving the stage below, and y^* is the mole fraction CO₂ in equilibrium with the liquid leaving the stage. This is illustrated in figure 3.

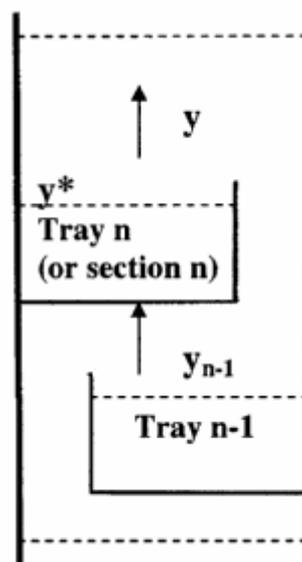


Figure 3: Definition of Murphree efficiency

Most process simulation programs have models for implementing Murphree efficiency in a column model. The Amines property package in Aspen HYSYS has a special estimation method for predicting this Murphree efficiency. This is based on the work of Tomkej [12]. This model is based on experience with CO₂ removal from high pressure natural gas. In Aspen Plus it is also possible to specify these efficiencies explicitly in an absorption or distillation column model.

Aspen Plus has a column model (RateFrac) which can include kinetic rate expressions in chemical reactions at each stage. In the references [1-3] the MEA property insert model in Aspen Plus is used in a column model.

2.4 Column convergence

To converge a column model in a process simulation program, all the equations describing the equilibrium and gas and liquid flows must be solved for each calculation stage. Including rate expressions for chemical reactions complicates the calculations further. This often leads to convergence problems.

The column model in Aspen HYSYS has a default set of convergence criterias, and a default set of calculation parameters. Different calculation models are also available. The Inside-Out algorithm is default, and a Modified Hysim Inside-Out algorithm is also available. A damping parameter for the column iteration is adjustable, and the damping can be specified to be adaptive.

2.5 Flowsheet convergence

All process simulation programs are based on modules for calculating different unit operations like heat exchangers, pumps, distillation columns etc. Process simulation programs are traditionally divided into either sequential modular or equation based programs. In a sequential modular program, the in-streams of each calculation module must be known prior to the calculation, and the out-streams are the result of the module calculation. The programs Aspen Plus and ProVision are sequential modular. Equation based simulation programs can be able to calculate in-streams based on out-streams. Aspen HYSYS is an equation based simulation program.

However, also in Aspen HYSYS, the column models are based on specified in-streams. Because of this, flowsheets with columns in practice have to be calculated in a modular sequential manner.

In many cases, it is of interest to calculate in-streams also to columns. This can be done by iteration methods. In the case of recycle streams, the flowsheet can be solved including recycle blocks. A recycle block compares the in-stream to the block with the out-stream from the block in the former iteration.

In the case of convergence problems in a column model, recycle iterations complicate the calculations further. In some cases, a recycle block will not converge due to parameters of minor interest. An example of such a parameter is the concentration of a trace component. In such cases, a possibility is to iterate manually on the main parameter (eg. the CO₂ concentration) by replacement, and accept the errors in the parameters of minor importance.

3. Calculations

3.1 Power plant simulation

Figure 2 shows the principle of a combined cycle power plant based on combustion of natural gas. A real plant will be much more complex with many heat exchangers, recycle pipes and tanks to optimize the overall efficiency of the power plant. However, a process as in figure 2 is practically realistic, but it will not have an optimum overall efficiency.

The main purpose of the power plant simulation is to make a simple but realistic model to evaluate the influence of heat for CO₂ removal on the overall power plant efficiency.

A simplified combined cycle 400 MW gas based power plant has been simulated with Aspen HYSYS. Pure methane is used as natural gas, air is 79 % nitrogen and 21 % oxygen, 100 % combustion is assumed, and traditional temperatures and pressures are used in the process. The Peng Robinson model has been used for the thermodynamic properties in the power plant. Specifications for the calculation are listed in table 1. A flowsheet of the Aspen HYSYS model is presented in figure 4. This model has been developed during the Master Thesis work of Kristin Vamraak [13] and Bjørn Moholt [14].

