

MODELING OF THE CO₂ CAPTURE IN POST-COMBUSTION

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Received: 30/10/2006

Accepted after revision: 17/01/2007

Abstract: The aim of this study is to assess the energy consumption of an alkanolamine CO₂ removal process integrated downstream from a power plant. The CO₂ capture has been simulated with the Aspen plus™ software for two types of power plants: a Natural Gas Combined Cycle (NGCC) and a Pulverised Coal Fired power plant (CF). Two amines were studied: monoethanolamine (MEA) and methyldiethanolamine (MDEA). The amine concentration in the aqueous solvent ranges from 10 to 30 wt.% for MEA and from 30 to 50 wt.% for MDEA. The results show that the more the flue gas is concentrated out of CO₂ the less the energy consumption per ton of captured CO₂ is high. Moreover, although MDEA is easier to regenerate than MEA, an aqueous solvent of MDEA requires as much energy as an aqueous solvent using MEA. Moreover, the kinetics of CO₂ with MDEA is lower than with MEA. This means that the installation size will be bigger and thus its investment cost more expensive.

Keywords: *CO₂ capture, process simulation, natural gas combined cycle, coal fired, monoethanolamine, methyldiethanolamine*

INTRODUCTION

After the Kyoto summit in 1997, most industrialized countries decided to take constraining measures on the CO₂ emissions to stabilize its concentration in the atmosphere between 450 and 500 ppm in 2100. Indeed its concentration keeps on rising since 1850, by 1.5 ppm per year these last decades. Given the gap of wealth and development between the industrialized countries and the underdeveloped countries, the first ones have to make a large effort to reduce their emissions. The anthropogenic CO₂ comes mainly from hydrocarbons combustion and particularly from power plants. There, the CO₂ emissions are very large and localized, which will ease its recovery. This particular point makes it possible to target the action on the more concentrated sources rather than on diffuse sources like transport, heating... Thus, priority has been given to the largest sources of CO₂ emissions: steel and cement factories, refineries and power plants. Technically, the CO₂ capture using an amine absorption process is a viable short to medium term strategy. However, the costs of such systems are currently perceived to be too high to be economically feasible. This is why these last years, much of studies have been focused on these problems. Some studies deal with the performance of an amine scrubbing process [1-5]. And other ones focus on the development of advanced processes for CO₂ recovery [6-10].

Concerning the absorption processes, chemical solvents are selected for their best performances with a low partial pressure of CO₂ (absorption capacity and reaction rate). Indeed the CO₂ partial pressure in the flue gas of a Natural Gas Combined Cycle power plant (NGCC) or a Coal-Fired power plant (CF) is quite low, between 40 and 150 mbar. Processes with physical solvents do not take advantage of these conditions of pressure. The solvents usually used under these conditions are primary alkanolamines and particularly monoethanolamine (MEA) [11-13].

The aim of this study is to assess the energy consumption of a CO₂ recovery process using two types of amine: MEA and MDEA. Two flue gases have been studied: one typical of a NGCC and the other coming from a CF.

STUDY BACKGROUND

We simulated the process with the Aspen PlusTM 2004.1 software. This software is described as a modular code with sequential resolution adapted to engineering processes. It allows the simulation, design and optimization of processes in steady conditions. It has been used to evaluate the energy consumption for different operating conditions. Two amines were used: monoethanolamine (MEA) and methyldiethanolamine (MDEA). Thus, four cases have been studied: NGCC-MEA, NGCC-MDEA, CF-MEA and CF-MDEA.

Flue gas

In this study, two flue gases have been used (Table 1). The first one corresponds to a 480 MWe NGCC and the second one comes from a 600 MWe CF with

desulphurization. The CO₂ concentration is 14.0 mol.% for the CF and 4.97 mol.% for the NGCC. The flue gas resulting from natural gas combustion contains less carbon per unit of mass than the one coming from coal combustion.

Table 1. Characteristics of the flue gases

	NGCC	CF
Composition (mol.%)		
H ₂ O	11.02	6.4
CO ₂	4.97	14.0
O ₂	9.73	3.8
N ₂	74.28	75.6
Flow (kmol.s ⁻¹)	574	646
Pressure (bar)	1.015	0.913
Temperature (°C)	100	96

Calculations

To calculate the energy consumption, a basic flow sheet for alkanolamine acid-gas removal process has been used (Figure 1).

