Evaluation of carbon capture technologies and integration into a pulp mill in Finland

Lappeenranta–Lahti University of Technology LUT

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ABSTRACT

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The thesis studied different options for integrating carbon capture into pulp mills. Several carbon capture technologies suitable for integration are under development, but commercial technologies still rely on chemical absorption with amines. However, in evaluating these technologies, it was found that amine-based capture is not the best option for integration according to the criteria examined in this work. The analysis identified molten carbonate fuel cells, the calcium looping process, potassium carbonate-based chemical absorption, and adsorption-based technologies as the most promising challengers to amine technology in the future.

The thesis compared HPC and MEA technologies integrated into a pulp mill located in Finland. The integration was assessed using the Aspen Plus modelling software. Integration was analysed for the recovery boiler and lime kiln, the lime kiln alone, and the recovery boiler alone. The results showed that the HPC process is more energy-efficient and cost-effective in all integration cases. Integration also has several impacts on the pulp mill. Changes would likely be needed in the pulp mill's steam network and cooling water circulation system. In addition, when integrated into the recovery boiler, the mill would become a net electricity consumer with both technologies. Also, If the capture technologies only consume steam, utilising all carbon capture potential in the pulp mill would require a new auxiliary boiler or significant savings in steam consumption.

TIIVISTELMÄ

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Hiilidioksidin talteenottoteknologioiden arviointi ja integrointi sellutehtaaseen Suomessa

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Avainsanat: CCS, CCU, Hiilidioksidin talteenotto, Sellutehdas

Diplomityössä tutkittiin eri vaihtoehtoja hiilidioksidin talteenoton integroinnille sellutehtaisiin. Tällä hetkellä kehitysvaiheessa integraatioon soveltuvia hiilidioksidin talteenottoteknologiota on useita, mutta kaupalliset teknologiat perustuvat edelleen kemialliseen absorptioon amiineilla. Kuitenkin teknologioiden arvioinnissa huomattiin, että amiineihin perustuva talteenotto ei ole työssä tarkastelluilla kriteereillä paras vaihtoehto integraatiolle. Analyysissä todettiin, että potentiaalisimmat amiiniteknologian haastajat ovat tulevaisuudessa sulakarbonaattipolttokennot, calcium looping -prosessi, kaliumkarbonaattiin perustuva kemiallinen absorptio ja adsorptioon perustuvat teknologiat.

Työssä vertailtiin HPC ja MEA teknologiaa integroituna Suomessa sijaitsevaan sellutehtaaseen Aspen Plus -mallinnusohjelmalla. Integrointikohteet sellutehtaassa olivat soodakattila ja meesauuni, pelkkä meesauuni ja pelkkä soodakattila. Työn tulokset näyttivät, että HPC prosessi on energiatehokkaampi ja kustannuksiltaan edullisempi vaihtoehto kaikissa integrointikohteissa. Integraatiolla on myös useita vaikutuksia sellutehtaaseen. Muutoksia joudutaan todennäköisesti tekemään sellutehtaan höyryverkkoon ja jäähdytysvesikiertoon. Soodakattilan integroinnin myötä tehdas muuttuu molemmilla teknologioilla sähköntuotannon osalta nettokuluttajaksi. Lisäksi jos talteenottoteknologiat kuluttavat pelkästään höyryä, koko hiilidioksidin talteenottopotentiaalin hyödyntäminen sellutehtaalla vaatii uuden apukattilan tai merkittäviä säästöjä höyryn kulutuksessa.

SYMBOLS AND ABBREVIATIONS

Roman characters

a	cost coefficient	
A	coefficient for equilibrium constant equation	
b	cost coefficient	
B	coefficient for equilibrium constant equation	
C	coefficient for equilibrium constant equation	
D	coefficient for equilibrium constant equation	
E	activation energy	cal/mol
I	investment cost	€, $, £
k	pre-exponential factor for reduced power-law equation	
K	annual cost	€
K_{eq}	equilibrium constant	
L	solvent loading	
n	scaling factor	
p	pressure	bar, Pa
q_m	mass flow rate	kg/s, t/a
R	gas constant	J/(kg K)
r	reaction rate	
S	capacity	t/h, t/a
T	temperature	°C, K

Subscripts

0	reference

b	boiler
CaCO3	calcium carbonate
cap	capital costs
CO2	carbon dioxide
e	electricity
fg	flue gas
fixed	fixed costs
LCO2	liquified carbon dioxide
O&M	operation and maintenance
primary	primary energy
variable	variable costs
water	process water

Abbreviations

ADt	air dry ton, bleached (90%) pulp
ASU	air separation unit
BECCS	bioenergy with carbon capture and storage
BECCU	bioenergy with carbon capture and utilisation
CAPEX	capital expenditures
CCS	carbon capture and storage
CCU	carbon capture and utilisation
CLOU	chemical looping with oxygen uncoupling
CPU	carbon dioxide purification and compression unit
DACCS	direct air carbon capture and storage

DCC	direct contact cooler
eq	equivalent
GHG	greenhouse gas
HPC	hot-potassium carbonate
LCOC	levelized cost of carbon capture
LK	lime kiln carbon capture case
LULUCF	land use, land use change and forestry
MCFC	molten carbonate fuel cells
MEA	monoethanolamine
NCG	non-condensable gases
OPEX	operational expenditures
PCC	precipitated calcium carbonate
ppm	parts-per-million
PSA	pressure swing adsorption
RB & LK	recovery boiler and lime kiln carbon capture case
RB	recovery boiler carbon capture case
TPSA	temperature pressure swing adsorption
TRL	technology readiness level
TRS	total reduced sulphur gases
TSA	temperature swing adsorption
TVSA	temperature vacuum swing adsorption
VCM	voluntary carbon market
VSA	vacuum swing adsorption
wt.%	mass fraction

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1 Introduction

Carbon dioxide, constituting over 80% of greenhouse gases, is a pressing issue contributing to global warming (Khan, 2021). The urgency of the climate crisis, one of the most significant international challenges, is evident in its impact on nature and people. Global warming is causing an increase in sea levels and the loss of ecosystems, requiring prompt action (Khan, 2021).

In the Paris Agreement on climate change, it has been internationally recognised that global warming should be limited to 1.5 ℃ compared to pre-industrial times to mitigate the adverse impacts of climate change (IPCC, 2023). However, according to the 6th IPCC (2023) climate report, the probability of reaching the target has diminished. Limiting it to 2 ℃ warming compared to pre-industrial times will also require significant reductions in greenhouse gas (GHG) emissions by 2100. Falling short on climate targets would mean permanent changes to the climate and ecosystems. The climate reports have highlighted technical carbon sequestration as one solution, which is assumed to play a significant role in keeping to the global carbon budget and enabling compliance with climate targets. The carbon budget means the maximum amount of net global anthropogenic carbon dioxide emissions, which can be cumulatively emitted to reach climate targets. The current carbon budget is approximately 500 Gt CO_2 with over 50% probability for the 1.5 °C target and 1150 Gt CO_2 with a 67% probability for the two-degree target. When more GHG emissions are released, the importance of using technical carbon sequestration to stay within the current carbon budget increases.

There are two main alternatives to technical carbon removals: bioenergy with carbon capture and storage (BECCS) and direct air carbon capture and storage (DACCS). In BECCS, carbon capture is done by capturing biogenic CO_2 and storing it in geological storage. In DACCS, carbon dioxide is captured from the air and stored in geological storage. The most cost-effective option is currently BECCS, which is why its research and role in technical carbon sequestration is significant (IPCC, 2023, p. 86).

Carbon capture has also been featured in Finland's national climate strategy. Finland aims to achieve carbon neutrality by 2035 and negativity by 2050. In Finland, carbon capture is expected to bring emission reductions of 9 Mt CO_2-eq by 2050 (Huttunen et al., 2022, p. 70).

The emission reductions are expected to come mainly from carbon capture from BECCS and steam reformers due to low costs compared to other capture options. However, the main problem with implementing this plan is the high cost of carbon capture technologies.

1.1 Background of the thesis

Background for this master's thesis is the increased need for carbon capture and its identified potential in kraft pulp mills. The implementation of the work has been influenced by the need for more detailed studies on carbon capture at kraft pulp mills, which focus on not only one technology. Due to their commercial availability, most prior research has focused on modelling chemical absorption with monoethanolamine (MEA) solvent in pulp mills. However, for example, a study by Kuparinen et al. (2019) mentioned that carbon capture technologies have yet to have a winning technology in cost and performance. More emphasis is also needed on clear criteria for integrating carbon capturing into a pulp mill.

1.2 Objectives of the thesis

The objective of this master's thesis is to determine the best carbon capture technology options for a kraft pulp mill, determine the main criteria influencing the choice of the technologies and highlight limiting factors for the integration. The objective is also to evaluate the applicability of the best technology option and the costs of integrating carbon capture technology into a pulp mill.

The master's thesis answers the following research questions from a kraft pulp mill’s perspective:

- What are the potential carbon capture technologies?
- What are the impacts when carbon capture is integrated?
- What are the costs of selected carbon capture technologies?

1.3 Scope of the thesis

The thesis is limited to existing kraft pulp mills in Finland, and only technologies that capture carbon dioxide from flue gas streams will be considered. Only the carbon capture process is considered, while further carbon dioxide utilisation is excluded from the study. Logistics and value chains outside a kraft pulp mill are not investigated. Only technologies with at least a technology readiness level (TRL) of 6 are considered.

The thesis briefly reviews carbon sequestration methods on a general level. After this, the pulp mill process is explained, and the primary sources of carbon dioxide in a pulp mill and their flue gas properties are assessed. The reasons for carbon capture in pulp mills and technologies that can be integrated into a pulp mill will be presented. The carbon capture process principles, benefits, disadvantages, the current TRL, the needed utilities and the cost of carbon capture are estimated based on the literature. After presenting different carbon capture technology options, the essential criteria from the pulp mill's point of view are presented, and based on the criteria, different carbon capture technologies are evaluated. The most promising technologies have been selected for further assessment, and their performance has been assessed using Aspen Plus process modelling software. Finally, the costs of carbon capture in an example kraft pulp mill are evaluated.

2 Carbon removals

Carbon removals are mechanisms to reduce carbon dioxide entering the atmosphere. One example is carbon dioxide sequestration. Carbon dioxide sequestration is needed to achieve climate goals, as reaching the current climate targets (1.5-degree target) requires an estimated 160-370 Gt CO_2 of negative carbon dioxide emissions by 2100 (IPCC, 2023, p. 87). There are two different options for carbon dioxide sequestration: natural or technical. This chapter introduces the basics of these two options.

2.1 Natural carbon removals

Natural carbon dioxide sequestration refers to the absorption of carbon dioxide from the atmosphere by vegetation, oceans, sediments, soil, and detritus related to nature's normal carbon cycle (Wigley & Schimel, 2000). Natural carbon removals cannot store all the carbon dioxide humans emit (IPCC, 2023). However, natural carbon sequestration is essential in decreasing the need for technical sequestration. Reforestation has been considered to have the best potential in sequestering carbon dioxide due to its low cost compared to technically sequestered carbon (IPCC, 2023, p. 4). Nevertheless, the scale of reforestation is limited.

Climate factors such as temperature, humidity, and solar radiation affect the carbon stored by land and vegetation (Wigley & Schimel, 2000). This means climate change can lead to changes in the future levels of naturally sequestered carbon, including in the ocean (NASA, 2004). In addition, changes in land use can impact sequestered carbon, and increasing land use changes threaten to reduce natural carbon sinks in the future (Wigley & Schimel, 2000).

2.2 Technological carbon removals

Natural carbon removals alone cannot cover all the necessary carbon sinks presented in the IPCC's 6th assessment report. The report estimates that most additional carbon removals must come from technical carbon removals to achieve climate goals. The possible reduction of natural carbon removals due to climate and land use changes also increases the importance

of utilising technical carbon removals. Due to the highest carbon removal potential, technical carbon removal usually refers to carbon capture and storage by DACCS or BECCS.

The basic principle in DACCS is capturing carbon dioxide directly from ambient air. DACCS has excellent potential for capturing carbon dioxide, which is mainly limited by the adequacy of geological storage (Fuss et al., 2018, p. 20). One of the challenges related to DACCS is that capturing carbon dioxide from the air requires significantly more energy than capturing it from concentrated point sources. The reason behind this is that carbon dioxide concentrations, for example, in the flue gases of power plants, are 100–300 times higher than the carbon dioxide concentrations in the air. Another challenge is the high cost of carbon capture. Fuss et al. (2018) mentioned that up-front expenses of sourcing supply chains and resolving infrastructure issues, combined with a lack of experience with the technology, resulted in high initial costs estimated to be 600–1000 €/t_{CO2} for current projects. When these challenges are overcome, according to Fuss et al. (2018), the cost of carbon capture at DACCS could fall from current costs to 100-300 €/t_{CO2}.

The second technological option for carbon dioxide sequestration is BECCS. The fundamental concept of BECCS is that carbon dioxide is first naturally absorbed into biomass. When biomass is combusted, the carbon of biomass forms carbon dioxide, which ends up in the atmosphere. In BECCS, carbon dioxide released in the combustion process is captured from flue gas streams and stored in geological storage. BECCS leads to decreased carbon dioxide from the natural cycle and, thus, from the atmosphere. The BECCS concept is shown in Figure 1.

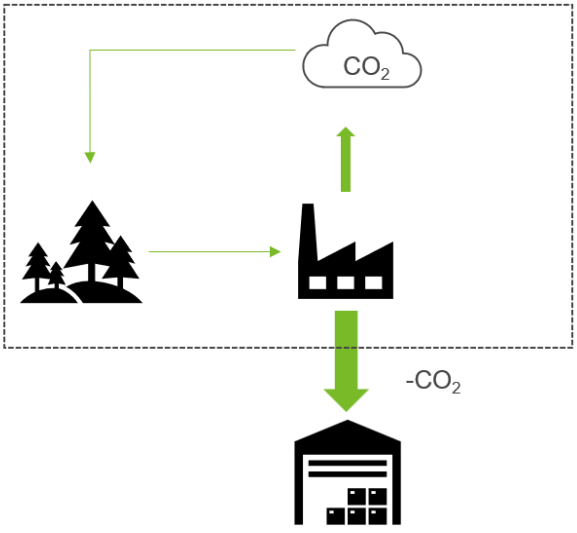

Figure 1. Concept scheme of BECCS.