Inlet air/gas temperatures	25 °C
Inlet natural gas pressure	30 bar(a)
Combustion temperature	1500 °C
Steam high pressure	120 bar(a)
Steam medium pressure	3.5 bar(a)
Steam low pressure	0.07 bar(a)
Pressure to stack	1.01 bar(a)
Stack temperature	100 °C

Table 1: Specifications for Power Plant Simulation

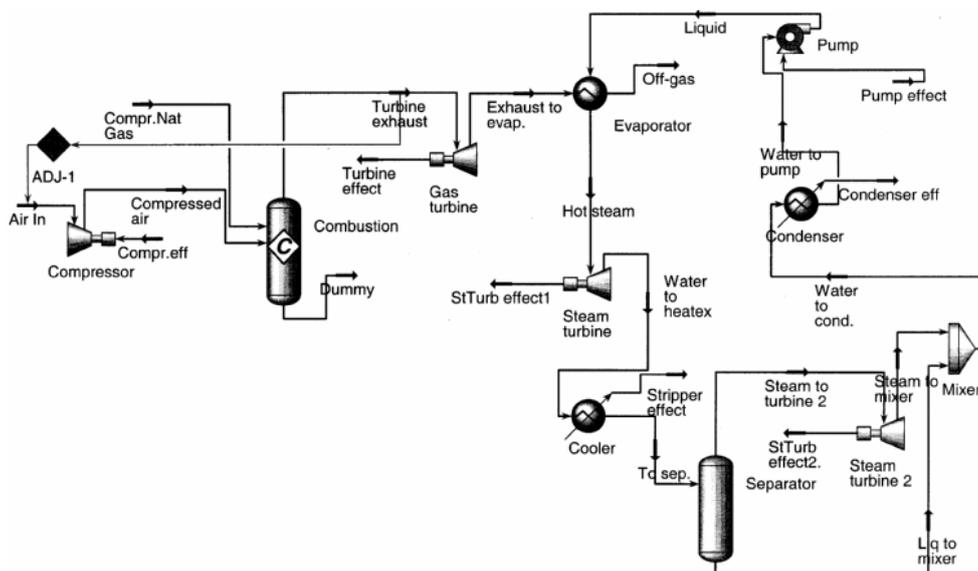


Figure 4. Aspen HYSYS model of simplified gas power plant.

To have a physically possible process, the flue gas temperature (680 °C from gas turbine to exhaust at 100 °C) has to be higher than the steam temperature through all the steam heat exchanger. This results in a maximum superheated steam temperature of 540 °C (at 120 bar). The chosen pressure is not optimized.

The efficiencies in compressors, gas turbines and steam turbines have been fitted to achieve a total thermal efficiency of 58 % (which is traditional) in the gas based power plant. The total efficiency is calculated as the turbine effects (minus compressor and pump) divided by the lower heating value of natural gas. The compressor efficiency (adiabatic) was adjusted to 90 %, the expander part of the gas turbine (expander) and the steam turbines were adjusted to 85 % (also adiabatically). These values are high compared to actual efficiencies for such equipment. A real power plant would be more energy optimized, and have lower equipment efficiencies.

In this part of the power plant simulation, the steam delivery (for CO₂ removal) is set to zero.

3.2 Simulation of CO₂ removal base case

An absorption and desorption process for CO₂ removal with an aqueous MEA solution has been simulated. The exhaust gas from the power plant model is used as the feed to this model. The absorption column is specified with 10 stages each with a Murphree efficiency of 0.25. (An estimated HETP (Height Equivalent to a Theoretical plate) of 4 meter, is about equivalent to 0.25 efficiency for each meter of packing.) Traditional concentrations, temperatures and pressures are used in the base case simulation. The thermodynamics for this mixture is

described by an Amines Property Package available in Aspen HYSYS. The Kent Eisenberg [10] model is selected in the Amines Property Package. Specifications for the calculation are listed in table 2. The Aspen HYSYS CO₂ removal model is presented in figure 5. Different versions of this model have been developed in several student projects. The version in figure 5 is based on a Master Project work by Trine Amundsen [15].

