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A comparison of post-combustion capture technologies for the NGCC

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Abstract

Four different post-combustion capture technologies are compared for the same NGCC (electric efficiency 58.1% without CO₂ capture). The technologies are aqueous amine, polymeric membranes, a low temperature sorbent and Ca-looping. The effect of 40% Exhaust Gas Recirculation (EGR) is compared for all technologies except Ca-looping. At 90% CO₂ capture rate, polymeric membranes (as expected) give a very poor efficiency (34.9-46.2%). Ca-looping efficiency spans over a wide range, 45.6-53.1 %, depending on process configuration and Ca-looping sorbent applied. The NGCC electric efficiency with aqueous amine capture (MEA or a Novel Generic Solvent) spans from 49.5-51.8%. The low temperature sorbent electric efficiency was found to be 50.8-52.3%. Aqueous amines, with their maturity, relative ease of process integration and potential for performance improvement when reducing reboiler approach, improving solvent performance and applying EGR appears to be a good option for CO₂ capture from the NGCC. The performance of the low temperature sorbent appear interesting enough to merit further investigations. Polymeric membranes could be interesting to evaluate further when aiming for lower capture rates, whereas Ca-looping mainly should be of interest to develop further for base load NGCC applications.

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Keywords: NGCC, CO₂ capture, aqueous amine, Ca-looping, polymeric membranes, low-temperature solid sorbent, EGR

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1. Introduction

The Natural Gas Combined Cycle (NGCC) may have a role to play as a load-follower in future energy systems with a high share of intermittent wind and solar power. In order to minimize the emission of greenhouse gases for such future energy systems, the NGCC should be equipped with CO₂ capture. Post combustion carbon capture is an attractive choice for the NGCC compared to other carbon capture routes (pre-combustion and oxy-combustion) because it can be retrofitted more easily into an existing plant. Several options are available or under development with various maturity levels, the most mature being CO₂ capture with an aqueous amine solvent.

Among the less mature technologies that are relevant to investigate with respect to integration in the NGCC are for instance Calcium looping (Ca-looping), low temperature solid sorbents and polymeric membranes. The purpose of this paper is to compare these three technologies against the more mature CO₂ capture with aqueous amines, in terms of net electric efficiency and capture technology integration with the NGCC. The default CO₂ capture rate (CCR) investigated is 90%, but additional (lower) capture rates were investigated for polymeric membranes. The targeted purity of captured CO₂ is minimum 95% molar composition for all considered technologies.

Nomenclature

ASU	(cryogenic) Air Separation Unit
CCR	CO ₂ capture rate
EBTF	European Benchmarking Task Force
EGR	Exhaust Gas Recirculation
FSC	Fixed Site Carrier
HRSG	Heat Recovery Steam Generator
MEA	Monoethylamine
NGCC	Natural Gas Combined Cycle
NGS	Novel Generic Solvent
PEI	Polyethyleneimine
SRD	Specific Reboiler Duty

2. Reference NGCC and CO₂ capture integration

An NGCC model was set up in Aspen HYSYS, using primarily the input parameters employed by the European Benchmarking Task Force (EBTF) [1]. The NGCC consists of a standard industrial gas turbine, from which the exhaust gas enters a steam bottoming cycle in a heat recovery steam generator (HRSG) in Fig. 1. This setup was previously described in publications investigating CO₂ capture with Ca-looping and aqueous amine capture in [2–4]. The nominal electric output without carbon capture is 416.4 MW_e from a LHV-based fuel input of 716.3 MW_e, resulting in an overall electric efficiency of 58.1%. The NGCC plant without CO₂ capture emits 41.1 kg CO₂/s (~1 Mt CO₂/year at 7500 operating hours/year).