From the perspective of carbon dioxide emissions, BECCS is an important carbon sequestration method. It has been estimated to have the potential to sequester 20 Gt_{CO2}/a globally, requiring several Mkm^2 of occupied land (Creutzig et al., 2021, p. 510-513). However, when applying BECCS, one has to consider the potential risks and impacts on biodiversity, livelihoods and intertemporal carbon balances. The impacts are mainly due to the previously mentioned high land use and changes in land use. Considering these risks, sustainable BECSS use is estimated at 0.5-5 Gt_{CO2}/a (Creutzig et al., 2021, p. 510-513).

Another reason to use BECCS is the lower cost compared to DACCS. Many studies have indicated costs under 200 €/t_{CO2} (Fuss et al., 2018, p. 12). In many studies, costs have been significantly lower depending on the carbon capture scale (Fuss et al., 2018, p. 12).

3 Kraft pulp mill

The kraft mill process is the most common way to produce pulp in Finland. Kraft pulp mills consist of fibre processing and chemical recovery. Figure 2 depicts a schematic process of a kraft pulp mill.

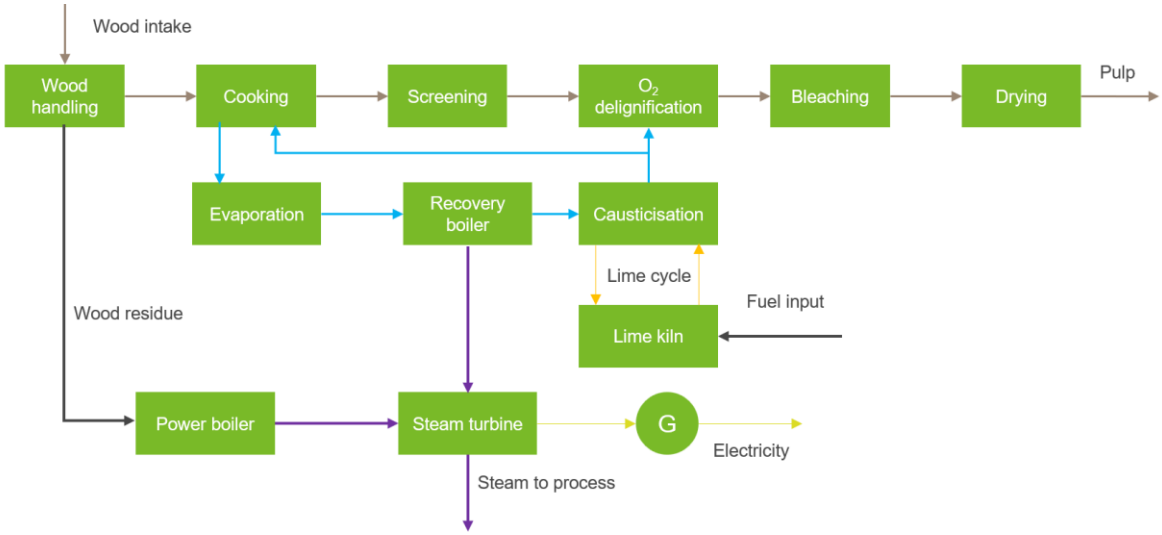

Figure 2. Schematic process of kraft pulp mill.

KnowPulp (2024) describes the fibre processing as follows. The fibre process is emphasised in Figure 2 with brown arrows. The pulping process begins with wood handling, where the wood is received, possibly de-iced, debarked, chipped into as homogeneous wood chips as possible, screened and stored. The wood chips are transported via conveyors to pulp cooking, where the chemicals and heat are used to remove lignin, which binds wood fibres. After the pulp is cooked, the brown stock pulp is washed. The purpose of brown stock washing is to separate dissolved substances from the pulp and recover organic and inorganic matter. After washing, the pulp is screened, and the delignification is continued with oxygen in the oxygen delignification stage. Following the oxygen delignification, the pulp is usually bleached in several stages before being dried, sheeted and baled in the drying machine.

Chemical recovery's primary role is regenerating cooking chemicals and generating steam. The chemical recovery process is emphasised in Figure 2 with blue arrows. KnowPulp (2024) describes the recovery line process as follows. The recovery line starts when weak

black liquor comes from the pulp-washing process to the evaporation plant. The evaporation plant's primary role is to evaporate the water from the weak black liquor so it can be combusted in a recovery boiler. The dry solid concentration is only 15-16% when the black liquor comes to evaporation. During evaporation, the goal is to increase the dry solid concentration of black liquor in modern pulp mills by up to 80-85%, and the absolute minimum is 58% to avoid smelt explosions. In addition to removing water, different by-products, such as methanol, turpentine, and lignin, can be separated.

After evaporation, concentrated black liquor is combusted in a recovery boiler, forming an inorganic smelt and generating steam. Steam is utilised in various pulp mill processes, such as cooking, evaporation, and electricity production. Usually, kraft pulp mills produce more electricity than they consume.

After combustion, the smelt is dissolved into weak ash, forming green liquor. Green liquor is fed to the causticising plant. At the causticising plant, green liquor is transformed back into white liquor through reactions with lime. Also, before white liquor is fed back into the cooking process, calcium carbonate has to be separated from white liquor. The separated calcium carbonate is combusted in a lime kiln, producing lime for causticising. As a result of the chemical cycle, all the chemicals at the kraft pulp mills are recycled with small losses.

3.1 Carbon dioxide sources in kraft pulp mills

Carbon capture at the kraft pulp mill can be integrated into multiple sources. These sources in kraft pulp mills include recovery boilers, lime kilns, and auxiliary boilers. Carbon dioxide is formed mostly by the carbon-oxidation process, which has the following equation.

$$C + O_2 \rightarrow CO_2 \quad (1)$$

3.1.1 Recovery boiler

Recovery boiler is the kraft pulp mill's primary source of carbon dioxide emissions, making it a primary source for carbon capture. Black liquor combustion forms biogenic CO_2 as a one-flue gas product, excluding boiler start-ups, which usually use fossil fuel, natural gas, and heavy fuel oil (Vakkilainen, 2005).

The amount of CO_2 and flue gas compositions from recovery boilers vary based on the properties of the combusted black liquor. The specific details of the mill also affect the amount of CO_2 formed. Due to different yields, the carbon content in black liquor varies slightly by the wood species used in a kraft pulp mill, which slightly impacts the formation of CO_2. According to Gullichsen & Fogelholm (1999), the carbon content of Scandinavian hardwood (birch) is around 31-35 wt.%, while the softwood (pine) carbon content is around 32-37 wt.%. If lignin extraction is utilised in a kraft pulp mill, the carbon content of black liquor will decrease depending on the lignin extraction percentage. The extraction amount can be 10-20% of lignin without significantly impacting chemical recovery (Jardim et al., 2022, p. 2). The extraction of 10-20% of lignin would mean a 1-2%-unit reduction in the carbon concentrations of black liquor (Vakkilainen & Välimäki, 2009).

3.1.2 Lime kiln

The lime kiln converts lime mud ($CaCO_3$) to burnt lime (CaO) by drying and heating the lime mud, with CO_2 formed as a by-product. Burnt lime is then used to produce white liquor for the pulp mill's cooking process. Lime mud is converted to burnt lime according to the following equation.

$$CaCO_3 + \text{heat} \rightarrow CaO + CO_2 \quad (2)$$

To produce CaO, a temperature of at least 850 ℃ is needed to start the calcination process. In industry, temperatures typically vary between 850-1150 ℃ (Manning & Hart, 2021, p. 38). The calcination process has typical efficiency ranging from 80-85% (Manning & Hart, 2021, p. 38). Commonly used fuels include natural gas and oil, while syngas derived from bark, tall oil pitch, and pulverised biofuels are also utilised in pulp mills in Finland (Heikkilä, 2023).

In a lime kiln, the share of biogenic CO_2 depends significantly on the fuel used and the amount of CO_2 released during the reaction. When using fossil fuels, the biogenic source of carbon dioxide is only the carbon dioxide released in the causticisation reaction. In contrast, the carbon dioxide released when combustion of renewable fuels, such as biomass gasification product gas, tall oil pitch, or pulverised biofuels, is also biogenic. Typically, the amount of CO_2 formed ranges between 100-250 kg_{CO2}/ADt (Kuparinen et al., 2019).

3.1.3 Auxiliary boilers

In KnowPulp (2024), it was mentioned that many kraft pulp mills in Finland also use auxiliary boilers to produce steam if the recovery boiler cannot generate all the steam required for all interconnected processes, especially at large integrated pulp mills or during start-ups and shutdowns. Also, it is common to produce heat for combined heat and electricity production.

KnowPulp (2024) mentioned that auxiliary boilers can use different fuels, such as biomass or fossil fuels. The most common boiler type used at pulp mills in Finland is a bubbling fluidised boiler that burns high-moisture-content fuel. High-moisture-content fuel can be wood residues from wood handling, bark or other solid fuels from the pulp mill process. Auxiliary boilers primarily use bark as their fuel source, producing typically biogenic CO_2 emissions. However, auxiliary boilers often use fossil fuels during start-ups and shutdowns. The CO_2 amounts depend on the fuel properties.

3.2 Flue gas properties in kraft pulp mill

Carbon dioxide concentrations and flue gas properties impact carbon capture technology selection. Table 1 presents typical flue gas concentrations at kraft pulp mills in different point sources.

Table 1. Wet flue gas concentration in point sources at a kraft pulp mill (Onarheim et al., 2017a).

Point source	CO_2 [mol-%]	H_2O [mol-%]	O_2 [mol-%]	N_2 [mol-%]	Other [mol-%]
Recovery boiler	13.0	17.0	2.3	67.6	<0.1
Lime kiln (Natural gas)	20.4	30.9	1.2	47.4	<0.1
Bark boiler	12.1	32.7	1.7	53.4	<0.1

Table 1 shows the typical concentration of CO_2 in flue gases when undergoing combustion in the presence of air. The share of biogenic CO_2 in lime kilns is around 66% when natural gas is used as a fuel (Kuparinen et al., 2019). Other flue gas compounds comprise 0.1% of flue gases. Other flue gas properties that have to be considered when carbon capture

technology is selected are the amounts of acid components, mainly NO_x, SO_x, and total reduced sulphur gases (TRS), as well as particles in flue gases. Knowing the properties is important because these components can impact the carbon capture performance negatively, for example, by causing degradation, leading to the need to remove these components (Teir et al., 2011). Typical NO_x, SO_x, TRS and dust concentrations in different carbon dioxide sources are presented in Table 2.

Table 2. NO_x, SO_x, TRS and dust emission in recovery boiler, lime kiln and auxiliary boiler (Roudier et al., 2015).

Point source	NO_x [mg/Nm^3]	SO_x [mg/Nm^3]	TRS [mg/Nm^3]	Dust [mg/Nm^3]
Recovery boiler	120-250	1-100	0-50	5-190
Lime kiln gas firing	200-350	1-20	0-20	5-25
Lime kiln oil firing	100-200	5-50	0-20	5-25
Lime kiln with oil and NCG	100-350	100	0-20	5-25
Auxiliary boiler (biomass)	80	1-20	0	0.5-10
Auxiliary boiler (mixed fuel, e.g. bark, coal, fuel oil, sludge and gas)	150-300	6-75	0	0.5-10

Table 2 shows that the acid components and dust emissions from kraft pulp mills differ based on the type of boiler, combustion technique, and fuel utilised. It is important to consider the impact of the combustion of non-condensable gases (NCG), as it can substantially increase NO_x and SO_x levels. These emissions can be particularly high during start-ups and shutdowns (Roudier et al., 2015). However, compared to coal combustion plants equipped with carbon capture technologies, pulp mills generally have lower sulphur emissions and similar levels of dust and nitrogen oxide emissions (Neuwahl et al., 2018). This indicates that these emissions from pulp mills are unlikely to be a limiting factor for carbon capture technology integration.

If, however, it is necessary to control these impurity levels, there are multiple ways to do so. Roudier et al. (2015) mentioned several techniques. An electrostatic precipitator or fabric filters typically remove particles such as ash and dust. If these methods are not sufficient, an additional scrubber can be installed. A flue gas scrubber can also eliminate SO_x and NO_x emissions from flue gas streams. Removing the SO_x and NO_x compounds in a flue gas

scrubber also allows for heat recovery from the flue gases. The secondary heat can be utilised for pulp mill operations or district heating production.

Roudier et al. (2015) described also other techniques. To some extent, NO_x and SO_x contents in the flue gas can also be reduced by optimising the combustion processor or utilising secondary methods to reduce NO_x emissions, primarily in larger auxiliary boilers. Secondary methods involve using urea or ammonia injections to flue gas flows, with or without a catalyst. These techniques are selective non-catalytic reduction and selective catalytic reduction. Additionally, SO_x emissions can be decreased by lime or dolomite injections in larger steam boilers in addition to scrubbing.

An important factor for carbon capture is the exit temperature of flue gas because, in many carbon capture technologies, flue gases must be pre-cooled before entering the carbon capture unit (Kvamsdal et al., 2011, p. 1552). Cooling can be done with the flue gas scrubbers and additional pre-coolers. The flue gas temperatures in boiler outlets are typically around 180 °C due to the sulfuric acid dew point. In the case of lime kilns, the flue gas exit temperatures usually range from 200 to 350 °C (Tran, 2007, p. 7).

4 Reasons to implement carbon capture in Finnish kraft pulp mills

The forest industry has the highest potential for biogenic carbon capture in Finland (Official Statistics of Finland, 2024a). The sector's bio-based carbon dioxide emissions are approximately 20 Mt CO_2 per year, and the most significant carbon capture potential is in Finnish kraft pulp mills (Official Statistics of Finland, 2024a). The target for implementing carbon capture in a kraft pulp mill is BECCS, bioenergy with carbon capture and utilisation (BECCU), or a combination of both. Several factors, including environmental and economic aspects, influence the drivers of carbon capture implementation in pulp mills.