Inlet gas temperature	40 °C
Inlet gas pressure	1.1 bar(a)
Inlet gas flow	85000 kmole/h
CO ₂ in inlet gas	3.73 mole-%
Water in inlet gas	6.71 mole-%
Lean amine temperature	40 °C
Lean amine pressure	1.1 bar(a)
Lean amine rate	120 000 kmole/h*)
MEA content in lean amine	29 mass-% *)
CO ₂ in lean amine	5.5 mass-% *)
Number of stages in absorber	10
Murphree efficiency in absorber	0.25
Rich amine pump pressure	2 bar
Heated rich amine temperature	104.5 °C *)
Number of stages in stripper	6 (3 + 3)
Murphree efficiency in stripper	1.0
Reflux ratio in stripper	0.3
Reboiler temperature	120 °C
Lean amine pump pressure	2 bar
Minimum deltaT in heat exch.	10 °C
*) In first iteration	

Table 2: Specifications for Base Case CO₂ removal

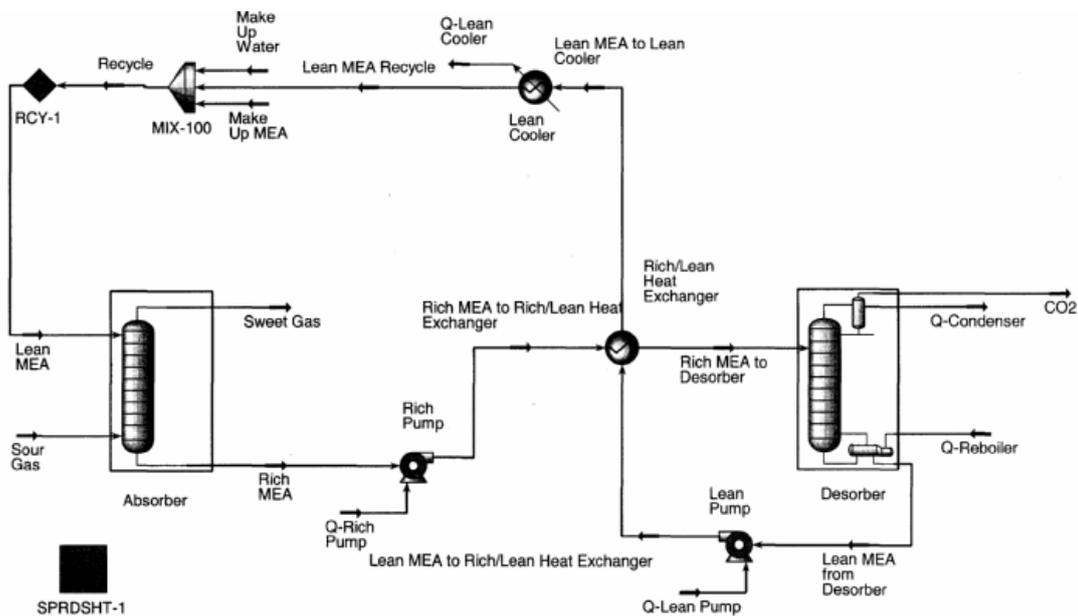


Figure 5. Aspen HYSYS model of CO₂ removal.

85 % CO₂ removal can be specified in the process. The Kent Eisenberg equilibrium model has been compared with the Li-Mather equilibrium model [11]. The CO₂ removal calculated by Aspen HYSYS was reduced from 85 to 82 %, and the heat consumption was reduced from 3.65 to 3.4 MJ/kg CO₂.

3.3 CO₂ removal sensitivity calculations

3.3.1 Variables held constant

The model has been used to evaluate the effects of changing the most important parameters.

In most of the calculations, the CO₂ removal and the stripping heat consumption were calculated, while keeping all the other parameters in table 2 constant. From a calculation viewpoint, this is probably the simplest.

Another possibility had been to keep the % CO₂ removal constant, and calculate the heat duty and the necessary column height. This would give the possibility to optimize the trade-off between operation cost (due to heat consumption) and capital cost (due to column height).

In the cases where the default Inside-Out algorithm did not converge, the Modified Hysim Inside-Out algorithm with adaptive damping was tried to obtain convergence.

3.3.2 Circulation rate

The effect of increased circulation rate, is that the removal grade increases. The results of the simulations are shown in figure 6. A minimum calculated steam consumption is calculated to 3.62 MJ/kg CO₂ removed.

