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The potential for improvement of the energy performance of pulverized coal fired power stations with post-combustion capture of carbon dioxide

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Abstract

The application of post-combustion capture processes in coal fired power stations can result in large reductions of the CO₂-emissions, but the consequential decrease in generation efficiency is an important draw-back. The leading PCC technology is based on an absorption process and the energy performance of this process is analysed. The analysis shows that the potential for improvement of the energy performance is quite large. In conclusion it is demonstrated that further development of the capture technology and the power plant technology can lead to generation efficiencies for power plants with 90% CO₂ capture which are equivalent to the current efficiencies without CO₂ capture, i.e. 0.4 (HHV).

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Keywords: post-combustion capture; power station; coal; absorption processes

1. Introduction

Capture and geological storage of CO₂ (CCS) is now widely recognised as an option that could contribute significantly to the prevention of greenhouse gas emissions. Globally coal fired power stations are emitting approximately 8 Gton CO₂ per annum from 2000 sources [1]. The typical lifetime of a coal fired power plant is more than 25 years. This means that once in place the power station will be emitting CO₂ for a long period. Only post-combustion capture (PCC) can effectively address the emissions of the existing power stations, often referred to as “locked-in carbon”. Other capture options can only be implemented in new power plants which will result in a more limited impact of CCS on the reduction of CO₂ emissions. It is often suggested that the potential for technology improvements is limited for a post-combustion CO₂ capture process. The underlying assumption is that this process is commercially available and that it is therefore fully developed. However, it must be emphasised that PCC has not been demonstrated on a full-scale power plant with optimal integration, nor has it been optimised. The objective of this contribution is to explore the potential for improvement in the energy performance of a coal fired

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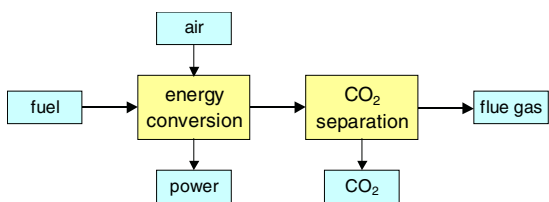
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power plant with post-combustion CO₂-capture based on an analysis of energy flows in both the capture process as well as the power plant.

2. Post-combustion capture of CO₂ (PCC)

In a conventional coal fired power station the fuel is mixed with air and burnt. The heat generated is used to generate steam at high pressure and power is produced by the expansion of steam through turbines. The flue gas undergoes a number of treatment steps, e.g. particulate removal and removal of nitrogen- and sulphur oxides before it is discharged to the atmosphere through the stack. The CO₂ concentration in the flue gas is typically around 12%. Flue gases are normally at atmospheric pressure but the temperatures might be between 45 °C and 120 °C, depending on the extent and type of contaminant removal. The general post-combustion process scheme is shown in Figure 1.

Power plants with post-combustion CO₂ capture involve two main process steps: an energy conversion step, during which power is produced, followed by a CO₂ separation process in which a concentrated stream of CO₂ is produced.



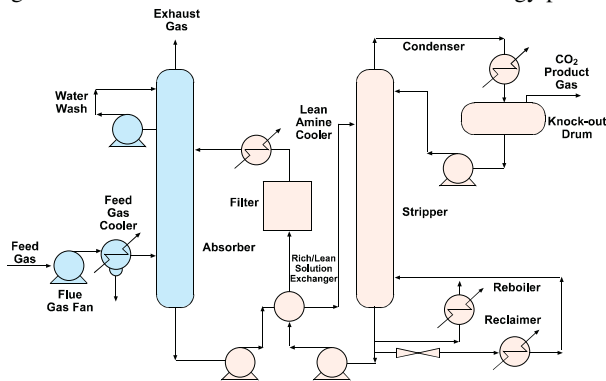
The separation task is to remove CO₂ from a mixture of mainly nitrogen and oxygen, but also the impact of flue gas impurities (SO_x, NO_x, particulates) needs to be taken into account. The post-combustion CO₂ capture process can be thought of as an add-on to a power plant and is similar to other flue gas treatment which might already be in place. This makes it the capture process which is the easiest to implement in existing power stations.