4.1 Environmental aspects

Environmental aspects regarding GHG emissions are one of the biggest drivers for implementing BECCS or BECCU. Figure 3 shows the development of Finland's GHG emissions, including land use, land use change and forestry (LULUCF) emissions, since 1990.

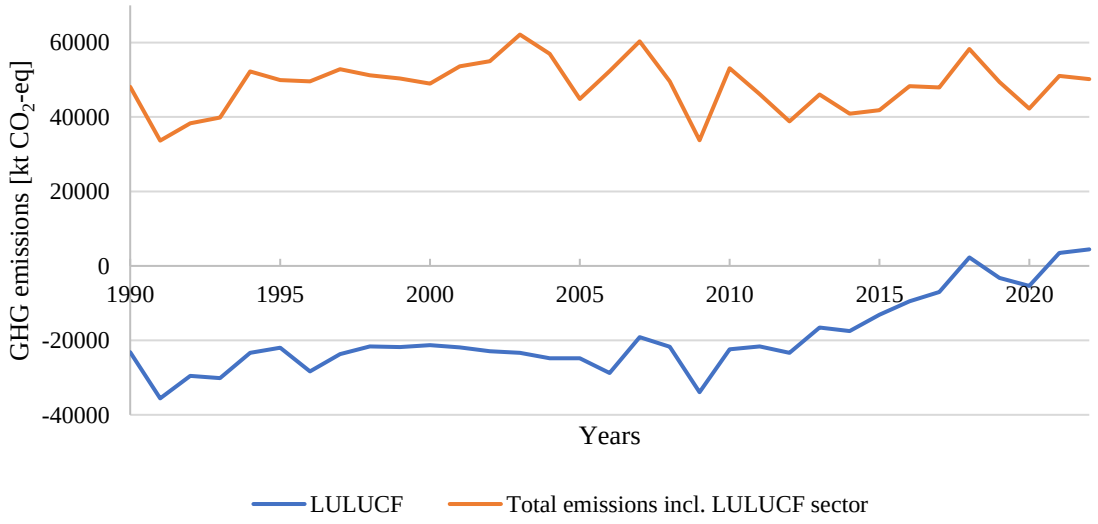

Figure 3. Total and LULUCF emissions (Official Statistics of Finland, 2024b).

Finland's current GHG emissions are about 51 Mt CO_2-eq, considering the LULUCF sector. Figure 3 shows that significant emission reductions are needed to achieve Finland's emission

targets and to achieve carbon negativity by 2050. One method for mitigating GHG emissions involves the implementation of BECCU. In this method, emission reductions can be achieved by replacing fossil-based production by utilising carbon dioxide that is part of the natural cycle, such as biogenic CO_2 from pulp mills.

Figure 3 also indicates that, in recent years, the LULUCF sector has not provided carbon sinks in Finland. If the LULUCF sector does not achieve negative CO_2 emissions, reductions in fossil-based CO_2 emissions alone will not meet carbon negativity targets, requiring technical carbon removals. In Finland, there is significant potential for BECCS, specifically at pulp mills, due to the substantial availability of biogenic carbon dioxide. Also, pulp mills have large point sources, enabling large-scale carbon capture (>1 Mt CO_2 annually), which can lead to economical CCS.

Other environmental reasons for utilising carbon capture at existing pulp mills are neutral environmental impacts on land use (Kuparinen et al., 2019, p. 1227). Despite the neutral effects of land use, carbon capture in existing mills has an impact, in many cases, on increasing water consumption and energy use. Depending on the technologies being utilised, it might also have other harmful emissions and waste streams.

4.2 Economic aspects

The cost of capturing CO_2 must be lower than the market or incentive price of carbon dioxide to be realised. If CCS/U is economical, it could generate significant revenue, especially in the context of Finnish pulp mills, which have a significant potential for carbon capture. In the case of pulp mills, where biogenic emissions contribute the largest share of emissions, profitability can be achieved by storing or utilising captured biogenic CO_2.

For CCS, participating in the voluntary carbon market (VCM) is currently the only option for monetising biogenic CO_2 in Finland since the compliance carbon market, the European Union emissions trading system, does not yet offer incentives to capture and store biogenic CO_2. In VCM, carbon dioxide equivalents are traded the same way as in compulsory markets, but the difference is that compulsory markets are designed to achieve mandatory emission reductions (Leonard, 2009).

The second way to monetise carbon capture of biogenic CO_2 is through carbon capture and utilisation (CCU). The main concept of BECCU is to generate more valuable products from biogenic CO_2 and simultaneously reduce the use of fossil-based products. This can lead to significant market benefits because demand for replacing fossil-based products is high, around 640 Mt/y (Kähler et al., 2023). In the context of pulp mills, biogenic CO_2 is transported to processes outside the kraft pulp mill and either further processed into more valuable products or used directly. The utilisation of CO_2 leads to additional income from captured biogenic CO_2 and may enable profitable carbon capture. However, compared to BECCS, the amount of utilised CO_2 in BECCU is typically smaller (about 10-500 kt_{CO2}/a per application) (IEA, 2019; Kircher & Schwarz, 2023). The current challenge in BECCU is that fossil-based products are usually more competitive than CCU-based products, which is challenging for extensive commercialisation (Ahlström et al., 2023). Another challenge for commercialisation is the existing regulation that does not differentiate between fossil and biogenic-based CO_2 sources, meaning that, for example, CCU fuels from fossil or biogenic sources are treated equally (Koytsoumpa et al. 2021, p. 4).

4.3 Utilisation of captured CO_2 in pulp mill processes

Depending on the pulp mill's specific details, small amounts of captured carbon dioxide can be utilised in its processes. This utilisation can result in reduced chemical and utility needs and potential small cost savings in pulp mills. These possibilities include mainly using CO_2 in lignin extraction, tall oil manufacturing, precipitated calcium carbonate (PCC) production, and other minor processes (Kuparinen et al., 2019, p. 1223).

Kuparinen et al. (2019) suggested that separating lignin can be a way to utilise captured CO_2. Lignin separation can reduce the boiler's heat load and increase pulp production. The separated lignin can be sold, refined, or used as a fuel alternative. Lignin is separated in the acidifying process of black liquor, partly done by CO_2, which requires around 150-250 kg_{CO2}/t lignin.

Kuparinen et al. (2019) stated that carbon dioxide can be utilised in the acidulation process of tall oil manufacturing. It reduces the demand for sulfuric acid and can replace around 50%. The consumption of CO_2 is around 4-6 kg/ADt. Tall oil production varies depending

on the wood quality, with Nordic pine producing around 45 kg/ADt, Nordic spruce 25 kg/ADt, and Nordic birch less than 20 kg/ADt.

Kuparinen et al. (2019) also proposed that PCC production can be used at mills integrated with a paper machine. A paper machine in an integrated mill consumes calcium carbonate to smoothen the paper surface. PCC is made by blowing lime kiln flue gasses through green liquor. The demand for PCC is usually fixed and corresponds to a local market. PCC production levels of 33 kg_{CaCO3}/ADt correspond to around 20 kg_{CO2}/ADt consumption.

Carbon dioxide can reduce water use and chemical consumption in pulp mill processes such as brown stock washes, bleaching, biological treatment, and pulp drying. For instance, carbon dioxide can reduce washing water usage with 2 to 4.5 kg_{CO2}/ADt doses in brown stock washes (Bokström & Kontola, 1995). Additionally, in the bleaching process, CO_2 can minimise the consumption of sulfuric acid and bleaching chemicals with a reported consumption of 2-4 kg_{CO2}/ADt (Jokinen et al., 2001). Furthermore, in pulp drying, carbon dioxide positively affects dewatering with a dose of approximately 2 kg_{CO2}/ADt (Jansson & Steel, 2009). Rigaud (2001) mentioned that carbon dioxide could be added to the wastewater neutralisation phase to reduce the use of foaming chemicals, polymers, and precipitants with a recommended dose of 75 kg_{CO2}/tm^3.

5 Carbon capture methods and technologies

Four carbon capture methods can technically be used for pulp mill point sources: pre-combustion carbon capture, post-combustion carbon capture, chemical looping combustion and oxy-combustion carbon capture (Zhao et al., 2023). Inherent carbon capture is not possible in a kraft pulp mill due to the low CO_2 concentration of flue gas.

Pre-combustion carbon capture refers to removing CO_2 before fuel is combusted. In some existing pulp mills in Finland, pre-combustion carbon capture can technically be integrated into lime kilns that utilise bark gasification. However, this is ineffective since CO_2 released during causticisation cannot be captured (Greco-Coppi et al., 2021). Due to these reasons, pre-combustion carbon capture is not considered in the thesis.

5.1 Post-combustion carbon capture

Post-combustion carbon capture involves removing CO_2 from the flue gas stream after combustion. Post-combustion carbon capture can be executed in various ways, each with unique benefits and challenges. The current technologies studied for post-combustion carbon capture include absorption, adsorption, membrane separation, fuel cell and electrochemical separation, calcium looping and cryogenic carbon capture (Dziejarski et al., 2023). However, it is important to note that cryogenic carbon capture is not considered suitable for pulp mill processes due to the low CO_2 concentration in flue gas streams of point sources. Cryogenic carbon capture becomes uneconomical when CO_2 concentrations are below 50% (Dziejarski et al., 2023).

5.1.1 Absorption

Rackley (2017) described absorption as follows: CO_2 can be separated from a gas mixture through physical or chemical absorption. In the absorption process, absorbents, known as solvents, are used to remove CO_2 from flue gas. In chemical absorption, a reactive solvent with selectivity towards CO_2 is used. CO_2 dissolves in the solvent due to its weak chemical bonding with the solvent's functional group. On the other hand, physical absorption involves

the absorbent attaching to CO_2 without a chemical reaction. Using chemical or physical absorption depends on CO_2 partial pressure in flue gas. The solvent loading in both absorption processes, as a function of CO_2 partial pressure, is presented in Figure 4.

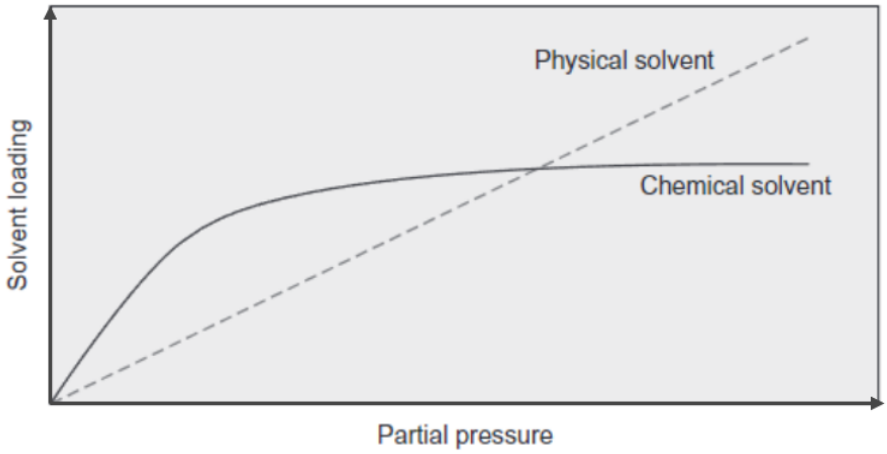

Figure 4. Solvent loading in the function of partial pressure (Rackley, 2017, p. 115-131).

Figure 4 illustrates that physical absorption is generally more effective at high CO_2 partial pressures in flue gas. Specifically, physical absorption is more effective when the partial pressure of CO_2 exceeds 10 bar (Teir et al., 2011). Chemical absorption is more suitable for pulp mill post-combustion where CO_2 partial pressures in the flue gas are low (10-20 kPa). The concept principle of chemical absorption is presented in Figure 5.

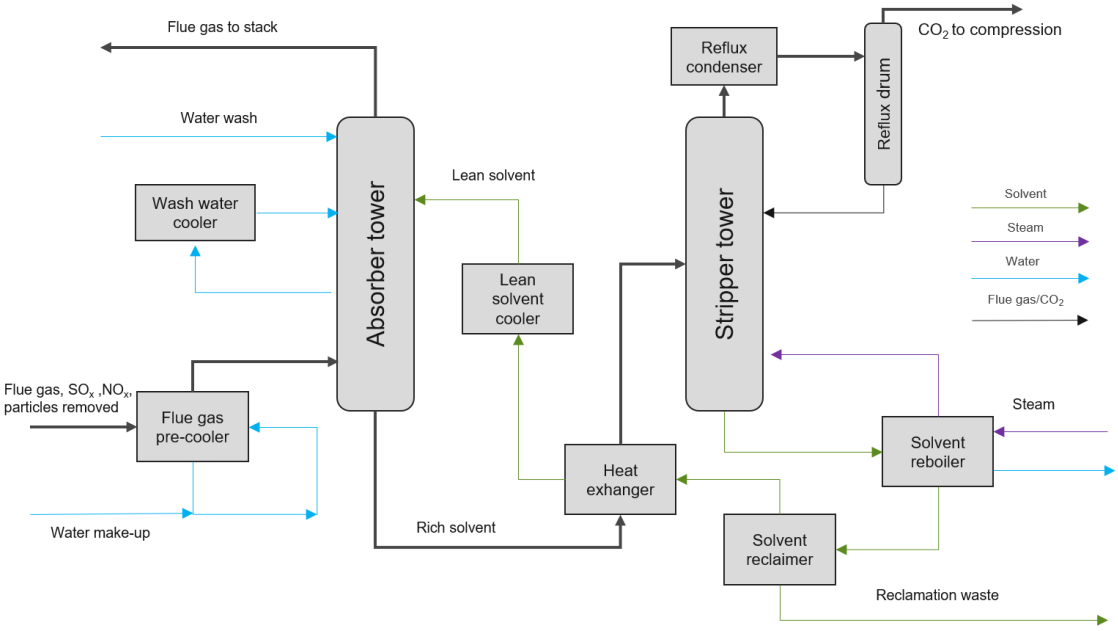

Figure 5. Chemical absorption process example based on amines (Rackley, 2017).

