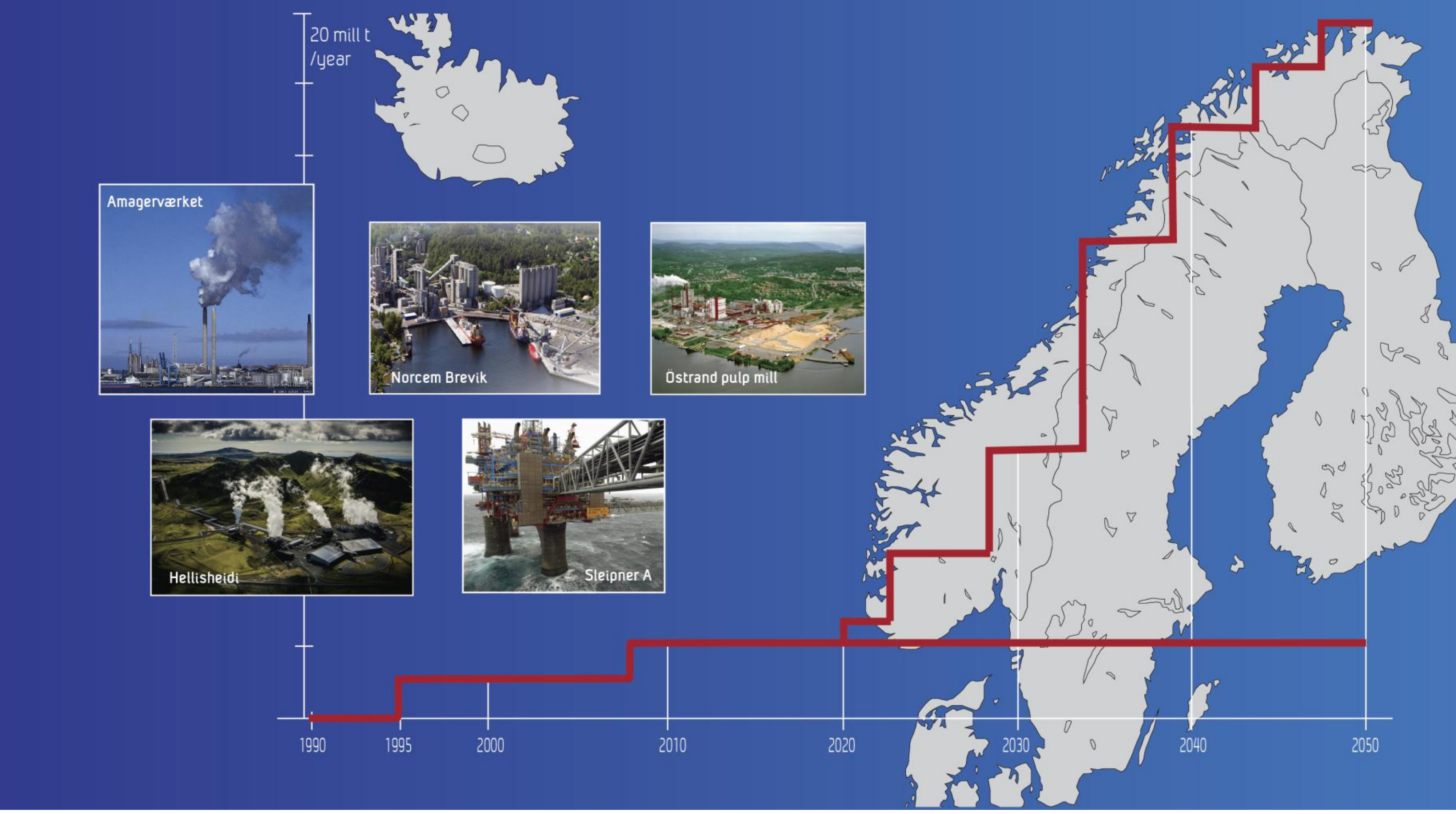

Industrial implementation of Carbon Capture in Nordic industry sectors

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Anette Mathisen, Lars O. Nord, David Berstad (eds.)

NORDICCS Technical Report D4.2.1501/D18

November 2015

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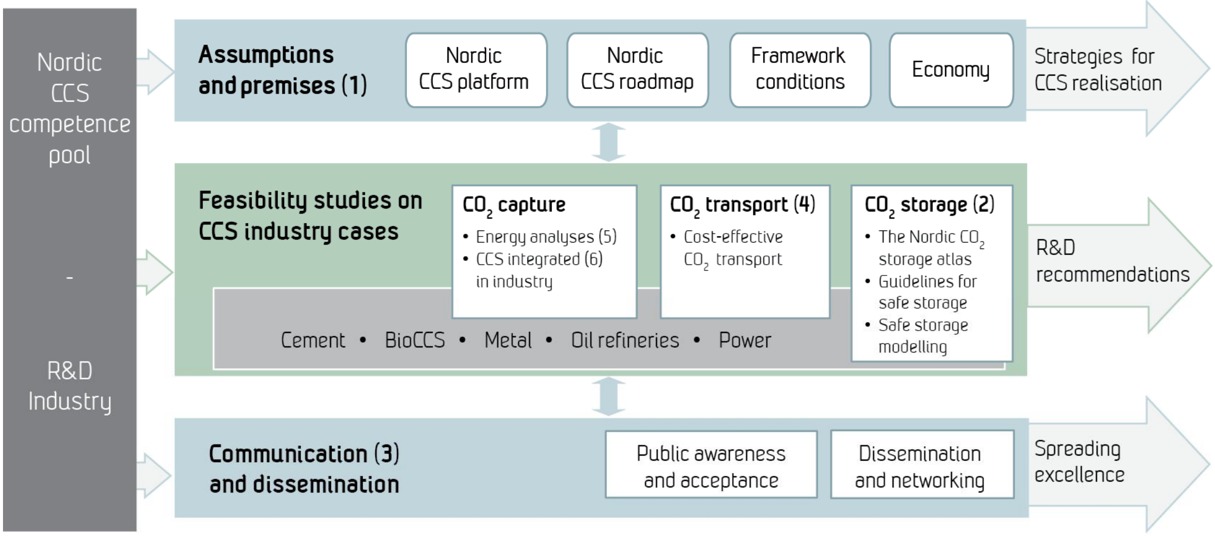

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Summary

This report investigates the feasibility of separating CO_2 from industrial gas streams for carbon capture and storage (CCS) in the Nordic countries. Detailed case studies were developed for iron and steel production, cement production, pulp and paper production, oil and gas refineries, and geothermal power production. Available technologies for separating CO_2 have been reviewed and detailed process simulations on the technologies feasible for implementation for the specific cases have been performed. The work shows it is feasible to apply carbon capture technologies to a broad set of process conditions. However, it is crucial to consider the specific conditions of each case as they strongly affect the performance of the technologies and the choice of technology. Considering process and site specific conditions as well as possible developments of the industrial process simplifies the implementation and increases the efficiency of carbon capture considerably. The specific findings for each case study are summarized below.

Iron and steel production

The steel mill case study showed that replacing the conventional gas powered steam cycle in the power plant with a low-BTU gas turbine improves process performance. Adding a water-gas shift reactor and a CO_2 capture process further increases the top gas fuel value. An additional 70 MW electric output can be achieved from this process configuration with a CO_2 reduction of about 80% - 85%. Furthermore, the design allows for a staged deployment of the concept, which minimizes the investment risk and burden of implementation.

Cement production

The case study on cement production favored a retrofit of the existing process by implementing an amine-based CO_2 capture plant. The capture unit can be added without any major updates of the cement plant. The most important installations required are the exhaust gas waste heat recovery unit, which will cover part of the steam needed to separate the CO_2, and the installation of exhaust gas cleaning equipment for SO_X, NO_X, and dust before the capture unit. Oxy-fuel combustion is also a feasible option for the cement case. In general, few modifications of the cement plant are required to implement oxy-fuel technology. To avoid air in-leakages in the cement plant, which has a considerable influence on the energy consumption of the compression and CO_2 purification unit, is one of the largest concerns for a retro fit of oxy-fuel combustion.

Pulp and paper production

The pulp production case considers possibilities with bio-energy with CCS (BECCS) as pulp mills mainly have biogenic CO_2 emissions. This work investigated the possibilities of implementing carbon capture to pulp mills using a conventional recovery boiler or black liquor gasification (BLG) technology. The latter scenario using either the Selexol process together with a combined cycle for electricity production or the Rectisol process together with dimethyel ether (DME) biofuel production proved a high performance. The results show that the pulp and paper industry is suitable

for BECCS. The combination of BLG technology and CO_2 capture would require low additional utility, compared with the conventional post-combustion process.

Oil and gas refineries

The case study on a refinery highlights the complexity of the refinery process that gives a large variety in size and quality of the many CO_2 sources. It is therefore of importance to investigate the sub-processes of the refinery individually. The most favorable CO_2 sources is the steam-methane reformer for hydrogen production due to high CO_2 content in the flue gas and being the largest point source of CO_2. The refinery also includes CO_2 sources that are not suitable for CO_2 capture, mainly due to their relatively small size, which will have the consequence that the overall capture efficiency of the refinery will be below the 85-90% that is possible to achieve on an individual stream.

Geothermal power production

The case study on geothermal power in Iceland is a unique example of a CO_2 source that is highly suitable for capture. The plant is already required to recover the high amounts of H_2S in the gas and there are several technology options for removing the H_2S and CO_2 from the turbine off-gas. Absorption in water as well as in amines was investigated with promising results. Also cryogenic gas separation would be technically feasible. Furthermore the results show the possibility of achieving a pure hydrogen stream if an extra pressure swing adsorption (PSA) unit is added.

Keywords CCS implementation, Nordic industry, CO_2 emissions

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Date
November 2015

About NORDICCS

Nordic CCS Competence Centre, NORDICCS, is a networking platform for increased CCS deployment in the Nordic countries. NORDICCS has 10 research partners and six industry partners, is led by SINTEF Energy Research, and is supported by Nordic Innovation through the Top-level Research Initiative.

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Contents

INTRODUCTION

During the last two centuries the concentration of CO_2 in the atmosphere has increased dramatically which is the likely cause of current global climate changes such as increased surface temperatures and extreme weather phenomena [1]. Since the start of the 1970s, the global anthropogenic greenhouse gas (GHG) emissions have increased by 70% and annual emissions of CO_2, the most important greenhouse gas, have increased by approximately 80%. The staggering increase in CO_2 emissions over this relatively short time period can be directly related to human activities.

A long-term goal is to stabilize the CO_2 emissions in the atmosphere to a level that limits the global average temperature to less than 2°C above the preindustrial level. As the CO_2 has a long lifetime in the atmosphere once released it is an urgent matter to stabilize the emissions and it is estimated that a 50 – 85% reduction of GHG gases is needed by 2050 in order to reach this goal [2,3]. The International Energy Agency (IEA) and the International Panel on Climate Change (IPCC) have identified Carbon Capture and Storage (CCS) as one of the required technologies for mitigating climate change. In the BLUE Map scenario [4], IEA considers CCS a key technology for reducing CO_2 emissions, constituting 19% of the total mitigation effect, see Figure 1.1.

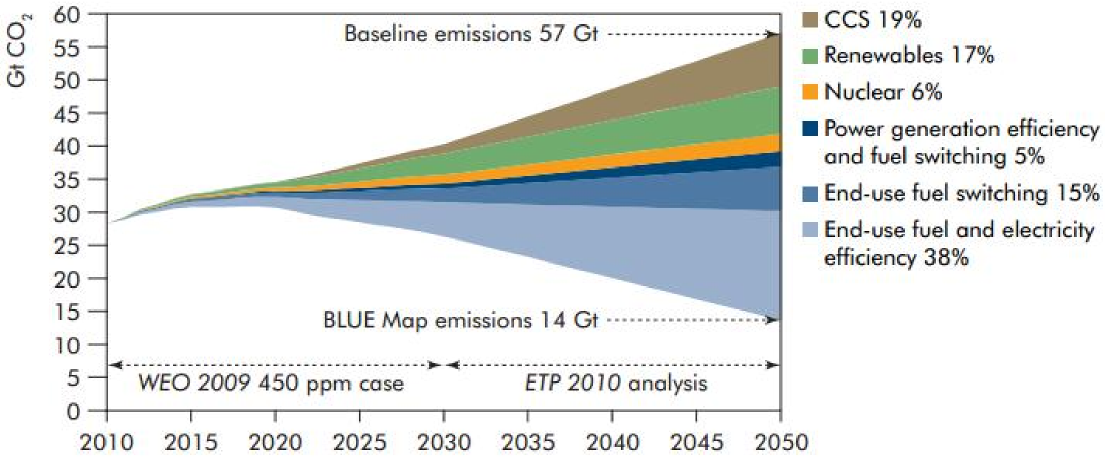


Figure 1.1 Key technologies for reducing CO_2 emissions under the IEA BLUE Map scenario [4].

The main part of anthropogenic CO_2 emissions is related to combustion of fossil fuels for energy conversion and alterations in land use [5]. Another large share originates from manufacturing processes such as cement, steel, paper and oil refining production processes. According to IEA [2], 21% of the total global CO_2 emissions originate from industry and an additional 18% originates from energy (electricity and heat) production. Implementing CO_2 capture and storage in industrial applications can therefore significantly reduce the total global CO_2 emissions.

Nordic industrial CO_2 emissions

The total Nordic CO_2 emissions from large industrial point sources (> 100 000 t/a) was 152.8 Mt in 2011. Nordic point source CO_2 emissions are dominated by power and heat production, pulp and paper production, oil and gas activities, iron and steel production and cement and lime production. A sectorial overview of the total industrial point source emissions for 2011 is illustrated in Figure 1.2, which illustrate the importance of industrial CCS in the Nordic countries in order to curb the anthropogenic CO_2 emissions.

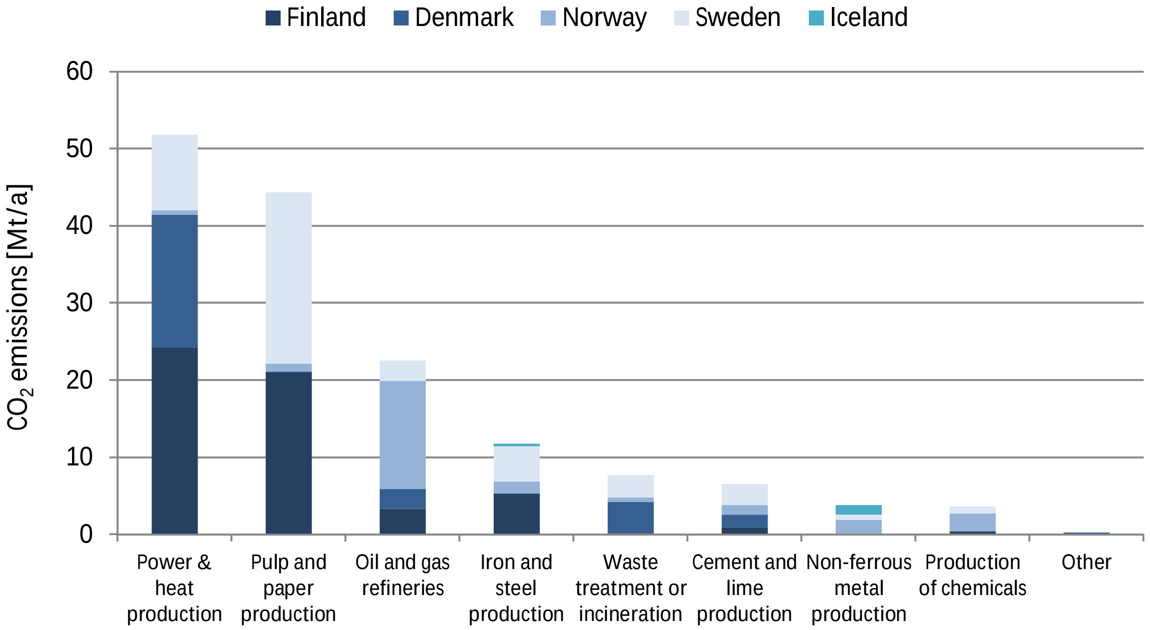


Figure 1.2 Nordic industrial CO_2 emissions by sector. CO_2 emissions from pulp and paper production include biogenic emissions as well as fossil emissions. Numbers are based on emissions from 2011.

Pulp and paper production is the second largest industrial point source emission sector after power and heat production. The forest industry plays an important role in the society and economy of the Nordic countries, especially in Finland and Sweden where it accounts for 16-20% [8] and 11-13% [9] of the country's industrial employment, exports, sales and added value, respectively. The pulp and paper industry is energy intensive, however the largest part of the industry´s CO_2 emissions originate from biogenic sources. Looking at Sweden as an example, the total CO_2 emissions from fossil fuels in 2011 were 49 Mt CO_2 while biogenic CO_2 emissions from the pulp and paper industry amounted to 22 Mt CO_2 [10].

The oil and gas industry is an important industrial sector in the Nordic countries, especially in Norway where it has played a major role in the country's economy since the 1970s. Offshore oil and gas extraction installations are operated both in Denmark and Norway. These activities are on a considerably larger scale in Norway, being the third largest exporter of natural gas in the world and fifth largest producer.

The oil refining industry is generally regarded as energy intensive and contributes considerably to the global CO_2 emissions [11], obviously not only from the industry itself but also from the end-use of their products (e.g. vehicle emissions). The oil and gas industry is an especially important emitter in

Norway where it today is responsible for around 28% of the domestic greenhouse gas emissions. Emissions include both offshore and onshore activities.

Iron- and steelmaking is an energy-intensive industry with significant CO_2 emissions (approx. 1 t CO_2/t steel). In 2011 more than 12 Mt of the Nordic point source CO_2 emissions originated from iron and steel production and currently all the CO_2 emissions from Nordic steel mills are fossil based.

An estimated 3.6 Gt of cement was produced globally in 2011 resulting in around 2 Gt of CO_2 emissions [6]. For the EU countries, the total 2010 CO_2 emissions for the cement sector were close to 100 Mt [7]. The annual production of cement in Europe has fallen over the last couple of years due to the economic situation and the resulting decline in construction activities; however, it continues to be an important industrial sector.

With energy production based almost exclusively on geothermal power production, industrial CO_2 emissions from Iceland are relatively low compared to its Nordic neighbor countries, and originate mainly from metal production (steel + non-ferrous). Geothermal power production does emit limited rates of CO_2 and H_2S. As of 2014, these gases are, for the most part (some are captured in a gas separation station for testing), emitted to the atmosphere after separation from the steam. New, emerging environmental regulations in Iceland will put constraints on the emission of H_2S [12]. Additionally, the long-term goal is to also reduce CO_2 emissions related to geothermal power. Therefore, separation of H_2S and CO_2 from the non-condensed gases (NCG) in the steam will be necessary, followed by some measure to store them.

Aim of research

This report assesses the technical possibility for implementing carbon capture technologies at case specific industrial sites under conditions relevant to the Nordic region. The aim is to provide the performance basis required for the economical assessment of industrial CO_2 capture within the NORDICCS project. The cost performance is reported in NORDICCS Technical Report D3.13.1302/D13 *CCS case synthesis – Final report*.

The technical possibility for implementation of carbon capture in the Nordic region is addressed through five case studies. The case studies represent the largest stationary industrial CO_2 emitters in the Nordic countries (power and heat production is not included). The cases include implementation of carbon capture in:

I. Iron and steel production in the Gulf of Bothnia area

II. Cement production in Norway

III. Pulp and paper production in Sweden

IV. Oil and gas refineries in Sweden

V. Geothermal power production in Iceland

METHODOLOGY

The case studies in this report are to a large extent based on industrial data from the actual plants described. Design and performance parameters were developed based on input from the plants taking into consideration local conditions for utilities like cooling water, heat, fuel and logistics. The project designed and evaluated the performance of retrofitting carbon capture to these existing plants by detailed process modelling using Aspen Plus and Aspen HYSYS. A capture rate of 85% CO_2 was considered for all cases.

The assessments follow the principles of consequential impact assessment, and only the specific parts of the plants that are affected by the application of CCS have been considered.

CO_2 capture technology

Amine based post-combustion CO_2 capture, which is a mature commercially implemented process with origins in the oil and gas industry, is the default capture technology in all case studies. However, the optimal choice of CO_2 capture technology is specific to each type of industrial plant and a series of capture technologies have been considered, including oxy-fuel combustion in the cement industry and Selexol post-combustion CO_2 capture in the iron and steel industry. For one of the plants investigated, the geothermal power plant on Iceland, specific considerations toward the choice of CO_2 capture technology are made due to the flue gas composition. In addition to the traditional MDEA absorption process for H2S and CO_2, alternative technologies such as water absorption and low-temperature (cryogenic) separation have been considered.

In the following sections a description of the monoethanolamine (MEA) based post-combustion CO_2 capture model is presented. The process simulations of the CO_2 capture plant were performed using Aspen Plus. A CO_2 capture rate of 85% is used as the basis for the simulation with a 30 wt-% MEA solution as the solvent. The absorber and desorber columns are simulated using the RadFrac block for rate-based calculations with the Electrolyte NRTL property model, and the Redlich-Kwong equation of state.

MEA absorption for CO_2 capture

The flue gas enters at the bottom of the absorber with a temperature of between 30 – 50 °C. The liquid absorbent, MEA, enters the absorber from the top. The flue gas flows upwards in the column and is contacted with the absorbent flowing down in a countercurrent flow. The CO_2 reacts with the liquid absorbent and exits at the bottom of the absorber with the absorbent in a CO_2 rich amine flow. The clean flue gas exits at the top. The rich absorbent is heated in a heat exchanger with the regenerated absorbent leaving the desorber before it enters at the top of the desorber. Energy in the form of steam with a temperature of approx. 120°C is supplied to the desorber reboiler. The steam is used to heat the rich absorbent and the CO_2 is desorbed from the absorbent. The CO_2 exits from the top of the absorber, while the CO_2-lean absorbent exits from the bottom. The CO_2-lean absorbent is then returned to the absorber after being cooled in the lean-rich heat exchanger and in an additional cooler. The process is illustrated in Figure 2.1.