The effect of exhaust gas recirculation (EGR) on the efficiency penalty with CO₂ capture was investigated for aqueous amines, a low-temperature sorbent and polymeric membranes. With EGR, a fraction of the flue gas exiting the HRSG is recirculated to the gas turbine inlet to be compressed and burnt with the fuel stream (Fig. 1). EGR can reduce the efficiency penalty of the NGCC with CO₂ capture, due an increase in the concentration of CO₂ in the exhaust, which provides higher driving forces for the CO₂ separation process. However, there is a limit to the extent of EGR possible in order to ensure flame stability and a good combustion efficiency. In the present paper, based on [5], EGR of 40% is considered, with a corresponding flue gas CO₂ molar concentration 7%. EGR was not investigated for Ca-looping, since in this case CO₂ is captured between the gas turbine and the HRSG,- This means

that the exhaust gas recycle would have to be located downstream of the gas turbine, at a high temperature, which would be impractical and also unfavourable from a thermodynamic point of view.. It should be noted that compressor and expander efficiencies in the gas turbine are kept constant in this paper as well as in [4] when simulating EGR. Also steam turbine efficiencies were kept constant.

CO₂ capture inevitably leads to an efficiency penalty for a power plant [6]. The efficiency penalty arises because of two primary reasons – the equivalent shaft work spent to enable the separation of CO₂ from a gas mixture (which is an exhaust gas for post combustion capture) and the equivalent shaft work required for CO₂ compression, intercooling and dehydration of the captured carbon dioxide stream. For the NGCC, the work spent for CO₂ separation when considering aqueous amines or low temperature sorbents, largely comes from steam extracted from the steam turbine section, meaning that the low pressure section of the steam turbine will generate less work. For polymeric membranes, shaft work is spent by electrically driven compressors and vacuum pumps that create a pressure differential across the membranes. In Ca-looping, CaCO₃ releases CO₂ and is regenerated to CaO through the addition of heat from an oxycombustion process that requires fuel. This heat can from a thermodynamic perspective be considered as equivalent shaft work, and also the cryogenic ASU that provides oxygen to the combustion process requires shaft work.

The different origins of the equivalent shaft work required for CO₂ separation means that the CO₂ capture process is integrated in different ways in the NGCC (Fig. 1). CO₂ capture with aqueous amines or low temperature sorbents are integrated at points A (after cooling of the exhaust at the HRSG outlet) and B (steam extraction for regeneration). Polymeric membranes have a single integration at point A. Ca-looping capture is integrated with the NGCC at point C, where there is a good temperature match between the gas turbine exhaust and the carbonation reaction.

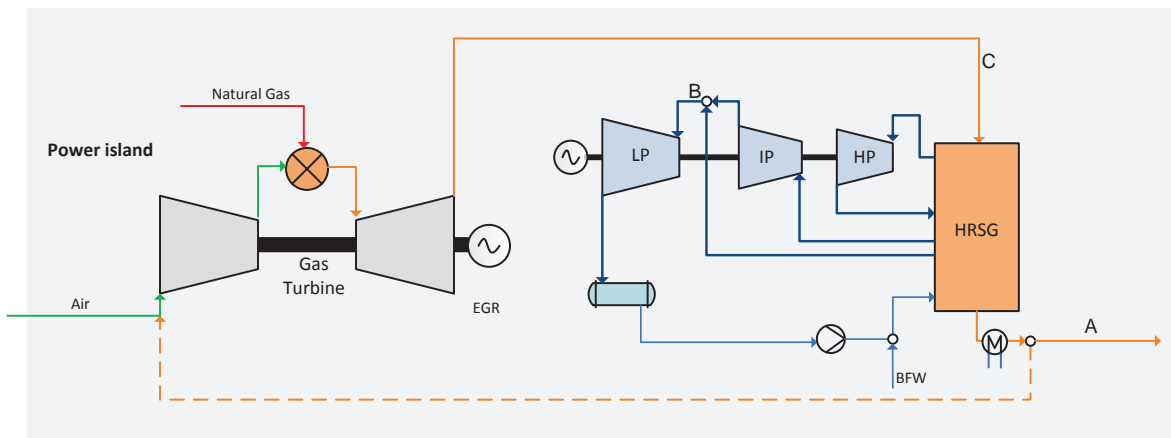


Fig. 1. Schematic of the NGCC, EGR illustrated with dashed line, points A-C indicate where capture technologies are integrated.