Rackley (2017) described the chemical absorption in detail in the following way. Before entering the absorber tower, flue gases and unwanted impurities, such as flue gas acid compounds and particles, are usually removed. Cleaned flue gas usually needs to be cooled to an operation temperature, for example, by a direct water contact cooler, which also helps to clean fine particulate and acidic matter from the flue gas stream. A lean solvent comes into contact with cooled flue gas in the absorber column. If using solvents that release emissions such as amines or ammonia, flue gas exiting from the top of the absorber is washed with water to reduce the entrainment and carryover of solvent droplets and vapour, and it is then vented to stack. CO_2-rich solvent exits from the bottom of the absorber and is pumped to the stripper tower. Heat for regeneration releasing CO_2 is supplied by a reboiler, which is integrated with the steam cycle of the steam source. Released CO_2 exits the top of the stripping tower after the water vapour is condensed from the CO_2 product stream. Finally, the product CO_2 stream is sent for further refining. Lean solvent from the bottom of the stripper tower passes the reclaim unit, where degradation products are settled out and are then fed into the absorber via cooling units.

Multiple solvents can be used in chemical absorption. Solvents have different properties and significantly impact the carbon capture process. Appendix 1 introduces these solvents and their advantages, disadvantages, and TRL in more detail.

Dziejarski et al. (2023) highlighted the advantages of chemical absorption, including mature technology with commercial technology options, retrofit easiness, simplicity, high absorption capacities, and easy attainment of product purity of more than 99%. Chemical absorption has been proven on a large scale and is suitable for flue gas volumes of recovery boilers. Multiple technology providers are available (Rezaei et al., 2023). Additionally, some solvents allow regeneration at low temperatures, making it possible to use only electricity, which can benefit pulp mills with limited steam availability.

Dziejarski et al. (2023) listed several drawbacks of chemical absorption, including high energy requirements for solvent regeneration, toxicity, and solvent losses of mature amine-based solvents. They also mentioned the large equipment size needed, possible negative environmental impacts, and the high cost of carbon capture.

5.1.2 Adsorption

Rackley (2017) described carbon capture through the adsorption process in the following way. In chemical or physical adsorption, carbon dioxide is attached to the surface of the solid adsorbent. In physical adsorption, the process is based on weak Van der Waals forces. In chemical adsorption, the process is based on covalent bonding. The process usually consists of two: adsorption and desorption beds. Regeneration typically occurs through temperature or pressure variation. The process principles of most typical adsorption processes are presented in Figure 6.

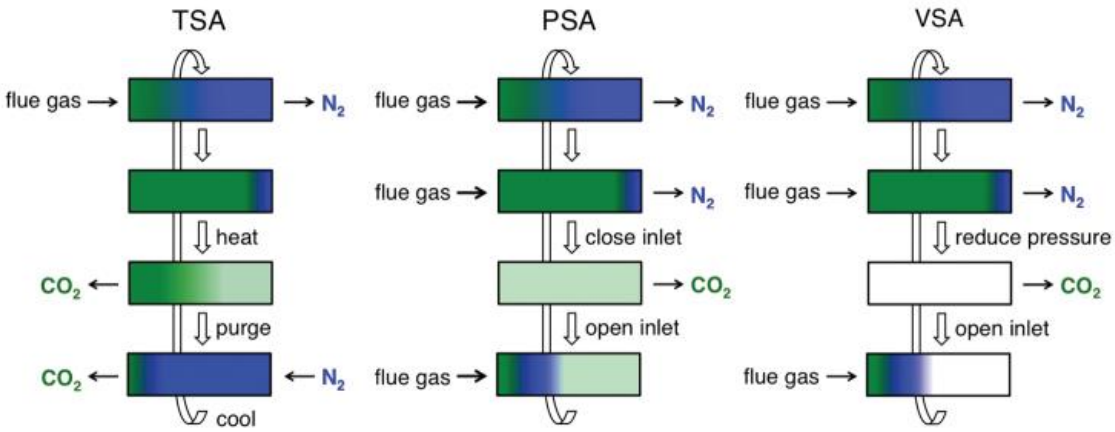

Figure 6. Concept of adsorption process (Sumida et al., 2012).

Clausse et al. (2011) stated that adsorption-based carbon capture offers three promising technologies for industrial-scale carbon capture. These technology options are pressure swing adsorption (PSA), vacuum swing adsorption (VSA) and temperature swing adsorption (TSA). In PSA, adsorption is done at higher than atmospheric pressure, and desorption is done at atmospheric pressure. VSA adsorbs CO_2 at atmospheric or higher pressure, and desorption occurs at lower than atmospheric pressure assisted by vacuum pumps. In TSA, adsorption is done in lower temperatures and desorption in higher temperatures by heating the bed with hot gas or steam.

In single-stage PSA or VSA processes, only low carbon capture rates and purities are achieved. Multiple stages and adsorption beds are required to achieve high purity of CO_2 and high carbon capture rates (Clausse et al., 2011, p. 1206-1207). On the other hand, while

a TSA process can achieve higher carbon capture rates and CO_2 purity levels in a single step, it requires more energy due to higher regeneration times from the heating and cooling of large masses of sorbent material, limiting effectiveness and increasing costs (IEAGHG, 2019). TSA regeneration times can decrease by combining temperature and pressure or vacuum swing processes, leading to lower heat demand (Liu et al., 2023). These processes are known as temperature pressure swing adsorption (TPSA) or temperature vacuum swing adsorption (TVSA), depending on whether the cycle is pressure or vacuum-assisted.

Different adsorbents can be used in these adsorption processes. The choice of adsorbent depends on characteristics such as selectivity, adsorption capacity, ease of desorption, and energy required. Appendix 2 details the most promising chemical and physical adsorbents and their advantages and disadvantages.

Adsorption-based carbon capture offers several advantages, including generating no liquid waste, being usable at multiple temperatures, requiring low energy for regeneration, and avoiding corrosion issues (Vaz et al., 2022, p. 5). Various adsorbent materials are available, allowing for high CO_2 uptake and separation efficiencies of over 90% with 95% purity at low capture costs (Dziejarski et al., 2023, p. 22-24).

Dziejarski et al. (2023) listed several challenges in adsorption processes. These include low TRL at the demonstration phase (TRL 5-7), CO_2 selectivity, scalability, and flue gas moisture and impurities sensitivity. Furthermore, there are concerns about the durability of adsorbents and the uncertainty of how adsorption works under real flue gases on a large scale.

5.1.3 Membrane separation

Rackley (2017) described membrane separation in the following way: the principle of membrane separation is to filter specific components from the flue gas stream. Depending on the membrane design and materials, the separation process can involve a combination of chemical and physical phenomena. There are two main types of membranes: gas separation membranes and membrane contactors. The process principles of these membrane types are presented in Figure 7.

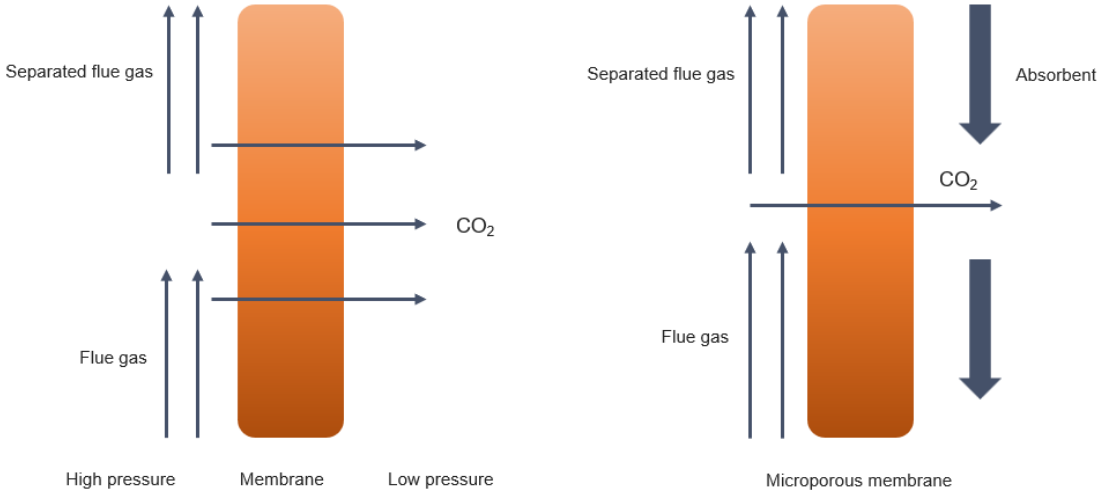

Figure 7. Principle scheme of carbon dioxide separation from flue gases with gas separation membrane (left) and membrane contactor (right) (Dziejarski et al., 2023, p. 18).

Dziejarski (2023) described membrane separation in different membrane types as follows. In the gas separation membrane, flue gas enters the module at high pressure, and carbon dioxide exits downstream towards lower pressure. The gas transport mechanism is based on dissolution and diffusion. Flue gas components undergo separation based on variances in their solubility within the membrane material and variations in the permeation rates. In membrane contactors, CO_2 is transported through a microporous solid membrane to absorption liquid, which captures CO_2. The separation depends on the membrane's pore size and the absorbing liquid. Before separation, impurities in the flue gas, such as SO_x and NO_x, must be removed from the flue gas because they can adversely impact performance and selectivity (Hekmatmehr et al., 2024, p. 22). Several different membrane types have been developed for post-combustion. These are, for example, Thin-film composite membranes, fixed-site carriers that are gas separation membranes and gas-liquid membrane contactors (Hou et al., 2023). Appendix 3 presents the advantages, disadvantages and TRL of these membrane types.

Membranes offer several advantages. They are easy to control, easily scaled up due to their modular nature, and have no moving parts (Bazhenov & Lyubimova, 2016, p. 892). Gas separation membranes do not require additional chemicals or produce waste streams and have low energy consumption (Dziejarski et al., 2023).

Disadvantages in membranes are related to the low-pressure difference between the feed and the permeate side (Nakao et al., 2019, p. 65). Low-pressure difference necessitates a large membrane area, which in turn increases the costs of carbon capture. Due to low-pressure differences, permeability is commonly preferred in gas separation membranes, leading to lower capture rates (60-80%) than in other carbon capture applications (Rezaei et al., 2023). Dziejarski et al. (2023) have identified additional challenges associated with scaling up membrane systems for large-scale deployment. These challenges include low resistance to flue gas impurities and water vapour, low TRL at the early demonstration phase (TRL 5-7), limitations in achieving higher permeability and selectivity for large-scale deployment, and difficulties maintaining long-term performance. Additionally, there is a need for multiple stages and recycle streams to achieve high CO_2 purity, which can negatively impact the economics of large-scale operations. Furthermore, these systems require mechanical and thermal stability at elevated temperatures and pressures (Hekmatmehr et al., 2024, p. 23).

5.1.4 Electrochemical separation

Carbon capture can be done utilising electrochemical potential differences in fuel cells. According to Slater et al. (2019), fuel cells are devices that directly convert the chemical energy of a gaseous fuel such as natural gas, biogas, hydrogen or syngas into electricity and heat. The conversion is achieved through an electrochemical oxidation process. Fuel cells are generally categorised based on the electrolyte's material and operating temperature. The most promising electrochemical technology for post-combustion carbon capture is high-temperature molten carbonate fuel cells (MCFC). MCFC has high electrical efficiency, scaling from hundreds of kW to a few MW sizes, and heat and electricity co-generation ability. The process principle of MCFC is presented in Figure 8.

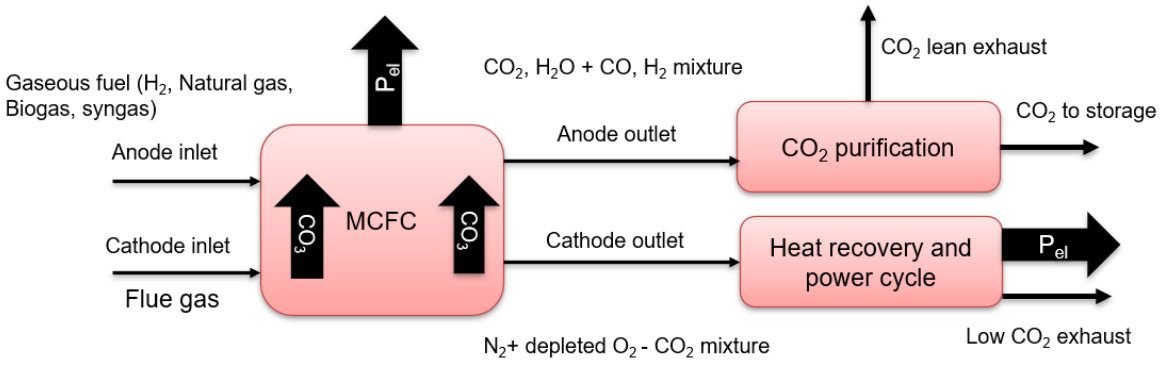

Figure 8. MCFC process principle (Spinelli et al., 2018).

In MCFC, the anode and cathode are commonly made of nickel. The electrolyte is harmless molten salts of lithium, potassium, and sodium carbonates suspended in a porous ceramic matrix. Fuel cell operates at high operating temperatures of 550-650 °C to create optimal conditions for reforming and shift reactions (Slater et al., 2019, p. 3). These reactions first convert hydrocarbon species and water into hydrogen, then convert water and carbon monoxide into additional hydrogen and carbon dioxide. The oxygen required for the final conversion of hydrogen is transported across the electrolyte in the form of CO_3 ions, generating an electric current.

MCFC offers several advantages. The process is highly efficient and has a low impact on net electricity production because of increased power capacity and low energy consumption (Slater et al., 2019). Another advantage is that MCFC is modular and can be scaled for different point sources (GCI, 2023). MCFC is flexible compared to the thermochemical carbon capture process due to the fast and easy control of electrical potential. Another major advantage is that it does not produce emissions, has low costs, and can achieve high capture rates (IEAGHG 2019).

MCFC also has several disadvantages, including excess gaseous fuel need, which has limited availability in some pulp mills in Finland since all pulp mills are not connected to the gaseous fuel network. In addition, high operating temperatures and a separate purification step are needed because anode exhaust has only around 70% carbon dioxide concentration (IEAGHG, 2019). Additionally, disadvantages include sensitivity to flue gas impurities, short fuel cell stack lifetimes, and currently high fuel cell prices (Slater et al., 2019, p. 14).

Another major drawback is the low technical readiness level (TRL 4-6), meaning that technology is still at the development and early demonstration level (IEAGHG, 2019).