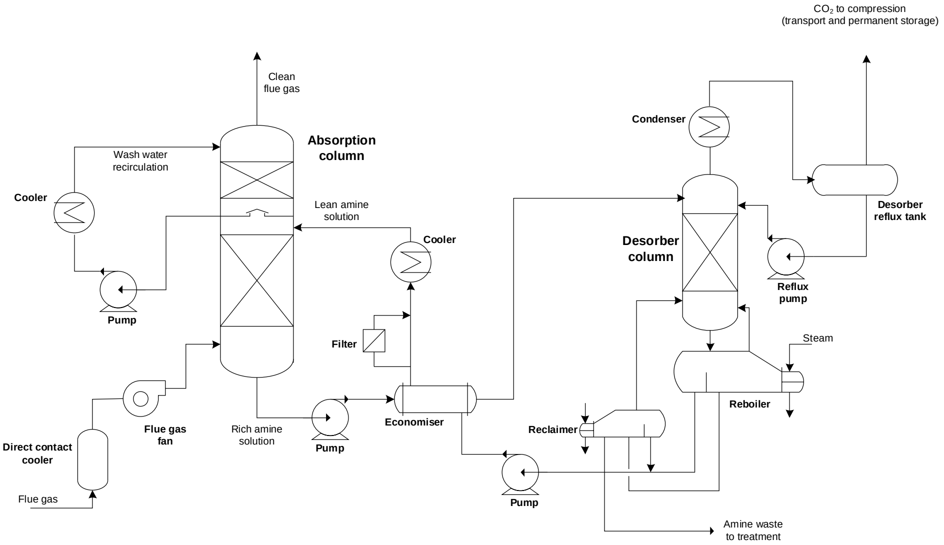


Figure 2.1 MEA post-combustion CO_2 capture

Assumptions for MEA post-combustion capture

Table 2.1 gives an overview of the assumptions made for the MEA post-combustion CO_2 capture modelling. All the case studies apply the same assumptions.

Table 2.1 Assumptions for MEA post-combustion CO_2 capture modelling

Area	Assumption	
	Packing material	Mellapak 250Y
PROCESS GAS	Temperature	40°C
	Impurities	None
ABSORBER	Pressure, top stage	1.012 bar(a)
	Pressure drop	0.012 bar
DESORBER	Operating pressure	1.9 bar
	Pressure drop	None
REBOILER	ΔT	10°C
	Regeneration temperature	120°C
STEAM	Temperature	130°C
	Pressure	2.7 bar
HEAT EXCHANGER	ΔT	5°C
CO_2	Final pressure	70 bar

The oxy-combustion process in the cement production case study was modeled and simulated in Aspen HYSYS. The chosen property package was Peng-Robinson-Stryjek-Vera EOS (PRSV). The assumptions made for the oxy-combustion simulations are presented in table 2.2.

Table 2.2 Assumptions for oxy-combustion CO_2 capture modelling

Area	Assumption	
ASU	O2 purity	95 mol-%
	N2 content	<3 mol-%
	Argon content	<3 mol-%
HEAT EXCHANGER	Type	Sea water
	Temperature (average)	12°C
	Pinch	> 10 K
	Pressure drop	0.3 bar
CO_2	Final pressure	110 bar(a)

Water absorption process for CO_2 and H_2S capture

The non-condensed gases from the steam turbines (see Chapter 7) are cooled before entering the compression train, which consists of three stages. The compressor stages include intercooling and after-cooling. The water for the absorber is pumped up to the selected absorber pressure (15 bar) before entering the top of the absorber. 10 stages were used in the absorber. Packing material with a diameter of 0.09 m was used. The process was modeled with Aspen HYSYS version 8.3 using the Predictive Soave-Redlich-Kwong property method was used for the water absorption process simulations. A simplified flow sheet of the water absorption method is shown in Figure 2.2.

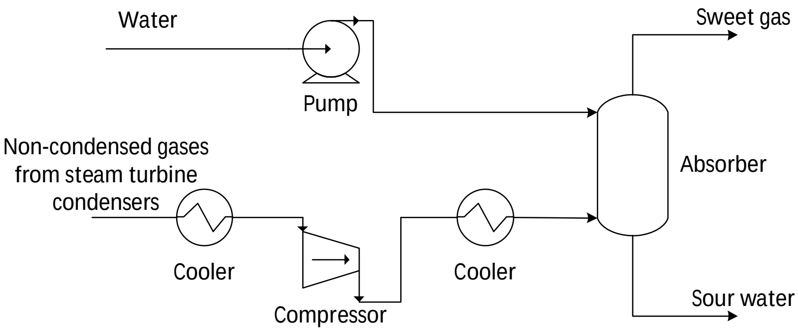


Figure 2.2 Simplified water absorption flow sheet

MDEA absorption process for CO_2 and H_2S capture

The MDEA unit was first simulated in Aspen HYSYS 8.3 using the amine package and assuming equilibrium conditions. However, as H_2S-selective absorption processes can be operated in non-equilibrium mode, more CO_2 can pass through the absorber without reaching equilibrium in reaction with the solvent. This mode of operation was investigated in the more advanced Acid Gas Cleaning package available in Aspen HYSYS. The basic process flow diagram of the MDEA unit is shown in Figure 2.3 and the main assumptions for the simulations are given in Table 2.3.

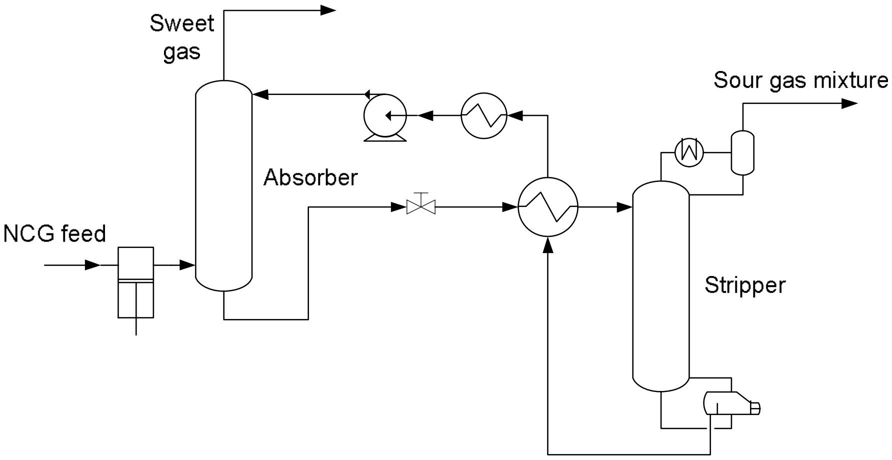


Figure 2.3 Amine absorption acid gas separation scheme

Table 2.3 Main assumptions for MDEA absorption process in the Iceland case

Area	Assumption	
Absorber	Pressure, top stage	19.5 bar(a)
Desorber	Operating pressure	1.2–1.5 bar
Heat exchanger	ΔT minimum	10°C
Pump	Adiabatic efficiency	75 %

Low-temperature (cryogenic) process for CO_2 and H_2S capture

Low-temperature gas separation units were simulated in Aspen HYSYS 8.3 using Peng–Robinson EOS. More details and process illustrations are given in the relevant sections in Chapter 8 Carbon Capture in Geothermal Power Production in Iceland.

Energy supply and integration of CO_2 capture

The energy consumption in a MEA based CO_2 capture plant is mainly in the form of steam supplied to the desorber reboiler to provide the heat for regeneration of MEA. In addition, energy in the form of electricity is needed for the process/flue gas fans and CO_2 compression.

For some industrial plants, heat can be recovered from the process gas to provide steam for the reboiler. However, not all industrial plants have sufficient process gas or the required process gas quality to cover all the energy needed. In those cases the remaining energy requirement can be provided by a combined heat and power (CHP) plant. Alternatively, the electricity can be bought from the electricity grid.

An onsite CHP plant can be fueled by natural gas, coal or biomass. The burning of these fuels will increase the CO_2 emissions from the overall industrial process, and must also be taken into account regarding CO_2 capture.

Post-combustion CO_2 capture is considered to be less intrusive on the industrial/power plant than oxy-combustion and pre-combustion. Integration of CO_2 capture with the industrial/power plant is considered individually under the specific case studies, this includes heat recovery from the industrial plant.

CARBON CAPTURE IN IRON AND STEEL PRODUCTION

Authors: Kristin Onarheim and Antti Arasto (VTT Technical Research Centre of Finland, Ltd.)

Process overview

Steel is mainly produced in a primary steelmaking process where iron is extracted from raw iron ore. The principle of extraction is to combust the combustible fractions of the ore and simultaneously smelt the metallic fractions in a blast furnace (BF). The extracted pig iron is further refined in a basic oxygen furnace (BOF) and blended to different steel grades.

The BF + BOF route uses carbon as a reducing agent in the blast furnace for transforming raw iron ore into pig iron alloy (iron making) and further into low-carbon steel. A simplified schematic illustration of the typical integrated iron and steel production process is shown in Figure 3.1.

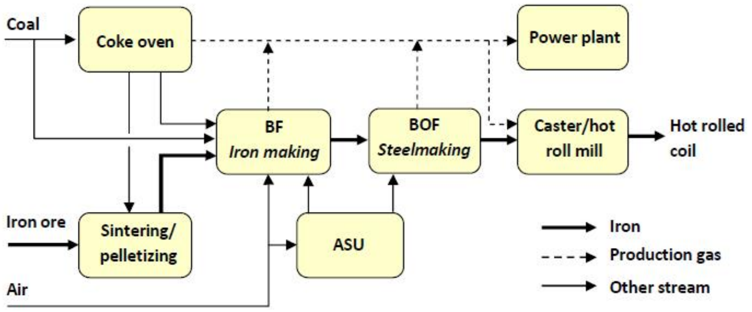


Figure 3.1 Simplified block flow diagram of an integrated iron and steel production process based on blast furnace and basic oxygen furnace route via pig iron. Reproduced from [1].

Coal or lignite is pyrolyzed into coke in a coke oven consisting of tall, narrow oven chambers at about 1 000°C. The pyrolysis process, also known as dry distillation, drives off the volatile matters from the coal in an oxygen-free environment to result in pure carbon. The conversion rate is typically 75% coke and 25% gas. The heat required for the coking process is usually provided by the coke gases themselves after they have been cleaned, and in some cases partly also by blast furnace gases. Coke oven gas can also be used in the blast furnace [2,3].

The produced coke is used in the sintering or pelletizing process to agglomerate iron ore (Fe_2O_3 or $Fe_{3}3O_{4}$) into small clusters. Iron ore typically consists of 60–65 wt-% iron (Fe). Coke is also used as a reducing agent and fuel in the blast furnace [2,3].

Iron agglomerates, or sinters, are charged at the top of the blast furnace together with coke, flux (e.g. limestone) and sometimes also lump ore at alternating frequencies. Sinters typically contain 55 – 60 wt-% pure iron. Also pulverized coal can be added to the blast furnace instead of coke [2]. In addition, hot air, or sometimes recycled flue gas, is added to the blast furnace. At the top of the blast furnace the feed is dried by the hot gases blowing through the furnace. As the feed travels downwards, the temperature increases. As a consequence, the carbon is burnt in a reducing reaction and the increasing heat causes the iron ore to melt. The combustion process takes place in the freeboard above the furnace bottom and the molten iron (also called pig iron) drips to the bottom of the vessel. The average temperature of the blast furnace is 1 500°C [2].

Besides melting the metal fractions in the iron ore, another important task for the blast furnace is to get rid of the oxygen in the iron ore. The additional coke fed directly to the blast furnace ensures an efficient reduction reaction producing mainly carbon monoxide, which again reduces the iron oxides to iron. In order to produce one ton of pig iron approximately 1.5 ton of iron ore and 450 kg of coke is needed [2].

The pig iron (hot metal, HM) produced in the blast furnace still contains some carbon after the reduction process, typically 4–4.5 wt-% [2]. This residual carbon makes the metal fragile and breakable and needs to be removed.

The top gas from the blast furnace exits at about 2–3 bar and contains +/- 20 vol-% CO_2. Gas from the blast furnace is typically used as fuel for the power plant and the hot stoves. As part of the fuel preparation of the gas, dust, and possibly also sulfur components, are removed in a wet scrubber and the gas is expanded in a turbine train [1,4].

The carbon enriched pig iron from the blast furnace is processed further in the basic oxygen furnace (BOF), also called converter. In addition to the pig iron usually also metal scrap is added to the BOF. The ratio is about 4:1 [2]. In the BOF a jet of almost pure oxygen is blown through the charge, removing most of the residual carbon and impurities in a range of reducing reactions. After the BOF the molten steel tapped and in some cases other alloys are added before it is casted and usually rolled [2].

The combustion of coke oven gases and blast furnace gases together with hot stove flue gases in the power plant result in relatively high CO_2 emissions. The layout of an integrated steel mill today also shows that the CO_2 emissions are distributed over several emission points on site, and this has to be taken into consideration when developing carbon capture solutions for the steel industry.

CO_2 capture in iron and steel production

All CO_2 emissions from a steel mill are related to combustion processes, and about 80–90% of the emissions originate from iron making [5] due to the high carbon intensity and large energy requirement of the processes. As a consequence, all main CO_2 capture technologies (post-combustion, pre-combustion and oxy-combustion) are in principle applicable. The majority of the carbon flow into an integrated iron and steel mill is emitted through the blast furnace, the coke oven and the basic oxygen furnace [6]. Outside the core iron and steel production routes, in a fully integrated steel mill, most of the carbon is emitted through the power plant.

Major efforts have been undertaken the last years to reduce the CO_2 emissions from iron and steel mills. The most conventional initiatives are increased energy efficiency by mill improvements and heat integration/heat recovery, especially for the blast furnace, recycling of by-product fuels, maximizing scrap steel recycling and increase in use of renewable energy [7] such as for instance biomass, and changing to a fuel or reducing agent with lower carbon emission factor [8]. In addition, major effort on the utilization of pure oxygen and top-gas recycling in the blast furnace has been made [9,10,11].

The *CO_2 Breakthrough Programme* was launched by the World Steel Organization in 2003 as an initiative to exchange information on regional activity on CO_2 reductions within the iron and steel industry. A number of sub-projects were initiated; among them the EU ULCOS program (Ultra-Low CO_2 Steelmaking), the US AISI Breakthrough Program, the Japanese COURSE50 project together with many others.

The European Ultra Low CO_2 Steelmaking program (ULCOS) is a European consortium of 48 European companies and organizations including major steel producers. The main purpose of the cooperative R&D initiative is to reduce the CO_2 emissions from iron and steel production. The overall target is a 50% reduction of CO_2 emissions [11].

The ULCOS initiative has identified four breakthrough technologies that can serve the purpose of the project, but only when combined with conventional carbon capture technology:

- Top Gas Recycling Blast Furnace (TGR-BF) – recycling of BF top gas enables a reduction of coke input to the BF. An oxygen-blown BF further facilitates CO_2 capture compared to an air-blown furnace.
- HIsarna – feeding of powdered coal and iron ore directly into the furnace, thereby omitting the need for coking, sintering and pig iron production in a blast furnace prior to steelmaking as in conventional processes
- ULCORED - gas-based Direct Reduced Iron technology based on for instance natural gas or syngas produced from coal gasification
- ULCOWIN/ULCOLYSIS (electrolysis)

Case study – Modified blast furnace and power production processes

The technological option for reducing CO_2 emissions in this case study is replacing the existing power plant gas boiler and conventional steam cycle with a gas turbine combined cycle (GTCC) supplemented with pre-combustion CO_2 capture. The case study is based on investigations on CCS in Nordic iron and steel industry with input data from the CCS Finland and CCSP projects [4,12,13]. Four cases were established:

- Case 0: Base case – conventional integrated steel mill based on the conventional blast furnace production route
- Case 1: Modified blast furnace concept – improved iron production process where the blast furnace serves both as a hot metal producer (primary role) and a fuel producer (secondary role).
- Case 2: Modified blast furnace + MEA – Modified blast furnace concept with MEA CO_2 capture
- Case 3: Modified blast furnace + Selexol – Modified blast furnace concept with Selexol CO_2 capture

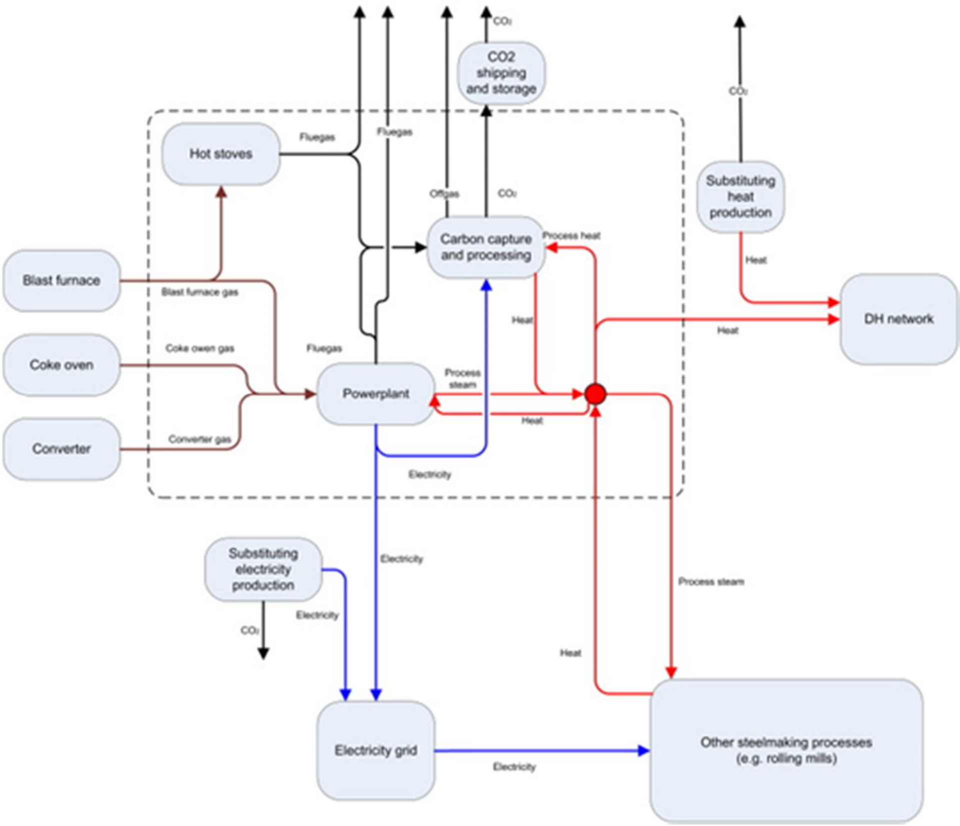


Figure 3.2 A simplified flow sheet illustrating the boundary limits for the base case [4]

Case 0: Base case

The annual production rate is 2.6 Mt of hot metal. The blast furnace is supplied with coal, pulverized coal injection (PCI) and enriched oxygen blast. The blast furnace top gas is utilised on site for firing the hot stoves in order to heat up the hot blast for the blast furnace and in the power plant in order to produce power and heat.

The power plant consists of a gas boiler and a steam cycle with steam extraction from the steam turbine to supply the mill with electricity and process steam. The mixture of fuel gases utilised in the gas boiler consists of blast furnace top gas, coke oven off-gas and converter off-gas. The steam cycle power production process typically has an efficiency of maximum 29% [14]. In addition to electricity, district heat for the surrounding premises is also produced in the power plant.

The oxygen content in the blast furnace feed is 21-29 mol-%. The calorific value of the conventional blast furnace top gas is rather low, typically between 3-4 MJ/Nm^3 while the coke rate is 300-360 kg/Mt HM [14]. The heating value of the base case blast furnace top gas was 2.7 MJ/kg. The low heating value is mainly a consequence of the high concentration of nitrogen (45.1 vol-%) and CO_2 (22.1 vol-%). The base case serves as a comparison to the cases investigated in this work. A simplified flow sheet of the base case is illustrated in Figure 3.3.

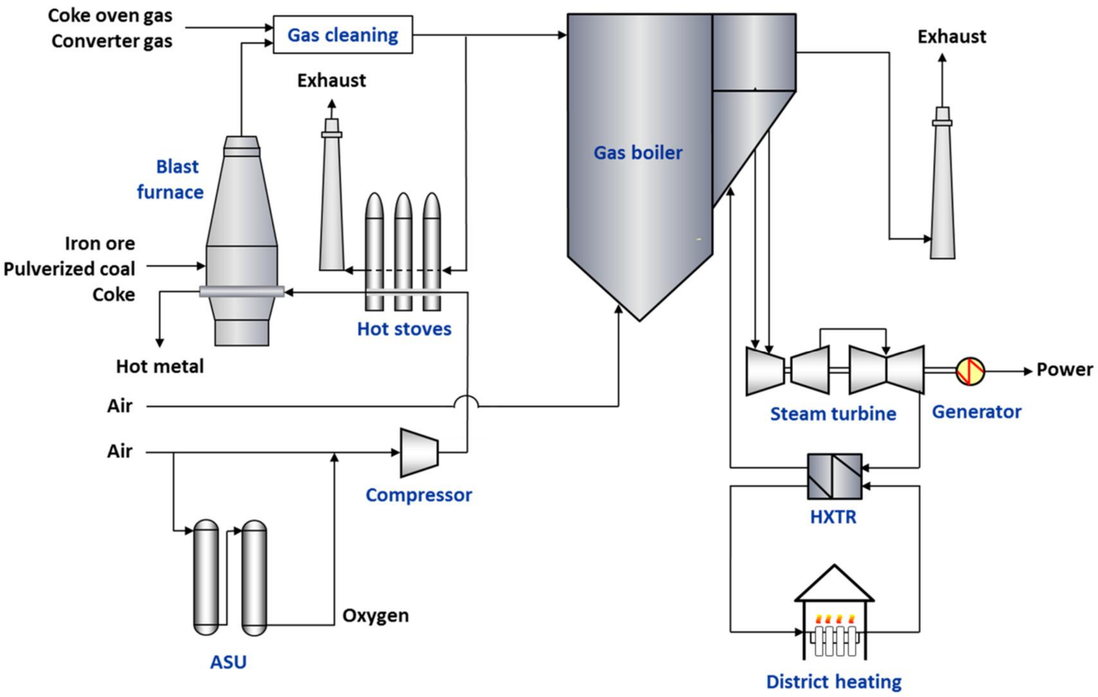


Figure 3.3 Conventional blast furnace with gas boiler power plant

Case 1: Modified blast furnace concept

Case 1 is based on the Blast Furnace Plus process, a patented concept developed by Air Products and Danieli Corus [14]. The basis for this concept is to increase calorific value of the blast furnace top gas used in the power plant. This is done by replacing part of the more expensive coke with increased pulverized coal injection (PCI) and enriched oxygen to the blast furnace. The resulting higher-calorific value top gas can be applied in a high-efficiency combined cycle for power production.