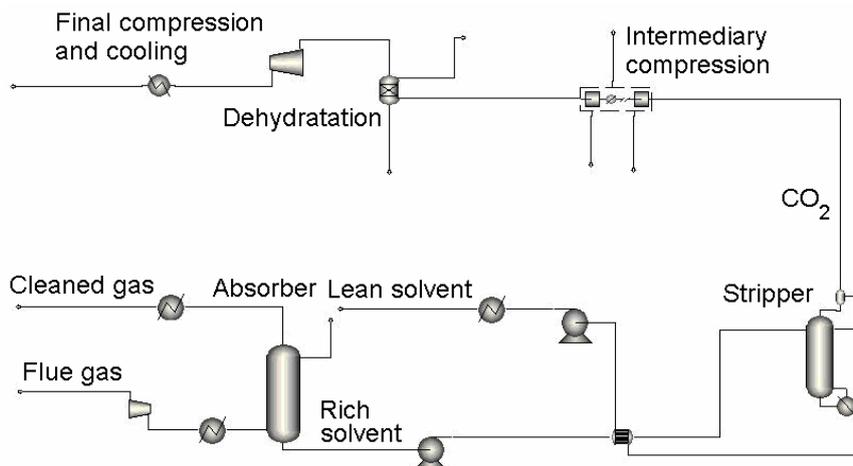


Figure 1. Alkanolamine CO₂ removal process

First the flue gas is compressed. Then it enters a heat exchanger where it is cooled until 40 °C. The flue gas is injected in the lower part of the absorber and washed with a counter-current solvent. During the absorption, the CO₂ loading in the solvent rises from a lean value (α_{lean}) to a high value (α_{rich}). The clean gas recovered at the top of the column is directed to the stack. The rich solvent recovered at the bottom of the absorber is pumped to compensate for the pressure loss in the stripper and minimize the final compression work of CO₂. Then this stream enters a heat exchanger where it is preheated by the regenerated solvent recovered at the bottom of the stripper. For this heat exchanger, a 10°C temperature pinch has been chosen between the hot stream and the cold stream. The rich solvent is regenerated in the stripper with heat duty supplied by the condensation of low-pressure steam in the stripper reboiler. During the regeneration, the CO₂ loading decreases from the rich value to the lean value. The

regenerated solvent has the same residual concentration of CO₂ than the one which enters the absorber. So it can be redirected to the absorber. The stream at the top of the stripper is mainly composed of CO₂ and water. The condenser at the top of the stripping column withdraws the bulk of water present in this stream by cooling it at 30 °C. The water is reinjected into the stripper while the CO₂ stream is compressed to 65 bars in a four-stage intercooled compressor. Then, it is dehydrated by a triethylene glycol process. The residual water fraction has been fixed at 20 ppm to minimize the risk of hydrates formation and corrosion during transportation. The CO₂ is then compressed to 150 bars. It is cooled to 37 °C to increase the density of the CO₂ stream. With such a process, the CO₂ stream has a very high purity (≈ 99.9 mol.%).

Steam supply

The steam required for the reboiler is not available at the same pressure for the cases NGCC and CF. For the first one, low pressure steam can be extracted at 3.2 bars and at 4.14 bars for the second one. Considering the pressure losses between the extraction location and the reboiler, the condensation temperature of this steam is 123.30 °C for the case NGCC and 133.5 °C for the case CF. This temperature level sets the maximum temperature in the reboiler since this latter must be smaller than the steam temperature.

Thermodynamic models

The choice of the thermodynamic model is very important. Aspen PlusTM provides packages for electrolyte systems. Two packages have been tested: “EMEA” and “MEA”. The solubility of CO₂ in a 30 wt.% MEA solution and in a 15.5 wt.% MEA solution has been evaluated. The results obtained have been compared with literature data. The data of Jou et al. [14] have been used for an aqueous solution of 30 wt.% MEA and those of Austgen and Rochelle [15] for an aqueous solution of 15.5 wt.% MEA. The models match very well the data of Austgen and Rochelle [15]. But for a solvent more concentrated in amine, the models underestimate the CO₂ solubility at 40 °C (Figure 2). This will result in an increase in the calculated solvent flow and a decrease in the rich CO₂ loading. At 120 °C, the EMEA package is in good agreement with the data of Jou et al. [14]. For our simulations, we have used this package. It uses the NRTL electrolyte model, which is suited for electrolytic aqueous solutions.

Similarly, we have studied three packages for MDEA: EMDEA, MDEA and PMDEA. The results have been compared with the data of Jou et al. [16] and Austgen and Rochelle [15]. The different packages give quite good results for aqueous solutions with low MDEA concentration (25 wt.%). They match very well the data of Jou et al. [16] and those of Austgen and Rochelle [15]. For an aqueous solution of 50 wt.% MDEA, there are more discrepancies (Figure 3). The PMDEA package underestimates the CO₂ solubility at 120 °C. This is not appropriated for calculating heat duty. The packages EMDEA and MDEA give practically the same results. But the first one seems to be closer to the literature data. For our simulations, we have used the EMDEA package. This package uses the NRTL electrolyte model.