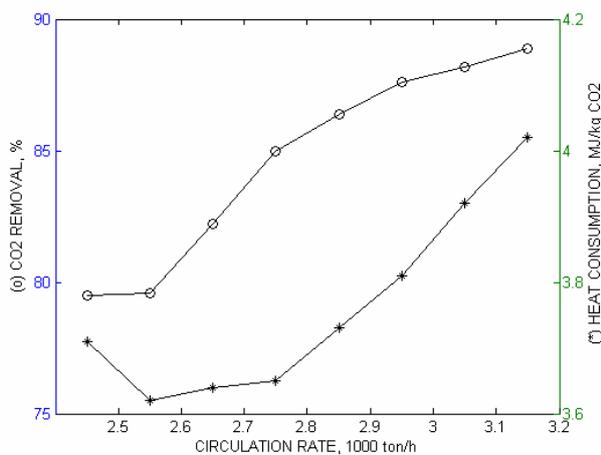


Figure 6: Circulation rate dependence

3.3.3 Number of absorption stages

The height of the absorption column is varied by changing the number of stages. The Murphree stage

efficiency for CO₂ is kept constant at 0.25. The height can also be changed by varying the stage efficiency. As expected, removal grade increases and heat requirement decreases with increased column height. The result is shown in figure 7. The calculation diverges using more than 12 stages in the column.

Hysys has calculated estimated E_M (equation 6) for a plate, and the resulting E_M for a plate varied between 0.08 and 0.13.

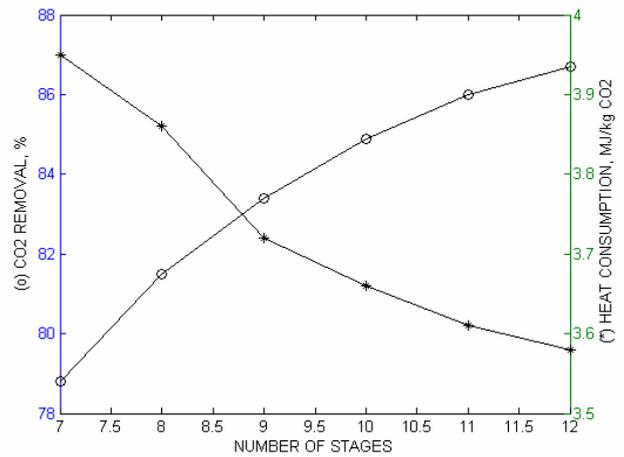


Figure 7: Number of stages dependence

3.3.4 Absorption temperature

An increase in gas and liquid inlet temperature leads to reduced absorption at equilibrium. Simulation results based on a constant stage efficiency are shown in figure 8. In practice, a higher temperature will give a higher absorption and reaction rate, but the equilibrium results will not show this effect.

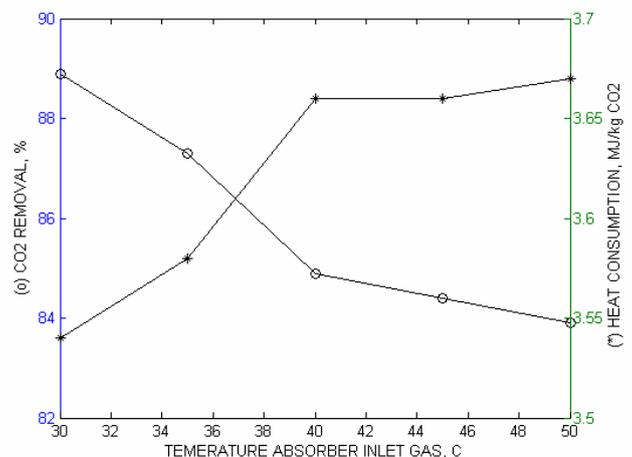


Figure 8: Absorber temperature dependence

3.3.5 Absorption pressure

The absorption pressure is set to atmospheric pressure at the outlet and atmospheric pressure plus pressure drop at the inlet. The pressure inlet in the base case was 1.1 bar. In the case of a pressure drop of 0.5 bar, the % CO₂ removal increased to 87.1 % and the energy consumption was reduced to 3.59 MJ/kg CO₂ removed.

3.3.6 Reboiler temperature

Increased reboiler temperature gives purer amine solution and better CO₂ removal efficiency. However, amine degradation problems arise above 120 °C. The temperature was varied between 118 and 121 °C. It was difficult to get converged solutions outside this range. At 121 °C, an outside range warning was given. The results up to 120 °C are shown in figure 9.