In the modified blast furnace concept the PCI and air separation (ASU) units are larger in order to serve the increased injection of PCI. The increase in enriched oxygen is necessary to maintain the required flame temperature in the blast furnace. Typically 12-19 %-point of additional enriched oxygen is required for the process. The flame temperature can also be lower than for the conventional blast furnace, which would give an additional advantage. The top gas is cleaned in the existing base case gas cleaning system. In the modified blast furnace process the main part of the top gas is further purified in an electrostatic precipitator (ESP) before it is dried and compressed to high pressure fuel gas prior to combustion in the combined cycle power plant. The flue gas exiting the gas turbine is used in a steam boiler where it produces steam for a steam turbine. The top gas calorific value from the modified blast furnace process typically increases with 1.5-2 MJ/Nm^3 compared to the base case. The minimum calorific value of a top gas fuel to the gas turbine is typically from 4 MJ/Nm^3. The overall efficiency of this concept is approximately 43% compared to around 29% in the conventional steam cycle [14]. Applying a low-BTU fuel in a gas turbine requires severe process modifications and installation of gas turbines that are specifically designed for this type of fuel.

The modified blast furnace concept offers in this way a decrease in the specific CO_2 emissions associated with iron production through allocating the emission on both blast furnace products; hot metal and power. A comparison between the main iron production values of conventional steel making in the base case. The modified blast furnace concept is illustrated in Figure 3.4.

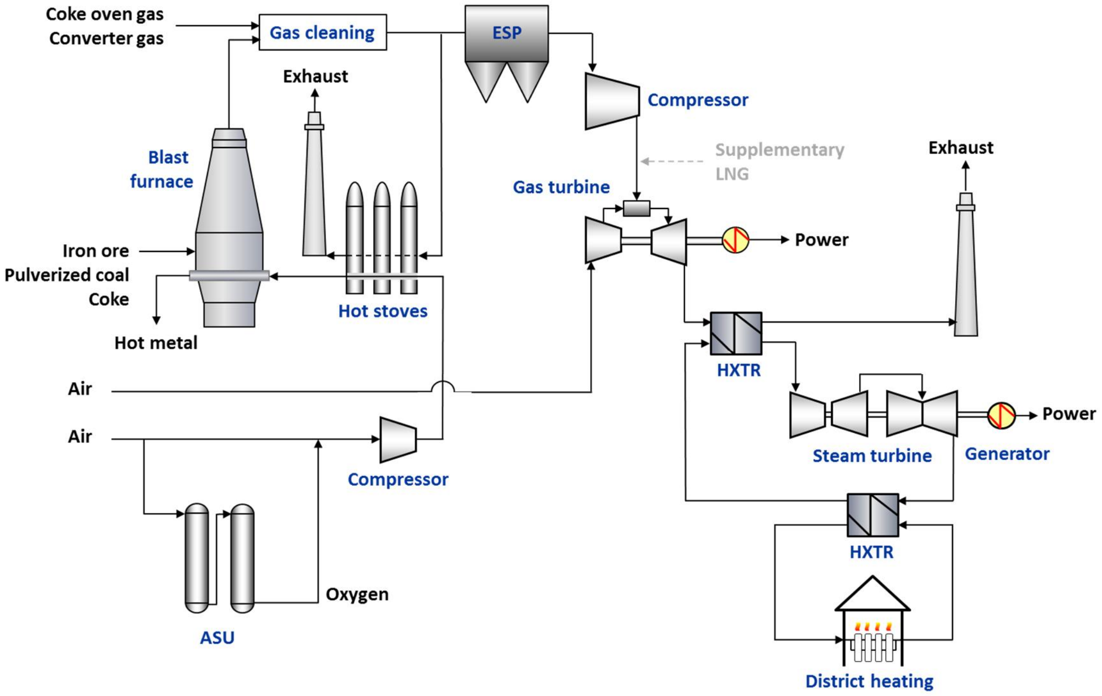


Figure 3.4 Modified blast furnace concept

Case 2 and 3: Modified blast furnace with carbon capture

Case 2 and 3 are a continued development of the modified blast furnace concept where a carbon capture process has been implemented prior to burning the high-calorific blast furnace top gas in the combined cycle gas turbine. After compression the fuel gas is clean and under high pressure, and this can be taken advantage of in terms of CO_2 capture. Removing a large part of the CO_2 in the fuel gas further improves the calorific value of the fuel.

In order to further increase the heating value, the purified top gas is sent through a water-gas shift (WGS) reactor which increases the CO_2 and H_2 content of the gas and facilitates the CO_2 capture downstream. After the water-gas shift reactor the fuel gas is sent to the CO_2 capture unit. Because of the high pressure of the gas after the WGS reactor, a physical solvent could be considered. Two post-combustion capture configurations were calculated; chemical absorption by MEA (Case 2) and physical adsorption by Selexol (Case 3). Figure 3.5 illustrates a simplified process flow sheet of Case 2, modified blast furnace with MEA-based capture.

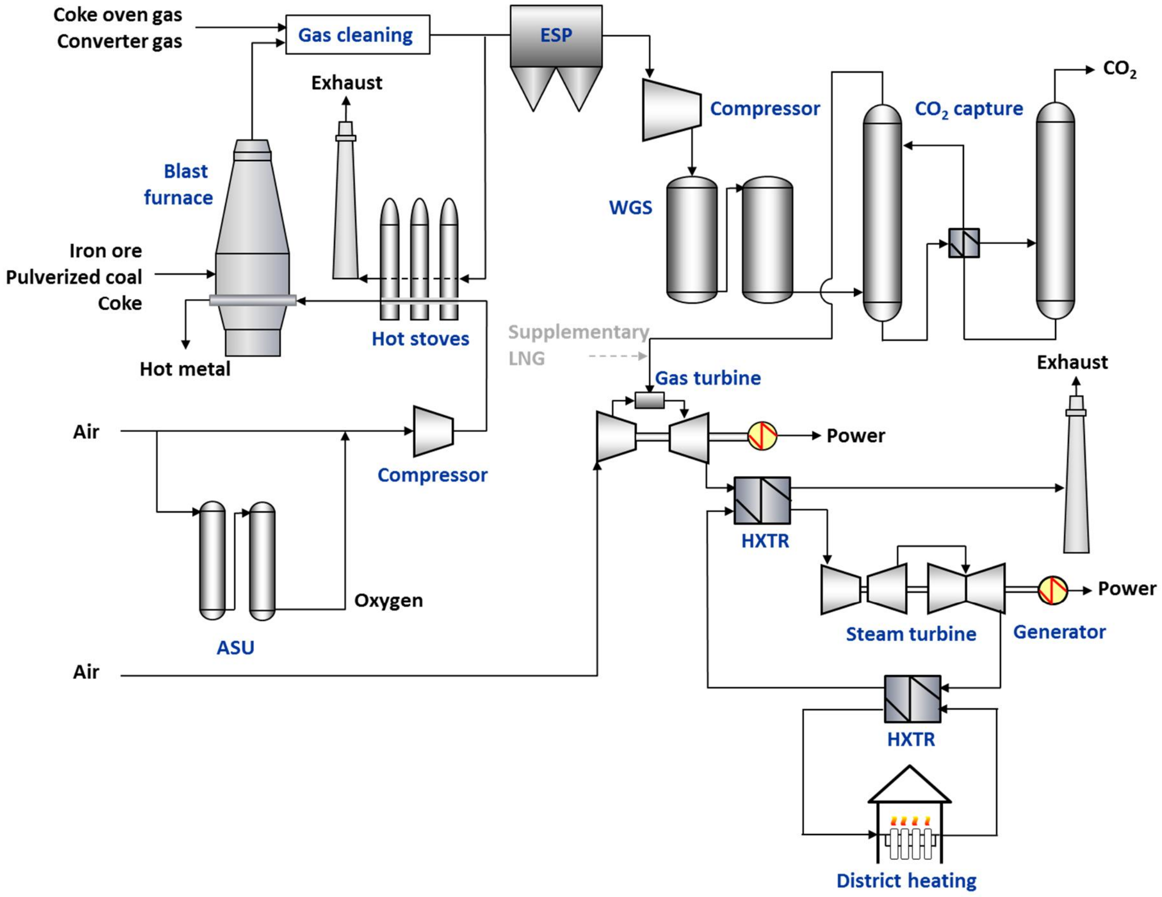


Figure 3.5 Modified blast furnace process with CO_2 capture

After the CO_2 removal H_2 rich gas from the absorber is fed to the gas turbine with a heat recovery steam generator to produce power and heat. The captured CO_2 is then shipped to a permanent storage.

Based on the consequential impact assessment only the specific parts of the steel mill that are affected by the application of CCS have been considered. The results of the assessment show the feasibility in electricity production and CO_2 emissions of the different application stages. Ultimately the price of CO_2 mitigation is assessed from the plant owner point of view to evaluate the future viability and development potential of the concept. The main results are shown in Table 3.1.

Table 3.1 Main results from the staged implementation

Parameter	Unit	Base case	Case 1 Modified BF	Case 2 Modified BF + MEA	Case 3 Modified BF + Selexol
CONSUMABLES					
Coke consumption	kg/t HM	287	242.8	242.8	242.8
PCI consumption	kg/t HM	180	240	240	240
Oxygen production	Nm^3/t HM	70	153.3	153.3	153.3
LNG	MW	0	0	201.7	0
ELECTRICITY CONSUMPTION					
Oxygen production	MW$_e$	11.0^a	15.6^b	15.6^b	15.6^b
Turbo blower	MW$_e$	8.6	8.6	8.6	8.6
Auxiliaries^c	MW$_e$	0	83.0	84.0	50.3
CO_2 compression	MW$_e$	0	0	15.8	17.0
TOTAL	MW$_e$	19.6	107.2	124.0	91.5
Top gas net electricity output	MW$_e$	96.7	232.1	243.5	172.4
HM production	kg/s	83	83	83	83
Captured CO_2	kg/s	0	0	58.6	56.0
CO_2 emission after capture	Mt/a	2.5	2.5	1.1	0.8

[a] Existing ASU with electricity consumption of 400 kWh/t O_2 [b] Assumed implementation of new ASU with electricity consumption of 260 kWh/t O_2 [c] Including compressors, pumps and auxiliaries, not including compression of captured CO_2

In the base case, the net electricity output is 96.7 MW (30% conversion efficiency in the gas boiler). Replacing part of the coke with coal and firing the resulting top gas in a gas turbine in the modified blast furnace concept yields 232.1 MW net electric output for the same amount of hot metal produced. Modified blast furnace Cases 2 with MEA-based CO_2 capture has a higher net electricity output than the modified blast furnace case, but this process needs a higher fuel input to the gas turbine in order to produce the steam needed for the MEA reboiler. Figures 3.6 and 3.7 compare the energy input, CO_2 emissions and electricity output for all cases.

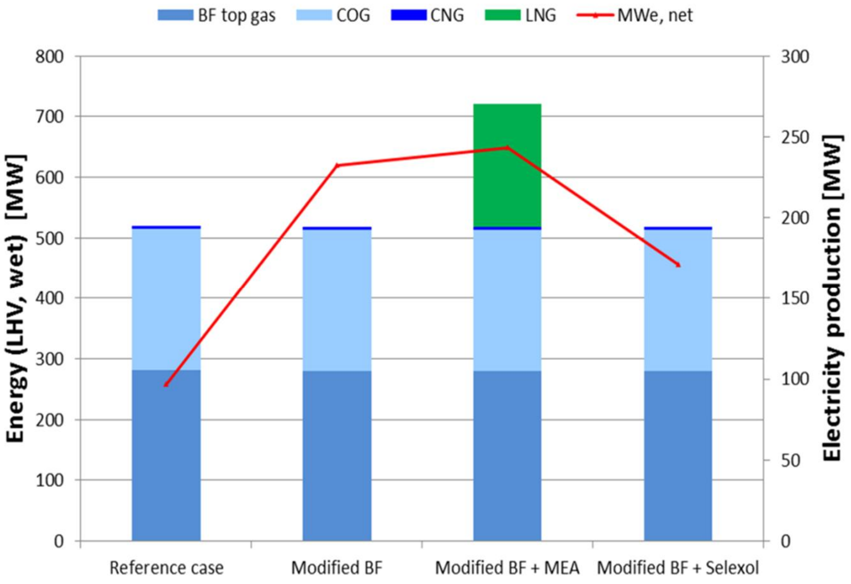


Figure 3.6 Energy input and electricity production from system boundary

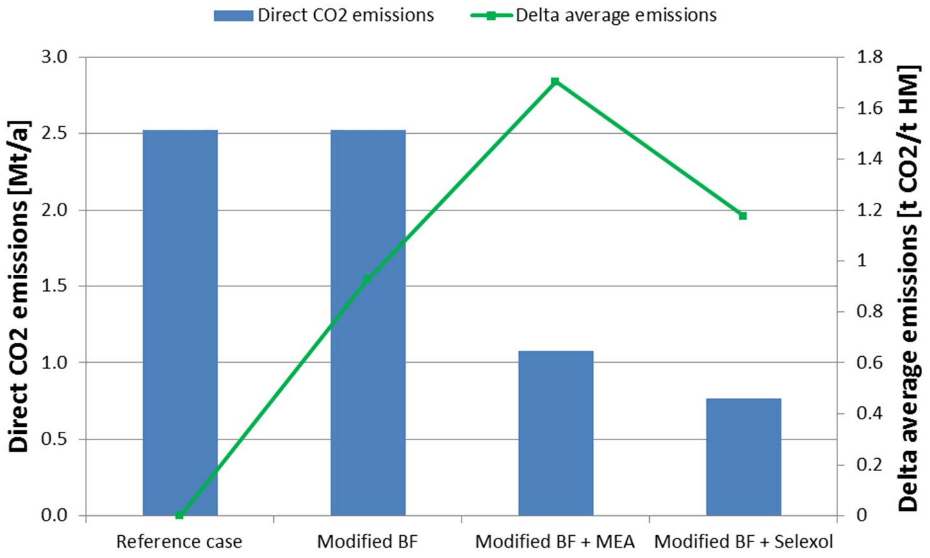
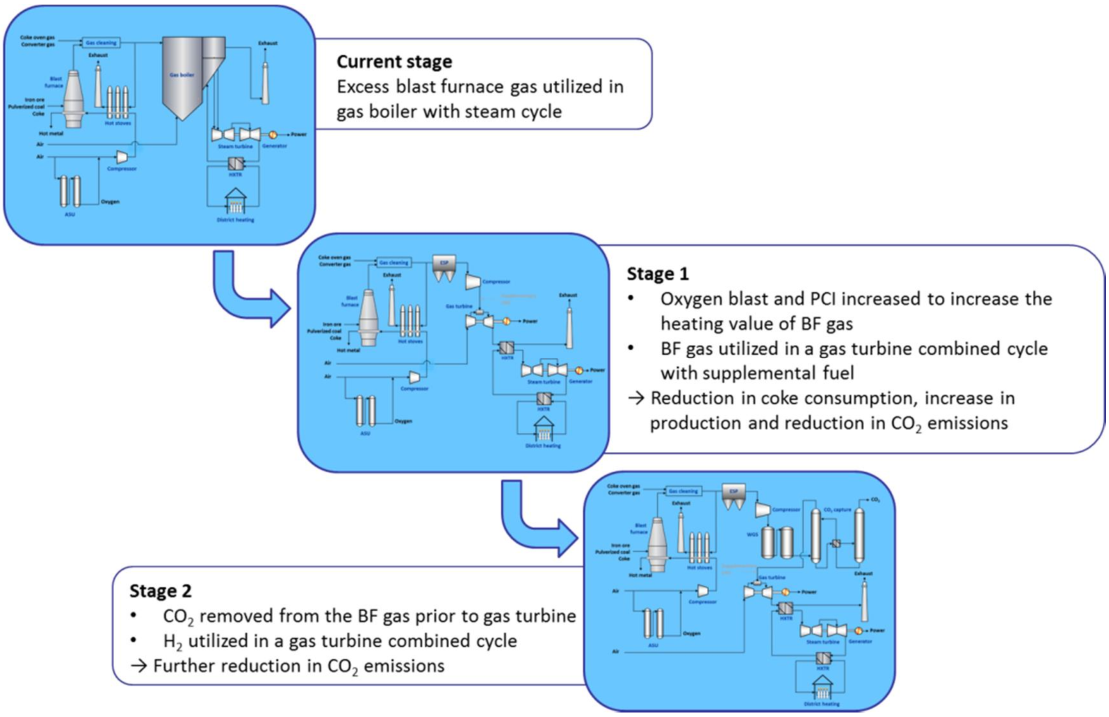


Figure 3.7 CO_2 emissions and difference in CO_2 emissions per ton hot metal compared to the reference case

No CO_2 emissions are avoided in the modified blast furnace Case 1. In this case, the emissions are allocated differently compared to the base case due to the increase in net electricity output, and the specific CO_2 emissions per ton produced hot metal decrease.

One important aspect with the modified blast furnace concept is the possibility for staged deployment in order to reduce the technology implementation and investment risks. The concept could be deployed by first applying the gas turbine without the CCS process step. The investment in a shift reactor and CO_2 removal could then be made after the CO_2 prices have increased in the future. The first implementation step would probably already bring benefits such as reduced nominal emissions, higher electricity production and reduced coke consumption if higher PCI and oxygen enrichment in the blast is deployed. Staged implementation is described in Figure 3.8.



Current stage
Excess blast furnace gas utilized in gas boiler with steam cycle

Stage 1
- Oxygen blast and PCI increased to increase the heating value of BF gas
- BF gas utilized in a gas turbine combined cycle with supplemental fuel

→ Reduction in coke consumption, increase in production and reduction in CO_2 emissions

Stage 2
- CO_2 removed from the BF gas prior to gas turbine
- H_2 utilized in a gas turbine combined cycle

→ Further reduction in CO_2 emissions

Figure 3.8 Description of staged application of blast furnace gas fired gas turbine with CCS

Conclusions

A concept of increasing the top gas calorific value of the blast furnace for more efficient power production has been investigated. Replacing expensive coke with less expensive pulverized coal increases the heating value of the blast furnace top gas and enables combustion in a combined cycle gas turbine for low-BTU fuels instead of in a low-efficiency gas boiler. It has been shown that power production increases significantly when replacing part of the reducing agent coal with increased Pulverized Coal Injection (PCI) and oxygen blast furnace. The effects of the modifications include decreased coke consumption, increased power to heat ratio and reduced CO_2 emissions. Implementing a CO_2 capture process will further increase the calorific value of the top gas while reducing the CO_2 emissions from the system boundary.

Application of post-combustion carbon capture and oxygen blast furnace with CCS to an integrated iron and steel mill are shown to be alternatives for significant CO_2 reductions in the industry. However, as the costs of CO_2 mitigation are currently generally understood to be rather high, the implementation of these technologies will probably take several decades. As a consequence, the interest has been directed to technologies that enable a staged implementation of CCS. This might also prove to have additional advantages that could possibly benefit the investor before carbon prices reach levels that would enable a more widespread application of CCS technologies.

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CARBON CAPTURE IN CEMENT PRODUCTION

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Process overview

The objective of this chapter is to evaluate retrofitting CO_2 capture at the Norcem Heidelberg Cement plant in Brevik, Norway. Both post-combustion capture with chemical absorption (MEA) and oxy-combustion CO_2 capture were considered in the study.

The main ingredient of making cement is limestone ($CaCO_3$). Limestone in powder form is mixed with different correction materials in order to achieve the right quality for the cement. This powder mix is pre-heated to 1 000°C, at this temperature the limestone is reduced to calcium oxide (CaO) and CO_2. The mixture then enters a rotating furnace where further heating to 1 450°C takes place. In this process, the powder mixture is sintered together and clinker is formed. After cooling the clinker is ground to cement in a mill. The process steps in cement production are illustrated in Figure 4.1.