3. Aqueous amine capture

The process integration when capturing CO₂ with an aqueous amine solution is straightforward and known: The flue gas exiting the HRSG at point A (Fig. 1) is cooled and ducted to an absorber. CO₂ reacts with an amine, the resulting rich amine is circulated in the liquid phase to a desorber, where CO₂ is released from the amine through heat addition from condensing steam in a reboiler. In the NGCC, the steam is typically supplied from the crossover between the IP and LP steam turbines. After being released from the amine, the CO₂ can be dried and compressed for transport and storage.

Lindqvist et al. [4] investigated the integration of CO₂ capture from the NGCC using an aqueous solution of either MEA or a Novel Generic Solvent (NGS). The NGS was developed by SINTEF/NTNU within the SOLVit

R&D program [7]. The specific reboiler duty (SRD) at 118°C and 1.85 bar for regeneration of MEA is 4.0 MJ/kg CO₂ and the corresponding NGCC efficiency is 49.5%. The SRD for NGS at 120°C and 1.85 bar is 3.37 MJ/kg CO₂ and the corresponding efficiency is 50.4%. Lindqvist et al varied the desorber pressure for NGS from 1.85 bar to 3.5 bar, and the reboiler approach temperature from 20°C to 5°C for both MEA and NGS. EGR as a means to increase CO₂ concentration and reduce SRD was also investigated. It should be noted that for the calculations in [4], it was assumed that the LP steam turbine efficiency was identical with and without CO₂ capture.

It was found by Lindqvist et al that replacing MEA with the NGS, introducing Exhaust Gas Recirculation and decreasing the reboiler pinch point from 20 to 10 or 5°C all lead to improvements of the NGCC efficiency. Increasing the reboiler pressure from 1.85 bar to 3.5 bar for the NGS did not increase the NGCC efficiency, since the reduced power requirement for the CO₂ compressor is balanced by increased steam pressure for regeneration. For further benchmarking with other capture technologies, it is thus sufficient to consider the NGS cases with reboiler pressure of 1.85 bar.

4. Polymeric membranes for CO₂ capture

4.1. Process configuration

CO₂ capture from the flue gas stream exiting the HRSG (point A) can be achieved by passing it through a membrane that is selective to CO₂ relative to other components, primarily nitrogen. The driving force for membrane separation is the partial pressure difference of carbon dioxide between the feed ($p_{feed}^{CO_2}$) stream and the permeate stream ($p_{permeate}^{CO_2}$). A schematic of a single stage of the membrane separation process, together with its associated rotational equipment (compressors, vacuum pump, and expanders) is shown in Fig. 2. The membrane is considered with a cross-flow regime. The rotational equipment that sets up the partial pressure differential requires electrical energy. This together with the energy required in compression of the captured stream is extracted from the power plant resulting in a reduction in net power plant efficiency. The retentate leaves through the exhaust stack, while the permeate is piped to the next membrane separation stage. 2-3 sequential membrane stages are typically simulated. After the last separation stage the permeate is equal to the captured CO₂ of targeted purity. This stream is dehydrated and compressed for transportation and storage.

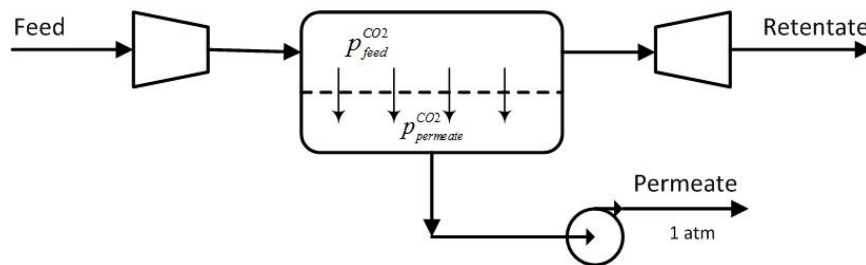


Fig. 2. Schematic of a single stage of the membrane capture process. The membrane is represented as the dotted line.