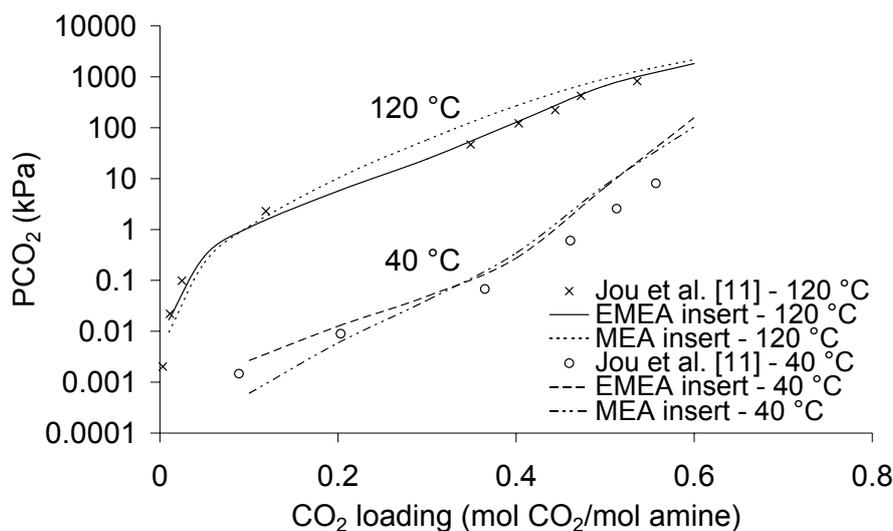


Figure 2. Solubility of CO₂ in a 30 wt.% MEA solution at 40 °C and 120 °C

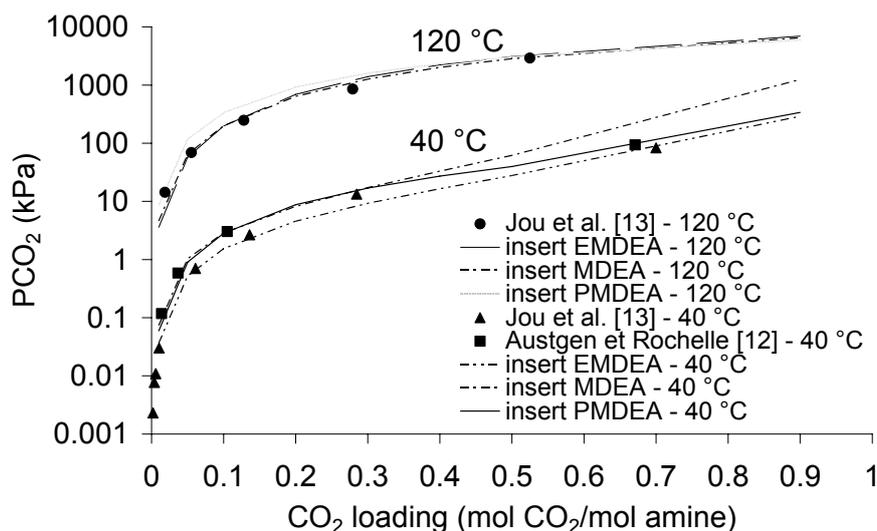


Figure 3. Solubility of CO₂ in a 50 wt.% MDEA solution at 40 °C and 120 °C

The Redlich-Kwong-Soave equation of state represents the gas phase. The gaseous components H₂, CO₂ and nitrogen follow the Henry's law.

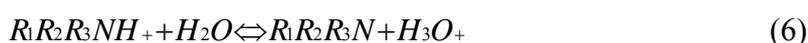
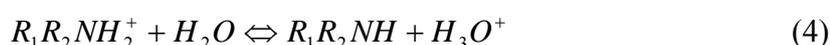
Chemical reactions

Within Aspen Plus™, there are two ways of simulating absorption: with kinetic reactions and by assuming equilibrium. We have only considered the second approach. The CO₂ absorption is modeled by a system of equations representing equilibrium between the various components.

MEA is written in the form R₁R₂NH, where R₁ = C₂H₄OH and R₂ = H. The CO₂ absorption mechanism with a primary or secondary amine in presence of H₂O results in

the formation of a zwitterion $R_1R_2NH^+COO^-$. This is a fast reaction but with a finished rate. It is followed by the deprotonation of this zwitterion by a base to produce a stable carbamate $R_1R_2NCOO^-$. This exchange of proton is considered instantaneous. The system is represented by the equations (1), (2), (3), (4) and (5).