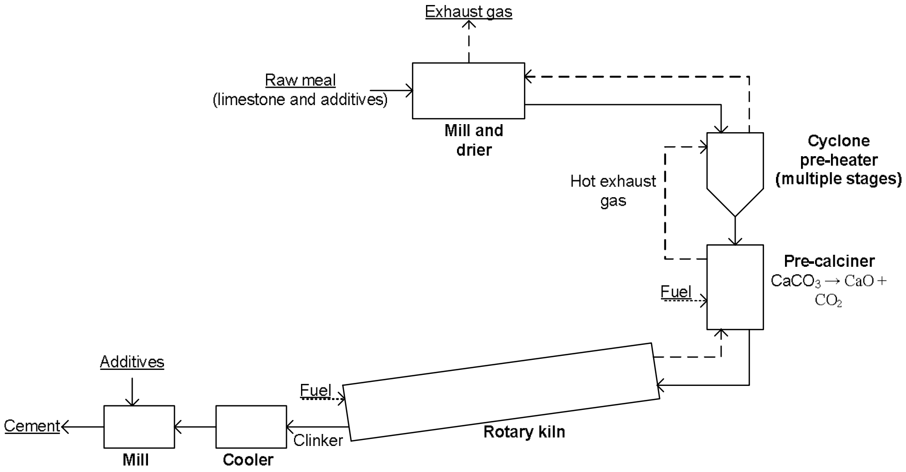

Figure 4.1 Illustration of cement production

CO_2 capture in cement production

Post-combustion and oxy-combustion technologies are considered to be the most applicable for CO_2 capture in a cement plant. Pre-combustion technologies have a disadvantage as only 40% of the emissions are due to fuel consumption in a typical cement plant. The remaining emissions come from the calcination process (i.e., originating from the raw material). Post-combustion capture would be easier to implement on existing plants as no major changes to the cement production are necessary (although SO_x, NO_x, and dust reduction would likely be needed). Implementation of oxy-combustion leads to modifications in the cement production plant, but is still considered to be interesting.

In 2013, Norcem AS, the Heidelberg Cement Group and the European Cement Research Academy (ECRA) initiated a small-scale test center at the Norcem plant in Brevik, Norway [1]. The project will benchmark four different post-combustion CO_2 capture technologies for capture from cement and test step 1 will be concluded in the second quarter of 2017. Three technologies will be tested on site and one will be tested off-site:

- Amine - Aker Solutions
- Solid Sorbent - Research Triangle Institute (RTI)
- Membrane - DNV GL, NTNU, Yodfat Engineers
- Regenerative Calcium Cycle - Alstom Power (off-site)

ECRA's CCS Project (Phase IV) continues with focus on oxy-combustion. Their findings so far show that the products from small-scale oxy-combustion pilots have negligible differences in physical properties when compared to conventional products. In addition, the tests indicate that the refractory materials used to build cement kilns can handle oxy-combustion conditions. The study aims to be concluded by second quarter 2015 [2].

Case study A – Retrofit application of carbon capture

MEA based post-combustion CO_2 capture from the exhaust gas is the focus of this case study.

Norcem's Brevik cement process is characterized by an exhaust gas stream divided into two strings after the pre-calciner. Both strings are fed through a series of cyclone pre-heaters (PHs) and cooled down by condensing towers (CTs). In the raw mill (RM), ambient air is supplied for fluidizing the raw material to prevent clogging. This leads to significant process air in-leakage in String 2 which is used for this purpose. The exhaust gas system is equipped with electrostatic precipitators (ESPs) in both strings, a gas suspension absorber (GSA) system in string one and bag filters (FF) in both strings. In addition, the pre-calciner is equipped with a selective non-catalytic reduction (SNCR) to lower NO_x emissions. An illustration of the current exhaust gas configuration is provided in Figure 4.2.

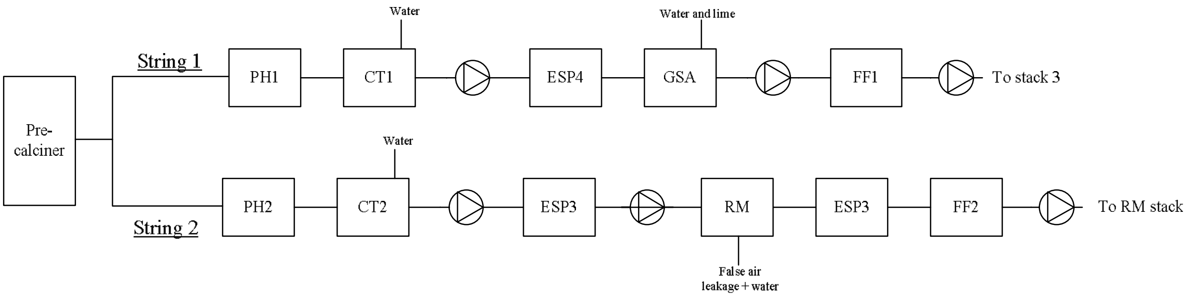

Figure 4.2 Current exhaust gas configuration at Norcem, Brevik

Exhaust gas data for the two strings at the stacks are given in Table 4.1. In addition, modified exhaust gas data is also provided for String 2; here String 2 is no longer utilized in the RM which means reduced air in-leakage. This was included as it would be the most likely configuration of String 2 if post-combustion CO_2 capture is to be adopted.

Table 4.1 Exhaust gas parameters after the preheaters [3]

Properties	Unit	String 1	String 2	Modified String 2
Volume flow	Nm^3/h wet	130 000	264 000	127 000
Temperature	°C	80	80	80
O_2	Mole frac	0.073	0.14	0.068
H_2O	Mole frac	0.089	0.053	0.093
CO_2	Mole frac	0.221	0.137	0.283
N_2	Mole frac	0.618	0.67	0.555
Total mole flow	kmol/h	5 800	11 800	5 700

The integration between the cement, CHP and CO_2 capture plant is illustrated in Figure 4.3. In the illustration, the cooling towers (CTs) are replaced with waste heat boilers (WHBs) for recovery of excess energy from the exhaust gas.

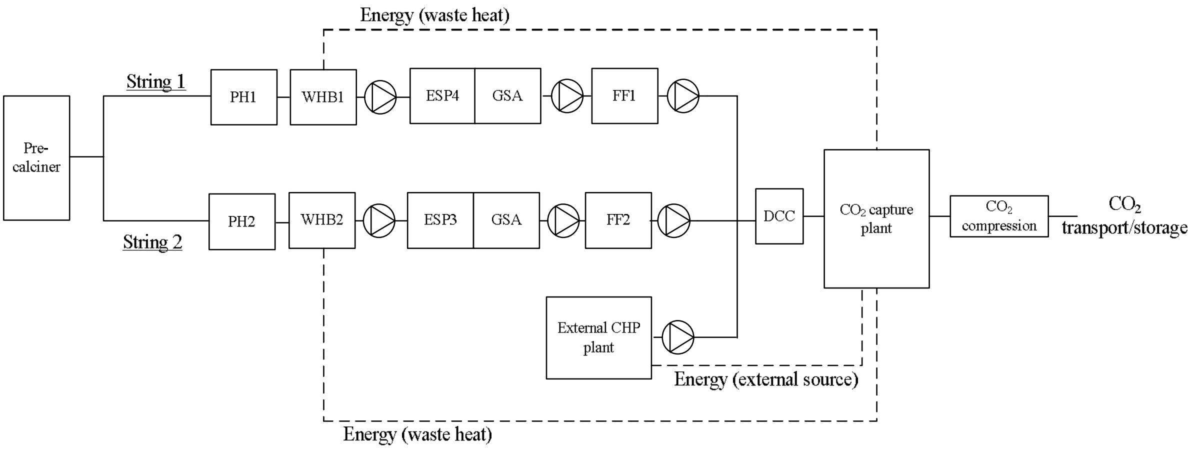

Figure 4.3 Integration between the cement, energy and CO_2 capture plant

The energy consumption in the capture plant is mainly due to the desorber reboiler for regeneration of MEA. In addition, there is also a need for electricity for operation of exhaust gas fan and compression of CO_2 to 70 bar. The initial Aspen Plus simulation of CO_2 capture from the cement plant gave a total reboiler duty of 111 MW and 12 MW for the operation of exhaust gas fan and CO_2 compression when capturing 85% of the CO_2.

Some of the energy needed in the desorber reboiler could be covered by waste heat that could be made available from both exhaust gas strings. In this study the cooling towers are therefore assumed replaced with waste heat boilers (WHBs) for recovery of this waste heat. It should be noted that this is not straight forward, as a WHB has a considerable area and maintenance need. The heat recovery potential from String 1 is from 389°C down to 169°C (the current operational temperature of the GSA). It could be argued for an even lower temperature of the exhaust gas into the GSA, however, this is not considered further here. As String 2 is assumed to no longer be utilized in the raw mill, it is considered equal to String 1 but, with a slightly lower temperature out of the pre-heater, resulting in heat recovery from 382°C to 169°C. The estimated energy that could potentially be recovered from the two strings was 24.5 MW.

The energy recovered from the exhaust gas can only cover a part of the total energy needed for capturing the CO_2. The rest must be covered by an energy plant. The energy needed for CO_2 compression and exhaust fan(s) was also generated in the energy plant. Here, energy is generated by burning natural gas in a boiler system.

The iterative Excel model of the energy plant described above was used to estimate the final capacity of the CO_2 capture plant needed to capture the CO_2 generated in both the cement and the energy plant. A capture rate of 100% of the CO_2 generated in the energy plant was needed in order to achieve an 85% capture rate from the cement plant. The results of the iteration calculations and the Aspen Plus simulation of the increased CO_2 capture plant are given in Table 4.2. The exhaust gas from the cement plant (String 1 and modified String 2) and the flue gas from the energy plant are included. The results presented in Table 4.2 show that a considerable amount of CO_2 (32 t/h) is generated in the natural gas fired energy plant. The inclusion of this CO_2 amount for capture together with the CO_2 produced in the cement plant increases the desorber reboiler duty from 111 MW to 165.5 MW.

Table 4.2 Results for the post-combustion capture case

Property	Unit	Value
CO_2 from cement production	t/h	127
CO_2 from energy plant	t/h	32
Actual capture rate	%	88
CO_2 captured	t/h	140
Natural gas consumption	Sm^3/h	1.7e4
Reboiler duty covered by waste heat	MW	24.5
Reboiler duty not covered by waste heat[1]	MW	141
Total reboiler duty needed[1]	MW	165.5
Exhaust gas fan energy consumption[2]	MW	1.5
CO_2 compression energy consumption[2]	MW	17.5

[1] A boiler loss of 15% based on LHV was included. [2] A boiler loss of 15% based on LHV and a turbine loss of 2.5% (mechanical) were included

Case study B – Oxy-combustion

Implementation of oxy-combustion CO_2 capture at a cement plant would require the following additional equipment, illustrated in Figure 4.4:

- Air Separation Unit (ASU) for production of oxygen.
- CO_2 Compression and Purification Unit (CPU) to purify the exhaust gas and compress the CO_2 to required CO_2 quality and pressure.
- Recirculation pipe to recirculate some of the exhaust gas back to the kiln to avoid too high temperatures in the kiln.

Oxy-combustion CO_2 capture causes significant auxiliary power consumption, compared to normal operation without CO_2 capture. The compressor work in the ASU and the CO_2 CPU are the main causes of the increased power consumption. In addition, process modifications to the cement plant, particularly the kiln burners and the clinker cooler, would be necessary.

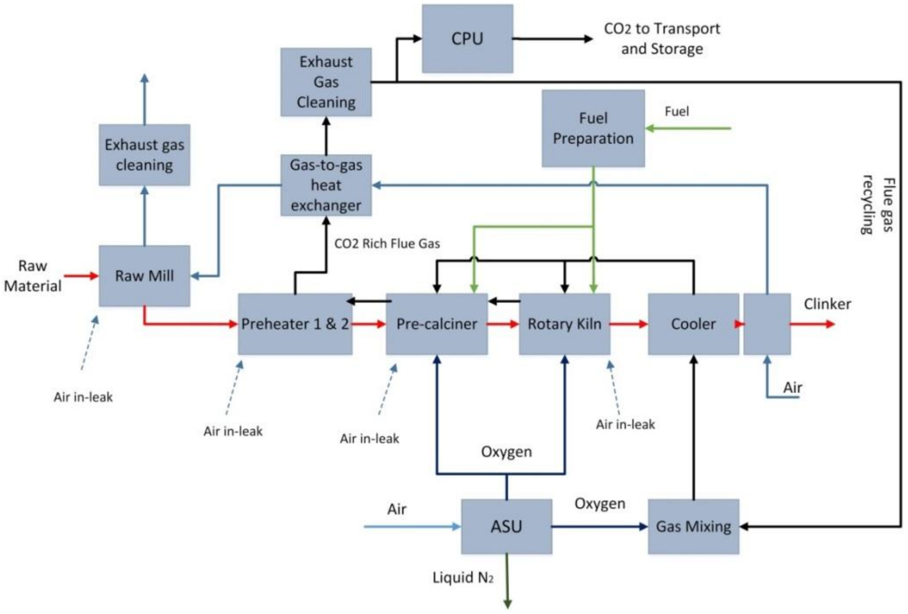

Figure 4.4 Oxy-combustion cement plant layout as modelled

The simulation layout chosen for the oxy-combustion setup is shown in Fig. 4.4. A summary of the results of the process simulations is shown in Table 4.3. The CPU power increase is compared to the best case scenario of 2 % air in-leakage. Table 4.3 shows that the power consumption of the CPU increases with increasing air in-leakage. This is because the increased air in-leakage causes a lower CO_2 concentration in the flue gas, and thus causes a higher work of separation. In addition, higher air in-leakage gives a higher volumetric flow of the flue gas. The energy consumption of the ASU remains unchanged, as the air in-leakage only affects the cement plant process and the CPU process. Table 4.3 highlights the importance of preventing air in-leakages in the system.

Table 4.3 Sensitivity of air in-leakage on results for oxy-combustion capture

Property	Unit	Case 1	Case 2	Case 3
Air in-leakage	%	2	4	6
CO_2 in flue gas before capture	mol-%	85	81	77
CO_2 capture rate	%	96	94	92
CO_2 captured	kmol/s	0.81	0.79	0.77
CO_2 captured	t/h	128	125	122
CPU power	MW	21.3	21.4	23.5
CPU power	kWh/t_{CO2}	166	171	192
Energy consumption ASU	MW	12.5	12.5	12.5

The technologies evaluated for use on the cement plant are based on two inherently different CO_2 capture processes. Both technologies have advantages and disadvantages. In Table 4.4, the two technologies are assessed against technical parameters that are important when evaluating CO_2 capture technologies.

Table 4.4 Assessment of post-combustion MEA and oxy-combustion CO_2 capture on an existing cement plant against relevant technical parameters: (o) neutral, (-) somewhat negative, (--) negative, (---) very negative.

	Post-combustion MEA	Oxy-combustion
Plant complexity	- -	- -
Area needed	- - -	- -
Energy consumption	- - -	- -
Chemicals	- -	o
Emissions	-	-
Cement production modification	-	- - -
Shut down of cement production during construction	-	- - -
Process gas pre-treatment	-	-
Fresh water consumption	- -	o
Fuel supply chain	- - -	o
Known technological solutions	-	- - -

The assessment of the technologies shown in Table 4.4 is indicative only, as the final conclusion is likely to mainly be based economic considerations.

Conclusions

Integration between cement production and two different CO_2 capture technologies, MEA based post-combustion and oxy-combustion has been assessed technically. When retrofitting the plant with an oxy–combustion system, an ASU for production of the oxygen is required for the combustion in the kiln and pre-calciner. In addition, installation of a CPU for processing of the flue gas is required. Additional space for both these units is necessary. For retrofitting of a cement plant, the clinker cooler and the kiln burners would require a new design. Reduction of air in-leakages, for example, by improvement of sealing would be critical. Evaluations of the electricity consumption of oxy-combustion technologies identified the CPU as a major consumer. The amount of air in-leakage becomes an important factor for the CPU performance. Air in-leakage should be kept to a minimum to ensure effective CO_2 capture.

Compared to oxy-combustion post-combustion CO_2 capture requires few modifications to the existing cement plant. The major modifications include the addition of WHB's for recovery of waste heat and if not already in place exhaust gas treatment (NO_x, SO_x and dust). CO_2 capture using amines (MEA) is an energy intensive process, and the energy is supplied from exhaust gas waste heat and a dedicated energy plant. The CO_2 generated in the energy plant is also included for capture leading to a larger CO_2 capture plant than if only the CO_2 from the cement plant is to be capture.

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CARBON CAPTURE IN PULP AND PAPER PRODUCTION

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Process overview

The Östrand pulp mill has a production capacity of 425 000 tons Kraft pulp and around 95 000 tons of chemi-thermomechanical CTM pulp. In addition, the mill may produce up to 75 MW of electricity. SCA Östrand is one of the largest pulp mills in Sweden and it is also the tenth largest industrial emitter of CO_2 in Sweden with around 1.3 Mt CO_2 emitted yearly. Figure 5.1 gives an overview of the Kraft pulping process at Östrand. The process may be divided into two major parts which are described in detail below: 1) the pulp producing process and 2) the chemical and energy recovery system.

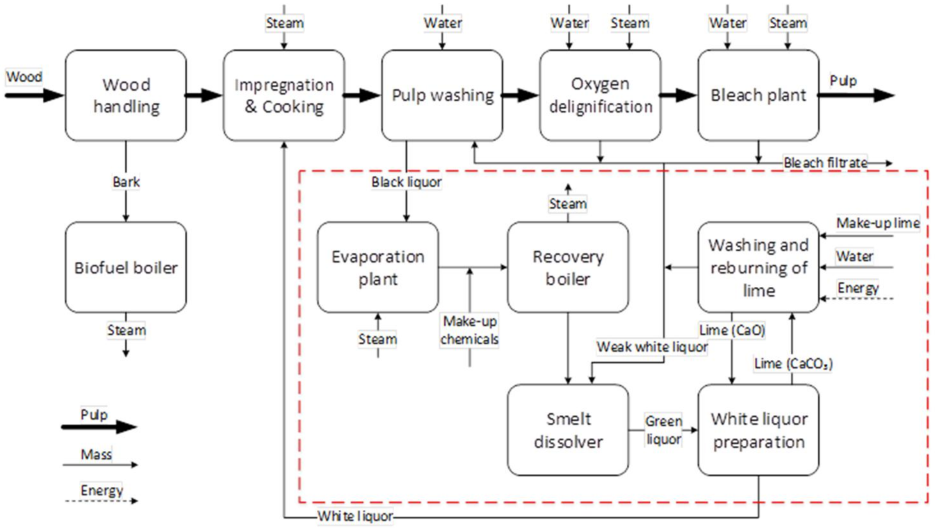

Figure 5.1 Overview of the Kraft pulping process. The process units within the red dashed box in the figure make up a chemical and energy recovery system.

Pulp production

In a chemical pulping process the fibers are extracted from the wood by cooking the raw material in a mixture of chemicals. The wood is received as wood chips or logs. Logs need to be debarked, chipped and screened before they can be cooked. After the wood handling, the wood chips are preheated with steam in order to remove any air before they are cooked in a chemical mixture, referred to as white liquor, mainly consisting of sodium hydroxide (NaOH) and sodium sulfide (Na_2S). During the cooking process, lignin and a part of the hemicellulose in the wood chips is dissolved in the white liquor. The mixture coming out of the cooking section is washed to remove the spent cooking chemicals, referred to as black liquor, and dissolved organic materials from the pulp. Generally, over 99% of the black liquor is removed from the pulp in the washing system.

Further delignification by oxygen and washing may be performed in one or two stages after the cooking process. Finally, bleaching is generally required to remove or oxidize the remaining lignin in order to fulfill the quality requirements for the pulp. Bleaching can be carried out in several stages and different chemicals can be used for this purpose. The most common ones are oxygen (O_2), hydrogen peroxide (H_2O_2), chlorine dioxide (ClO_2) and sodium hydroxide (NaOH). The waste stream from a bleaching plant is typically released from the plant instead of being recovered. The amount of chemicals released from bleaching plants has decreased with an increased use of extended cooking and oxygen delignification techniques.

As Östrand is a stand-alone pulp mill the pulp is dried and pressed before being transported to a paper mill or alternative industrial user. At an integrated pulp and paper mill, the wet pulp is transported to the paper-making process.

Chemical and energy recovery system

The chemical and energy recovery system is an important part of the pulping process. The process units which make up this system are marked in Figure 5.1 with a red dashed box. The chemical and energy recovery system has the following main purposes:

- To recover the inorganic pulping chemicals
- To combust dissolved organic material in order to recover and make use of its energy content in the form of electricity and process steam
- To control pollution, especially with respect to waste water which is released to a waste water treatment plant
- To recover valuable by-products such as tall oil soap, crude tall oil and crude sulfate turpentine

In addition to recovery of the inorganic chemicals within the pulping process the recovery boiler also generates electricity and steam by combusting organic material dissolved in the black liquor. The electricity and steam generated in the boiler usually covers the demand of the pulp mill and in some cases electricity and/or heat is exported to the grid or nearby district heating systems. However, integrated pulp and paper mills generally rely on imported electricity and fuels in order to cover their electricity and process steam demand.

Future process development – Black liquor gasification

The traditional recovery boiler suffers from several drawbacks such as corrosion, fouling and smelt-water explosions, which can be costly with respect to production losses [1]. Improvements have been made over the years but the recovery boiler and its Rankine steam cycle have some inherent limitations such as low thermal efficiency and low power-to-heat ratio. In addition, modern pulp mills have a steam surplus which motivates a better use of the energy contained in the black liquor, such as electricity generation or biofuel production. The issues stated above have been incentives for examining other alternatives to recover the cooking chemicals [2].

Black liquor gasification is an alternative technology that has the potential to solve many of the stated problems. Gasification of spent cooking liquor was examined as early as in the 1950s and 1960s. Since then, a number of research projects and pilot plants have examined this process. The gasification process considered in this study is based on the Chemrec oxygen-blown pressurized gasification. A schematic description of the gasification process is presented in Figure 5.2.