For a given feed carbon dioxide composition, the attained permeate composition is a function of the CCR, the available membrane area, the pressure differential across the membrane, as well as membrane properties such as permeance and selectivity. In addition, the number of membrane stages can be varied, to obtain a target composition of the captured CO₂ rich stream. Thus, design of the most energy efficient membrane capture process is a complex task as there are several parameters to consider. SINTEF Energy Research has developed a graphical methodology for systematic design of this process, based on minimizing energy consumption. This methodology is based on the 'attainable region' approach as presented in detail in [8–11].

A simplification is made by modelling the feed flue gas as a binary stream comprising only carbon dioxide and nitrogen components. Thus, the membranes are chosen by their selectivity to CO₂ compared to nitrogen. The membranes considered in the present paper are listed in Table 1 together with their characteristics. There exists a trade-off between membrane selectivity and permeability, and their theoretical limits are given by the Robeson plot [12]. Membranes A, B and D data are typical for what is currently being developed on a research scale while membranes C and E represent theoretical limits according to the Robeson plot.

A numerical version of the graphic SINTEF methodology, similar to that in [8] is used to determine the ideal design parameters optimized with respect to energy spent. Membrane capture processes with two or three stages were studied, and the number of membrane stages resulting in the higher power plant efficiency is reported. Single stage membranes are not considered because these are insufficient when a permeate CO₂ purity of 95% is to be obtained [9]. Minimum vacuum pressure is set to 0.2 bar, and maximum compression pressure is set to 50.0 bar, to provide practically feasible results. The total energy spent to create the pressure differential over the membranes is subtracted from the NGCC net power without CO₂ capture to calculate the net NGCC efficiency with CO₂ capture.

Table 1. Characteristics of the membranes considered for capture from the NGCC.

Membrane	Permeance [$\text{m}^3_{(\text{STP})}\text{m}^{-2}\text{h}^{-1}\text{bar}^{-1}$]	Selectivity [-]
A	5.94	50
B	13.9	50
C	5.94	79
D	2	135
E	4	135

4.2. Simulation results for polymeric membranes

Simulations were done for CO₂ concentrations representing the NGCC both without and with EGR. Since (as expected) membrane separation of CO₂ has a high power consumption at 90% CO₂ capture, lower capture rates were investigated, down to 60%.

Fig. 3 shows the variation of the power plant efficiency with different capture rates using the 5 investigated membranes. The results for membranes A and D are not visible in the plots because the power plant efficiency is only dependent on the membrane selectivity. Therewith, membranes A and B have similar efficiencies and so do D and E. This effect is due to the low CO₂ concentration of the NGCC exhaust. When it comes to determining the cost of membrane capture, the permeance, shown in Table 1, will indeed play a role also for the NGCC.

Lowering the CCR from 90% to 60% has a significant impact on decreasing power consumption for CO₂ capture, primarily because the flowrate of the second and third membrane feed streams decreases significantly when the flow through the first membrane stage is reduced. Also the pressure ratio across all membrane stages decreases. The decrease for the NGCC without EGR is ~7.5 to 10% points while the decrease with EGR is ~5% points. The increase in energy efficiency with reduced CO₂ capture rate is higher for membranes A and B compared with membranes D and E.

With a CO₂ capture rate (CCR) of 90%, membranes D and E have the best performance, although the performance is poor compared to the other CO₂ capture technologies investigated in this paper. The NGCC efficiency is only 39.0% without EGR and 46.2% with EGR. Reducing the CCR to 60% gives an NGCC efficiency of 46.5% without EGR and 51.0 % with EGR for membranes D and E. This electrical efficiency with EGR is in the same range or lower than what is possible to obtain for the other technologies in this paper at 90% CCR and *without EGR* (i.e. at a lower CO₂ concentration in the exhaust). Hence, from a pure efficiency point of view, polymeric membranes are not relevant for CO₂ capture for the NGCC, but are of more interest for CO₂ capture from streams with higher inlet CO₂ concentrations.