MDEA is written in the form $R_1R_2R_3N$, where $R_1 = R_2 = C_2H_4OH$ and $R_3 = CH_3$. The alkalinity of the tertiary amines in aqueous solution allows the hydrolysis of CO_2 and the formation of the bicarbonate ion HCO_3^- and of the protonated amine $R_1R_2R_3NH^+$, which returns to a simple proton exchange. The CO_2 absorption by a solvent containing H_2O and MDEA is represented by the system (1), (2), (3) and (6).



Lean CO_2 loading

At 40 °C, for a CO_2 partial pressure of 10 kPa, the CO_2 loading at equilibrium in 30 wt.% MEA aqueous solution is approximately 0.56 mol CO_2 / mol MEA [14], and 0.48 mol CO_2 / mol MDEA in 30 wt.% MDEA aqueous solution [17]. The CO_2 loadings which it is possible to reach are higher with MEA than with MDEA. If a 90 % CO_2 recovery is wished, the CO_2 in the cleaned gas will approximately have a partial pressure equal to 1 kPa. At equilibrium, the corresponding CO_2 loading in the aqueous solvent is 0.38 mol CO_2 / mol MEA and 0.1 mol CO_2 / mol MDEA. It corresponds to the maximum value of the lean CO_2 loading in the solvent entering the absorber. The CO_2 loading in the lean solvent must be small enough to allow CO_2 absorption. Thus, when using MEA, a solvent with high lean CO_2 loading could be used. But, when using MDEA, the lean CO_2 loading must be small enough to reach the specified CO_2 recovery rate.

Characteristics of the absorber

The pressure of the flue gas is close of the atmospheric pressure. It must be compressed to make up for the pressure loss in the absorber. A sensitivity study has been made to assess the compression impact on the solvent flow and on the compression work of the flue gas. Three pressures have been tested: 1.14, 1.2 and 1.3 bar for a 85% CO_2 recovery rate. The case NGCC-MEA has been chosen for this study. A lean solvent of 30 wt.% MEA with $\alpha_{lean} = 0.2$ mol CO_2 / mol MEA has been used. A pressure increase from 1.14 bar to 1.3 bar leads to a 12 MW extra consumption, that is to say an increase of 117%. Concerning the solvent flow, it benefits from the pressure increase. But the

variation of flow is very small. Increasing the flue gas pressure is not favorable to the performance of the process. In the simulation, the compressor outlet pressure has been fixed at 1.14 bar.

A sensitivity study was carried out on the theoretical number of trays of the absorber (Figure 4). This is done in order to minimize the solvent flow. By setting a 85 % CO₂ recovery rate, the solvent flow and the thermal energy required in the stripper were determined according to the number of trays. For the case CF-MEA, the solvent containing 30 wt.% MEA with a lean CO₂ loading of 0.2 mol CO₂ / mol MEA has been used. The increase in the theoretical number of trays is beneficial to the process. Indeed the thermal energy required for regenerating the solvent decreases similarly with the solvent flow. However, beyond a certain number of trays, the variation becomes negligible. This can be observed for two types of flue gas and for all solvents. In the simulation, we have considered eight trays for the solvents using MEA and six for the solvents containing MDEA.

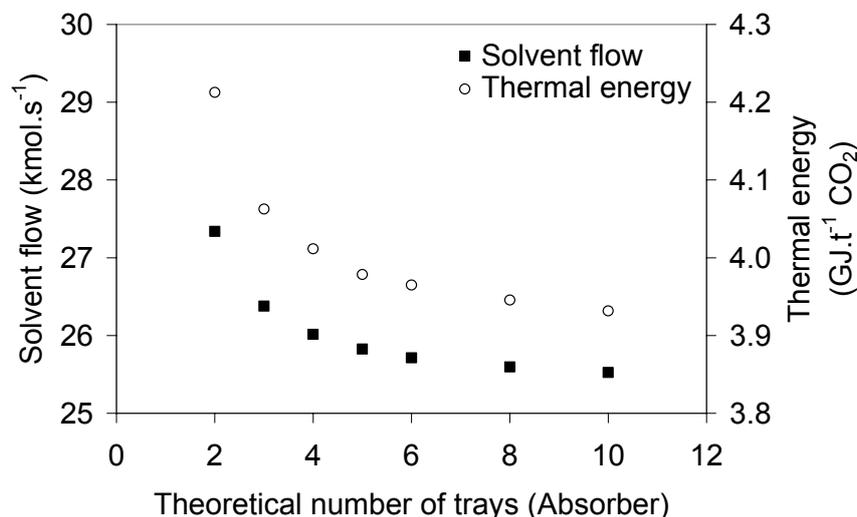


Figure 4. Optimization of the number of trays for the absorber (CF-MEA)