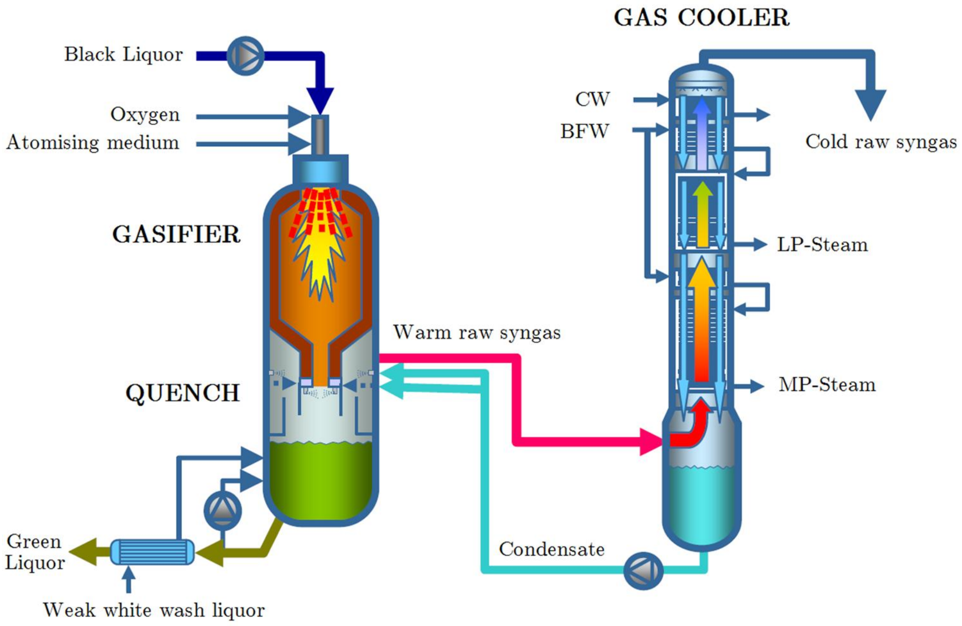

Figure 5.2 The pressurized, oxygen-blown black liquor gasification technology developed by Chemrec. Adapted from Lindblom and Landälv [3].

The gasification takes place at elevated pressure and the black liquor is gasified with oxygen. Melted inorganic salts are separated from the syngas in the gasifier and are dissolved in condensate from the gas cooling section to form green liquor. The produced green liquor will contain some dissolved gases that will be released when the pressure is reduced and are mixed with the raw syngas. The gas formed in the gasification process is a mixture mainly composed of CO, H_2, CO_2, H_2O and H_2S but it also contains minor amounts of N_2, CH_4 and COS. After the quench section the produced gas enters the counter-current gas cooler where the gas releases heat to generate medium pressure (MP) steam and low pressure (LP) steam. To achieve the final cooling in the top section cooling water is utilized. For a more detailed description of the process, see Ekbom et al. [4].

The investigation includes two alternative uses for the syngas produced. One alternative is to combust the syngas in a gas turbine to produce electricity and steam, referred to as black liquor gasification combined cycle (BLGCC). The other alternative uses syngas as a feedstock for production of motor fuel, in this case dimethyl ether (DME), a gaseous compound at atmospheric pressure but is usually treated as a low-pressure liquid which can easily be transported and stored. This alternative is referred to as black liquor gasification with motor fuel production (BLGMF). Schematic overview of the BLGCC and BLGMF processes are presented in Figures 5.3 and 5.4, respectively. Here follows a brief description of the two concepts and how CO_2 capture can be implemented in the processes.

Black liquor gasification combined cycle

The syngas produced in the gasification unit contains a considerable amount of H_2S (around 2 mol-% on a dry basis [4]), originating from the cooking chemicals. To produce the cooking liquors needed in the pulp production processes and to avoid large emissions of SO_X from the gas turbine, the H_2S is separated from the syngas through physical absorption using the Selexol process. The Selexol process may be modified to capture CO_2 by letting the sulfur-free syngas pass through a second Selexol absorber. The CO_2-rich solvent is regenerated by lowering the pressure. The separated CO_2 may then be compressed and transported to storage. The clean syngas is combusted in a gas turbine to produce electricity. The flue gases from the gas turbine passes through a heat recovery steam generator to produce steam which is partly used in the pulping process and partly used to produce electricity. The H_2S stream leaving the Selexol process must be further processed, in order to produce the desired cooking chemicals and is treated in so-called Claus and SCOT process units to recover elemental sulfur up to 99.7%. For an overview and a detailed description of the Selexol process concept, see Field and Brasington [5].

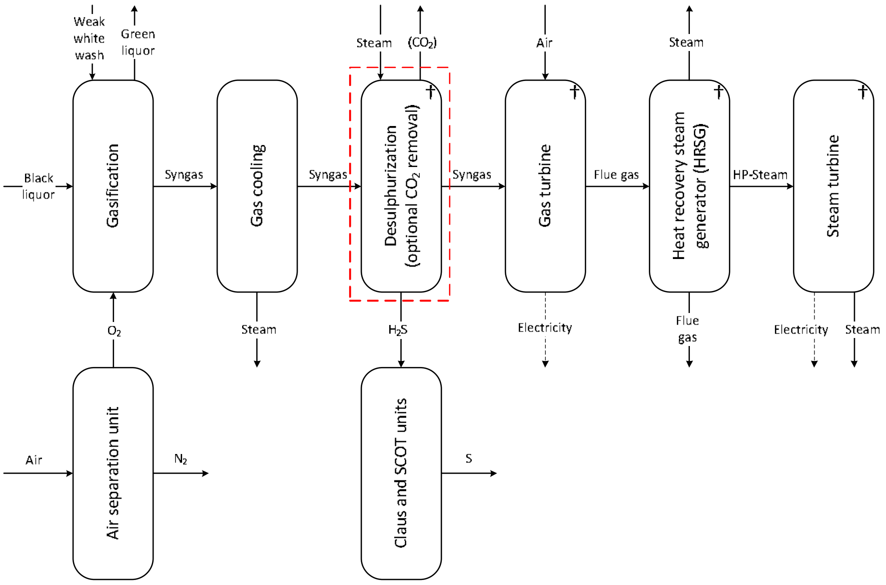

Figure 5.3 Schematic overview of the BLGCC process. Process units marked with a cross are simulated in this study and the red dashed box marks the CO_2 capture process

Black liquor gasification with motor fuel production

In the BLGMF alternative, the DME synthesis process put stringent requirements on the sulfur level in the syngas, the required sulfur levels are below 0.1 ppm compared to the 20 ppm requirement in the BLGCC alternative. In addition, the DME production is inhibited by the presence of CO_2 and the concentration of CO_2 in the syngas should be reduced to below 3 mol-%. These requirements can be satisfied by using the Rectisol physical absorption process to separate H_2S and CO_2 from the syngas. The H_2S stream removed from the syngas in the Rectisol process is passed through identical Claus and SCOT units as in the BLGCC alternative. The clean syngas leaving the Rectisol process enters the DME synthesis. Conventionally, DME has been produced in an indirect two-step synthesis route, where methanol is produced via methanol synthesis and then converted to DME by dehydration. A new technology was developed by Ohno et al. [6] which utilizes a direct synthesis where DME is produced from the syngas according to the overall reaction:

$$3\,CO + 3\,H_2 \rightarrow CH_3OCH_3 + CO_2$$

When using the new direct DME synthesis technology, CO_2 is formed in the process. However, this work is limited to investigating the possibility to capture the CO_2 separated from the syngas in the Rectisol process. The possibility to capture the CO_2 formed in the DME production is, thus, not considered. For an overview and a detailed description of the Rectisol process, see Gatti et al. [7].

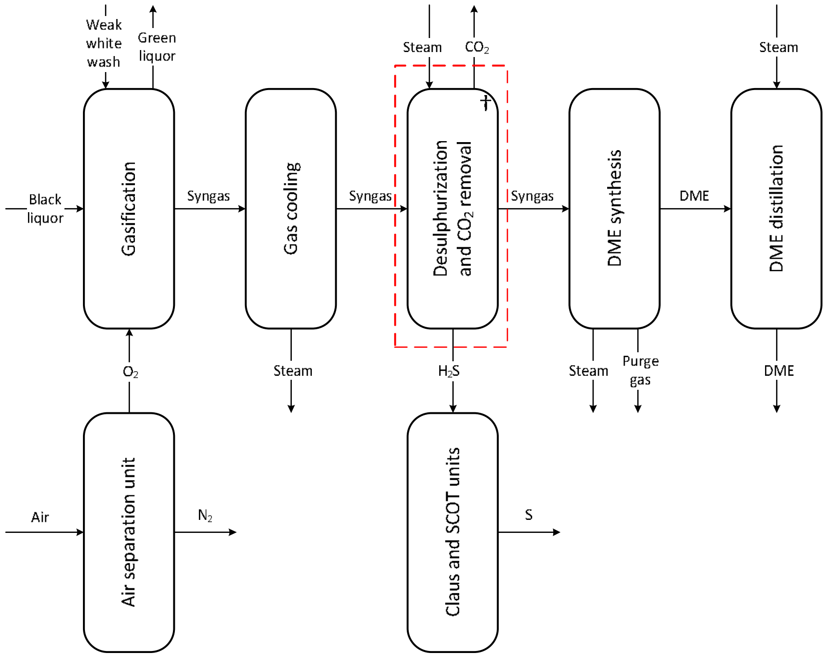

Figure 5.4 Schematic overview of the BLGMF process. Process units marked with a cross are simulated in this study and the red dashed box marks the CO_2 capture process

CO_2 capture in pulp and paper production

Carbon capture in Kraft pulp mills has been investigated by [8-10]. The focus has mainly been on CO_2 capture from combustion of black liquor in the recovery boiler and combustion of bark and fuel oil in the bark boiler. CO_2 concentration in a flue gas stream from a black liquor recovery boiler is typically in the range of 13-14 mol-%, which is similar to what can be found in coal fired boilers. Möllersten et al. [8] investigated several possibilities for CO_2 capture from stand-alone pulp mills and integrated pulp and paper mills. Four different carbon capture technologies for combustion of black liquor are included in these studies. The main results from these studies show that post-combustion as well as oxy-fuel combustion CO_2 capture from the recovery and the bark boiler has a high CO_2 reduction potential. However, the studies conclude that the implementation would lower the mill's electrical efficiency considerably and, thus, increase the dependency on imported electricity.

Implementation of BLGCC with CO-shift has been shown to increase the CO_2 reduction potential and reduce the negative effects on the electrical efficiency. However, it should be noted that the BLGCC technology has not yet been commercially demonstrated on a full scale and that there are no current pilot or demo scale CCS initiatives related to the pulp and paper industry.

A black liquor gasification unit has been operating commercially in New Bern pulp mill in North Carolina, USA, since 1996. Another gasification unit, producing dimethyl ether (DME) biofuel is operating at Smurfit Kappa Kraft pulp mill in Piteå, Sweden [10]. Even though these units are not operated in connection to CO_2 capture they illustrate the possibility for implementing such technology and can provide the necessary knowledge needed for further research in this field.

The characteristics of Kraft pulp mills that will be of importance to carbon capture include:

- A large share of the CO_2 emission from pulp and paper mills originates from biogenic sources, which gives the possibility to achieve negative CO_2 emissions by implementing carbon capture technology
- The recovery boiler is the largest point source of CO_2 in Kraft pulp mills and is also special in its design and function
- The lime kiln is the largest fossil CO_2 source in the Kraft pulping process
- Different capture technologies are possible, including post-combustion chemical absorption with different solvents, oxy-combustion in the recovery boiler and pre-combustion from a black liquor gasification unit

The present work aims to evaluate the possibilities for CO_2 capture at an existing Swedish Kraft pulp mill for different development strategies at the mill, and thus, the investigation is largely based on site-specific conditions. The work is based on detailed process modelling of the investigated CO_2 capture processes as well as an evaluation of the various sub-processes associated with the development strategies investigated to determine their global CO_2 reduction potential.

Case study – Three scenarios for future development in the pulp mill

The recovery boiler is the largest point source of CO_2 at the pulp mill and is responsible for nearly 70% of the emissions, see Figure 5.5. Therefore, this work focuses on these emissions. However, CO_2 capture may also be applied to the biofuel boiler and possibly to the lime kiln. Three near-future scenarios of the pulping process development have been identified and are presented in Table 5.1. The first one is the "business as usual" case where the conventional recovery boiler continues to be the technique used for regeneration of the cooking chemicals. In the second and third scenario the recovery boiler is replaced with a gasifier which generates syngas. In the second scenario the syngas is used to produce electricity in a combined cycle, or a BLGCC. In the third scenario the syngas is used to synthesize DME, referred to as BLGMF. A detailed description of the modelling methodology and assumptions applied in the second and the third scenario is found in the report by Hedström [11]. The implementation of carbon capture to the pulp mill was evaluated for the three scenarios. MEA based post-combustion capture and Selexol and Rectisol pre-combustion capture is applied to the first, second, and third scenario, respectively.

Table 5.1 The three scenarios for future development of the pulp mill investigated

Scenario	Recovery system	Additional product	Capture technology	Capture technology
RB	Recovery boiler	n/a	Post-combustion	MEA
BLGCC	Black liquor gasification	Electricity	Pre-combustion	Selexol
BLGMF	Black liquor gasification	DME	Pre-combustion	Rectisol

The composition and characteristics of the gas streams entering the capture processes in the three investigated scenarios are presented in Table 5.2. The data was obtained from various sources, including SCA Östrand pulp mill for the RB scenario as well as literature sources [4,12,13] for determining the specifications for the syngas stream in both the BLGCC and BLGMF scenarios.

Table 5.2 Process stream data used in the simulations. The composition is specified as mol-% except the COS-concentration which is specifies in ppmv

Scenario	$\dot{n}$ [mol/s]	P [bar]	T [°C]	H_2	CO	CO_2	H_2S	CH_4	O_2	N_2	H_2O	COS
RB	5014	1	110	0	0	13.3	0	0	4.4	63.3	19.0	0
BLGCC	865	31.5	30	39.2	38.1	19.0	1.9	1.3	0	0.2	0.2	10
BLGMF	865	31.5	30	39.2	38.1	19.0	1.9	1.3	0	0.2	0.2	122

The simulations result in an estimation of utility demand for the three scenarios investigated. To make a fair comparison, an overall balance of the pulp mill, including the major in and outflows of energy and mass, is constructed for all six cases, i.e. for all the three scenarios investigated, both with and without CO_2 capture. The focus of the analysis is thus to determine the differences in energy and mass flows between the two cases (with and without capture) for each scenario and subsequently estimate the cost/utilities coupled with capturing the CO_2.

As indicated in the process descriptions the BLGCC and BLGMF scenarios include several additional processes compared to today's market pulp mill, the primary one being a full replacement of the recovery boiler by a gasification unit. To determine the utility consumption or production of these processes various sources are consulted and the obtained information is scaled to match the pulp production rate of SCA Östrand. However, some of these processes are simulated in Aspen Plus, namely the gas turbine, the HRSG and the steam cycle, see Figures 5.3 and 5.4. The steam cycle is modelled to determine the amount of external fuel needed to balance the energy demand of the complete pulp mill.

Figure 5.5 gives an overview to the origin of the CO_2 emissions at Östrand pulp mill in 2010. The recovery boiler and bark boiler dominates the CO_2 formation. These emissions are generated from biomass and are in that sense not contributing to increasing the concentration of CO_2 in the atmosphere. Fossil fuels are mainly used in the lime kiln. It should be noted that the study is based on data from the year 2010. In the end of the year 2011, the two fossil fueled lime kilns at the pulp mill were replaced with a biomass fueled kiln. Since then, the fossil-based emissions at the pulp mill have decreased by around 80% and fossil fuels are at present mostly used in start-up conditions and in shortage of biofuel to the new lime kiln [14]. SCA Östrand offsets part of their waste heat to a nearby district heating system. Besides that, the available data on waste heat streams are limited; however most of the waste heat produced at the mill is believed to be of too low quality to be used directly in the capture process.

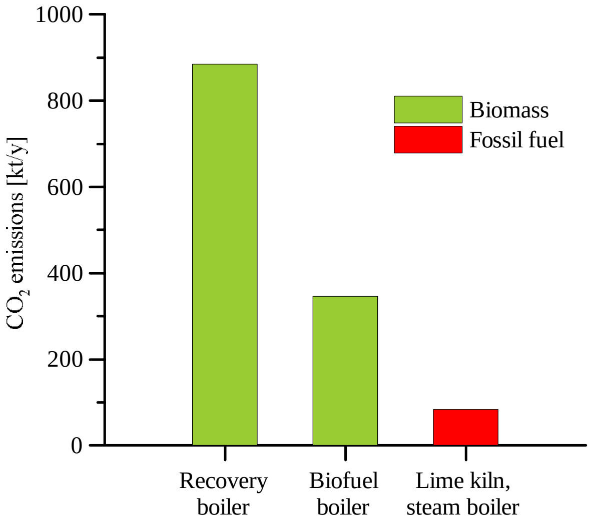

Figure 5.5 CO_2 emitting sources at the pulp mill (2010). The emissions do not include emissions allocated to the electricity used in the processes

Figure 5.6 shows the utility consumption for each of the three scenarios investigated, both with and without CO_2 capture. The different range on the Y-axis in the figure should be noted, the primary reason for this large difference is that the MEA-based process handles a gas flow about five times larger than the other two processes.

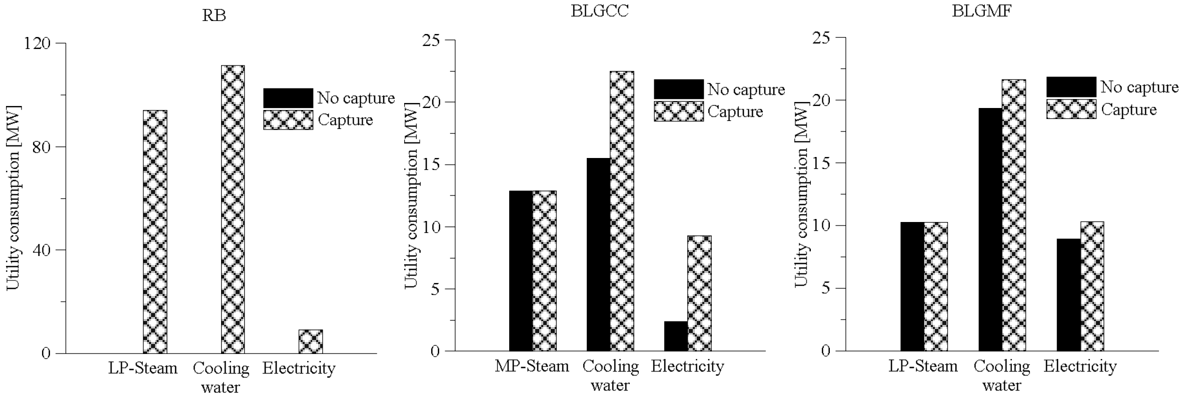

Figure 5.6 Utility consumption of the three scenarios, with and without CO_2 capture. The capture rate was 85% in all scenarios.

If the capture cases of the BLGCC and BLGMF scenarios are compared, it can be observed that the utility demands are similar. The Selexol process has lower electricity demand compared to the Rectisol process, but uses MP-steam instead of LP-steam. However, the loss in electricity production in the Selexol process due to the use of MP-steam instead of LP-steam outweighs the lower electricity demand. Thus, considering utility demand the Rectisol process is the most favorable alternative for carbon capture.

The difference in utility consumption between the capture case and the case without capture divided with the amount of CO_2 captured is presented in Table 5.3. The MEA-based post-combustion process applied to the existing recovery boiler has the highest utility consumption per kg CO_2 captured with respect to both LP-steam and cooling water while the Selexol capture process applied to the BLGCC process requires the highest amount of electricity.

Table 5.3 The additional utility consumption in kJ per kg CO_2 captured for the three scenarios investigated, 85% capture rate

Utility [kJ/kg CO_2 captured]	RB	BLGCC	BLGMF
LP-Steam	3760	0	0
Cooling water	4460	1130	370
Electricity	360	1110	220

From the overall mass and energy balances of the three scenarios with and without carbon capture the pulp mills influence on global CO_2 emissions are evaluated. A number of assumptions were made for this evaluation, such as regarding the source of electricity, emission factors for the fossil fuels as well as the bark and solid wood fuels and substitution of biofuels for diesel. A detailed listing of assumptions made can be found in [11]. It was also assumed that the LP-steam needed to power the CO_2 capture process (notably only in the RB scenario, see Table 5.3) is produced by combustion of additional solid wood fuel in the biofuel boiler. The CO_2 balances of each of the carbon capture cases are presented in Figure 5.7. The top bar represent emission reduction through capturing CO_2, substitution of electricity and diesel while the bottom bar represents the gross emissions originating from fuel combustion and electricity consumption. The black color represents emission originating from fossil sources while the green color represents emissions from biofuels, which are considered CO_2-neutral. The results show that the RB scenario has the largest CO_2 reduction potential, almost double the amount of CO_2per year compared with the other two scenarios, both based on BLG. This is primarily due to the large flue gas flow in the RB scenario since a large share of the carbon fed to the combustion process leaves with the syngas in the BLGCC and BLGMF scenarios and forms CO_2 during combustion in a gas turbine or when DME is combusted in vehicles.

Compared with the present day situation, the RB case has significantly larger amount of emissions originating from the biofuel boiler due to the increased amount of biomass combusted in order to supply steam to the capture process. The large amount of emissions originating from solid wood fuels in the BLGMF case is due to that the energy contained in the black liquor cannot be used to satisfy the demand for heat and electricity in the pulp mill but instead leaves the system in the form of DME. Hence, additional fuel and electricity has to be purchased from an external source to satisfy the demand.