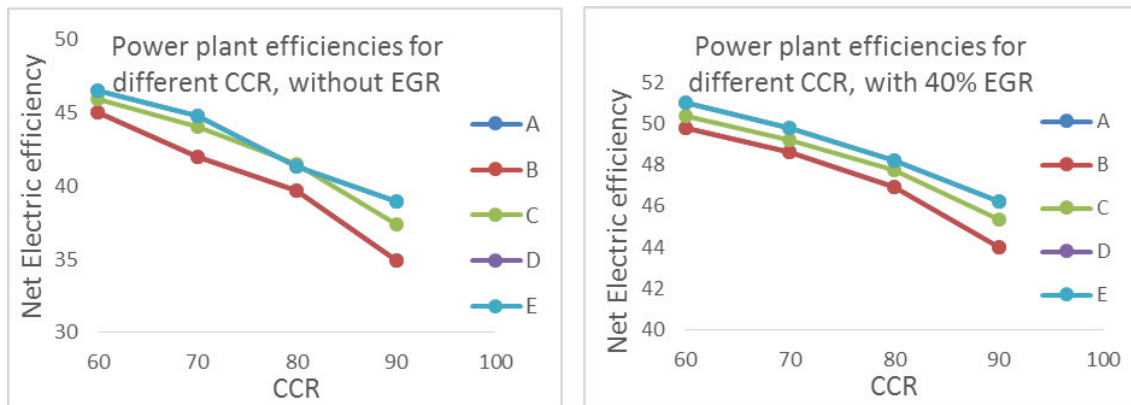


Fig. 3. Plot of the variation of power plant efficiencies with CCR for the 5 different membranes. Membranes A and B overlap, and also D and E overlap, meaning that the curves for membranes A and D are not visible in the graphs.

5. Low temperature sorbents

5.1. Background for study and process configuration

As an alternative to aqueous amines, a solid molecular basket adsorption sorbent was developed at Pennsylvania State University [13]. This sorbent is composed of a silica support material impregnated with polyethylenimine (PEI). PEI consists of primary, secondary and tertiary amines that adsorb CO_2 through the carbamate and bicarbonate reaction pathway as listed in [14]. In a study investigating CO_2 capture from the NGCC with this sorbent [13] the regeneration energy is claimed to be 2.69 GJ/tonne CO_2 , at a regeneration temperature of 110°C for an NGCC process having a CO_2 concentration of 4.04%. The pressure drop over the sorbent system (direct contact cooler, absorber and baghouse filter), that must be overcome by a fan 0.1724 bar. In the present paper, these data provided for the PEI solid sorbent CO_2 capture process have been combined with the simulation of the reference NGCC described in section 2, to provide a basis for comparison with aqueous amines, polymeric membranes and Ca-looping. The authors recognize that the investigation of the PEI solid sorbent is less in-depth than the other investigations presented in this paper. Nevertheless, it is thought that this comparison could have a value as an indicator of whether this type of solid sorbents merits further evaluation as a potential CO_2 capture technology for the NGCC.

A schematic of the low temperature sorbent process as simulated in the present work is shown in Fig. 4. The flue gas stream first passes through a pre-cooler (included in some of the simulations in the present work, but not in [13]) before being compressed by an exhaust gas fan. Excess water is knocked out, before it passes through a re-cooler to reduce the temperature to 40°C. The exhaust gas fan (with 85% isentropic efficiency) is used to compensate for the above mentioned pressure drop which is 0.1724 bar in [13]. The flue gas enters the adsorption tower at the bottom, in counter-current flow contact with the fluidized sorbent, whereby 90% of the CO_2 is adsorbed. The CO_2 is captured by the PEI solid sorbent in an exothermic reaction as the heat of adsorption is released. The adsorption tower is maintained at 70°C by a stream of cooling water that acts as heat sink for some of the released heat. The cleaned flue gas then leaves from the top, where it is filtered to remove entrained solid sorbent particles before being transferred to the exhaust stack.