Characteristics of the stripper

Before entering the stripper, the solvent is pumped in order to compensate for the pressure loss in the column and decrease the final compression work for CO₂ transportation. For the case CF-MEA, several pressure levels were studied (Figure 5) in order to see the impact on the thermal energy requirement. For this study, we have used a solvent of 30 wt.% MEA with $\alpha_{lean} = 0.2$ mol CO₂ / mol MEA and a CO₂ recovery rate of 85 %. The stripper has 15 trays. The thermal energy requirement decreases from 4.19 to 3.36 GJ.t⁻¹ CO₂ when the pressure ranges from 1.14 to 1.3 bar. But, in the same time, the temperature difference between the steam required for the solvent regeneration (at dew point) and the reboiler decreases. Since the steam temperature must be higher than the reboiler, the stripper pressure cannot be increased indefinitely. In the simulation, we have considered a 5 °C temperature pinch. This pump contributes to the

CO₂ compression for transportation. It is well known that using a pump is less expensive than a compressor. The compression work of the CO₂ stream recovered at the top of the stripper will be lower.

As for the absorber, a sensitivity study was made on the theoretical number of trays of the stripper to minimize the heat duty in the reboiler. This heat duty decreases to a certain extent with the number of trays. In the simulations, we have considered 15 theoretical trays for MEA and 10 for MDEA.

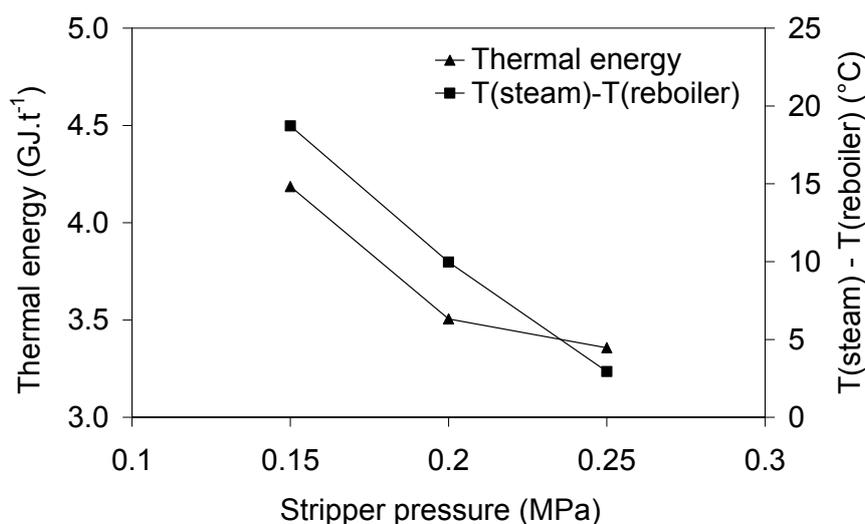


Figure 5. Influence of the stripper pressure (85% recovery rate – CF-MEA)

RESULTS

For each power plant, CO₂ recovery has been performed with MEA and MDEA. The recovery rate ranges from 85 to 98 %. For MEA, we have studied 3 amine concentrations: 10, 20 and 30 wt.%. The lean CO₂ loading has been varied from 0.1 to 0.3 mol CO₂ / mol MEA. It is no useful to decrease α_{lean} beyond 0.1 mol CO₂ / mol MEA because the heat duty becomes too high. The MEA concentration has been limited to 30 wt.% to take into account corrosion limitations. For MDEA, 3 amine concentrations have been studied: 30, 40 and 50 wt.%. Since there is no serious problem of corrosion, the amine concentration can be higher than with MEA. The lean CO₂ loading takes three values: 0.01, 0.05 and 0.08 mol CO₂ / mol MDEA. This last loading has just been associated with a MDEA concentration of 50 wt. %. The lean CO₂ loading must be low enough to complete the recovery rate.

Solvent flow

A sensitivity study was carried out on the CO₂ recovery for evaluating its impact on the solvent flow for the case NGCC-MEA (Figures 6 and 7). The solvent flow is as low as the amine concentration is high and as α_{lean} is low. This is a well-known result since the

solvent flow is highly dependent on the free amine quantity. But, the MDEA concentration in the solvent has a lower impact on the solvent flow since an increase in the MDEA concentration is accompanied by a reduction of the CO₂ solubility in the solvent (Figures 8 and 9). The absorption capacity of the solvent is thus reduced, offsetting the increase in the amine concentration.