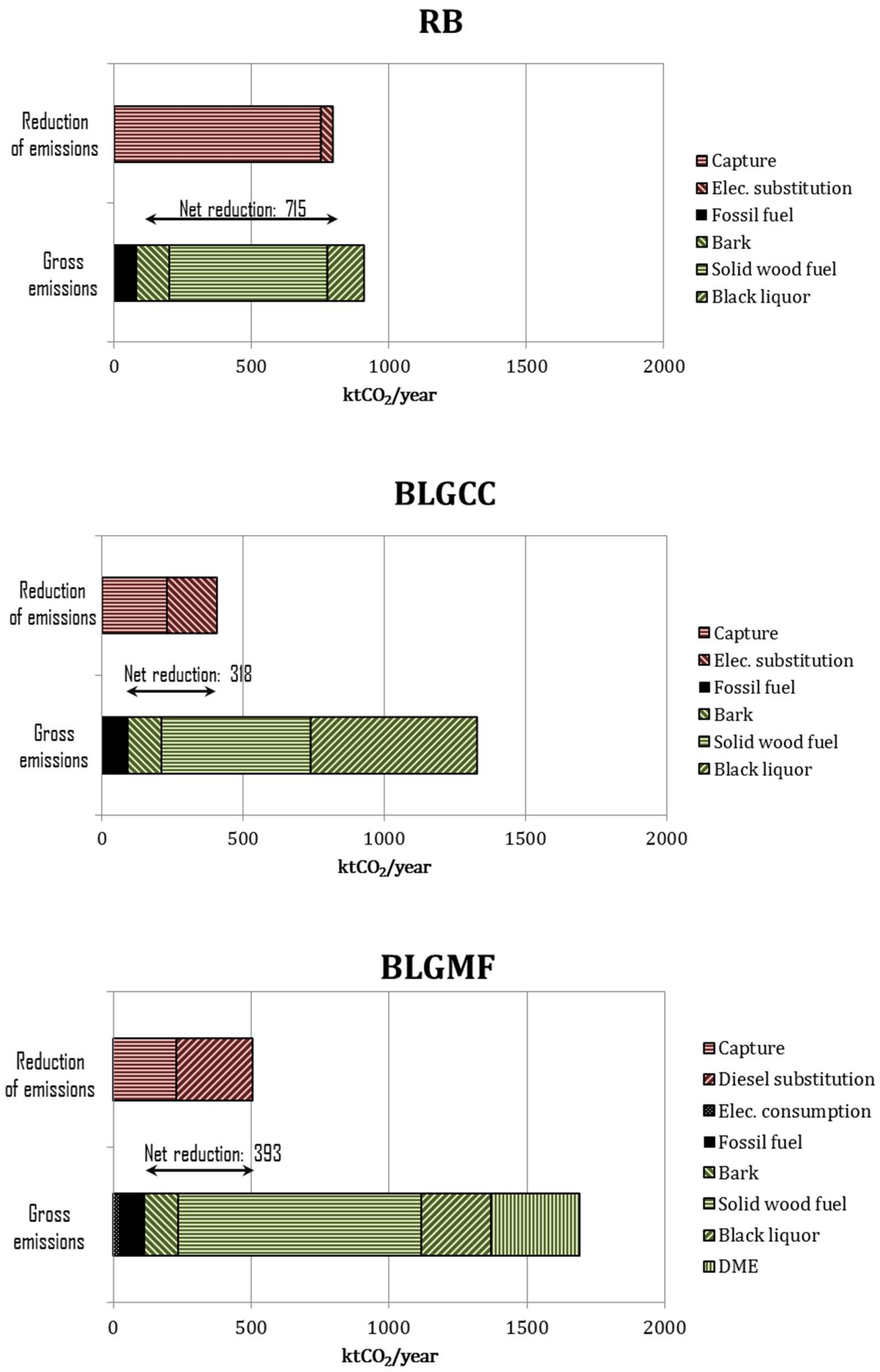

Figure 5.7 CO_2 emission balance for the CO_2 capture case in each of the three scenarios investigated

Conclusions

This work has investigated the possibilities with implementing carbon capture to pulp mills given different scenarios for the pulping industry. The first scenario assumes that the pulping process remains unaltered and hence uses the conventional recovery boiler to regenerate the cooking chemicals. For this scenario, post combustion capture using the MEA process was studied. The implementation of the capture process has large consequences on the overall balance of the process as additional electricity was generated when generating the steam required running the capture process. The steam required was produced by combusting relatively large amounts of additional fuel in the bark boiler. The potential effect on global CO_2 emissions by applying BECCS was the highest in this scenario out of the three, with a net reduction of global CO_2 emissions of 715 000 t/a CO_2.

The second scenario investigates a future where black liquor gasification technology has replaced the recovery boiler to enhance both pulp yield and electricity production. For this scenario the Selexol process was determined to be the most suitable capture process. The calculations resulted in rather low additional utility consumption, with the exception of electricity usage. However, the cost associated with adding on the CO_2 capture section to the Selexol process could prove to be considerable since it includes relatively large process units such as an absorber and flash tanks [5]. The potential effect of applying BECCS was lower for this scenario compared with the other two, with a net reduction of global CO_2 emissions by only 318 000 t/a CO_2.

Black liquor gasification technology combined with production of DME is investigated in the third scenario. The additional resource consumption associated with CO_2 capture using the Rectisol process was shown to be the lowest of the three scenarios. In addition, since only a CO_2 compression section prior to transport needs to be added for capture case [7] the investment cost for the third scenario is lower than for the other two. The potential effect on global CO_2 emission for this scenario is a net reduction of 393 000 t/a CO_2.

The results imply that the pulp and paper industry could be a suitable future candidate to which BECCS can be applied. The BLG technology has been successfully demonstrated on a small-scale and would it be commercialized for a full-scale application, the implementation of a CO_2 capture unit would require relatively low additional utility, compared with the conventional post-combustion process.

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CARBON CAPTURE IN OIL AND GAS REFINERIES

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Process overview

Preemraff Lysekil is a relatively complex refinery including crude distillation with a vacuum distillation, processes to remove sulfur from gasoline, diesel and other oil fractions and to reduce the aromatics content in diesel. In addition, there are process units that improve the octane rating of both light and heavy naphtha. Figure 6.1 shows a simplified process scheme of Preemraff Lysekil.

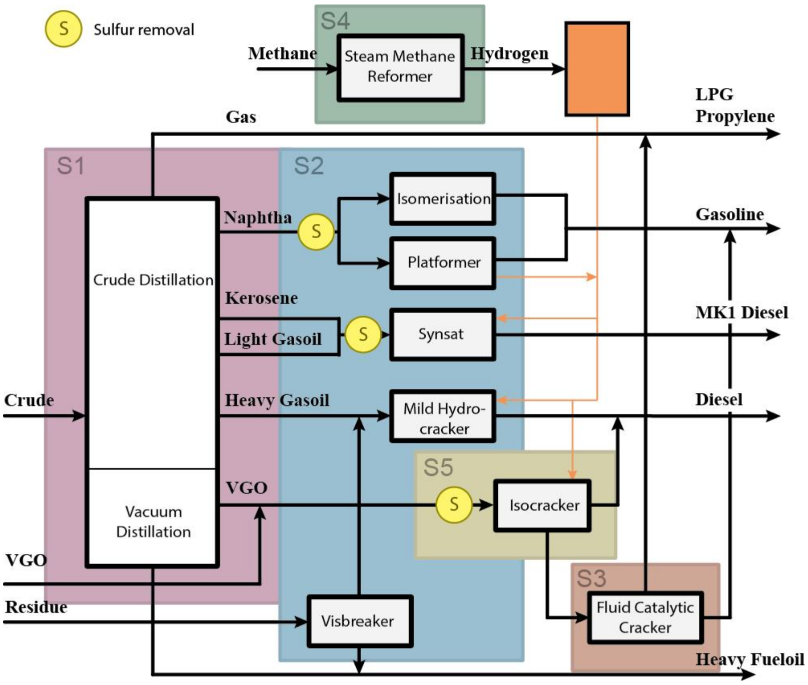

Figure 6.1 Overview of Preemraff Lysekil. The fields S1 to S5 indicates the clusters of unit operations with a common stack

The fractions of the crude oil are separated through distillation, which is the central process to any refinery. The desalted and dehydrated crude oil is heated until most of it has reached vapor phase before being fed to the distillation column. The temperature inside the column is carefully controlled with each tray slightly cooler than the one below it. As a result, each tray collects a particular group of substances with a specified range of boiling points. In this way the distillation tower separates the crude oil into several products: (from the top) gas, naphtha, kerosene and light and heavy gasoil. Some of the products are ready for consumption while others have to be processed further. The downstream processing is further discussed below. The top product from the atmospheric distillation consists mainly of small molecules that are processed into liquefied petroleum gases and propane.

The residual oil consists of hydrocarbons with boiling point above 370-380°C. At such temperatures the hydrocarbon molecules would decompose into fragments, a reaction which is called cracking, and this would plug up the distillation tower. However it is still possible to distill the residual oil in a vacuum distillation unit, which usually operates at pressures in the range of 0.07-0.13 atm.

Naphtha

The naphtha coming out from the distillation tower is cleaned from sulfur before being separated into a light and a heavy fraction. The light fraction goes through an isomerization process while the heavy naphtha goes through a reforming process. The objective of isomerization in oil refining is to increases the octane number of light naphtha for gasoline blending. This is done in reactors containing catalysts, often aluminum chloride or noble metals.

Similar to the isomerization the platformer aims at increasing the octane number of the gasoline blend and to produce aromatics for petrochemical processes. This is done by removing hydrogen atoms from cyclo-paraffin molecules in the presence of a catalyst and transforms them into aromatics with higher octane numbers. An important by-product of the reforming process is high purity hydrogen that can be recycled and used for hydrotreating processes.

Kerosene and Light Gasoil

In the SynSat (synergetic saturation) process, hydrogen is used to desulfurize and de-aromatize light gas oil (LGO) and kerosene in two separate reactors. The SynSat process has two operation modes. When in kerosene mode, sulfur is separated from the kerosene. The product is then used in EU diesel (sulfur content < 10 ppm). When in LGO mode, sulfur is removed from light gasoil where aromatics are saturated by hydrogen. The product is the Swedish “miljöklass 1” (environmental class - low-aromatics, low-sulfur) diesel (MK1) that besides low sulfur content also has a low contentment of aromatics (<5%).

Heavy Gasoil

The heavy gas oil goes through a mild hydrocracking (MHC) process that transforms longer chains of hydrocarbons into shorter and more valuable ones, such as diesel. Impurities as nitrogen and sulfur are to a great extent removed from the short hydrocarbon chains which are formed in the process. Both the impurities and the hydrocarbon chains are then saturated with hydrogen. The components are then blended in to e.g. EU diesel.

Vacuum Gasoil

The vacuum gasoil (VGO) is the hydrocarbon chains separated in the vacuum distillation unit. Preemraff Lysekil also imports VGO from other refineries. The VGO is processed in crackers to produce shorter and more valuable chains. The primary cracker is the iso-cracking plant which produces hydrocarbons that may be used in blending of diesel oil. The process is also called hydrocracking as the hydrocarbon chains react with hydrogen under high pressure. The heavy fraction remaining after the iso-cracking process is called unconverted oil (UCO). The UCO is taken to a fluid catalytic cracker (FCC) that can vaporize and break these long chains and thus increases the H/C ratio. The resulting components are mainly used in gasoline. The FCC increases the flexibility of the refinery in making gasoline and diesel according to demand.

Residual oil

The residual oil goes through a so-called vis-breaking process which includes converting the residual oil to more valuable fractions by heating the feedstock in a furnace. In the furnace the hydrocarbon molecules are thermally cracked and the viscosity of the residual oil is reduced. The product of this process is mainly used for heavy oil and bunker oil. A small amount of naphtha and gasoil are formed in the process.

Desulfurization - Hydrotreating

The most common desulfurization technology in refineries is hydrodesulphurization which removes up to 90% of the sulfur in different fractions of liquid petroleum. Sulfur is removed from the petroleum fractions to reduce the sulfur dioxide emissions that result from combustion of the fuel. Furthermore sulfur could damage equipment as well as the catalysts used in the refining process. In the process, the feed is treated with hydrogen at specific pressure and temperature conditions that minimize hydrocracking. In addition to remove contaminants the process also saturates the molecules that are treated with hydrogen so they can be used to upgrade middle distillate petroleum fractions into finished kerosene, diesel fuel, and heating fuel oils.

The sulfur rich gas formed in the various desulfurization processes in the oil refinery is treated in an amine plant where approximately 99.9% of the sulfur is recovered as hydrogen sulfide. The gas is then led to an incinerator where the rest of the impurities are combusted completely. The hydrogen sulfide is transformed to pure sulfur in liquid form in a sulfur recycling plant. The elementary sulfur formed is used as a raw material for e.g. manufacturing of fertilizer and other chemicals.

Hydrogen production plant

The production and consumption of hydrogen during different steps of the refining is important. With increasing demands for product flexibility, higher yields, and lower sulfur content the demand of hydrogen is increasing. In Lysekil the hydrogen from the reforming process is not sufficient for the hydrogen consumption within the refinery and an addition hydrogen production plant is operated to fulfill the need for hydrogen.

Hydrogen is produced by steam reforming of methane. Today the methane is produced from butane in a pre-reformer but an LNG terminal is under construction. In the steam methane reformer (SMR) the methane reacts with steam at high temperatures and in the presence of a metal-based catalyst to form a syngas, mainly consisting of hydrogen and carbon monoxide (CO). The syngas is then further treated in a water-gas-shift (WGS) reactor where CO reacts with steam to form CO_2 and hydrogen at lower temperatures and in the presence of a catalyst. Finally high purity hydrogen (99.99%) is separated from the other gases in a pressure swing absorber (PSA). The hydrogen gas is then used as a feedstock for hydrogen consuming processes inside the refinery, especially the iso-cracker.

CO_2 capture in oil and gas refineries

A series of reviews and overview studies are available in literature on possibilities for CCS in refineries - the IPCC report [1] being the one most commonly referred to. The IPCC concludes that the cost for CO_2 captured from refineries currently is in the range 70 and 110 USD/t but could be lowered to between 40 and 80 USD/t with foreseeable advances in capture technologies. A more recent review includes the assessment by Kuramochi et al. [2]. They conclude on similar costs for CCS as the IPCC report and stress the importance of site specific conditions for the capture cost. They propose that further research should focus on energy and economic performance of industrial CCS under site specific conditions, effects of capture on emissions of air pollutants, effects on the production process as well as potentials for integration with power production.

Furthermore, carbon capture in oil refineries has been investigated by [3-8]. All the studies reviewed estimated the cost of capture up to a delivery point for further transportation and storage, i.e. the capture plant and compression of the CO_2 was included in the cost estimations. As concluded in the reviews, there is a large difference in the estimated costs, which is strongly influenced by the heterogeneity of the refinery. Studies focusing on hydrogen production by steam-methane reforming are generally in the low-end of the cost scale. As expected most studies focus on post-combustion capture, however, the studies that include oxy-fuel combustion conclude that this capture technology is the most economical. It is reasonable to believe that the comparison between technologies is highly dependent on assumptions on technology maturity.

Studies focusing on the integration of a carbon capture unit with a hydrogen production process include the work by Collodi [6], who showed that carbon capture for the reformer flue gas would allow for an overall capture of roughly 90%. CO_2 capture from the hydrogen rich product from the steam methane reformer would allow for around 60% overall capture. The study by Meerman et al. [7] concluded that the optimal location of the CO_2 capture unit is between the shift reactor and the PSA and that this configuration results in around 60% CO_2 reduction based on using ADIP-X as a solvent.

The present work investigates CO_2 capture possibilities at an existing, complex refinery, thus considering site-specific conditions in detailed modelling of investigated CO_2 capture processes and a thorough evaluation of feasible CO_2 sources at the plant site. In addition, heat integration possibilities and potentials for integration with power production based on site-specific conditions are evaluated

Current carbon capture initiatives (pilot/demo)

There are currently two large scale demonstration projects for CCS related to refineries. These are summarized in Table 6.1. Common for both projects are that they capture CO_2 from existing SMR units and that they have offset for the captured CO_2 through enhanced oil recovery (EOR). One plant uses vacuum swing adsorption (VSA) and one plant uses absorption with an amine solvent.

Table 6.1 Existing CCS demonstration projects in oil refineries

	Port Arthur [9]	Quest [10]
Company	Air Products	Shell
Location	Texas, US	Alberta, Canada
Status	Operation 2013 to 2015	Under construction
Size	1 Mt/a	1.2 Mt/a
Capture type	Post-combustion (VSA)	Post-combustion (amine)
CO_2 source	Existing steam-methane reformers	Existing steam-methane reformers
Storage	EOR	EOR

Major technical challenges for carbon capture

The following characteristics of the refinery will be of importance to carbon capture:

- The refinery is a complex process with several flue gas streams
- The CO_2 concentration vary within the process from low to high (around 5 to 50%)
- There is a relatively large amount of excess heat available within the process
- The main product of the refinery is a fossil fuel and the refining process accounts for less than 10% of the well-to-wheel emission of CO_2.

The complexity of the refinery will favor different solutions to reduce CO_2 emissions within the process and it will be necessary to focus on the individual process units rather than the entire refinery. Especially the large variation in CO_2 concentration is important to the economy and competitiveness of different capture technologies. The refineries are of special interest for carbon capture as they have excess heat which can be utilized to power the capture process. This heat is however distributed over the refinery and it is important to assess how much of this heat that can be utilized. Furthermore, it is important to stress that emission of CO_2 from the end-use of the oil products is an even larger concern than the process emissions for the refining industry.

Case study – Partial CO_2 capture in oil refineries

To make an assessment of carbon capture possibilities in the Preem refinery in Lysekil, the refinery processes are mapped with respect to their CO_2 emission sources and excess heat. The CO_2 emission in the refinery is collected and released from several platforms and the available excess heat from the platforms are identified and characterized in order to evaluate possible heat integration with a carbon capture process. Two CO_2 capture technologies based on chemical absorption are investigated, using MEA (see methodology section) and ammonia as absorbents. For a detailed description of the ammonia based CO_2 capture process model, see Garðarsdóttir et al. [11].

Evaluation of excess heat and heat integration possibilities

A simple approach is used for estimating the available waste heat at the refinery, that is, only extractable heat from flue gases is considered for use in a waste heat boiler and potential heat integration within process units is not considered in this work. With this approach, interference with production in the refinery is avoided and production losses can thus be neglected which simplifies the analysis. In the study, the amount of waste heat with respect to the heat requirement of the capture process itself is evaluated in two ways, firstly, by estimating how much of the total heat requirement at 85% capture rate can be covered by the waste heat and secondly, by estimating how much CO_2 can be captured by waste heat alone. For the second alternative, linear dependence is assumed between the capture rate and energy consumption in the reboiler. In the case studies, the temperature difference of the system is assumed as ΔT_{system} = 30 K. In the model simulations, the reboiler operates at 120°C and 160°C in the MEA and ammonia-base process, respectively, meaning that the temperature of the exhaust gas leaving the waste heat boiler is 150°C and 190°C for the two capture processes.

External energy generation

The additional energy needed to power the capture process as well as to drive the CO_2 compression train has to be supplied via external energy source. In this case study, the energy is supplied by combusting natural gas in a boiler system, generating superheated steam. The reason why natural gas, LNG, is assumed as the fuel used is because a LNG terminal is present at the refinery site and it should thus have a high availability. For more detailed description of the external energy plant and assumptions made in the calculations, see section on methodology.

The results are divided into two chapters. The first chapter evaluates the CO_2 and heat sources at the Preem refinery in Lysekil and the second chapter discusses the results of the absorption process simulations and the heat integration study.

Mapping of CO_2 sources and available heat

The Lysekil refinery is divided into five platforms, each with an individual stack. The flue gas formed from a series of various process units are, thus, collected within each platform before emission. The largest part of the emission comes from gas fired process heaters. Some process heaters also use oil, as a backup fuel, and the sulfur reactor heaters and the incinerator in the sulfur recycling plant also burn hydrogen sulfide (H_2S). Figure 6.2 shows the total emission per year from each stack. The amount of NO_X and SO_X in the flue gas is low, which is important for capture technologies sensitive to sulfur, such as absorption with MEA. The low NO_X content is accomplished by using low NO_X and ultra-low NO_X burners. Only the FCC process has flue gas cleaning system. This information along with an examination of the formation of CO_2 in each unit is important for identifying an appropriate CO_2 capture technology for each unit or stack.

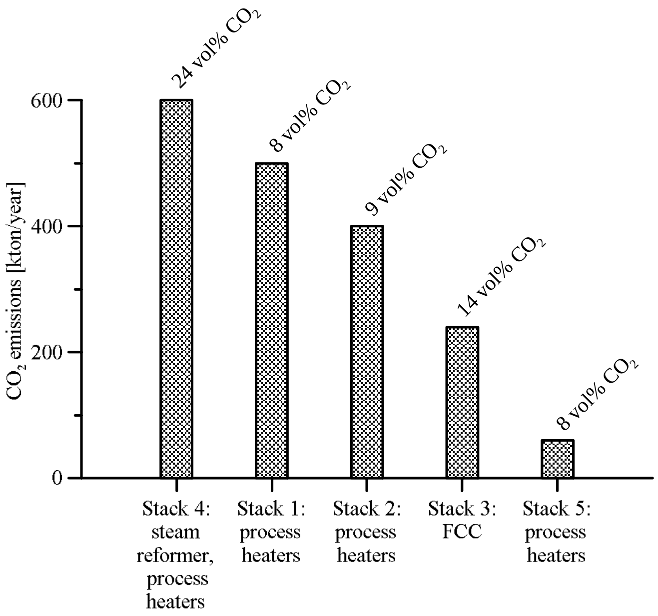

Figure 6.2 CO_2 emissions from stacks. The emissions do not include emissions allocated to the electricity used in the processes

Stack 1: Crude and vacuum distillation

The main CO_2 sources in stack 1 are two crude oil heaters, two vacuum oil heaters, three steam boilers, hot oil heater and an incinerator. Other emission sources to stack 1 are the vacuum pumps in the vacuum distillation unit. The share of CO_2 emission from the distillation units in Preemraff Lysekil is about 25% of the total CO_2 emissions. The CO_2 concentration in the exiting flue gases is around 5-10%.