5.1.5 Calcium looping

The calcium looping process consists of two fluidised bed reactors where carbon capture reactions occur, typically accompanied by a secondary steam cycle that generates additional power. The process principle of the calcium looping is presented in Figure 9.

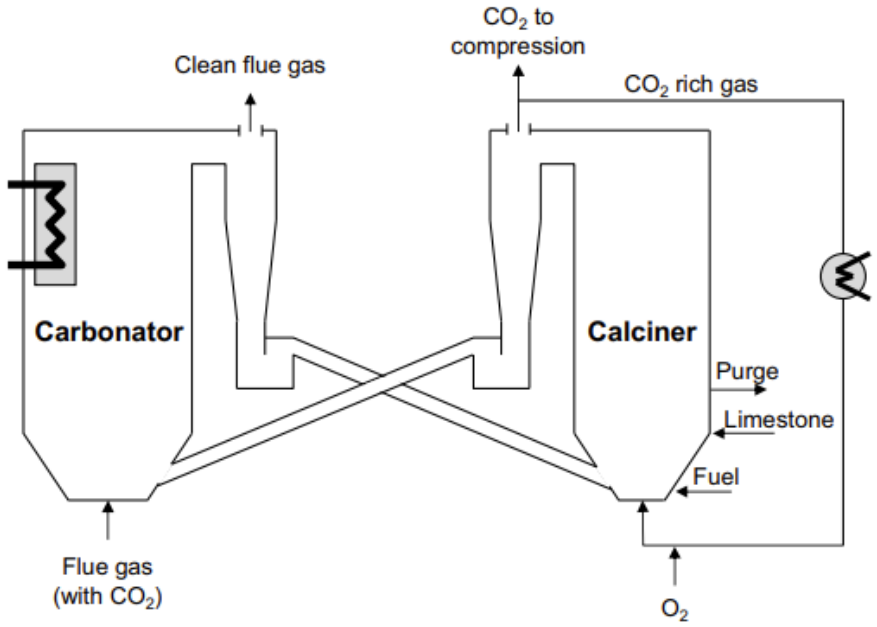

Figure 9. Post-combustion carbon capture with calcium looping process (Martínez et al., 2016, p. 273).

Martínez et al. (2016) mentioned that in the process, flue gases are in contact with a large flow of calcium oxide (CaO), also known as burnt lime, in the carbonator fluidised bed reactor at a temperature of around 650 °C. Carbon dioxide reacts with CaO in the reactor, forming calcium carbonate ($CaCO_3$) according to the following equation.

$$CaO + CO_2 \rightarrow CaCO_3 + \text{heat} \quad (3)$$

The reaction releases energy, which can be recovered to increase process energy efficiency. Heat recovered from the process can be used for electricity production in the secondary steam cycle. Solid carbonates formed in the reaction are sent to a calciner where calcium

oxide regeneration and carbon dioxide release occur at a temperature of 900-950 °C according to the calcination reaction (equation 2). The required heat for regeneration is produced by the combusting additional fuel under oxy-fuel conditions. Oxy-fuel conditions are used to avoid diluting the CO_2 stream, and these conditions require an air separation unit (ASU) that separates nitrogen from the oxidiser stream.

Ylätalo (2013) mentioned several advantages of calcium looping over traditional capturing technologies. It utilises well-known technologies such as oxy-combustion and limestone utilisation in fluidised beds, which can help with the industrialisation of processes. Also, the method utilises abundant and cheap resources, which can be obtained from lime kilns at kraft pulp mills, reducing the cost of carbon capture. Calcium looping retrofit can also reduce sulphur emissions from flue gas sources by more than 95%. A separate desulphuring unit may not be needed because the occurrence of CaO particle sulphation within the carbonator does not interfere with the formation of $CaCO_3$. High carbon capture rates (90%) can be achieved. The technology offers the potential for additional power generation through an energy recovery system.

Ylätalo (2013) also mentioned several challenges in calcium looping. The thermal power required for the post-combustion calcium looping is expected to require a substantial initial investment for the plant due to high specific energy demand. A steady supply of limestone and fuel is also needed, leading to new logistical solutions at the pulp mill. Technology also has unresolved issues, such as particle attrition, solid circulation rates, and sorbent degradation, which cause uncertainty about the technology. Achieving high purity targets (>99%) requires an additional purification unit (IEAGHG, 2019). The technology is still in the demonstration phase (TRL 6-7) (Dziejarski et al., 2023).

5.2 Chemical looping combustion

Chemical looping combustion is a carbon capture method in which solid material transfers oxygen to a fuel source, producing flue gases, primarily carbon dioxide. At existing pulp mills in Finland, chemical looping combustion can be utilised in a large auxiliary boiler combusting solid fuels using chemical looping with oxygen uncoupling (CLOU). CLOU can be implemented in large bark auxiliary boilers by replacing the current auxiliary boilers (Saari et al., 2023). CLOU is typically implemented by using two interconnected fluidised

bed reactors. It comprises an air and fuel reactor, where an oxide carrier circulates between them. The basic concept of CLOU is presented in Figure 10.

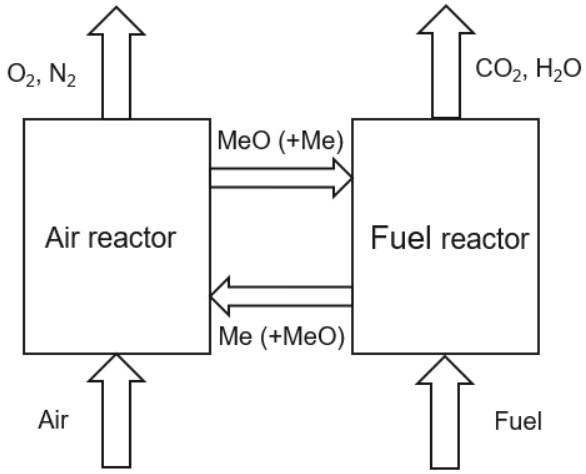

Figure 10. CLOU process loop (Peltola, 2014, p. 20).

As can be seen from Figure 10, a metallic oxide carrier moves between the reactors. It undergoes oxidation in the air reactor and is then transferred to the fuel reactor. Where the metal carriers release oxygen through decomposition at the right temperature range (800-1200 ℃) (Peltola, 2014, p. 20). After realising this, oxygen reacts with the fuel, oxidising it to carbon dioxide and water and releasing heat. The resulting hot flue gas is removed using a cyclone for heat utilisation, and the solid oxygen carrier is cycled back to the air reactor for another round of oxidation according to the equation.

$$2\mathrm{Me} + \mathrm{O}_2 \leftrightarrow 2\mathrm{MeO} \tag{4}$$

Peltola (2014) mentioned that air reactor flow mainly consists of nitrogen, excess oxygen, and small amounts of impurities from a combustion process. The fuel reactor gas flow consists primarily of combustion products, carbon dioxide, and water vapour. Carbon dioxide can be sequestered by condensing the water vapour from the flue gas stream.

CLOU offers several advantages. It enables highly efficient carbon capture with minimal energy consumption for auxiliary equipment, leading to low operational costs (Cormos, 2017, p. 930). Another benefit of the CLOU process is the reduced need for flue gas purification, as it produces significantly lower concentrations of SO_x, NO_x, HCl, mercury, and tar than regular combustion processes (Peltola et al., 2022, p. 9395). Additionally, the process can utilise wood residues from kraft pulp mills as a fuel source.

While the CLOU process offers significant advantages, it also presents several challenges. These include the need for substantial investments in a new boiler and low TRL. The TRL is estimated to be at the demonstration level (TRL 6) (Peltola et al., 2022, p. 9402-9403). In addition, the partial load operation of CLOU has yet to be thoroughly studied, which is crucial as larger amounts of variable renewable energy sources are introduced to the energy mix (Peltola, 2014). Dziejarski et al. (2023) also highlighted the challenges of CLOU, including the stabilities of oxygen carriers and slow redox kinetics.

5.3 Oxy-fuel combustion carbon capture

Oxy-fuel combustion is a carbon capture method in which fuel is combusted with pure oxygen to form flue gases, mainly carbon dioxide and water vapour. Figure 11 presents a process scheme of an oxy-fuel combustion system.

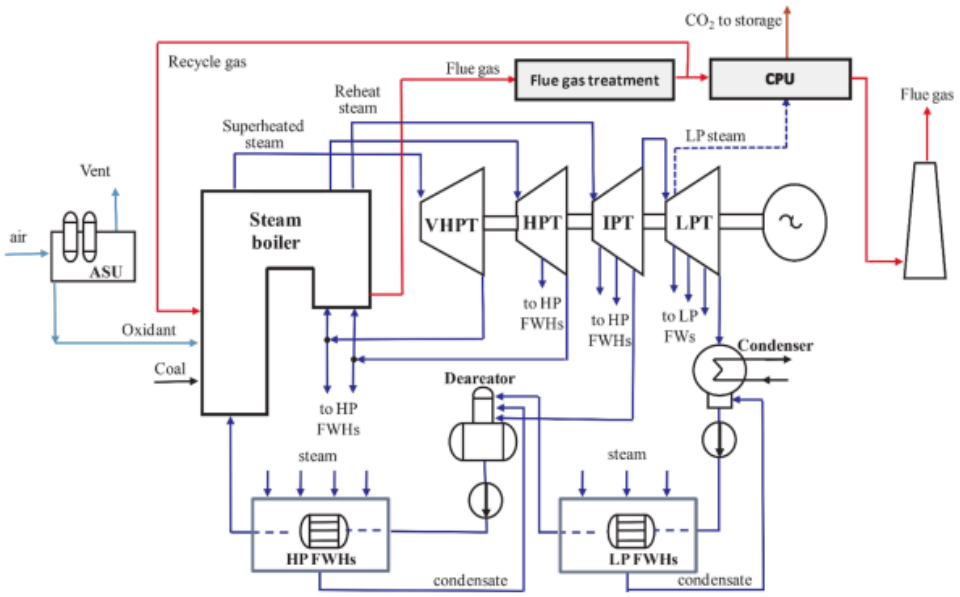

Figure 11. Oxy-fuel combustion process adapted into a steam boiler (Yadav & Mondal, 2022, p. 5).

Yadav & Mondal (2022) described oxy-fuel combustion in the following way. In oxy-fuel combustion, nitrogen is separated from the air, for example, by a membrane, cryogenic separation, or adsorption in an ASU. After nitrogen separation, almost pure oxygen (~95 mol-%) is used for combustion with fuel. The ASU unit is the largest energy-consuming unit in carbon capture, typically contributing 60-75% of electricity consumption.

Yadav & Mondal (2022) also mentioned that in oxy-fuel combustion, formed flue gases consist solely of carbon dioxide and water vapour. However, any sulphur compounds in flue gas must be removed in the flue gas pre-treatment before the separation of carbon dioxide. Carbon capture occurs in the carbon dioxide purification and compression unit (CPU). In the CPU, water is separated from the flue gas stream by cooling and condensing. After removing water, pure carbon dioxide is obtained by drying, cleaning, compressing, and distilling the flue gas. The CPU unit comprises several components, including multi-stage compressors, heat exchangers, flash evaporators, and valves. The CPU is also a major energy consumer, usually contributing 25-40% of carbon capture electricity consumption. Oxy-fuel combustion and carbon capture can be retrofitted to the kraft pulp mill's auxiliary boilers.

Yadav & Mondal (2022) have listed advantages when utilising oxy-fuel combustion. High CO_2 capture efficiency (90-98%) and purity (95-99.9%) can be obtained in oxy-fuel combustion. When nitrogen is separated using ASU, nitrogen oxide emissions are reduced by about half compared to the regular combustion process with air. When retrofitting oxy-fuel combustion, most of the existing equipment in the air combustion boiler can also be used in the oxy-fuel combustion process. Carbon dioxide separation requires little energy due to the high concentration of CO_2 in flue gases and the possibility of separating it by condensing water from the flue gas stream. Also, it has been studied that if integrated with methanol or hydrogen production process, which forms oxygen as a product, oxygen needs can at least be partially fulfilled. This decreases oxygen demand from ASU units and lowers the energy consumption of oxy-fuel combustion (Jeddizahed et al., 2024, p. 9).

Technology also has several disadvantages and challenges. The main disadvantage is related to the ASU unit, which increases the capital and operational costs of the integration (Yadav & Mondal, 2022, p. 4-16). Costs can be reduced by replacing a typical cryogenic ASU with membranes, as they consume less electricity (Garcia-Luna et al., 2024, p. 14). Another major disadvantage is the low technical readiness level of oxy-fuel combustion with solid fuels, which is at the demonstration level (TRL 7) (Dziejarski et al., 2023, p. 8). Yadav & Mondal (2022) noted other challenges in oxy-fuel combustion retrofits. Challenges arise from increased combustion temperature and potential air leaks, which can cause damage and reduce performance. Additionally, the presence of water vapour can lead to corrosion.

6 Integration criteria for carbon capture from a pulp mill point of view

Several integration criteria should be considered when integrating carbon capture into existing kraft pulp mills. This study includes the following criteria: retrofit easiness suitability and scalability possibilities, technology maturity, achievable CO_2 purity and carbon capture rate, possible energy and other flow limitations, emissions and waste streams, operational flexibility, and the costs of the carbon capture.

Carbon capture technology should be easily retrofitted to carbon dioxide point sources without major changes to the pulp mill. The overall suitability of different technologies for capturing carbon dioxide from various point sources in a pulp mill needs to be assessed, as each technology may have limitations and varying utilisation benefits depending on the point source. For instance, capture technologies best suited for large-scale capture from recovery boilers may not be ideal for smaller-scale carbon capture from auxiliary boilers or lime kilns.

It is important that technology has reached a high level of maturity and has been proven to perform well under expected conditions. Mature technology helps minimise project risk, thus reducing overall investment risk (Arroyo & Fortin, 2018, pp. 1-5). In this thesis, TRL of 8-9 indicates that the technology has been proven to perform under expected conditions.