It can be noticed that the solvent flow is higher when using MDEA than with MEA. This is due to a lower absorption capacity of an aqueous solvent using MDEA than of a solvent containing MEA when the partial pressure of CO₂ is low.

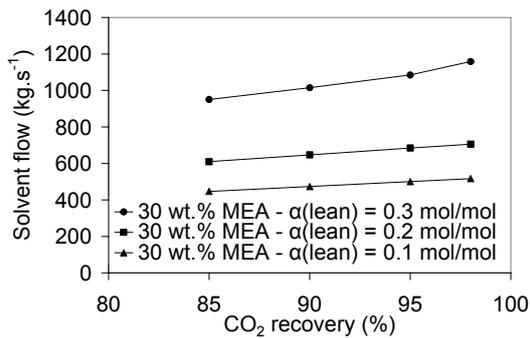


Figure 6. Influence of the lean CO₂ loading on the solvent flow (NGCC-MEA)

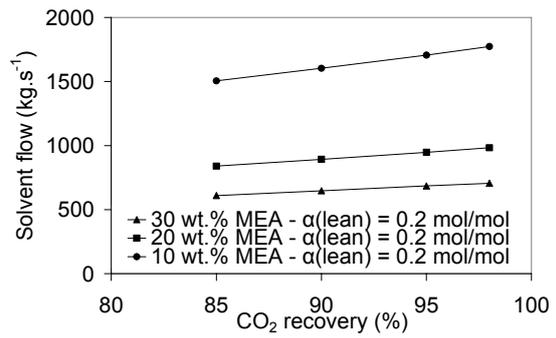


Figure 7. Influence of the MEA concentration on the solvent flow (NGCC-MEA)

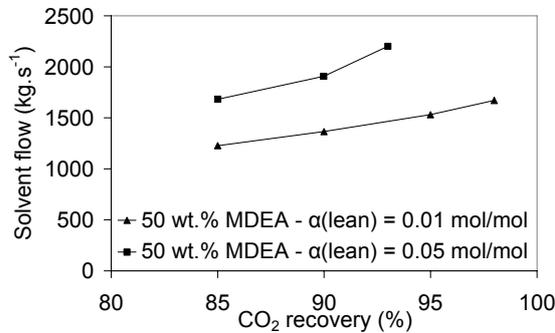


Figure 8. Influence of the lean CO₂ loading on the solvent flow (NGCC-MDEA)

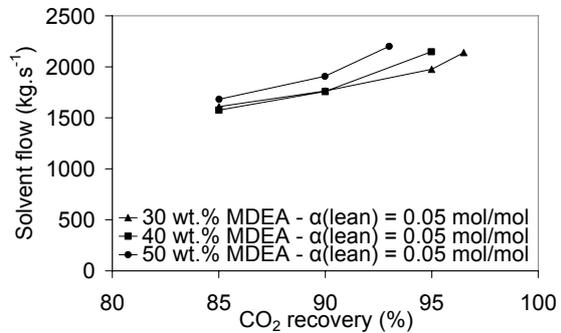


Figure 9. Influence of the MDEA concentration on the solvent flow (NGCC-MDEA)

Thermal energy requirement

A low solvent flow does not mean necessarily a low heat duty. Actually, it depends on the quality of the solvent regeneration too. For MEA, the thermal energy requirement increases quickly when the lean CO₂ loading decreases towards the lowest value (Table 2). The solvent for which the lean CO₂ loading is equal to 0.1 mol CO₂ / mol MEA is the one which requires the highest heat duty whereas the solvent flow is very low. This is true whatever the amine concentration is. The strong increase in the energy consumption with the decrease of the lean CO₂ loading has already been observed [18]. Above a certain CO₂ loading, the thermal energy consumption varies slightly with the

CO₂ loading. Whereas, below this value, the thermal energy requirement is very sensitive to the variations of the CO₂ loading, showing an effectiveness loss of the solvent regeneration. This is mainly due to the weak variation of the CO₂ partial pressure at low CO₂ loading, resulting in a higher requirement of vaporized steam to strip carbon dioxide. It is better using a solvent with a high amine concentration. The solvent flow is reduced as well as the thermal energy requirement. With an aqueous solution containing 30 wt.% MEA, the thermal energy requirement reaches a minimum at a lean CO₂ loading of 0.25 mol CO₂ / mol MEA for a NGCC power plant and at 0.2 mol CO₂ / mol MEA for a CF power plant. The thermal energy requirement is respectively 3.56 and 3.38 GJ.t⁻¹.