The atmospheric crude oil distillation process is a large energy consumer within the refinery and needs different types of energy inputs such as steam, fuel oil and cooling water. Both the atmospheric and vacuum distillation processes require heat to bring the feed oil up to high temperatures so the oil fractions can be separated. The heat required is generated by combustion processes in various process heaters.

Stack 2: Platformer, SynSat and Vis-breaker

The CO_2 emission from the Platformer is formed when coke is combusted with air off the catalytic surface in the catalyst regeneration process. CO_2 is also formed in six gas fired heaters that produce heat for the process. In the SynSat process, CO_2 is formed in two gas fired process heaters. All CO_2 emission allocated to the vis-breaking process is formed in two gas fired heaters. In Preemraff Lysekil, the Platformer along with the SynSat and vis-breaking processes account for about 20% of the total CO_2 emitted from Preemraff Lysekil with a CO_2 concentration of about 8% in the exiting flue gas stream.

The excess heat released from the Platformer is generated in a catalytic regeneration process and in process heaters. Excess heat released from the SynSat process is generated in process heaters and in the process reactions which are exothermic. Vis-breaking is a highly endothermic process and the excess heat from the process is therefore only generated in the process heaters.

Stack 3: Fluid Catalytic cracker

CO_2 in the FCC process is formed when air is used to regenerate the catalysts by burning coke off the catalytic surface. The heat generated by the coke combustion in the catalytic regeneration process is more than required to power the cracking process.

Stack 4: Hydrogen production unit

More than half of the total amount of CO_2 that leaves the unit is formed in the gas-shift reaction. The remaining CO_2 is formed in the combustion of additional fuel gas required by the steam methane reformer (SMR). About 30% of the CO_2 emission from Preemraff Lysekil comes from the hydrogen steam reformer. The flue gas stream has a CO_2 concentration of about 24%.

The overall reaction in the steam reformer is endothermic and requires heat from process heaters. The shift reaction is mildly exothermic and therefore produces small amount of heat.

Stack 5: Iso-cracker

Part of the CO_2 emissions from the iso-cracker are formed in two gas fired heaters. Other emissions from the process are from the regeneration of the catalyst where coke is combusted off the catalytic surface with air. CO_2 emissions from the iso-cracking process accounts for about 3% of the total emissions from Preemraff Lysekil with about 8% CO_2 concentration in the exiting flue gas stream. The overall iso-cracking process is exothermic but the feed has to be heated before it is sent to the reactors where the cracking occurs. The excess heat is therefore generated both from process reactions and process heaters.

Process simulation – three cases for carbon capture

Stack 4: Hydrogen production unit

The largest point source of CO_2 emission at the refinery is the exhaust gas from the hydrogen production unit through stack 4, see Figure 6.3 In addition, the exhaust gas has relatively high CO_2 content. As discussed in the previous chapter, the CO_2 released through the stack is formed in both a process heater and the steam reforming process itself. In this study, post-combustion CO_2 capture is the examined carbon capture technology. The CO_2 capture unit can be integrated with the SMR in several ways. Three locations, which all have different properties with respect to CO_2 concentration and flow rates, are usually considered in the literature. These are shown in Figure 6.3 as the flue gas from the steam reformer + furnace (1), between the WGS and the PSA units (2) and with the purge gas from the PSA unit (3).

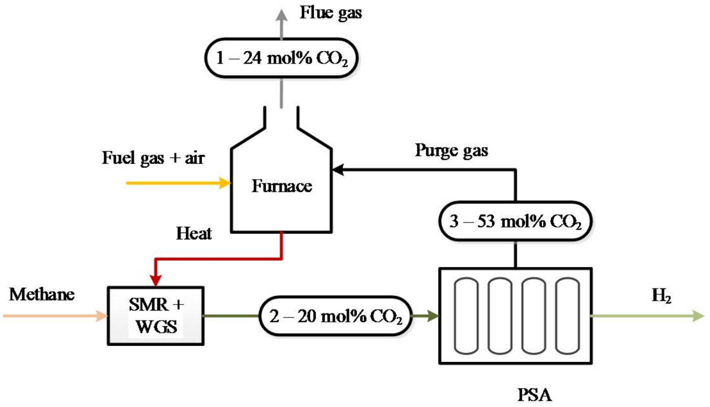

Figure 6.3 Schematic of the hydrogen production process with the three suggested locations of the carbon capture process, including CO_2 concentrations at each location

The location of the carbon capture unit is chosen so as to minimize interference with the hydrogen production itself while maximizing the overall capture potential. Thus, location 1 was chosen for this simulation.

Stack 1: Crude and vacuum distillation

The flue gases released through stack 1 originate from a number of process heaters and constitute the largest flue gas stream in the refinery. For this case the carbon capture unit is placed at the end of the flue gas train.

Stack 3: Fluid catalytic cracker

The fluid catalytic cracker is the second smallest source of CO_2 emission at the refinery. However, the flue gas stream has a relatively high CO_2 content compared with the other stacks, 14 mol-%. As mentioned previously, the CO_2 released through stack 3 originates from coke combustion when regenerating the catalyst. Similarly as for the other two cases, the post combustion unit is located at the end of the flue gas train; the properties and the composition of the flue gas stream for all three stacks investigated are presented in Table 6.2.

Table 6.2 Flue gas properties and composition at position 1 in the SMR, stack 1 and 3

Location	CO_2	N_2	H_2O	O_2	Temperature [°C]	Mass flow [kg/s]
Position 1, SMR	24	59	15	2	170	55
Stack 1	8	73	15	4	160	126
Stack 3	14	70	15	1	270	36

Simulations of the capture process

The specific heat requirement of the two absorption processes investigated is presented in Figure 6.4 as well as the total reboiler duty for the three cases investigated. All cases were simulated with a CO_2 capture rate of 85%, resulting in 510, 425 and 204 kt CO_2/a captured from stack 4, 1 and 3, respectively. Due to high temperatures in the MEA-based absorber in the case of stack 4, the SMR, the temperature profile was optimized using intercooling, which was previously shown to improve the process performance at relatively high flue gas CO_2 concentration [12]. Intercooling to 50°C was applied in the column at the optimal position of 1/2th of the column height and is assumed to be achieved with on-site cooling water. The application of intercooling resulted in a decrease in the specific heat requirement by 7%.

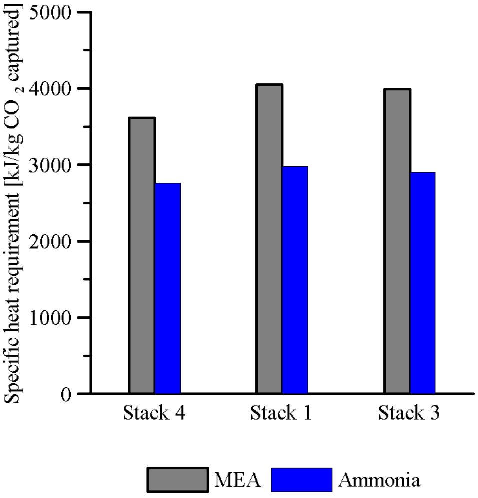

Figure 6.4 Specific heat requirement and reboiler duty of CO_2 capture with a) MEA and b) ammonia when applied to the three stacks investigated

Heat integration possibilities

The most easily accessed excess-heat is the heat present in the flue gas leaving the stacks. This heat can be used to meet a part of the energy requirement of the carbon capture process. The available excess heat at each stack is characterized in Table 6.3 for both the MEA and ammonia based absorption process.

Table 6.3 Available waste heat in the flue gases leaving the stacks and share of total heat required in both MEA and ammonia based absorption integrated with the hydrogen production process

	Temperature	Mass flow	Available waste heat [MW]	
	[°C]	[kg/s]	MEA	CAP
Stack 1	160	126	1.5	0
Stack 2	180	90	3.1	0
Stack 3	270	36	5.0	3.6
Stack 4	170	55	1.3	0
Stack 5	130	15	0	0
Total			10.9	3.6

Results from the heat integration study are summarized in Figure 6.5. Due to the relatively high operating temperature in the ammonia desorber, a considerably lower amount of waste heat is available for the ammonia process than for the MEA process. Thus a larger amount of CO_2 could be captured with waste heat alone using MEA-based absorption than ammonia. However, more than half of the heat needed in all of the three cases needs to be supplied from an external energy source. It could also be noted that considerably less energy is needed to power the CO_2 compression train in the ammonia-based absorption due to higher operating pressure in the desorber compared to the MEA-based absorption.

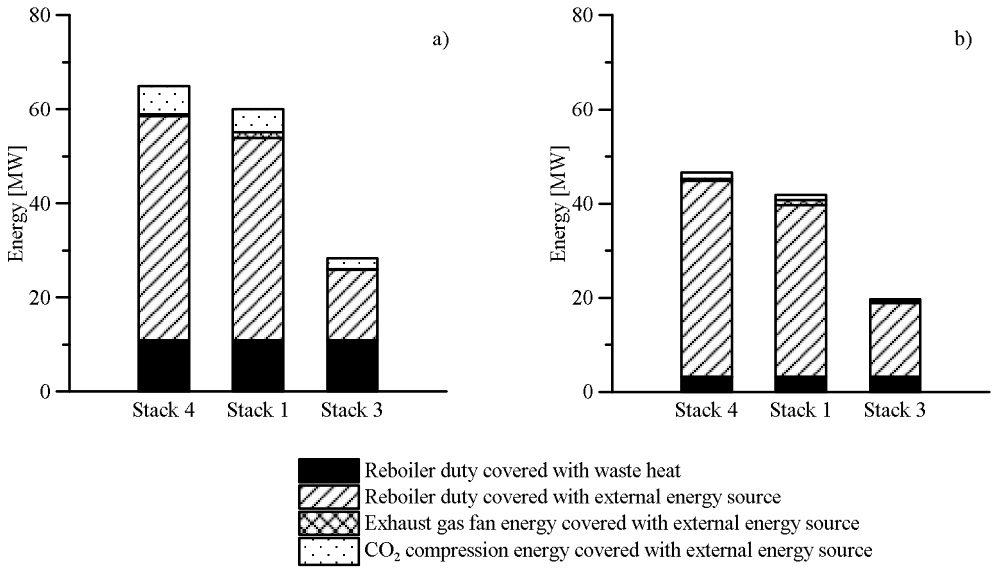

Figure 6.5 The energy needed to cover the requirements for a) MEA and b) ammonia based absorption

<u>External energy generation</u>

Since CO_2 is released during the combustion process in the boiler system the net CO_2 capture rate from the refinery site is decreased. However, when evaluated on a plant level, the rate of CO_2 capture does not decrease drastically since each of the stacks investigated does not, individually, account for a majority of the CO_2 emission, or around 30%, 28% and 13% of the total plant emissions for stacks 4, 1 and 3, respectively. The capture rate on a plant level when accounting for the emissions from the energy plant is presented in Figure 6.6 and as a comparison, the capture rate without accounting for the energy plant is plotted in the figure.

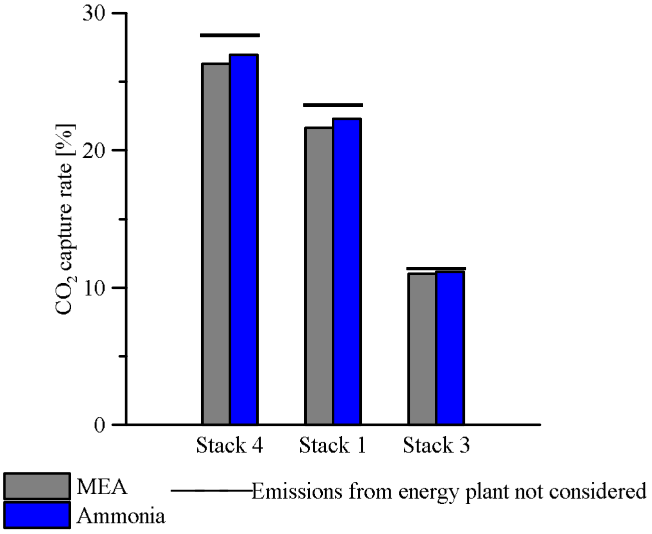

Figure 6.6 Capture rate on a plant level from the different stacks investigated when emissions from the energy plant are accounted for compared with the capture rate when emissions from energy plant are not considered

Conclusions

The evaluation of the refinery found that the largest point source of CO_2 emissions at the refinery, exhaust gas from the hydrogen production unit, also has the highest CO_2 content of the five refinery stacks. Capture from the largest stack, stack 1 was also investigated as well as capture from the FCC unit, stack 3, which is the second smallest emission source but has a relatively high CO_2 content in the flue gas stream.

Two post-combustion capture processes applied to three different stacks at the refinery were investigated, using MEA and ammonia as solvents. The process simulations show that the ammonia process is more favorable from heat requirement point of view. The evaluation also shows that a share of the total heat required in the post combustion processes can be covered with waste heat from the exhaust gases of the refinery processes. The higher share of heat recovered for the MEA based process is due to lower operating temperature in the reboiler compared with the ammonia based process.

A drawback regarding the refinery is that is consists of a number of CO_2 sources and even though the hydrogen production unit (stack 4) is the single largest point source at the refinery, it only accounts for roughly 30% of the refinery´s total emissions. A significant part of the capture process heat demand can be covered by waste heat, and thus there is not a drastic decrease in the net capture rate on a plant level when emissions from the external energy plant are considered.

From the case study on Preem refinery in Lysekil it is concluded that:

- The complexity of the refinery process gives a large variety in size and quality of the CO_2 sources. It is therefore of importance to investigate the sub-processes of the refinery individually.
- The refinery includes CO_2 sources that are favorable for CO_2 capture. The hydrogen production through steam-methane reforming being the most beneficial due to high CO_2 content in the flue gas and being the largest point source of CO_2 at the refinery site.

The refinery also includes CO_2 sources that are not suitable for CO_2 capture, mainly due to their relatively small size, which will have the consequence that the overall capture efficiency of the refinery will be below the 85-90% that is possible to achieve on an individual stream.

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CARBON CAPTURE IN GEOTHERMAL POWER PRODUCTION IN ICELAND

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Process overview

Detailed information on the Hellisheiði plant can be found in [1]. The plant is located in Iceland around 20 km from Reykjavik and cogenerates power for energy-intensive industry and hot water for district heating. The plant consists of six 45 MW_e high-pressure steam turbines (HPT) and one 33 MW_e low-pressure steam turbine (LPT) for a total of around 300 MW_e. The thermal capacity is, as of 2014, 133 MW_{th}, but the plant has a plan to extend it to 400 MW_{th}. The power plant has 30 production wells located in an 8 km^2 area around the power plant. The steam at Hellisheiði is not pure H_2O, but also contains H_2S, CO_2, H_2, N_2, and CH_4.

From the production wells, the saturated steam is transported to gas liquid separators with a pressure of 10 bar; the liquid is pumped to the low pressure liquid gas separator which operates at 2 bar. The gas from both the high and low pressure liquid gas separators is taken to moist separators to prevent liquid from entering the turbines. After the HPT, the steam is condensed. The condensers preheat the fresh water for the district heating system. Because the fresh water is saturated with dissolved oxygen and becomes corrosive when heated, the heated water is deaerated before leaving the plant. The NCG are extracted from the condensers. The NCG from this power plant vary in composition between the different turbines, but all streams contain CO_2, H_2S, CH_4, H_2 and N_2. The mass fraction varies from 59% CO_2 to 83% CO_2 as is shown in Table 7.1. The gas component mass flows of the incoming geothermal steam are shown in Table 7.2. As can be observed the emission rate is very low compared to fossil power generation of comparable power output. The flowrates of CO_2 and H_2S are maximum 51 kt/a and 21 kt/a, respectively.

Table 7.1 Mass fraction of non-condensed gases exiting the steam turbines at Hellisheiði. Turbines 1–6 are high-pressure turbines and turbine LP is low-pressure. Table based on numbers from Aradóttir [4]

	CO_2 [wt-%]	H_2S [wt-%]	H_2 [wt-%]	N_2 [wt-%]	CH_4 [wt-%]
Turbine 1	59.37	38.60	1.03	0.89	0.13
Turbine 2	70.56	28.03	0.69	0.59	0.12
Turbine 3	67.28	31.12	0.94	0.57	0.08
Turbine 4	59.44	38.49	1.07	0.87	0.14
Turbine 5	82.69	12.77	0.32	4.07	0.14
Turbine 6	73.61	24.70	0.78	0.80	0.11
Turbine LP	58.65	40.34	0.33	0.66	0.03

Table 7.2 Component mass flows (kg/s) of the Hellisheiði geothermal steam. Numbers based on Table 7.1 and Hallgrímsdóttir et al. [1]

	H_2O	CO_2	H_2S	H_2	N_2	CH_4	Total
Mass flow [kg/s]	1177.67	1.61	0.67	0.017	0.03	0.0027	1180.0

As mentioned, there are two projects for acid gas re-injection in Iceland: CarbFix and SulFix. Both of them focus on pumping CO_2 or H_2S or both down into the ground. Tests done in the CarbFix project, where pure CO_2 dissolved in water are re-injected, have been successful. The CO_2 have been injected at a depth of 400 m. The SulFix project started in January 2013; here CO_2 and H_2S are injected with water to a reservoir below 800 m depth.

Case study – Different options for acid gas removal

There are several different H_2S and CO_2 capture methods used in the industry [5]. Some of those have been considered for H_2S capture for geothermal power plants [6,7]. Four different capture systems were selected and subsequently modeled and simulated in this work:

- Water absorption. This is the reference process and tested on site.
- Amine absorption with MDEA.
- Amine–low temperature hybrid concept.
- Stand-alone low-temperature (cryo) separation.

Water absorption

According to the ongoing research of the CarbFix and SulFix projects, the two gases CO_2 and H_2S have to be dissolved in water in order to be injected back into the ground [2,3]. An absorption process consists of an absorber in which the specific gases are absorbed by a solvent and a desorber wherein these gases are stripped from the solvent. The gases leave the top of the desorber whereas the solvent leaves the bottom and is recirculated. However, in the special case where a solution of water and gases is injected directly into the ground in the second part of the process, the desorber is not needed.

The solubility of CO_2 and H_2S are dependent on pressure and temperature. The lower the temperature and the higher the pressure, the better is the solubility, which means less water is required to absorb the gases. In Iceland, the available fresh water has a temperature of around 5°C leading to a rather low absorber temperature. The increase in pressure brings a higher pumping power for the water as well as a higher compressor power for the gases.

The water absorption process model was validated against Hellisheiði plant data from the installed gas separation station. The validation was done by setting the pressures and temperatures of the streams close to the plant data and subsequently calculating the required mass flow rate of water to achieve the equal absorption rate, as in the tested water absorption process at the plant, mass flow rates of captured and cleaned gas flows. The calculated mass flow of water in the process simulation was within 1% of the plant data. The validated flow sheet is shown in Figure 7.1.

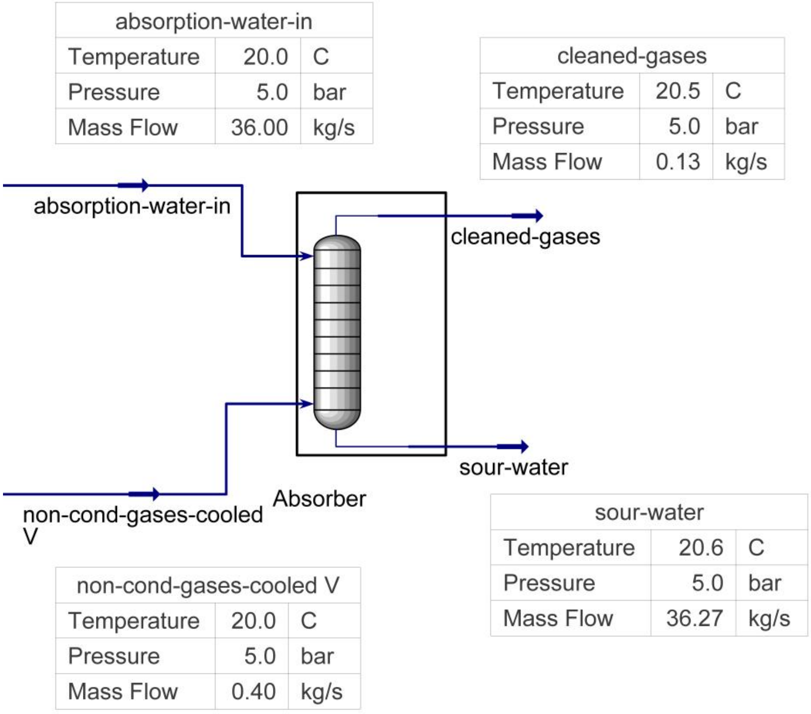

Figure 7.1 Validated water absorption process

As the main criterion for the Hellisheiði plant was to capture H_2S according to the new Icelandic law, the amount of captured CO_2 was not the main focus. The selected solution was to capture almost all H_2S (99%) and a substantial amount of CO_2 (78.5%). To reach a reasonable compromise between absorber pressure and water consumption, an absorber pressure of 15 bar was selected. This led to a water mass flow of 64 kg/s which gave a mass flow ratio of 26.5 between the water and NCG. The results are summarized in Table 7.3. The power penalty which came along with these parameters was 1.7 MW (vacuum pumping, water pumping and compression power). Excluding the vacuum pumping power the power penalty is approximately 0.75 MW. For all case studies in the rest of the acid gas removal options, the vacuum pumping power has been excluded, but would be equal for all cases. Hence, 0.75 MW is the figure to which the results for other acid gas removal options should be compared.