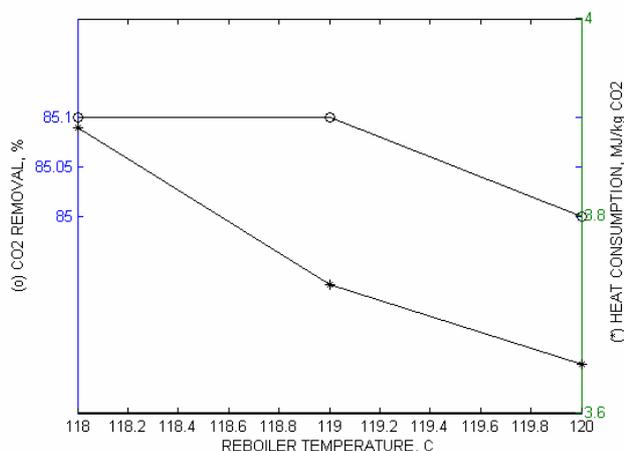


Figure 9: Reboiler temperature dependence

3.3.7 Stripper pressure

The stripper pressure was specified to 2 bar(a) in the calculation. It was very difficult to get a converged solution at other pressures. A solution with a warning (outside range) was achieved with a pressure at 1.9 bar(a).

3.3.8 Minimization of heat consumption

There have been performed many Aspen HYSYS calculations at different conditions. One aim is to reduce the heat duty as much as possible. The lowest heat consumption calculated was 3.39 MW/kg CO₂ removed. The CO₂ removal efficiency was then 93.8 %. This was obtained with gas temperature at 30 °C, 16 absorption stages, stripper pressure 2.0 bar and reboiler temperature at 200 °C. At lower temperatures or more absorption stages, the calculation did not converge.

3.4 Power plant efficiency reduction due to CO₂ removal

The steam consumption in the CO₂ removal process is delivered from the gas power plant as shown in figure 2, and the total thermal efficiency is reduced. Steam at 3.5 bar is delivered at 139 °C. When the base case heat consumption was used, the total efficiency was reduced from 58 % to 53 %. If the efficiency loss due to exhaust gas fans and circulation pumps was included, the resulting total efficiency would be approximately 50 %. If energy for CO₂ compression and condensing was to be delivered from the power plant, the total efficiency would be below 50 %.

The calculated alternative with the lowest duty was with 3.39 MJ/kg CO₂ removed. This gives a reduced reduction in total efficiency of 0.4 % (%-points).

The effect of a possible lower temperature for heat stripping can be calculated. The medium steam pressure can be reduced slightly. The same duty is used, and the total effect should give a slight increase. This effect is so marginal, that it is within the uncertainty of the calculations.

4. Discussion and Conclusion

4.1 Accuracy

With the same specifications, the calculated results vary slightly dependent on initial values. The accuracy in the calculations is normally within 1 % (absolute) in the CO₂ removal and a few percent (absolute) in heat consumption. This accuracy can probably be improved with tighter convergence limits.

The uncertainty due to equilibrium is probably higher. This is indicated by the CO₂ removal changing with 3 % (%-points) when changing equilibrium model from Kent Eisenberg to Li-Mather. The uncertainty in the power plant calculation due to the Peng Robinson equation of state has not been checked, but is expected to be less than the uncertainty in the amine models.

4.2 Evaluation of the simplified process

The calculated CO₂ removal process is a simplified process. Heat losses and some pressure losses are neglected. A real process contains more equipment, pipes and valves, and all this equipment also have heat losses and pressure losses.

A real process with MEA will probably include a water wash section to reduce MEA emissions and a reclaiming unit to recover MEA from thermally de-

generated MEA. These additional units will increase the energy consumption.

There are possibilities to reduce the energy consumption in the process by different stream configurations (eg. split stream). These possibilities will however normally increase the investment.

The calculated heat consumption of 3.65 MJ/kg CO₂ is regarded as realistic. This is slightly lower than normally found in literature, eg. Desideri [1] which has a list of references with values mostly above 4.0 MJ/kg CO₂. If water wash is included in the highest value, this can explain the difference. The lowest calculated value of 3.39 MJ/kg CO₂ is regarded as an optimistic value. It might be regarded as a potential value for improvements. The cost optimum heat consumption will probably be a trade-off between investment and operational cost.

4.3 Convergence problems

There are many problems with convergence in these calculations. The problems normally occur in the absorption or stripping column. One problem is numerical. It is found that the Modified Hysim Inside-out algorithm with adaptive damping gives the best convergence.

If there are too many stages specified in the columns, they tend to diverge. That is traditional for column stage calculations in typical process simulation tools.