The rich sorbent is mechanically conveyed (green dashed line in figure 3) from the bottom of the adsorption tower to the desorption tower. The desorption tower is maintained at a pressure of 1.85 bar, and temperature of 110°C as suggested in [13]. The desorption reaction to regenerate lean low temperature sorbent is endothermic. Regenerating steam is extracted from the NGCC low pressure turbine. It is used to supply the heat required to drive the desorption reaction as well as provide the sensible heat for the rich sorbent to attain the temperature of the desorption tower. After regeneration, the lean sorbent is returned to the adsorption tower, while the captured CO₂ stream is compressed and conditioned for transportation and storage.

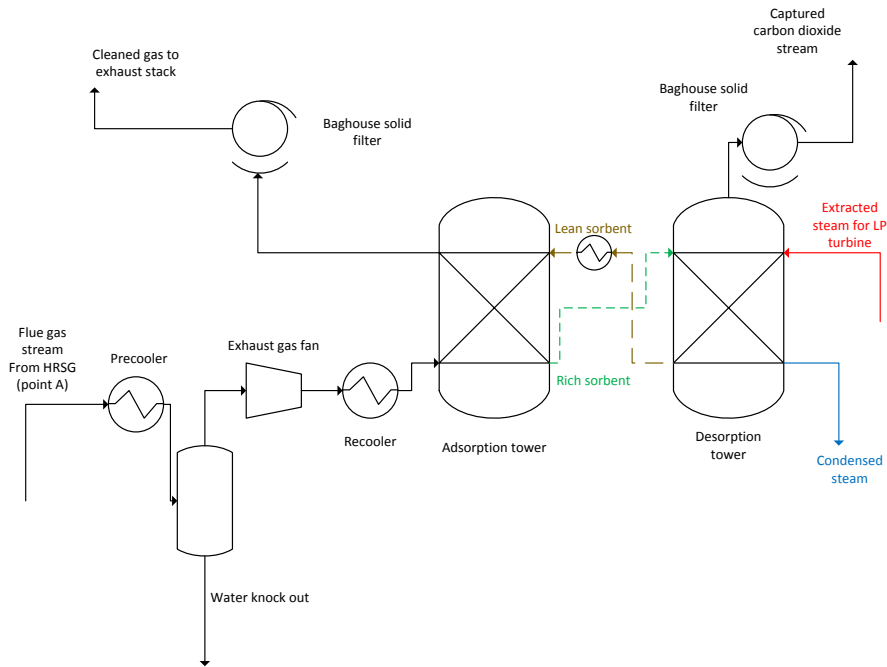


Fig. 4. Schematic of the low temperature sorbent capture unit, based on [13], with the addition of a precooler.

The temperature difference between the desorption tower and the condensing steam is analogous to the reboiler approach temperature in the aqueous amine capture unit and is set to 20°C, which was the largest temperature difference in [4], meaning that the regenerating steam enters the desorption tower, and is set to condense at 130°C. A pressure drop of 0.5 bar is assumed from LP steam turbine extraction point to for the condensing steam. For this pressure drop, steam has to be extracted at a pressure of 3.27 bar. This steam is desuperheated using LP water before being added to the desorption tower. The specific regeneration energy provided by the condensing steam is analogous to the specific reboiler duty (SRD) in the aqueous amine capture unit.

A rough attempt was made by the authors to estimate the efficiency of the NGCC with 40%EGR for CO₂ capture with the PEI solid sorbent, and the specific regeneration energy was roughly estimated by the authors to be in the range of 2.48 – 2.55 MJ/kg CO₂, when looking at how SRD decreases with increasing CO₂ concentration for aqueous amine capture. The approach for the rough EGR estimate was thereafter to calculate the NGCC efficiency with EGR and CO₂ capture for both 2.48 and 2.55 MJ/kg CO₂, and thereafter calculate the average.

The pressure drop in the adsorption tower in one of several parameters that merit a sensitivity analysis, due to the electric power consumption of the exhaust gas fan. The impact of pressure drops of 0.05, 0.10 and 0.15 bar was therefore investigated.