Table 7.3 Key results for the water absorption process simulation

Pressure	Captured gas		Mass flow water	Power penalty
[bar]	[mol-%]		[kg/s]	[MW]
Absorber	H_2S	CO_2	H_2O	Total
15.0	99.0	78.5	64.0	1.7[a] 0.75[b]

[a]Including vacuum pumping of turbine off-gas [b] Excluding vacuum pumping – for benchmarking with energy results for the other acid gas removal options

MDEA absorption

The acid gas from all seven steam turbines was assumed to be mixed to make a single, consolidated stream, with about 55 mol-% CO_2 and 30 mol-% H_2S concentrations. As this stream was available at atmospheric pressure, the suggested process included compression of this stream to 20 bar in order to increase the acid gas partial pressure. The absorber in the equilibrium-based model was simulated with 15 theoretical stages using a 42 wt-% MDEA solution. The rich MDEA loading, for H_2S and CO_2 in aggregate, leaving the bottom stage was 0.55. The desorber column was set to operate at 1.2–1.5 bar with a reboiler temperature of 113.6°C. The resulting lean MDEA loading was 0.019. Preheating of the rich solvent stream was carried out in a liquid-liquid heat exchanger configured to operate with a minimum temperature approach of 15°C. In the non-equilibrium simulations the desorber operated at 1.5–1.75 bar pressure and the reboiler temperature was 116.9°C. The lean and rich loading of the solvent were 0.044 and 0.53, respectively.

Results for the equilibrium and non-equilibrium simulations are given in Table 7.4. This table also includes additional product stream data for the MDEA/low-temperature hybrid process scheme. A first important difference to observe between the two simulation cases is the increased H_2S selectivity for the non-equilibrium case. The amount CO_2 co-captured with H_2S is significantly lower, resulting in higher CO_2 concentration in the sweet gas (70.7 mol-% vs. 57.1 mol-%), higher acid gas purity (58.4 mol-% H_2S vs. 41.4 mol-%) and higher flowrate of the CO_2 stream separated in the low-temperature separation stage (71.2 kmol/h vs. 37.1 kmol/h).

Table 7.4 Stream compositions for the two simulated cases of the two-stage hybrid gas separation scheme (MDEA + low-temperature). Columns with grey background indicate equilibrium results while those with white background are from non-equilibrium simulations

	Unit	Feed gas		Sweet gas		Acid gas (H_2S)		CO_2		Raw H_2	
Flowrate	kmol/h	206.1	206.1	72.5	106.6	143.0	105	37.1	71.2	35.3	35.2
Temperature	°C	25.0	25.0		26.12		40				
Pressure	bar	20.5	20.5	20.0	19.5	1.2	1.5	7–100+	7–100+	57	57
Composition											
H_2O	mol-%	0.0	0.0	0.21	0.19	6.75	5.0	0.0	0.0	0.0	0.0
CO_2	mol-%	55.18	55.18	57.10	70.74	50.71	36.51	99.81	99.80	12.48	12.47
H_2S	mol-%/ppm	29.74	29.74	0.01	0.02	42.43	58.44	179 ppm	285 ppm	19 ppm	30 ppm
CH_4	mol-%	0.26	0.25	0.72	0.48	0.0	0.0	0.07	0.07	1.41	1.31
N_2	mol-%	1.62	1.62	4.59	3.12	0.01	0.0	0.08	0.08	9.36	9.27
H_2	mol-%	13.21	13.21	37.36	25.45	0.09	0.04	0.02	0.02	76.75	76.95

The energy requirement is shown in Table 7.5. The main driver for power requirement was that for the front-end sour gas compression from 1 atm to 20 bar assumed for the MDEA process, which amounts to around 0.77 MW (two intercooled stages with 75% isentropic efficiency). In addition to power, around 3 MW of regeneration heat was required for the non-equilibrium case.

Table 7.5 Key results for the amine absorption of H_2S and CO_2. Power penalty does not include compression of the acid gas product.

	Pressure [bar]	Captured gas [mol-%]		Power penalty [MW]	Thermal energy [MW]
	Absorber	H_2S	CO_2	Total	Reboiler
Equilibrium	20	98.9	63.7	0.8	4.6 (114°C)
Non-equilibrium	20	99.9	33.6	0.7	3.0 (117°C)

Amine – low-temperature hybrid concept

A two-stage hybrid gas separation concept, as shown in Figure 7.2, has been investigated for the NCG. The first stage was an MDEA chemical absorption process configured to selectively remove H_2S from the feed gas. The sweetened and CO_2-rich gas from the MDEA absorber was passed to a CO_2 condensation unit in which the main portion of CO_2 was separated from the volatile components. The gaseous product from this separation stage had high hydrogen concentration, which may be further processed to produce high-purity hydrogen for use, e.g., in fuel cell vehicles in the immediate area.

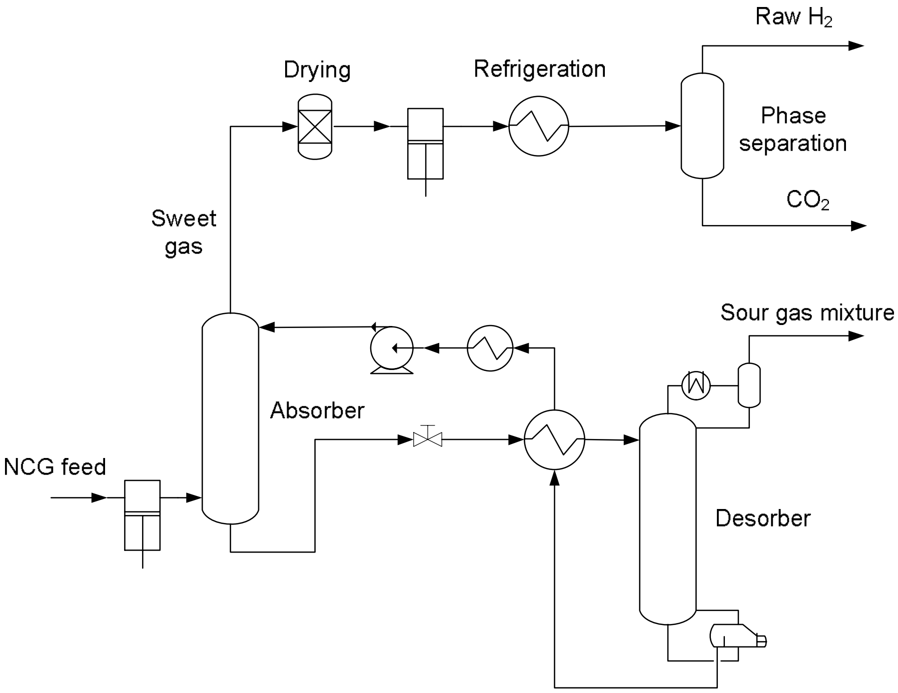

Figure 7.2 Simplified hybrid two-stage acid gas separation scheme

The purpose of this unit was to separate the CO_2 from the volatile components H_2, N_2 and CH_4. For high initial CO_2 concentrations such as those obtained for the sweet gas (57% and 71%), condensation and phase separation is an effective process. Prior to entering the low-temperature separation unit, complete dehydration is required, typically by molecular sieve adsorption. Subsequently, the dried sweet gas was assumed to be compressed to around 60 bar and cooled to a separation temperature somewhere between -56°C and -55°C. Depending on the exact CO_2 concentration, pressure and separation temperature, 90–95% of the CO_2 was condensed and separated in liquid phase, while a hydrogen-rich gaseous product was extracted from the top of the phase separator. The disposal of H_2S and CO_2 products were not further considered in the process simulation work, but there are various options for handling of these separation product streams.

The separation processes have by no means been optimized. The choice of process and process sequencing will be subject to preferred separation product specifications (purity/composition, pressure, phase, etc.). This applies in particular to the H_2S and CO_2 products. There seem to be two typical options for the hydrogen stream – combustion/flaring/purging or purification and sale – and the option of hydrogen purification and sale will be dependent on the processing cost in relation to commercial value.

The energy requirement for the amine part of the hybrid process is identical to that shown in Table 7.5. Product stream data is given in Table 7.4. The additional power requirement in form of compression to 60 bar and cooling in the low-temperature process was very modest in comparison, and amounts to roughly 0.2 MW. It is very important to emphasize that this figure includes pressurization of the CO_2 product to around 110 bar by liquid pumping. It should also be noted that the energy requirement associated with gas drying before the cooling process (for instance hot nitrogen gas for periodic regeneration of drying bed) were not included in any of the low-temperature results. The total energy requirement is shown in Table 7.6.

Table 7.6 Key results for the amine–low temperature absorption of H_2S and CO_2. The non-equilibrium MDEA simulations are used as basis for the low-temperature part.

Pressure [bar]		Captured gas [mol-%]		Power penalty [MW]	Thermal energy [MW]
Absorber	Low-T. separator	H_2S	CO_2	Total	Reboiler
20	57.5	99.9	96.0[a]	0.9	3.0 (117°C)

[a] 33.6% (entrained in H_2S product) + 62.4% (99.8% pure CO_2 product stream at 100+ bar pressure)

Stand-alone low-temperature (cryogenic) separation

A direct cooling and phase separation of the NCG can be an interesting alternative if re-injection of a mixed H_2S/CO_2 product is acceptable. The liquid separation process will be more or less identical to the low-temperature process in the hybrid concept, and the liquid product will consist of mainly CO_2 and H_2S while the volatile components will remain in the gaseous separation product. A process flow diagram is shown in Figure 7.3. Pressurisation of the liquid H_2S/CO_2 product is obtained by liquid pumping, which is very energy efficient.

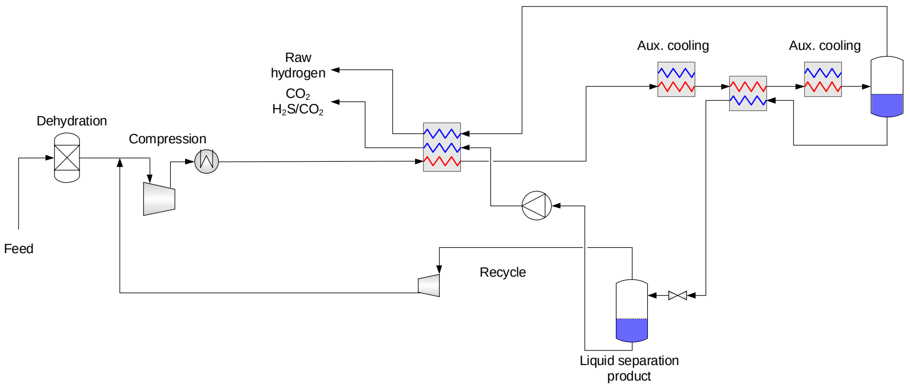

Figure 7.3 Low-temperature gas separation scheme

CO_2 can be separated from H_2S and purified to a large extent by cooling and phase separation. There will however still be trace impurities in the CO_2 and the product will not conform to the strict standards of food-grade CO_2. It could still be investigated whether this product can be of any value for local customers such as greenhouse growers, as the purity is very high.

Hydrogen is another potentially valuable product which as a by-product is available at 70–80% purity. This product is likely of no value, but if a PSA purification unit is added to the process, hydrogen with very high purity, for instance for fuel cell vehicles, can be supplied at a rate of roughly 1 ton per day. The H_2S/CO_2–H_2 component split depends on the separation pressure and its impact on purities of the liquid phase (H_2S+CO_2) and gaseous components (H_2, N_2, CH_4), as well as H_2S/CO_2 separation ratio, is shown in Figure 7.4.

For the two selected cases, 17.5 and 37.5 bar separation pressure, overall results are summarised in Figure 7.4 and Table 7.7. The estimated power penalty was around 1.0 MW and 1.15 MW, respectively. It is important to emphasize that these power figures included compression of H_2S and CO_2 to around 110 bar. In addition, the raw hydrogen is available at high pressure and can be purified, flared or purged.

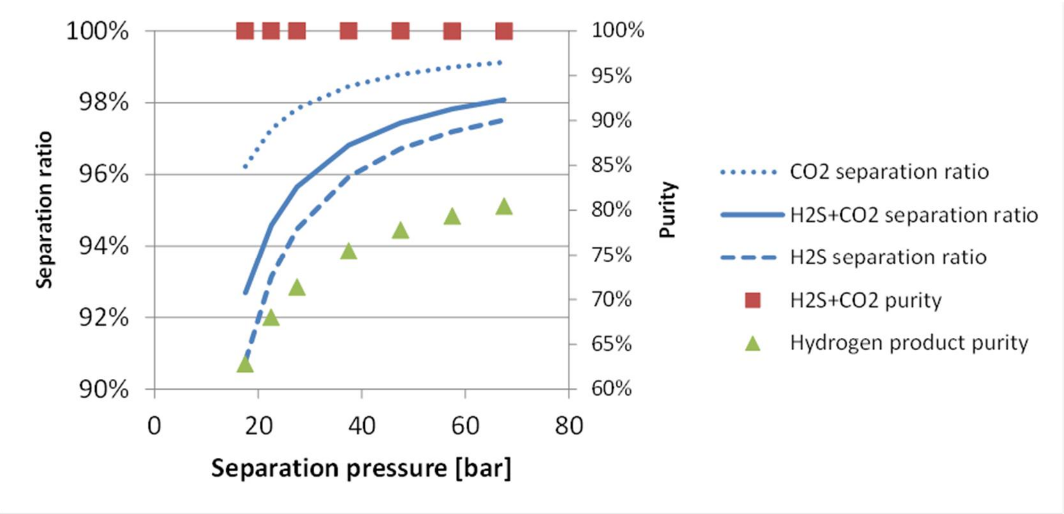

Figure 7.4 Results for low-temperature separation of H_2S and CO_2 from non-condensable components (H_2, N_2, CH_4)

Table 7.7 Selection of key results for the low-temperature separation of H_2S and CO_2 from non-condensable components (H_2, N_2, CH_4)

Pressure [bar]	Captured gas [%]		Power penalty [MW]
Main separator	H_2S	CO_2	Total
17.5	90.8	96.2	1.0
37.5	95.9	98.5	1.15

Conclusions

There are several technology options for removing the H_2S and CO_2 from the non-condensed gases in the Hellisheiði geothermal plant. Of these technologies, the following processes for H_2S and CO_2 removal have been simulated in order to determine energy requirement and separation capabilities: water absorption (reference process being tested), MDEA absorption, MDEA in combination with low-temperature (cryo) separation, and stand-alone low-temperature separation.

Simulation results for water absorption corresponded well with Hellisheiði plant data. An absorber pressure of 15 bar was considered a reasonable trade-off between compressor and pumping power requirement. A water/gas mass flow ratio of 26.5 resulted in 99% H_2S removal, 78.5% CO_2 removal and inflicted a power penalty of about 0.75 MW. This figure excludes vacuum pumping, and this applies to all other power requirement estimates. For MDEA absorption, 20 bar was selected as absorber pressure. 99.9% H_2S removal was obtained along with 33.6% CO_2 removal with an energy penalty of 0.7 MW power and 3.0 MW heat at 117°C reboiler temperature. This penalty figure did not include compression of the acid gas product stream from the desorber pressure (around 1.5 bar). In the MDEA/low-temperature hybrid process the energy requirement was equal to the MDEA case, with an additional 0.2 MW power penalty from the low-temperature unit. In addition to the H_2S and CO_2 removal from the MDEA process, an additional 62.4% CO_2 was removed in a separate stream at high pressure. A hydrogen stream – in one of the examples at 77% purity and 57 bar pressure – could become a valuable product provided that further purification is carried out, for instance by PSA. In the stand-alone low-temperature case, 95.9% H_2S removal and 98.5% CO_2 removal were obtained with a power penalty of 1.15 MW, including compression of the H_2S/CO_2 stream to 110 bar. A hydrogen stream of 75% purity and 37 bar pressure was also produced, which can be upgraded by PSA.

For co-removal of H_2S and CO_2, low-temperature separation seems to be an attractive alternative to the conventional water absorption process due to the low power penalty. Ultimately, the energy calculations must be complemented by cost estimations in order to get a more complete basis for comparing the different technologies.

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CONCLUSIONS

This report discusses the feasibility of separating CO_2 from industrial gas streams by conducting case studies on five types of industries: coal fired heat and power generation (as reference), iron and steel, cement, pulp and paper, oil and gas refining, and geothermal power generation. The work has reviewed available technologies for separating CO_2 and performed detailed process simulations on the technologies most feasible for short-term implementation. Table 8.1 summarizes the technologies investigated to capture CO_2 from the investigated industries. In total eight technologies have been investigated in detail, including pre-combustion, post combustion as well as oxy-fuel technologies. The broad set of technologies proposed shows that there is no technology that holds a clear advantage over the others, but that the circumstances for the specific industry is crucial to the choice of technology. The case studies on the cement production and oil and gas refining even show that there are important differences within the specific processes that strongly affects the suitability to separate CO_2 from different process streams, and the case studies on iron and steel, and pulp and paper production shows that the implementation of specific capture technologies may affect the favored development of the process. However, absorption based post-combustion technologies could be pointed out to have an advantage to other capture technologies in that it may be applied as an “end-of-pipe” technology with minimal interference on the process to any of the process streams.

Table. 8.1 Summary of investigated capture technologies

Industry	Capture Technology
Coal fired heat and power	Absorption (MEA)
Iron and Steel	Pre-combustion (MEA and Selexol)
Cement	Absorption (MEA)
	Oxy-Fuel Combustion
Pulp and Paper	Absorption (MEA)
	Pre-Combustion (Selexol and Rectisol)
Oil and Gas Refining	Absorption (MEA and NH3)
Geothermal Power	Absorption (Water and MDEA)
	Cryogenic separation

Even though the specific conditions of the different industries may favor certain capture technologies the performance of the capture technologies is similar to the performance when applied to a coal-fired power plant, for which most technologies have originally been developed. The most important difference with applying capture to an industrial process instead of heat and power generation is that the industrial processes do not have access to the large amount of low value heat required to power some capture technologies. To acquire and operate a unit to generate heat on a hundreds-of-

MWs scale is a large task. This fact may in turn favor technologies like oxy-fuel combustion which is mainly driven by power rather than heat.

This work shows that it can be technically feasible to apply carbon capture technologies to a broad set of process conditions. However, it is important to consider the specific conditions as they strongly affect the choice of technology as well as the performance of the technology. Considerable savings in capture cost could be made by considering the process and site specific conditions as well as the possible development of the process when designing for carbon capture. The related costs are published in NORDICCS Technical Report D3.13.1302/D13 *CCS case synthesis – Final report*.

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Abbreviations

AISI	American Iron and Steel Institute
AP	Constant net electricity production
ASU	air separation unit
BECCS	bio-energy with carbon capture and storage
BLG	black liquor gasification
BLGCC	black liquor gasification combined cycle
BLGMF	black liquor gasification with motor fuel production
BF	blast furnace
BOF	basic oxygen furnace
BTU	British thermal unit
CCF	cyclone converter furnace
CCS	carbon capture and storage
CCSP	Carbon Capture and Storage Program
CPU	compression and purification unit
CST	crude sulfate turpentine
CT	condensing tower
CTM	chemi-thermomechanical
DCC	direct contact cooler
DEPG	dimethyl ether of polyethylene glycol
DH	district heating
DME	dimethyl ether
DRI	direct reduced iron
e	electric
EAF	electric arc furnace

ECRA	European Cement Research Academy
EDRP	experimental direct reduction pilot
EOR	enhanced oil recovery
EOS	equation of state
ESP	electrostatic precipitators
EU	European Union
FCC	fluid catalytic cracker
FF	bag filter (fabric filter)
FGD	Flue gas desulfurization
FI	Finland
GHG	greenhouse gas
GJ	gigajoule
GSA	gas suspension absorber
GT	gas turbine
GTCC	gas turbine combined cycle
h	hour
HM	hot metal [kg/s]
HP	high pressure
HPT	high pressure turbine
IEA	International Energy Agency
IP	intermediate pressure
IPCC	Intergovernmental Panel on Climate Change
IS	Iceland
kg	kilo
kmol	kilo mole
kWh	kilowatt hour

LGO	light gas oil
LHV	lower heating value
LNG	liquefied natural gas
LP	low pressure
LPG	liquefied petroleum gas
LPT	low pressure turbine
MDEA	methyl diethanolamine
MEA	monoethanolamine
MHC	mild hydrocracking
MJ	megajoule
mol	mole
MP	medium pressure
Mt	megaton
MW	megawatt
NCG	non-condensable gases
Nm^3	normal cubic meter
NO	Norway
NORDICCS	Nordic CCS Competence Centre
NRTL	Nationally Recognized Testing Laboratory
NTNU	Norges teknisk-vitenskapelige universitet (Norwegian University of Science and Technology)
OHF	open heart furnace
PCI	pulverized coal injection
PH	pre-heater
ppm	parts per million
ppmv	parts per million volume

PR	Peng-Robinson
PRSV	Peng-Robinson-Stryjek-Vera
PSA	pressure swing adsorption
RB	recovery boiler
RDF	refused derived fuels
RM	raw mill
RTI	Research Triangle Institute
s	second
SCOT	Shell Claus off-gas treating
SE	Sweden
SMR	steam methane reformer
SNCR	selective non catalytic reduction
SR	smelting reduction
t	ton [SI]
TGR	top-gas recycling
th	thermal
TOS	tall oil soap
TWh	terawatt hour
UCO	unconverted oil
ULCOS	Ultra Low CO_2 Steelmaking
VGO	vacuum gas oil
vol	volume
VPSA	vacuum-pressure swing adsorption
VSA	vacuum swing adsorption
WGS	water-gas shift
WHB	waste heat boiler

wt	weight