Figure 1: Post-combustion CO₂ capture

3. Absorption processes for post-combustion CO₂ capture

Although several different processes are currently under development for the separation of CO₂ from flue gases, absorption processes using aqueous solutions of chemical absorbents are the leading technology. The typical flow sheet of CO₂ recovery using chemical absorbents is shown in Figure 2.

After cooling the flue gas, it is brought into contact with the chemical absorbent in the absorber. A blower is required to pump the gas through the absorber. At temperatures typically between 40 and 60 °C CO₂ is then bound by the chemical absorbent in the absorber. After passing through the absorber the flue gas undergoes a water wash section to balance water in the system and to remove any droplets or vapour carried over and then leaves the absorber. The “rich” absorbent solution, which contains the chemically bound CO₂ is then pumped to the top of a stripper, via a heat exchanger. The regeneration of the chemical absorbent is carried out in the stripper at elevated temperatures (100 – 140 °C) and pressures between 1 and 2 bar(a). Heat is supplied to the reboiler to maintain the regeneration conditions. This leads to a thermal energy penalty as a result of heating up the solution, providing the required desorption heat for removing the



chemically bound CO₂ and for steam production which acts as a stripping gas. Steam is recovered in the condenser and fed back to the stripper, whereas the CO₂ product gas leaves the condenser. The CO₂-product is a relatively pure (> 99%) product, with water vapour being the main other component. Due to the selective nature of the chemical absorption process, the concentration of inert gases is low. The “lean” absorbent solution, containing far less CO₂ is then pumped back to the absorber via the lean-rich heat exchanger and a cooler to bring it down to the absorber temperature level. CO₂ removal is typically around 90%.

Figure 2: Process flow diagram for CO₂ recovery from flue gas with chemical absorbents

4. The thermal energy requirement in chemical absorption processes

Various novel chemical absorbents are being investigated, with the object of achieving a reduced energy use for solvent regeneration. This has the following contributions:

- The energy required to break the bond between CO₂ and the active component in the solvent
Reducing this energy requirement can be achieved by using amines with a lower binding energy for CO₂. This has to be balanced with the reaction rates, which might be lower. This will then lead to a larger absorber.
- The heat required for the bringing the solvent up to the reboiler temperature
A solvent with a high cyclic loading will lead to a lower heat requirement, as there is less solvent to heat up.
- The evaporation enthalpy for the stripping steam which leaves the stripper together with the CO₂
The amount of water vapour leaving the stripper is controlled by the operating conditions of the stripper and the type of solvent.

Taking a historical perspective it must be realised that the absorption process based on MEA has been patented in the 1930's and used for removal of acid gases from industrial gas streams. Operational issues related to corrosion have resulted in the development of alternative amines with a better performance. The development of corrosion inhibitors has resulted in renewed applications for CO₂ removal from flue gases in the 1980's, where the product CO₂ would be used for enhanced oil recovery. One may call these technologies first generation capture technologies (G1). More recently, the interest in post-combustion capture processes for reduction of CO₂-emissions has resulted in further process improvements and development of alternatives. These processes are either based on further improvements in MEA-based processes through e.g. use of intercooling in the absorber and use of a split-flow system [2] or novel chemical absorbents [3] These can be called second generation capture technologies (G2), where the CO₂-removal process is heat-integrated with a power plant. However, as it will be demonstrated further on, there is scope for further improvement in the overall process efficiency, leading to third and fourth generation technologies (G3, G4). The technological performance of the various capture technologies can be measured by the energy performance of the absorption process and the energy performance of the power plant with integrated CO₂ capture.

In Table 1 the heat of absorption of CO₂ for different absorbents, based on data from the literature, is given.