When choosing carbon capture technology, it is important to consider the purity of the resulting CO_2 product. Different applications require different levels of purity, and various technologies can effectively produce different purities of CO_2. Table 3 outlines the purity requirements for carbon dioxide in CCS shipments and CCU when using food-grade CO_2 as a reference. It also provides suggested product purities for CCS pipeline transport. Due to the carbon capture and storage law 2012/416/3 (2012), transportation requirements are considered because captured CO_2 in CCS needs to be transported outside of Finland due to a lack of geological storage.

Table 3. Maximum allowable molar concentrations of impurities in product CO_2 for CCS shipments, CCS pipeline transport, and CCU using a food-grade standard (Northern Lights, 2024; Abbas et al., 2013a; EIGA, 2016).

Product gas component	CCS shipments [mol-%]	CCS, pipeline transport [mol-%]	CCU [mol-%]
CO_2	>99.81%	>95%	>99.9%
H_2O	30 ppm	<50 ppm	20 ppm
O_2	10 ppm	<10 ppm	30 ppm
SO_x	10 ppm	<100 ppm	1 ppm
NO_x	1.5 ppm	<100 ppm	2.5 ppm
H_2S	9 ppm	<10-200 ppm	0.1 ppm
N_2	50 ppm	<4%	-
Ar	100 ppm	<4%	-
CO	100 ppm	<2000 ppm	10 ppm
NH_3	10 ppm	-	2.5 ppm
Amine	10 ppm	-	-
H_2	50 ppm	<4%	-

For all three CO_2 applications listed above, the requirements for CO_2 purity are very high and, in fact, close to each other. High purity requirements can lead to the need for additional purification. For a pulp mill, carbon capture is executed after combustion, which means that the primary impurities in CO_2 product streams are water and oxygen. Additional purification is often needed to meet the mentioned application requirements (Abbas et al., 2012, p. 2392). Razak et al. (2023) mentioned several things about primary impurities. Water is generally harmless but can cause issues when reacting with impurities or CO_2 in transport. Therefore, water concentrations should be as low as possible to prevent excessive corrosion and hydration. Oxygen, on the other hand, can reduce the density of the product CO_2, potentially limiting CO_2 storage capacity. Additionally, the presence of O_2 may threaten storage integrity as it can dissolve caprock and cause corrosion in transport. Other impurity levels are usually insignificant in post-combustion or oxy-fuel carbon capture.

Carbon capture rate targets impact the selection of carbon capture technology. Different technologies have different efficiencies, impacting the volumes that can be captured. The aim is to maximise captured biogenic carbon dioxide economically.

Emissions and waste streams should be considered when comparing and evaluating different technologies. Some technologies produce toxic emissions and liquid waste streams that need

to be handled, for example, by secondary biological treatment or incineration (Nurrokhmah et al., 2013, pp. 13644-13651). Waste and emission streams increase the environmental impacts and can increase the cost of carbon capture.

Possible limitations from a pulp point of view should be evaluated, such as utilities and energy. Steam availability at pulp mills can be one limiting factor because some carbon capture technologies utilise low-pressure steam. Maximal steam availability should be assessed so the carbon capture technology does not impact the steam needs of the pulp mill process. The steam availability could limit the maximum carbon capture rates if investing in the new boiler is not cost-effective. The water circulation system at the pulp mill may require modifications with the integration of carbon capture, as it increases water consumption. Therefore, it is essential to estimate the increase in water consumption. Pulp mills are usually net electricity producers, which can change due to the high energy consumption of carbon capture. Energy and utility needs vary between technologies and should be investigated and compared. Possible energy and utility limitations and heat utilisation options should be explored by simulating the selected carbon capture technologies in a pulp mill.

Operational flexibility and dynamics of the carbon capture technology are important factors in evaluating and understanding the system's flexibility and ability to adapt to load changes. Abdilahi et al. (2017) mentioned in their study that high operational flexibility also enables the adjustment of carbon capture according to electricity prices. Some technologies utilise low-pressure steam, which can be used for additional electricity production when electricity prices are high, depending on how the carbon capture unit is connected to a steam network. Also, the technologies that utilise only electricity should have characteristics that adapt to electricity price changes to increase carbon capture's profitability. Electricity prices can vary significantly between hours, which requires fast operational flexibility of carbon capture technology to adapt to load changes. Abdilahi et al. (2017) mentioned that operational flexibility for additional electricity production could increase operating profit by up to 0-35% depending on technology, electricity price and the cost of CO_2. The need for operational flexibility and ability increases when more renewable electricity is added to the energy system and electricity price changes increase.

The final evaluation should be based on the cost of capturing CO_2. To estimate the costs, a techno-economic analysis of selected carbon capture technologies should be conducted to determine the most cost-effective solution to integrate into the pulp mill.

The integration analysis method is based on the integration criteria. Table 4 summarises the integration criteria outlined in Chapter 6 and demonstrates how these criteria are analysed in this thesis.

Table 4. Integration criteria and thesis methods.

Integration criteria	Impacts	Analysis method
High retrofit easiness and scalability	Does not require major modifications to a pulp mill. Easy to retrofit to different point sources.	Literature reviews
High technology maturity (TRL 8-9)	Reduces the project's overall risk. Carbon capture performs under expected conditions.	Literature reviews
High capture rate and purity of CO_2	Increases capture scale and can decrease specific costs of CO_2. Decreases additional purification requirements.	Literature reviews
Emissions and waste streams	Increases a pulp mill's environmental impacts and can increase carbon capture costs.	Literature reviews
High operational flexibility	Enables adaption to load changes. Enables to optimise carbon capture according to electricity prices.	Literature reviews
Energy and utility consumptions	It can be a limiting factor for integration. Impacts operational expenses. Impacts on energy balances.	Literature reviews and modelling
Costs of carbon capture	Impacts application profitability	Literature reviews and techno-economic analysis

7 Comparison of carbon capture technologies from a kraft pulp mill point of view

Based on the criteria determined in Table 4, carbon capture technologies are compared to find the most potential technologies to be integrated into the pulp mill. The comparison is made by presenting how technologies perform under integration criteria.

7.1 Technology maturity and retrofit easiness

Table 5 provides an overview of carbon capture technologies' current TRL, retrofit easiness and development trajectory. Development trajectory uses indicators: low, moderate, and high. A low rating indicates that the technology development is in the lower stages than the current TRL. Moderate demonstrates that the ongoing development is at the current TRL, and there are no plans for larger-scale demonstrations. A high rating signifies that the ongoing development surpasses the current TRL or that there are plans for higher-level evaluations or demonstrations beyond the present TRL.

Similarly, the retrofit easiness category uses low, moderate, and high indicators. A low rating indicates that integration requires large modifications in the pulp mill, such as new equipment. A moderate rating indicates that some smaller changes are needed in the pulp mill. High means that technology is an end-of-pipe solution, and integration does not require significant changes in the pulp mill.

Table 5 also presents the suitability of these technologies for different carbon dioxide point sources. The chemical absorption technologies in Table 5 are listed according to the solvent used.

Table 5. Current TRL of technologies, development trajectory, retrofit easiness in the pulp mill and integration suitability for different point sources.

Technology	TRL	Development trajectory	Retrofit easiness	Point sources
CLOU	6	Moderate	Low	Auxiliary boilers
Amine solvents	6-9	Moderate	High	All
Phase change solvents	5-6	High	High	All
Aqueous ammonia solvents	6-7	Moderate	High	All
Potassium carbonate solvents	7-8	High	High	All
Water-lean solvents	5-6	High	High	All
Chilled ammonia solvents	6-7	Moderate	High	All
PSA and VSA	6	Moderate	High	All
TSA and TVSA	5-7	High	High	All
Gas separation membranes	5-7	High	Moderate	All
Membrane contactors	5-6	Moderate	Moderate	All
MCFC	4-6	High	Moderate-high	All
Calcium looping	6-7	Moderate	Moderate-high	All
Oxy-fuel combustion	7	Moderate	Low-moderate	Auxiliary boiler

Currently, the only commercial carbon capture technologies from assessed technologies are amine-based chemical absorption. The next most mature technology is the hot-potassium carbonate process (HPC) based on potassium carbonate-based solvents at TRL 8. From these technologies mentioned in Table 5, the TSA adsorption process, gas separation membranes, MCFC, and chemical absorption technologies based on potassium carbonate, phase change, and water-lean solvents have been announced to have higher demonstration-level projects coming, and their TRL can be increased quickly in the future (Rezaei et al., 2023).

The ease of retrofitting in the pulp mill varies depending on the technology. All post-combustion technologies can be easily retrofitted. However, membrane contactors and gas separation membranes require modifications to the flue gas flow paths, making them moderately difficult to integrate. MCFC is considered at a moderate-high level, as it requires access to a gaseous fuel network, which may not be possible in all pulp mills in Finland. Calcium looping requires new logistical solutions for a steady supply of lime mud and fuel and is considered to have retrofit easiness of moderate-high. CLOU is deemed to have a low retrofitting ease as it requires boiler replacement. Oxy-fuel combustion is ranked low-moderate due to the need for substantial modifications to the existing auxiliary boiler.

Some technologies can have some limitations when scaling technologies on a large scale. Although membranes and MCFC can theoretically be integrated into all point sources, they are naturally modular. When integrated into the largest point sources, they require a lot of land area due to the multiple modules needed to utilise all carbon capture potential. The land area requirements can limit integration to smaller point sources, such as lime kilns or smaller auxiliary boilers. PSA can also be integrated into all point sources. However, PSA adsorption is more suitable for lower flue gas volumes, which can limit its integration to smaller point sources such as lime kilns or smaller auxiliary boilers (Rezaei et al., 2023).

7.2 Energy consumptions, capture rates & purities and costs

Different carbon capture technologies are compared regarding energy consumption, capture rates, product purity, and costs to identify the most promising technology for further assessment. Table 6 presents a detailed comparison of these factors for each technology. The chemical absorption technologies in Table 6 are listed according to the solvent used.

Table 6. Carbon capture rates and purities, carbon capture energy consumption and CO_2 cost avoided achieved with different carbon capture technologies.

Technology	CO_2 capture rate/purity	Energy consumption	Cost
CLOU [1], [7], [24], [27]	75-100% / 90-99.9%	0.6-0.7 GJ_e/t_{CO2}	24-27 €/t_{CO2}
Amine solvents [10], [14]	90-95% / 95-99%	2.5-3.8 GJ/t_{CO2}	40-65 €/t_{CO2}
Phase change solvents [10], [14], [25], [33]	90-95% / 95-99%	2.2-2.4 GJ/t_{CO2}	40-45 €/t_{CO2}
Aqueous ammonia solvents [10], [14], [18], [20]	90-95% / 95-99%	2.4-3.6 GJ/t_{CO2}	50-70 €/t_{CO2}
Potassium carbonate solvents [3], [10], [11], [14]	90-95% / 95-99%	2-5.3 GJ/t_{CO2} 0.7-1.5 GJ_e/t_{CO2}	30-45 €/t_{CO2}
Water-lean solvents [10], [12], [14], [19]	90-95% / 95-99%	2-2.8 GJ/t_{CO2}	35-55 €/t_{CO2}
Chilled ammonia solvents [10], [14], [30]	90-95% / 95-99%	2.5-3.2 GJ/t_{CO2}	66 €/t_{CO2}
PSA and VSA [10], [14], [16], [26]	75-90% / 90-95%	> 2.3 GJ_e/t_{CO2}	30-45 €/t_{CO2}
TSA and TVSA [5], [9], [10], [14], [21]	80-90% / 90-95%	2.8-3.2 GJ/t_{CO2} 2.4 GJ/t_{CO2}; 1.1 GJ_e/t_{CO2}	30-45 €/t_{CO2}
Gas separation membranes [4], [10], [13], [14]	60-90% / 80-95%	0.5-1.2 GJ_e/t_{CO2}	40-60 €/t_{CO2}
Membrane contactors [2], [10], [14], [29]	85-90% / 90-95%	2.9 GJ/t_{CO2}	45-57 €/t_{CO2}
MCFC [22], [23]	90% / 95%	0.9-1.3 $GJ_{primary}/t_{CO2}$	20-45 €/t_{CO2}
Calcium looping [8], [17], [28], [32]	85-95% / 90-95%	3.8-5.7 $GJ_{primary}/t_{CO2}$	27-39 €/t_{CO2}
Oxy-fuel combustion [6], [15], [31]	90-100% / 90-99.9%	1.3-2.0 GJ_e/t_{CO2}	48-60 €/t_{CO2}

References: [1] Adánez et al. (2018); [2] Bazhenov & Lyubimova (2016); [3] Borhani et al. (2015); [4] Chiwaye et al. (2021); [5] Clausse et al. (2011); [6] Cormos (2016); [7] Cormos (2017); [8] De Lena et al. (2017); [9] Duarte et al. (2017); [10] Dziejarski et al. (2023); [11] GCI (2023); [12] Heldebrant et al. (2017); [13] Hou et al. (2022); [14] IEAGHG (2019); [15] Kolster et al. (2017); [16] Krishnamurthy et al. (2021); [17] Kuparinen et al. (2019); [18] Li et al. (2016); [19] Linjala & Kajolinna (2023); [20] Liu et al. (2020); [21] Liu et al. (2023); [22] Mastropasqua et al. (2017); [23] Mastropasqua et al. (2019); [24] Peltola (2014); [25] Rackley (2017); [26] Riboldi & Bolland (2017); [27] Saari et al. (2023); [28] Santos et al. (2021); [29] Scholes (2023); [30] Valenti et al. (2012); [31] Vasudevan et al. (2016); [32] Ylätalo (2013); [33] Zhang et al. (2019).

Table 6 separates energy consumption into different rows for potassium carbonate-based chemical absorption, as it can consume only electricity in certain applications and heat in others. It also distinguishes TSA and TVSA technologies because TVSA has significant simultaneous electricity and heat consumption. Regarding CLOU and oxy-fuel combustion, the energy consumption is calculated based on the electricity consumption of their primary auxiliary equipment. Fuel consumption is not considered, as it has been integrated into an auxiliary boiler, assuming its fuel consumption will remain at the same level as before integration.