Table 2. Thermal energy requirement with MEA (85% recovery rate)

MEA (wt.%)	α_{lean} (mol CO ₂ /mol MEA)	Thermal energy requirement (GJ.t ⁻¹ CO ₂)	
		NGCC	CF
10	0.1	7.35	5.28
	0.2	4.89	4.62
	0.3	5.30	4.84
20	0.1	8.77	6.29
	0.2	4.00	3.79
	0.3	4.19	3.99
30	0.1	10.12	7.27
	0.15	5.97	4.40
	0.16	5.44	
	0.2	3.94	3.38
	0.25	3.56	3.44
	0.3	3.67	3.55

For MDEA, it seems that the amine concentration and the lean CO₂ loading have a slight effect on the thermal energy requirement (Table 3). However the best solvent seems to be the one with the highest amine concentration. This choice allows having the smallest heat duty. For NGCC, the lower thermal energy requirement is practically equal to the one found with MEA, with a value of 3.65 GJ.t⁻¹. Although MDEA is easier to regenerate than MEA, the higher solvent flow requires a higher energy consumption for preheating the solvent before the stripper. For the CF power plant, the thermal energy requirement is lower at 2.99 GJ.t⁻¹.

Table 3. Thermal energy requirement with MDEA (85% recovery rate)

MDEA (wt.%)	α_{lean} (mol CO ₂ /mol MDEA)	Thermal energy requirement (GJ.t ⁻¹ CO ₂)	
		NGCC	CF
30	0.01	3.93	3.33
	0.05	3.99	3.28
40	0.01	3.70	3.20
	0.05	3.80	3.12
50	0.01	3.65	3.17
	0.05	3.75	3.04
	0.08	4.03	2.99

If we only consider the thermal energy required for the solvent regeneration, a process using MDEA seems the best solution for the CF power plant. But the solvent flow will be bigger than with MEA. Moreover these results assume that the system is at equilibrium and that the kinetics of the different chemical reactions do not act any more. However, it is well known that the rate coefficient between CO₂ and MDEA is lower than the one between CO₂ and MEA. This means that the solvent flow will be even larger than the one found in this study. The size of a recovery process using MDEA will be bigger than with MEA and thus more expensive.

For the NGCC power plant, a solvent containing MDEA implies a higher energy consumption than with MEA. The thermal energy requirement is higher than in the CF power plant. This can be explained by the lower partial pressure of CO₂ in the flue gas, which acts unfavorably on the absorption.

Effect of the CO₂ recovery rate

For a CO₂ recovery rate ranges between 85 and 95%, the thermal energy requirement is practically steady, whereas a slight increase can be observed with MDEA (Table 4). With MEA, the solvent flow is proportional to the recovery rate. Consequently, the thermal energy required for preheating the solvent remains unchanged. With MDEA, the solvent flow increases more quickly when approaching the maximal CO₂ recovery rate. The thermal energy required for preheating the solvent increases with the CO₂ recovery. The global thermal energy requirement is thus higher.

Table 4. Influence of the CO₂ recovery rate on the thermal energy requirement

	NGCC-MEA		NGCC-MDEA	
	85	95	85	95
CO ₂ recovery rate (%)	85	95	85	95
Solvent flow (kg.s ⁻¹)	742	837	1227	1532
Thermal energy requirement (GJ.t ⁻¹ CO ₂)	3.56	3.58	3.65	3.89
	CF-MEA		CF-MDEA	
	85	95	85	94
CO ₂ recovery rate (%)	85	95	85	94
Solvent flow (kg.s ⁻¹)	1512	1713	3265	4037
Thermal energy requirement (GJ.t ⁻¹ CO ₂)	3.38	3.41	2.99	3.14

Power of compression

The power of compression has been assessed for different solvents, as shown on Figure 10. The flue gas flow has been normalized to 100 kg.s⁻¹ to compare the results for the NGCC and the CF power plants.