Table 1: Heat of absorption for CO₂ in different absorbents

Absorbent	Heat of reaction/absorption [GJ/ton CO ₂]	Reference
MEA – H ₂ O	1.92	[4]
DGA – H ₂ O	1.91	[4]
DIPA– H ₂ O	1.67	[4]
DEA– H ₂ O	1.63	[4]
AMP– H ₂ O	1.52	Derived from [5] at 313 K
MDEA– H ₂ O	1.34	[4]
TEA– H ₂ O	1.08	[4]
K ₂ CO ₃ – H ₂ O	0.64	[4]
H ₂ O	0.39	Clausius-Clapeyron fit to CO ₂ solubility data
N-Methyl-Pyrrolidone	0.37	[6]
Propylene Carbonate	0.36	[6]
Sulfolane	0.28	[6]

Table 1 shows that the commonly used MEA is the chemical absorbent with the highest binding energy. Physical solvents like propylene carbonate have a binding energy which is less than 20% of the value for MEA. There seems to be quite some potential for improvement of lowering this contribution to the energy required for releasing the CO₂ from the chemical absorbent. The value for the binding energy for chemical absorbent is determined by the overall reaction path which includes [7]:

- Hydration of CO₂ in the solution, which for water is around 25% of the binding energy in case of MEA (~ 20 kJ/mol CO₂)

- Carbamate formation depending on the amine which is around 15% of the binding energy in case of MEA (~ 10 kJ/mol CO₂)
- Protonation of the amine which contributes around 60% of the binding energy in case of MEA (~ 50 kJ/mol CO₂)

The summation of these contributions results in an overall binding energy of around 80 kJ/mol CO₂ (1.82 GJ/tonne CO₂), which is slightly lower than the value given in Table 1 (1.92 GJ/tonne CO₂ for MEA) and this is the representative value for a first generation solvent (G1).

The typical reaction paths for mono-amines are shown to be:

- Primary amines: $\text{CO}_2 + 2\text{R}_1\text{NH}_2 \rightarrow \text{R}_1\text{NH}_3^+ + \text{R}_1\text{NHCOO}^-$
- Secondary amines: $\text{CO}_2 + 2\text{R}_1\text{R}_2\text{NH} \rightarrow \text{R}_1\text{R}_2\text{NH}_2^+ + \text{R}_1\text{R}_2\text{NCOO}^-$

In these cases CO₂ reacts with the amine to form a carbamate and a protonated amine, limiting the theoretical capacity to 0.5 mol CO₂/mol amine. A second reaction path without the formation of a carbamate, is the following:

- Tertiary amines: $\text{CO}_2 + \text{R}_1\text{R}_2\text{R}_3\text{N} + \text{H}_2\text{O} \rightarrow \text{R}_1\text{R}_2\text{R}_3\text{NH}^+ + \text{HCO}_3^-$
- Sterically hindered primary or secondary amine: $\text{CO}_2 + \text{R}_1\text{NH}_2/\text{R}_1\text{R}_2\text{NH} + \text{H}_2\text{O} \rightarrow \text{R}_1\text{NH}_3^+/\text{R}_1\text{R}_2\text{NH}_2^+ + \text{HCO}_3^-$

In these cases CO₂ reacts with the amine to form a bicarbonate and protonated amine, allowing a theoretical capacity of 1 mol CO₂/mol amine.

As shown above the contribution of the protonation step on the overall binding energy is quite dominant. The enthalpy changes associated with the protonation can be derived from the temperature dependence of the pK_a [10]. Current developments have already resulted in the development of sterically hindered amines for post-combustion capture of CO₂ [3]. Through the avoidance of carbamate formation the binding energy is reduced by 10 kJ/mol CO₂ to 70 kJ/mol CO₂ (1.59 GJ/tonne CO₂). Further reductions through the identification of amines with lower enthalpy requirement for the protonation [8] might result in a further reduction of 15 kJ/mol CO₂ to 55 kJ/mol CO₂ (1.25 GJ/tonne CO₂) in a third generation chemical absorbent (G3). In Table 2 the enthalpy for protonation of several commercial chemical absorbents is given [9].

Table 2: Enthalpy of protonation for several commercial chemical solvents [9].

Chemical absorbents	Heat of protonation [GJ/ton CO ₂]
MEA	1.15
DEA	0.96
AMP	1.13
MDEA	0.79
DIPA	0.97
DGA	1.14
Piperazine	0.98

Fourth generation chemical absorbents (G4) might then involve chemical absorbents making use of bicarbonate formation, which is shown to have the lowest binding energy of any chemical absorbent. Examples of such solvents are aqueous solutions of ammonia or potassium carbonate.