The form and amount of energy consumed are important aspects of energy consumption and how it impacts the pulp mill's energy balance. Technologies that consume pulp mill’s low-pressure steam greatly impact the pulp mill’s steam balances. Steam availability can also be a limiting factor for large-scale integration. Technologies that primarily consume electricity can significantly impact the net electricity production of pulp mills. Possible negative net electricity production does not impact pulp mill processes, but it requires purchasing more electricity and increases the risk of electricity price fluctuations. Technologies that primarily use additional fuel for carbon capture increase the primary energy consumption in pulp mills. However, the increase in primary energy consumption is mostly related to fuel consumption, which does not greatly impact pulp mills' operations. Therefore, the main impacts on energy balance are electricity and steam consumption.

Due to low steam and electricity consumption, CLOU, MCFC and calcium looping have the most neutral impact on a pulp mill's energy balance. These technologies use additional fuel or fuel combustion with oxygen to enable carbon capture. Also, these processes increase power production, which partly compensates for the technology's electricity consumption.

Membranes and HPC technologies are the most energy-efficient among electricity-consuming technologies, with minor impacts on pulp mill electricity consumption. Among the primarily steam-consuming technologies, TVSA adsorption, absorption using phase change, aqueous ammonia, potassium carbonate-based, or water-lean solvents can reduce steam consumption slightly compared to most mature amine carbon capture.

Costs indicate that lower steam or net electricity consumption generally results in reduced capture costs. This applies to CLOU, MCFC, and calcium looping, which have the lowest carbon capture costs. Compared to the most mature technology, amine-based carbon capture, other potential technologies are adsorption and chemical absorption using water-lean or potassium carbonate-based solvents. However, it should be noted that most cost estimations are typically conducted for large-scale power production plants. Real cost estimations should be calculated with point source-specific details.

All mentioned technologies can produce high product CO_2 purities, and the variation between technologies is minor. However, to match application requirements, all technologies require additional purification, excluding technologies where flue gas purification is part of the process, such as oxy-fuel combustion and CLOU. Energy consumption increases if further purification is needed (Wetenhall et al., 2014).

It can also be noted that carbon capture rates can be high with every technology, nearly 90%, and there is not a huge variety between technologies. Only oxy-fuel combustion technologies and CLOU processes can have higher carbon capture rates. If the purity requirements for transportation and utilisation remain high, then the capture rates are closer to 90%. A lower capture rate is due to a possible need to utilise the distillation CPU to remove excess oxygen and water to the required level (Kolster et al., 2017, p. 135).

7.3 Emissions and waste streams

Emissions and waste streams related to the carbon capture technologies should be identified and compared between technologies. Table 7 presents emissions and waste streams of different carbon capture technologies.

Table 7. Emissions and waste streams of technologies.

Technology	Toxic emissions and waste streams
CLOU	No additional emissions or liquid waste is formed during the separation process.
Chemical absorption	Amine and ammonia-based solvents can produce toxic and acidic gaseous and aerosol emissions. In addition, amines form toxic degradation waste (Fang et al., 2020).
Adsorption	There are no additional emissions or waste during the operation, but the adsorbents must be changed. Materials can be reused with low waste generation (Dziejarski et al., 2023).
Membranes	No additional emissions or liquid waste formed in the capturing process
MCFC	No additional emissions or liquid waste formed in the capturing process
Calcium looping	Reduce SO_x emissions and not produce additional emissions or waste (Ylätalo, 2013).
Oxy-fuel combustion	No additional emissions or liquid waste is formed during the separation process.

Table 7 shows that only ammonia and amine-based chemical absorption technologies produce additional emissions during carbon capture by forming possible amine and ammonia emissions. When amines and ammonia are released into the air, they can react with rainwater and undergo further chemical reactions with NO_x, HNO_3, and O_3. This can lead to the formation of more harmful substances or strong carcinogens, such as nitrosamines (Fang et al., 2020, p. 1). Gaseous and aerosol emissions amounts vary by equipment size, solvent composition and system operating conditions. Amine emissions are reported to be around 1-4 ppm, and ammonia emissions around <1-28 ppm, depending on the solvent and control methods used (Fang et al., 2020, p. 3). These reported emissions amounts are generally small because the amine and ammonia emission recommendations are less than <6 ppm in clean flue gas, and total emissions are under 2.8 t/year. It is important to note that gaseous emissions can be effectively controlled using various methods, such as water or acid wash (Fang et al., 2020, p. 2). Nurrokhmah et al. (2013) mentioned that carbon capture waste amounts generated using conventional amine-based solvents have been estimated to be 3-3.5 kg/t_{CO2}. This waste stream needs biological treatment or incineration. Also, the possible utilisation of this waste stream as a scrubbing agent for NO_x emissions due to ammonia and nitrogenous compounds has been proposed. Other technologies do not produce additional emissions or waste streams, making them more attractive for carbon capture technology in a pulp mill.

7.4 Operational flexibility

Carbon capture technology's operational flexibility is assessed according to how fast it adapts to load changes, and how it can be operated at part load is presented in Table 8.

Table 8. Operational flexibility of carbon capture technologies.

Technology	Operational flexibility
Chemical absorption	Moderate. The chemical absorption unit cannot shut down and start rapidly due to rapid changes in electricity prices between hours. Desorption columns need 1-2 hours of preheating, even in a hot start-up. The minimum load of units is 30% of nominal capacity, and ramp rates are 3-8%/min. Solvent storage and flue gas venting can increase flexibility and allow the operation to be more flexible according to electricity prices. Solvent storage increases investment costs (Abdilahi et al., 2017).
Adsorption	High. Easy start-up, load following and can be shut down when needed (Rezaei et al., 2023, p. 5). Depending on the control method, response times are short for load changes, around 10%/0-13 min (Skjervold et al., 2023).
Membranes	High. Membranes have fast responsiveness and high operational flexibility when operating at part-load. They have very short response times. For example, 10% of the load change response times happen in seconds, and cold start-up times are less than 30 minutes to full capacity (Yuan et al., 2019, p. 159).
MCFC	High. The part load operation is possible with high efficiency. High efficiencies can be achieved at part load, even with a 50% nominal load (Sánchez et al., 2011, p. 10335). The MCFC can be operated quickly due to its controlled electrical potential.
Calcium looping	High. The sorbent can be easily stored, which helps it run with different loads. The process can be operated at 50% of the nominal operation flue gas load. High capture efficiencies can be maintained, but modifications to the make-up flow might be needed (Diego & Arias, 2020).
Oxy-fuel combustion	Low. ASUs are mainly designed to perform under design conditions. ASUs can be operated only with 60-100% load, and efficiencies decrease rapidly compared to the design load when operated under 80%. Efficiency in part load can be increased by oxygen buffer storage capacity. (Toftegaard et al., 2010, p. 589).
CLOU	Low. Part-load operation is challenging. Gas velocity in the air reactor cannot be significantly reduced because it will stop solids circulation. It can be controlled slightly by a separate solid circulation system or by staging the air reactor (Lyngfelt & Leckner, 2015, p. 379).

Table 8 shows high operational flexibility, especially with adsorption, membrane, calcium looping and MCFC carbon capture technologies. These technologies enable flexible carbon capture and a fast load following the point source. Also, it allows adjusting the carbon capture process according to electricity prices between hours without significantly impacting the yearly pulp mill's overall carbon capture rate. Chemical absorption was evaluated to have moderate-level carbon capture. Chemical absorption flexibility can be increased, but it will also increase capital investment costs, especially when investing in rich solvent tanks. When considering the investment in rich solvent tanks, it is important to determine the required storage capacity. According to Abdilahi et al. (2017), the optimal storage capacity for solvents should be such that it can effectively leverage high electricity prices for 15–30 minutes daily. However, flexibility does not necessarily increase chemical absorption profitability significantly. Flue gas bypassing can also improve flexibility but will impact carbon capture rates. Oxy-fuel combustion and CLOU show low performance operating at part-load compared to other technologies.

7.5 Potential carbon capture technologies

When comparing carbon capture technologies according to the integration criteria, two technologies could be integrated into an existing pulp mill. Amine-based carbon capture and HPC process based on potassium carbonate solvents were the only technologies with a TRL of at least 8, making these two technologies the only ones possible to integrate at the current moment. Therefore, these technologies have been chosen for additional integration and cost analysis. HPC performs better under the selected criteria of these two technologies because it has lower costs and less impact on energy balances. HPC also does not produce additional emissions or waste.

However, there is a potential for other technologies to develop quickly in recent years and challenge amine-based carbon capture and HPC in the future. Because of this, the potential of other technologies to reach TRL 8-9 is also evaluated. The criteria used in this evaluation are current TRL, retrofit easiness, existing markets, component availability and maturity, other technology applications, cost reduction potential and development needs.

The evaluation uses a scoring system ranging from 1 to 3 for each category, with all categories being equally weighted. Information in IEAGHG (2019) for existing markets,

component availability and maturity, other technology applications and development needs are used in the evaluation. Additionally, approximations of current TRL, retrofit easiness, and cost reduction potential compared to amine-based carbon capture found in Tables 5 and 6 are used for a more precise assessment. The scores for technologies are presented in Appendix 4. Other promising technologies expected to become widespread next in the future include calcium looping, adsorption technologies, aqueous ammonia-based chemical absorption, and MCFC carbon capture technologies. Compared to other technologies, these technologies encompass mature components, potential cost reduction compared to amines, easy retrofit, and other industry applications, leading to a high potential for widespread in the future.

These four other potential widespread technologies were compared to amine and potassium carbonate-based technologies using the criteria presented in Chapter 6 without considering current TRL to find the best integration option for a pulp mill in the future. Amine-based carbon capture is used as a reference for the impacts of costs and energy balance. The highest potential technology for integration was identified and detailed in Appendix 5 using the same grading system employed in the initial technology analysis.

It was found that, based on the selected criteria, amine-based carbon capture may no longer be a viable option for integration in the future. With determined criteria, the calcium looping process and MCFC show the highest potential for integration into pulp mills in the future. Both technologies exhibit strong performance across all categories, particularly regarding energy balance impacts with low steam and electricity consumption and potential cost reductions compared to alternative technologies. Despite slightly lower performance levels, adsorption and potassium carbonate-based technologies remain viable options due to small differences in analysis. The lower technology maturity of MCFC introduces uncertainty regarding its costs and performance under real flue gases, and more studies and demonstrations of the technology are needed to confirm integration suitability. It is also important to highlight that uncertainties remain regarding the costs and energy consumption of integrating calcium looping into a pulp mill. Most existing studies have been conducted in the cement industry using kilns similar to those used in pulp mills. Furthermore, regardless of the time frame or retrofitting, CLOU could be regarded as a promising technology if installed as an auxiliary boiler in a new pulp mill or to replace an old boiler due to lower capture expenses and its negligible effect on the energy balance of the kraft pulp mill.

8 Selected carbon capturing technologies in kraft pulp mill

Technologies selected to further energy balance and integration analysis into existing pulp mills are the HPC process utilising potassium carbonate solvent and amine-based chemical absorption utilising MEA solvent. MEA was selected among different amine solvents due to its high maturity, proven operation in commercial power plants, and availability of process data. The carbon capture process integration into an example pulp mill A in Finland is modelled, including flue gas preconditioning, chemical absorption process and liquefaction. Process modelling is done with a steady-state simulation approach using Aspen Plus V14 process modelling software. Pulp mill A is a pulp-only mill producing birch and softwood pulp. Pulp mill A has process values that are presented in Table 9.

Table 9. Process values of Pulp mill A.

Production parameters	Amount	Unit
Pulp production	800 000	ADt/a
Electricity production	720	GWh/a
Electricity consumption	570	GWh/a
Operational hours	8400	h/a
Lime production	600	t/d
Heat consumption	2600	GWh/a
Water consumption	8000	m^3/h
CO_2 from recovery boiler	1350	kt/a
CO_2 from lime kiln	290	kt/a

From Table 9, the pulp mill without carbon capture integration is self-sufficient in electricity consumption. The self-sufficiency of pulp mill A in electricity consumption may change with the integration of carbon capture technology and is analysed in simulation. An increase in water and steam consumption is also expected and considered in the detailed analysis. In this thesis, capturing carbon dioxide is modelled by considering two-point sources. The point sources are the pulp mill's recovery boiler and lime kiln because of their close locations and stable production according to chemical recovery needs in a pulp mill. For these reasons, operational hours for capture units are assumed to be the same as those for pulp mills. The

CO_2 emissions from the power boiler are excluded from capture considerations due to the substantial operational variability of the boiler.

Furthermore, the emissions from the power boiler are strongly affected by the operational dynamic of the interconnected processes at the mill site. The lime kiln at the pulp mill uses a tall oil pitch as fuel during normal operations, which means that during normal operations, CO_2 produced at the pulp mill is biogenic. Other times, fossil fuels are used mainly during the start-ups and shutdowns. This results that annually, around 99.8% of the time, emissions are from biogenic sources. Wet flue gas properties from considered point sources are presented in Table 10.

Table 10. Flue gas properties of pulp mill A point sources.

Point source	H_2O [mol-%]	CO_2 [mol-%]	N_2 [mol-%]	O_2 [mol-%]	q_m [kg/s]	T [°C]
Recovery boiler	20.4	11.4	65.6	2.6	250	180
Lime kiln	24.5	20.9	51.6	3	30	290

Only major flue gas components are considered in the analysis. Flue gas impurity levels are considered low and do not impact the MEA or HPC process after flue gas preconditioning, where acidulous components (SO_x and NO_x) and the rest of the particles are removed. This thesis considers three integration scenarios: separate carbon capture from the recovery boiler, lime kiln and full carbon capture from both point sources. When considering the full carbon capture scenario, flue gases are perfectly mixed before the absorber. Flue gas mixing is done because chemical absorption has high capital investments, and flue gases from lime kilns have low flue gas flow rates compared to recovery boilers. The following chapters present modelling methods, including flue gas preconditioning, chemical absorption process and CO_2 liquefaction. After the methods, simulation results are presented.