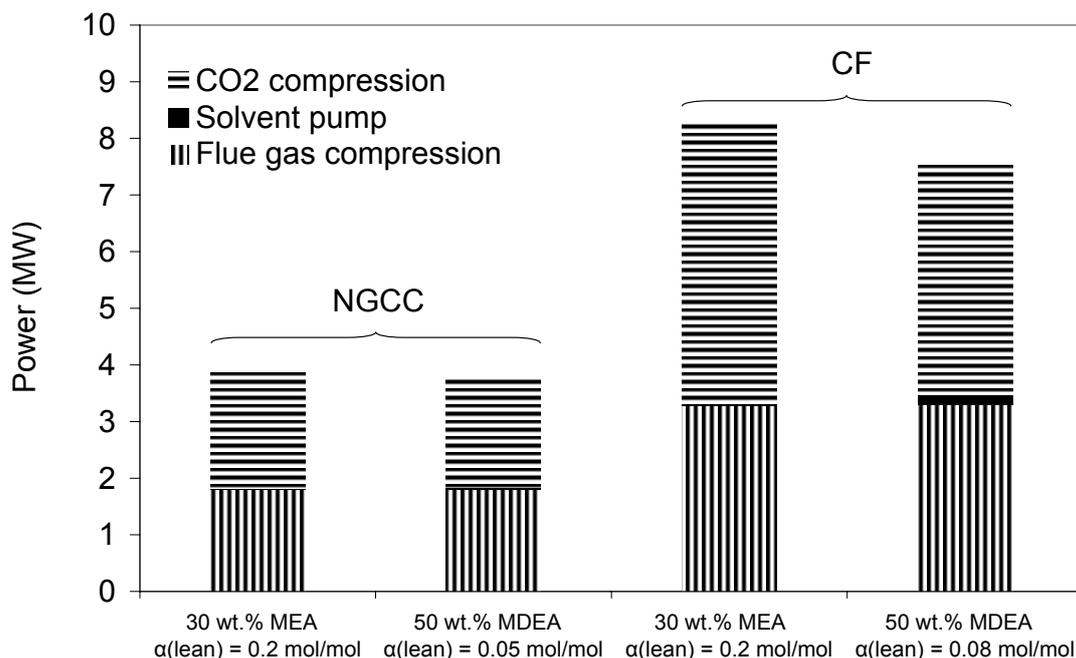


Figure 10. Compression power for a flue gas flow of 100 kg.s^{-1} (85 % CO₂ recovery)

The pressure of the flue gas is lower for the CF power plant than for the NGCC power plant (Table 1). That is why the flue gas compression requires more power: 3.3 MW for the CF power plant against 1.8 MW for the NGCC. Concerning the CO₂ compression, it depends on the pressure and the flow of the CO₂ stream. The difference between the two types of power plant comes from the fact that the CO₂ concentration in the flue gas is about three times higher in a CF power plant than in a NGCC. However, although the CO₂ stream flow is three times higher, the stripper pressure is higher too, which reduces the compression power. For the CF power plant, the CO₂ compression requires between 4.1 and 5.0 MW according to the type of amine. For the NGCC power plant, this compression needs approximately 2.0 MW. The power required to pump the rich solvent is very small compared to the two others powers. However, it can be noticed that this power is more important when using MDEA since the solvent flow is bigger.

CONCLUSIONS

We studied a CO₂ recovery process applied to a Natural Gas Combined Cycled power plant and to a Coal-Fired power plant. The process is based on a chemical absorption of CO₂ with an aqueous solvent containing either MEA or MDEA. This process has been built with Aspen PlusTM. The calculations are based on the equilibrium of the different chemical reactions. The influence of the amine concentration and the lean CO₂ loading was studied to assess their impact on the solvent flow and the thermal energy required when regenerating the solvent.

The solvents with the highest amine concentration are those displaying the lowest thermal energy requirement when the lean CO₂ loading was optimized. This last

parameter acts on both the solvent flow and the thermal energy requirement. For a Coal-Fired power plant, MDEA gives a lower energy consumption than MEA but, in practice, this type of solvent is penalized by its low CO₂ absorption rate. MEA seems better for a CO₂ recovery process. The solvent, which gives the best results, is an aqueous solution of 30 wt.% MEA with a lean CO₂ loading equal to 0.2 mol CO₂ / mol MEA for a coal fired power plant and 0.25 mol CO₂ / mol MEA for a natural gas combined cycle. The thermal energy requirement is respectively 3.38 and 3.56 GJ/t CO₂. When applied to a coal-fired power plant, the recovery process takes advantage on the higher CO₂ partial pressure. The recovery rate has only a slight influence on the thermal energy requirement, especially when using MEA.

The compression power of the process depends mainly on the type of power plant. The compression of the flue gas before the absorber accounts for 40-50% of the total power consumption. The compression of the CO₂ stream until 150 bars represents more than 50% of the power consumption. The power required by the solvent pump is negligible by comparison with the two previous powers.

ACKNOWLEDGEMENTS

The authors wish to thank ARMINES and EDF (Electricité de France) for their financial support to this investigation.

NOMENCLATURE

CF	Coal-fired power plant
MDEA	Methyldietanolamine
MEA	Monoethanolamine
NGCC	Natural Gas Combined Cycle power plant
P	Pressure (bar)
P(CO ₂)	CO ₂ partial pressure (kPa)
α_{lean}	Lean CO ₂ loading, mol CO ₂ / mol amine
α_{rich}	Rich CO ₂ loading, mol CO ₂ / mol amine

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