Next the contribution of the absorbent heating up in the stripper is analysed. For a G1 chemical absorbent the flow requirement is 20 m³/tonne CO₂ [10], a typical heat capacity of 4 MJ/(m³K) and an approach temperature of 15 K the contribution to the overall energy requirement is 1.20 GJ/tonne CO₂. At first it appears that this can be decreased quite simply by reducing the lean/rich heat exchanger approach temperature to 10 K. In addition it is clear that a switch from a primary amine to a sterically hindered amine (G2), doubling the molar capacity of the chemical absorbent, would also bring benefits. By combining these two approaches the contribution can be reduced by 2/3 to 0.4 GJ/tonne CO₂, which is a large improvement over a G1 absorbent. Further progress is still possible through reduction of the approach temperature to 5 K and a 20% reduction in the solvent flow rate associated with the G3 absorbent technology, resulting in a contribution of chemical absorbent heating of 0.16 GJ/tonne CO₂. G4 absorbent technology assumes a further halving of the absorbent flow rate, which might be achieved through the use of an aqueous ammonia solution [11]. Also the heat exchanger approach temperature is further reduced, by incorporating the heat exchanger partly in the stripper section. Such flow sheet alterations have been suggested in [12] providing

for a more efficient regeneration process. Approach temperatures as low as 3 K are deemed possible and have been realised in seawater desalination equipment. Other engineering approaches to reduce the overall energy requirement are split-flow [12] and intercooling. Intercooling results in a higher loading of the solvent by limiting the increase in temperature in the absorber [2, 13].

Finally the contribution of the evaporation enthalpy required for the generation of stripping steam was assessed. The stripping steam generated in the stripper bottom is needed to provide a high enough driving force for CO₂ desorption. It also provides the heat requirement of the overall stripper and releases this heat upon condensation in the stripper column. However, the CO₂ will leave the top of the stripping still saturated with water vapour and the heat of condensation will be lost in the cooling water. The condensate is returned to the stripper and the reflux ratio, expressed as ton H₂O/ton CO₂, for a G1 absorbent is typically 0.7. The reflux ratio can be reduced by the use of chemical absorbents which exhibit a higher CO₂-partial pressure at a given loading and temperature, leading to an increase in the CO₂/H₂O ratio in the gas stream exiting the stripper. An example is the KS1 chemical absorbent developed by Mitsubishi Heavy Industries [3]. For such a G2 chemical absorbent the reflux ratio might be reduced to 0.6. Further lowering of the reflux ratio is achievable by the use of an integrated heat exchanger in the stripper, which allows recovery of the evaporation enthalpy inside the regeneration process [14, 15], leading to a G3 solvent process with a reflux ratio equal to 0.4. Finally, this might be further reduced to 0.1 in G4, which is achievable by non-aqueous chemical absorbents, based on e.g. ionic liquids.

The improvement potential in the three contributions to the thermal regeneration energy requirement can now be combined to provide the overall results. This is shown in Table 3.

Table 3: Synthesis results for the thermal energy requirement for regeneration of chemical absorbents

Absorption process development status		G 1	G 2	G 3	G 4
Binding energy	MJ/kmol CO ₂	80	70	55	30
HTX approach	K	15	10	5	3
Solvent flow	m ³ /ton CO ₂	20	10	8	4
Reflux ratio	ton H ₂ O/ton CO ₂	0.7	0.6	0.4	0.1
Thermal energy	GJ/ton CO ₂	4.56	3.31	2.29	0.95
Contribution CO ₂ binding	GJ/ton CO ₂	1.82	1.59	1.25	0.68
Contribution solvent heating	GJ/ton CO ₂	1.20	0.40	0.16	0.05
Contribution steam stripping	GJ/ton CO ₂	1.54	1.32	0.88	0.22