There is also a problem with the range for the model. The Kent Eisenberg model is limited to below 30 weight-% MEA and below 120 °C. The simulation is calculated, but the program gives a warning.

4.4 Parameter variations

Different parameters have been varied to calculate the effects and to make a tool for optimizing the process.

The only parameter that is varied so that an optimum is found, is the recirculation rate. In figure 6 it is shown that a minimum heat consumption (of 3.62 MJ/CO₂) is achieved at a lean amine circulation rate of 2550 ton/h.

When the number of stages is increased, the performance of the process increases, but the calculation tends to diverge. However, when the column diverges due to too many stages, the % CO₂ removal and heat consumption is probably close to a maximum.

A reduction in absorption temperature leads to improved performance according to figure 8. According to literature, a temperature of about 40 °C is recommended. At lower temperature, the reaction rates

will be too slow. The calculations have been performed with constant column stage efficiencies of 0.25. The original Aspen HYSYS calculates efficiencies are temperature dependent. A temperature dependent stage efficiency is probably necessary to find an optimum absorption temperature.

It is not difficult to calculate the effects of absorption pressure drop. There is a trade-off between CO₂ removal efficiency and cost of column height and pressure drop.

Increased reboiler temperature gives purer amine solution and better performance. The optimum temperature is a trade-off between improved CO₂ removal and degradation of the amine solution. Aspen HYSYS limits the calculation to 120 °C as in the base case. The resulting lean loading (mole CO₂/mole amine) in the liquid to the absorption column is 0.26. This is close to 0.25 as reported as an optimum by Alie [3].

The stripper pressure is an adjustable parameter. It is difficult to get a converged calculation with a stripper pressure far away from 2 bar with 120 °C. According to Freguia [2], the pressure should be between 1.5 and 2 bar. It is not obvious whether the convergence difficulties are due to physical limitations, or if it is a numerical problem. The pressure must be consistent with the pressure of the amine solution leaving the reboiler. The problems of limited range of reboiler temperatures and stripper pressures are probably related.

4.5 Aspen HYSYS compared to other tools

There are other tools to simulate such processes. Other commercial process simulation tools like ProVision, ProMax or Aspen Plus can be used to simulate the process in a similar way. Aspen Plus also has the possibility to calculate rate expressions on an ideal mixing stage (simulating a column plate). This has the advantage of taking into account the reaction rates for different reactions simultaneously. It is possible (but difficult) to include rate expressions of absorption (transport of CO₂ from the gas to the liquid phase) in such a model. It is also a question whether this kind of a mixing stage model is a good model for continuous countercurrent operation as in structured packing.

The presented Aspen HYSYS model is based on a specified Murphree efficiency for each stage (or height of packing). It is possible to make this efficiency a function of rate expressions for the absorption rate and the reaction rates.

It is of course possible to simulate CO₂ removal processes without using commercial process simulation programs. It is however necessary to include at

least one reliable and robust equilibrium calculation model and one robust column model. It is difficult to compete with the commercial process simulation programs in these two matters. The commercial programs also normally have very good input and output facilities.

A problem with the commercial process simulation programs, at least from an academic point of view, is that some of the models of interest are not documented accurately.

4.6 Further development

There are available calculation models for the design of CO₂ removal plants based on MEA. The largest uncertainty is probably connected to the absorber stage efficiency. In the case of a structured packing column, this gives a large uncertainty in the necessary packing height. An improvement here will be of great value.

In the case of other amines than MEA, the limitation is in the equilibrium model or in the uncertainty in the equilibrium model parameters.

In the case of mixed amines, the importance of the reaction rates increases. Such models are probably best treated in a rate based model like RateFrac in Aspen Plus. It is however possible that a practical approach when using Aspen HYSYS, is to perform estimation of Murphree efficiency outside the process simulation tool.

4.7 Conclusion

The CO₂ removal model developed in Aspen HYSYS is useful for evaluating the effects of changing amine circulation rate, absorption column height, absorption temperature and reboiler temperature. The reduced total efficiency in a gas based power plant can be calculated.

The developed models can be developed further to improve amine absorption processes for CO₂ removal. The combination of one model for a power plant and another for CO₂ removal can be useful for total energy optimization and total cost optimization.

5. Acknowledgements

These models are the result of much trial and error work done by my students during many years. Thanks to all of you!

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