5.2. Low temperature sorbent results

Figure 6 shows the NGCC thermal efficiency results for the PEI solid sorbent Using the configuration suggested in [13] for an exhaust pressure drop of 0.1724 bar without a pre-cooler results in an NGCC efficiency of 50.8%, an improvement of 1.4 % points compared to MEA capture with a reboiler temperature difference of 20°C. Exhaust gas precooling (as indicated in Fig. 4) gives according to our calculations an increase the NGCC efficiency by 0.3% points. In order to evaluate the effect of the pressure drop through the direct contact cooler, adsorber and baghouse filter, simulations were also made with a reduced exhaust gas pressure drop, down to 0.05 bar, which is similar to what is assumed in [4]. This resulted in a net NGCC efficiency of 52.2% for the PEI sorbent in the present work. In other words, part of the efficiency gain from reduced SRD and reduced steam extraction pressure compared to MEA is offset by increased exhaust gas pressure drop over the absorber.

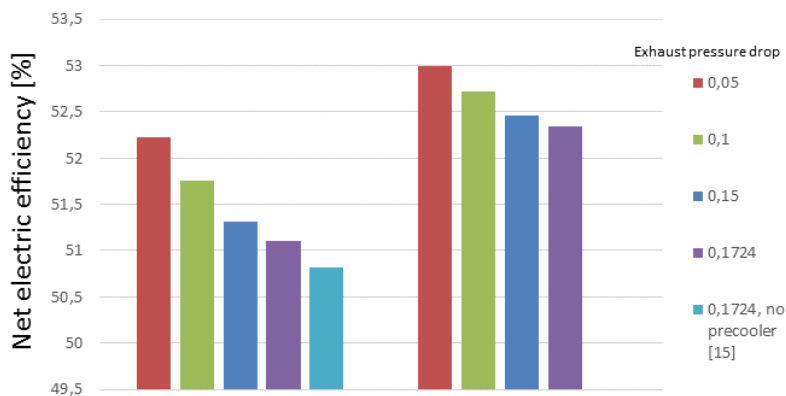


Fig. 5. Net electric efficiency of the NGCC with CO₂ capture using a low-temperature solid sorbent, input data from [13]

6. Calcium looping capture of CO₂

Berstad et al studied various process configurations with Ca-looping (previously referred to as CaO/CaCO₃ looping), and the corresponding impacts on net process efficiency [2,3]. Calcium looping uses a Calcium oxide solid sorbent (high temperature sorbent) in a carbonator through which the flue gas steam is passed as the 90% of the carbon dioxide stream is adsorbed to produce calcium carbonate. The required adsorption temperature for the carbonation stage in Ca-looping is above 600°C, and this temperature matches well with the temperature of the exhaust gas from the gas turbine, meaning that it is thermodynamically relevant to investigate Ca-looping as a CO₂ capture technology for the NGCC. Regeneration of the sorbent is done at 850-900°C in a calciner. The heat for regeneration is supplied though oxycombustion, using oxygen from an off-the-shelf cryogenic Air Separation Unit (ASU) and natural gas. The CO₂ leaving the calciner is therewith at an elevated temperature, meaning that heat is recovered for a secondary steam cycle.

Berstad et al. investigated both simple and more complex process configurations: Three different secondary steam bottoming cycle technology levels, and the addition of a solid-solid recuperator between the calciner and the carbonator were investigated in [2], where electric efficiencies ranged from 45.6% to 48.1%. Untreated calcite (limestone) was the only sorbent investigated in this work. Thereafter, in [3], a hot CO₂ recycle for calciner fluidization was investigated, as well as the impact of replacing untreated calcite with untreated dolomite or synthetic CaO. The three different secondary steam cycle technology levels were investigated also here, and electric efficiencies range from 50.0% to 53.1%. Concentration of the captured CO₂ varies from 94.8-96.6%.