For G1 chemical absorption technology the thermal energy requirement is higher compared to results from recent analyses executed for a 30% MEA solution [10]. It presents the status of commercially available processes in the 80/90's of the last century. The binding energy typically amounts to 40% of the energy requirement, whereas liquid absorbent heating and the overhead steam contribute around 25% and 35%. Going from G1 towards a G2 technology the overall thermal energy requirement is reduced by 27%, which is achieved predominantly by the increased CO₂-loading of the solvent and a closer temperature approach in the heat exchanger. The thermal energy requirement for regeneration is somewhat higher than e.g. the thermal energy requirement for MHI's KS-1 solvent [3]. Next going from a G2 technology towards a G3 technology the thermal energy requirement is reduced by an additional 22% compared to G1 technology. A typical example of G3 technology is the precipitating absorbent technology [15], which results in high absorbent loadings enabling an easier regeneration and incorporates a heat integrated stripper. Finally, going from a G3 to a G4 technology an additional reduction of 29% in the thermal energy requirement compared to G1 technology is projected. A typical example of a G4 technology is aqueous ammonia [16]. Overall the potential for improvement of the thermal energy requirement for chemical absorbent is nearly 80%.

5. Power requirement of the capture process

The power requirement in the absorption process is in general determined by:

- The power needed for the blower to guide the flue gases through the absorber
This is determined by the volumetric flow rate, the pressure drop over the absorption column and the percentage removal of CO₂.
- The power needed for the solvent pumps
This is determined by the achievable solvent loading in the absorption process and the level of solvent regeneration, combined into the cyclic loading.
- The power needed for the compression process
This is determined by the thermodynamic properties of CO₂ and the number of stages.

Table 4 gives an overview of the expected technology improvements for G1 to G4 technologies. Here it is assumed that the scope for improvements in the efficiency of compression technologies is limited. However, through the use of lower pressure drop absorber packing materials and solvent with higher absorption rates, it is expected that the power for rotating equipment can be reduced significantly.

Table 4: Power requirement of the capture process

Process development status		G 1	G 2	G 3	G 4
Power rotating equipment	MWh/ton CO ₂	0.040	0.030	0.020	0.010
Power compression	MWh/ton CO ₂	0.114	0.108	0.102	0.095
Power total	MWh/ton CO ₂	0.154	0.138	0.122	0.105

6. Efficiency improvement of conventional coal fired power plants

The generation efficiency of pulverised coal fired power plants continues to be improved as a result of the progression towards higher steam pressures and temperatures [17]. This requires the development of new and stronger materials for use in piping and tubing in the boiler, including their manufacturing methods. The generation efficiency, however, is also influenced by the temperature of the cooling water, the air to fuel ratio, the allowable stack gas temperature and the use of reheat in the steam cycle [18]. In addition, the definition of the heating value of the fuel - higher heating value HHV (USA, Australia) versus lower heating value (Europe) - affects the numerical efficiency value. The efficiencies are therefore also influenced by the local conditions and conventions. The average generation efficiency for coal fired power stations in Australia is currently 0.35 (based on the HHV). In this study it is assumed that the efficiency will undergo an improvement from 0.35 (G1) to 0.5 (G4). The latter efficiency is not unlike the highest efficiency mentioned in [19], translated to the Australian conditions and conventions, i.e. higher cooling water temperatures and the use of the HHV definition. The two intermediate points have been chosen somewhat arbitrarily but it is assumed it will get increasingly difficult to obtain further efficiency improvements. Table 5 shows the efficiency development path for different generations of power plant technology.

Table 5: Efficiency improvement for coal fired power stations (based on HHV=26 GJ/ton coal; Emission factor= 0.09 ton CO₂/GJ_{th})

Status power plant technology		G 1	G 2	G 3	G 4
Efficiency	[-]	0.35	0.41	0.46	0.50
CO ₂ -emission	[ton CO ₂ /MWh]	0.928	0.792	0.706	0.650

Table 5 shows that by jumping from G1 power plant technology to G4 power plant technology CO₂-emissions are reduced by 30%. Although this is sizeable emission reduction on its own, the reduction by CO₂ capture and storage would be much higher.

7. Integration of post-combustion capture plant with the power plant

The absorption liquids are regenerated by raising the temperature, upon which CO₂ evolves from the solution. The heat can be supplied by an external heat source, but because of the low temperature levels (typically 120 °C) it is more efficient to derive the heat from the power plant by the extraction of steam from the steam cycle. The capture plant can be considered to be a cogeneration plant with the capture process being the heat recipient. The steam is

typically extracted at the cross-over between the intermediate and low pressure steam turbine. The extracted steam condenses in the reboiler to provide the heat for the regeneration process, but then it is not used to generate electricity and hence leads to a reduction in the power output. The loss in output of the power plant can be determined by a detailed integration study of the capture process with the power plant. Such a study would also entail the use of waste heat for the preheating of boiler feed water in detail. For the purpose of the present explorative assessment a much simpler analysis, which can be used for the future power plants with a higher efficiency and hence different steam and water flow rates, is followed. A convenient way of assessing the impact of the steam extraction on the power plant output is the use of a power equivalent factor (PeF) which relates the steam needed for the reboiler duty to the power output reduction. In [20] this factor is given graphically as a function of steam temperature and pressure for a natural gas fired combined cycle. In [21] the impact of the steam extraction is analysed in further detail and a general description for the power equivalent factor is given which takes into account both the reduction in power output and the use of the reboiler condensate for feedwater preheating. A significant portion of the heat requirement of the solvent process is, however, still recoverable as it leaves the regenerator as steam and its heat of condensation can be used, but because of the large amounts its re-use in the power station might be limited. The power equivalent factor has been estimated, for the different technology generations, assuming that the thermal energy re-use fraction will increase if the thermal energy requirement of the absorption process will decrease. However the power equivalent factor is assumed to level out at 0.15. The results of the analysis are shown in Table 6.

Table 6: Results of thermal integration of absorption process with power plant

Status power plant technology		G 1	G 2	G 3	G 4
Efficiency	[-]	0.35	0.41	0.46	0.50
Thermal energy	GJ/ton CO ₂	4.56	3.31	2.29	0.95
Power equivalent factor	[-]	0.25	0.20	0.15	0.15
Equivalent power requirement	MWh/ton CO ₂	0.317	0.184	0.095	0.040

8. Synthesis of analysis results and conclusions

The results of the thermal analysis of the absorption process and its integration into the power plant can now be combined to provide the overall assessment of the improvement potential. The synthesis results are given in Table 7, which draws on Tables 3,4,5 and 6, assuming 90% CO₂ capture.

Table 7: Synthesis results from overall analysis of energy performance

Status PCC technology		G 1	G 2	G 3	G 4
Efficiency (no capture)	[-]	0.350	0.410	0.460	0.500
CO ₂ -emission (no capture)	ton CO ₂ /MWh	0.928	0.792	0.706	0.650
Thermal energy absorption process	GJ/ton CO ₂	4.56	3.31	2.29	0.95
Equivalent power requirement of solvent regeneration process	MWh/ton CO ₂	0.317	0.184	0.095	0.040
Power requirement of capture process and compression	MWh/ton CO ₂	0.154	0.138	0.122	0.105
Overall power loss due to capture	MWh/ton CO ₂	0.471	0.322	0.217	0.145
Efficiency (with 90% CO ₂ capture)	[-]	0.212	0.316	0.397	0.458
CO ₂ -emission (with 90% CO ₂ capture)	ton CO ₂ /MWh	0.153	0.103	0.082	0.071
Increase in coal use due to capture	[%]	65	30	16	9

The results presented in Table 7 give an indication of the large potential for improvement of the post-combustion CO₂ capture process. Going from a G1 to a G4 technology leads to a 70% reduction in the overall power loss due to the CO₂ capture process. Further development of both the absorption process technology and the conventional

power plant technology have the potential to achieve generation efficiencies for coal fired power plants with post-combustion capture equivalent to current technologies (G2) without CO₂ capture. The required performance is typically slightly better than a G3 power plant with PCC technology, which achieves an overall generation efficiency of ~0.40 with 90% CO₂-capture.

9. Acknowledgments

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