Capture process integration into the NGCC is more complicated than for aqueous amine capture, since the Ca-looping absorber must be located between the gas turbine and the HRSG. This addition of a large volume between the gas turbine exhaust and the HRSG is expected to affect the NGCC ability to respond rapidly to load changes, as well as the retrofitability of the capture technology to the NGCC. Also, some of the other modifications made to the process may be challenging to resolve. In brief, the Ca-looping technology may not be practical for CO₂ capture from the NGCC in energy systems with a high requirement on rapid load response from NGCC plants, but could be valuable to explore further in an energy system scenario where NGCC with CO₂ capture is relevant for baseload operation.

7. Comparison of CO₂ capture technologies

A summary table of the four investigated post-combustion capture technologies without EGR is provided in Table 2, and the equivalent results with EGR are listed in Table 3. Primarily, the cases with lowest and highest efficiency are reported in the tables, together with a short explanation of the characteristics of each case. However, for aqueous amines, the highest efficiencies are reported for 10°C reboiler approach temperature, since recent calculations at SINTEF have revealed that 5°C may be unfeasible. Also, due to the high uncertainty of the EGR results for the PEI solid sorbent, only one result is listed for EGR here.

First of all, as already mentioned in section 4.2, the low efficiency for polymeric membranes, due to the high power consumption was an expected result at 90% capture rate. Hence, from a pure efficiency point of view, polymeric membranes are not relevant for CO₂ capture for the NGCC, but are of more interest for CO₂ capture from streams with higher inlet CO₂ concentrations. However, membranes have the easiest integration with the NGCC, and may still be of relevance to investigate further for reduced capture rates or cases where it is desirable to be able to switch on and off the CO₂ capture. It should be of relevance to investigate the cost for membranes as well as the other CO₂ capture technologies at reduced capture rates in order to properly evaluate the value of the membrane performance at 60% capture rate.

Ca-looping has a more complicated process integration, but also the highest electric efficiency of all investigated cases. In brief, the Ca-looping technology may not be practical for CO₂ capture from the NGCC in energy systems with a high requirement on rapid load response from NGCC plants, but could be valuable to explore further in an energy system scenario where the NGCC with CO₂ capture is relevant for baseload operation.

Aqueous amines and the low temperature (PEI) solid sorbent have the same principle for process integration, capturing CO₂ downstream of the HRSG, and using steam at a suitable pressure for regeneration. It can be seen in Table 2 that compared to MEA, the performance of the solid sorbent is superior, but when reducing the reboiler approach temperature and replacing MEA with an aqueous amine of superior performance, the efficiency is similar for the solid sorbent and the aqueous amine.

Aqueous amines, with their maturity, relative ease of process integration and indications for performance improvement potential when reducing reboiler approach and improving solvent performance appears to be a good option for CO₂ capture from the NGCC. The performance of the solid sorbent investigated in the present work is however interesting enough to merit further investigations both without and with EGR.

Table 2. Characteristics of investigated CO₂ capture technologies without EGR.

Technology	Lowest efficiency		Highest efficiency	
	Efficiency [%]	Characteristics	Efficiency [%]	Characteristics
Aqueous amines	49.5	MEA, reboiler approach temp 20°C	50.8	NGS, reboiler approach temp 10°C
Polymeric membranes	34.9	Membrane selectivity: 50	39.0	Membrane selectivity: 135
Low temp sorbent	50.8	No fluegas precooler, Δp exhaust 0.1724 bar	52.2	Fluegas precooler, Δp exhaust = 0.05 bar
Ca-looping	45.6	Modern but not supercritical steam data, no internal heat integration, natural calcite sorbent	53.1	Internal heat integration, hot CO ₂ recycle, supercritical steam cycle, synthetic CaO sorbent

Table 3. Characteristics of investigated CO₂ capture technologies with EGR.

Technology	Lowest efficiency		Highest efficiency	
	Efficiency [%]	Characteristics	Efficiency [%]	Characteristics
Aqueous amines	50.5	MEA, SRD approach temp 20°C	51.8	NGS, SRD approach temp 10°C
Polymeric membranes	44.0	Membrane selectivity 50	46.2	Membrane selectivity 135
Low temp sorbent	52.3	Fluegas precooler, Δp exhaust 0.1724 bar	52.3	Fluegas precooler, Δp exhaust 0.1724 bar

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