8.1 Flue gas preconditioning

Pulp Mill A does not have a pre-installed flue gas scrubber for point sources. Therefore, flue gas preconditioning is required to remove impurities and decrease flue gas temperature. Preconditioning is modelled using a direct contact cooler (DCC). DCC removes leftover particles and acidic components from the flue gas stream and lowers the temperature of the flue gases to 40°C. This cooling and removal of acidic components are necessary for the

amine process to prevent the degradation of amines and increase the solubility of CO_2. Potassium carbonate-based solvents are well-resistant to impurities. However, DCC significantly decreases compressor electricity demand in the carbon capture process since high-temperature flue gases require more electricity to be compressed to the desired pressure. Also, in the reference study, it was observed that flue gases entered the HPC carbon capture units at a temperature range of 39-42.5 °C (Gustafsson et al., 2021). The temperature range is similar to the temperature required in the amine carbon capture process. As a result, the decision was made to implement the same DCC system in the HPC process. Both DCCs are modelled separately for point sources to limit building oversized columns in full carbon capture cases. Madeddu et al. (2019) mentioned that packed bed columns should have a smaller diameter than 12 m. After DCC, flue gases are combined in a full carbon capture case. DCC Aspen process model is presented in Figure 12.

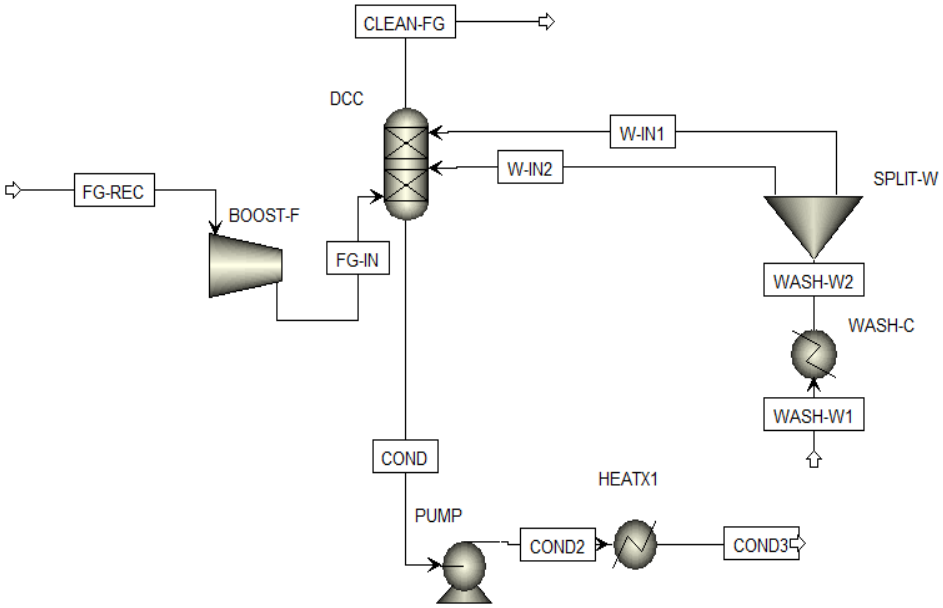

Figure 12. DCC Aspen model for recovery boiler.

The DCC is modelled using an electrolyte-non-random-two-liquid (ENRTL) with Redlich-Kwong (RK) equations of state (ENRTL-RK) property method, and the column is modelled using rate-based modelling to calculate the dimensions of the flue gas cooler column. In the model, hot flue gas is fed into the column at the bottom of the scrubber to direct contact with water. A flue gas booster blower compensates for pressure drops in the DCC column and the

absorber. The flue gas enters the blower at 1.1 bar, and the pressure is expected to increase slightly to 1.15 bars after passing through the blower.

The water is supplied to the DCC in two stages, as Verma et al. (2022) suggested. According to their suggestion, in a 10-stage DCC column system, 24% of the water is introduced at the top, while the majority (76%) is supplied to the columns at the fourth stage from the bottom. This adjustment increases the residence time of flue gas, effectively reducing SO_x and NO_x concentrations. In their study, DCC removed over 99.9% of SO_x and 92% of NO_x, meeting capture unit requirements for impurity concentrations typically found in flue gases from pulp mills. During simulations, water mass flow to the column is adjusted to achieve the desired flue gas temperature. Initially, water at 24 °C is introduced into the DCC, and as the flue gas cools, some of its water content condenses.

The condensed flue gas and water are removed from the bottom of the column and pumped back into the column through heat exchangers. Since some water content condenses from the flue gas in the DCC, a portion must be removed from the cycle before being returned to the column. In this thesis, the removed water is assumed to be the difference in mass flow between the condensate and the input wash water. Condensate exits at a temperature close to the flue gas dew point of around 65-70 °C. Recoverable heat from the condensate flow is studied in the first heat exchanger, where the condensate flow is cooled to 52 °C. This heat can be, for example, used to heat a district heat return water or heat warm water at the pulp mill. After the DCC unit, cleaned flue gases are fed to the chemical absorption units.

In the simulation, the dimensions for DCC columns used for recovery boiler flue gases were as follows: the column height was 18 m, and the diameter was 11.4 m. For lime kiln flue gases, the column height was 7 m, and the diameter was 4.2 m.

8.2 Chemical absorption with HPC

Chemical absorption with the HPC process is modelled using Aspen Plus with the ENRTL-RK property method and rate-based approach for absorber and stripper. The used property method is widely accepted among researchers when chemical absorption carbon capture is studied (Gustafsson et al., 2021). The ENTRL property model was used to assess the thermodynamics of the liquid phase. In contrast, the properties of the vapour phase were

described using the Redlich-Kwong equations of state. CO_2, N_2, and O_2 are selected as Henry's components for the process model. 30 wt.% potassium carbonate was used as a solvent. It is the recommended solution strength if unwanted precipitation of salts is to be avoided (Gustafsson et al., 2021). The following dissociation, kinetic, and equilibrium equations used in Aspen plus simulations that are considered in the model are as follows:

$$KOH \rightarrow K^+ + OH^- \tag{5}$$

$$K_2CO_3 \rightarrow 2K^+ + {CO_3}^{2-} \tag{6}$$

$$KHCO_3 \rightarrow K^+ + {HCO_3}^- \tag{7}$$

$$CO_2 + 2H_2O \leftrightarrow H_3O^+ + {HCO_3}^- \tag{8}$$

$${HCO_3}^- + H_2O \leftrightarrow H_3O^+ + {CO_3}^{2-} \tag{9}$$

$$2H_2O \leftrightarrow H_3O^+ + OH^- \tag{10}$$

$$CO_2 + OH^- \rightarrow {HCO_3}^- \tag{11}$$

$${HCO_3}^- \rightarrow CO_2 + OH^- \tag{12}$$

Equations (5-7) are dissociation reactions, equations (8-10) are equilibrium reactions, and equations (11-12) are kinetic reactions. Equilibrium constants are used to calculate reactant concentrations in equilibrium for defined equilibrium reactions using Aspen's built-in equilibrium constant equation.

$$\ln\left(K_{\mathrm{eq}}\right) = A + \frac{B}{T} + C \cdot \ln(T) + D \cdot T \tag{13}$$

where K_{eq} is the equilibrium constant, A, B, C, and D are coefficients, and T is temperature (K).

The kinetic model equations use Aspen's built-in reduced power-law equation to determine reaction rates.

$$r = k\exp\left(-\frac{E}{RT}\right) \tag{14}$$

where r is the reaction rate, k is the pre-exponential factor, E is activation energy (cal/mol), and R is the gas constant (J/(kg K)).

Table 11 includes the constants needed for the equilibrium constant equation, activation energies, and pre-exponential factors for reduced power-law equations.

Table 11. Equilibrium and kinetic constants for HPC system.

Equilibrium:	*A*	*B*	*C*	*D*
Equation 8	231.465	-12092.1	-36.7816	0
Equation 9	216.05	-12431.7	-35.4819	0
Equation 10	132.899	-13445.9	-22.4773	0
Kinetic:	*k*	*E* [cal/mol]		
Equation 11	4.32	13249		
Equation 12	2.38	29451		

The process is modelled according to suggestions by Gustafsson et al. (2021), Navedkhan et al. (2022) and HPC process improvement suggestions by Ayittey et al. (2020) with small modifications. The Aspen process model is presented in Figure 13.

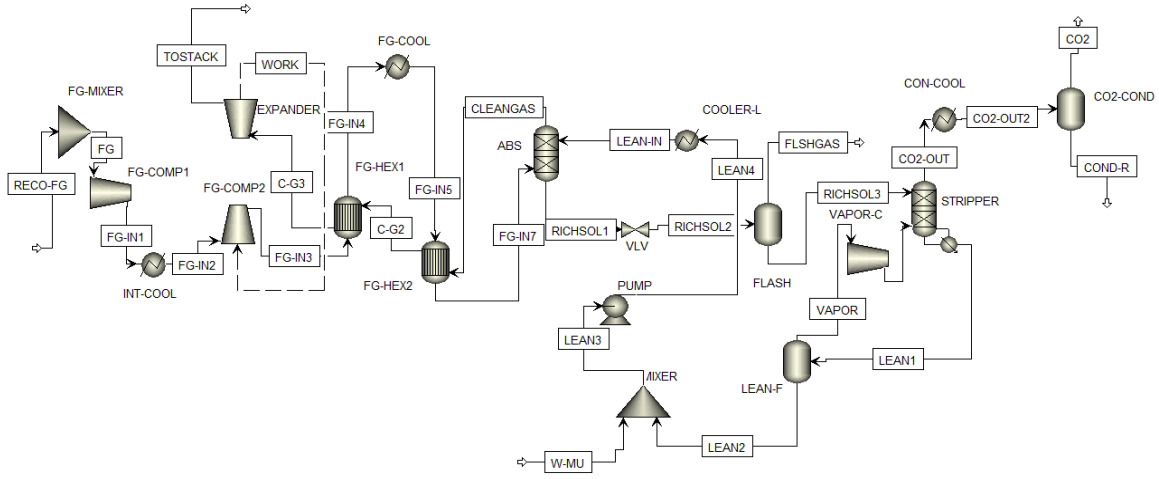

Figure 13. Chemical absorption by hot potassium carbonate process for recovery boiler.

In the HPC process, it is essential to precondition the flue gas after DCC using flue gas compressors powered by either a turbo expander and electric motor or high-pressure steam. In this thesis, the compressor is operated using electric power in conjunction with the turbo expander to reduce steam consumption. After the first flue gas compressor, the flue gas flow is cooled to 120 °C so the temperature is higher than its dew point. In the second compressor, operated by the expander, the flue gas is compressed to its final pressure, set at 8 bar

according to absorption column requirements. Gustafsson et al. (2021) suggest a 6-8 bar pressure range is optimal for flue gas absorption in the HPC system. They note that pressure increases have only a small effect on absorption capacity while simultaneously increasing the power consumption of compressors. Following the flue gas compressors, the flue gas is further cooled to around 80 °C through a heat exchanger and flue gas cooler. This improves mass transfer and CO_2 solubility, thus enhancing absorption (Ayittey et al., 2020).

After cooling, flue gases are fed into the absorber, where absorption reactions happen between compressed flue gas and solvent. Clean flue gas exits from the top of the absorber, is heated through heat exchangers and cooled via an expander before being sent to the stack. The flue gases enter to stack at 1-5 ℃. The CO_2-rich solvent exits from the bottom of the absorber and is then sent through the expansion valve, which expands the solvent to 3 bars. Next, the solvent is flashed to remove excess nitrogen and increase the purity of the CO_2 stream after a stripping column. After flashing, the rich solvent is heated to release CO_2 from the solvent in the stripper column at 1.3 bars. The CO_2-rich stream is cooled to 45 ℃ to remove water in the flash unit. Due to converge reasons in the model, excess condensate is removed from the capture unit. The surplus water is assumed to be wastewater and is supposed to be directed to a wastewater treatment plant.

To reduce the reboilers' heat duty, a process involving the flashing of lean solvent vapour and subsequent compression to a pressure of 1.5 bars is implemented. The compressed vapour is then reintroduced into a stripper at the bottom stages of the system. Flashed vapour mainly consists of water (~ 97%) and CO_2 (~ 3%). Flashed vapour flow is around 3-4% by mole of lean solvent flow.

After flashing, make-up water is mixed with a lean solvent whose mass flow equals the difference between the water outlet and inlet flows. Inlet flows are flue gas water content, and outlet flows are exiting flows after flashing, stripping, and absorption. After mixing, the lean solvent is pumped back into the absorber via a solvent cooler, where it is cooled to 65 ℃.

8.3 Chemical absorption with MEA

Carbon capture in the absorber and stripper was modelled using ENRTL-RK property methods and a rate-based approach. A 30 wt.% MEA solvent was used for the simulation without considering the solvent's degradation. In the simulation, N_2, O_2, and CO_2 are selected as Henry's components. This thesis uses kinetic and equilibrium reaction equations that AspenTech proposed for chemical absorption simulations with a 30 wt.% MEA solvent (Aspen Plus®, 2008). The reaction equations are the following.

$$2H_2O \leftrightarrow H_3O^+ + OH^- \quad (15)$$

$$HCO_3^- + H_2O \leftrightarrow CO_3^{2-} + H_3O^+ \quad (16)$$

$$MEAH^+ + H_2O \leftrightarrow MEA + H_3O^+ \quad (17)$$

$$CO_2 + OH^- \rightarrow HCO_3^- \quad (18)$$

$$HCO_3^- \rightarrow CO_2 + OH^- \quad (19)$$

$$MEA + CO_2 + H_2O \rightarrow MEACOO^- + H_3O^+ \quad (20)$$

$$MEACOO^- + H_3O^+ \rightarrow MEA + CO_2 + H_2O \quad (21)$$

Equations (15-17) are equilibrium reactions, and equations (18-21) are kinetic reactions. Equations 13 and 14 are used for defined reaction equations. For equilibrium calculations, Aspen computes constants used in the equilibrium constant equation (13). The Aspen built-in reduced power-law equation (14) uses the pre-exponential factors and activation energies presented in Table 12.

Table 12. Pre-exponential factors and activation energies of MEA absorption system.

Reaction equation number	*k*	*E* [cal/mol]
18	$1.33 \cdot 10^{17}$	13249
19	$6.63 \cdot 10^{16}$	25656
20	$3.02 \cdot 10^{14}$	9855.8
21 (Absorber)	$5.52 \cdot 10^{23}$	16518
21 (Stripper)	$6.50 \cdot 10^{27}$	22782

The process is modelled according to Li et al. (2015) suggested model due to their good accuracy (±4%) compared to the experimental plant data. Small modifications are made to scale it to full scale. The Aspen model is presented in Figure 14.

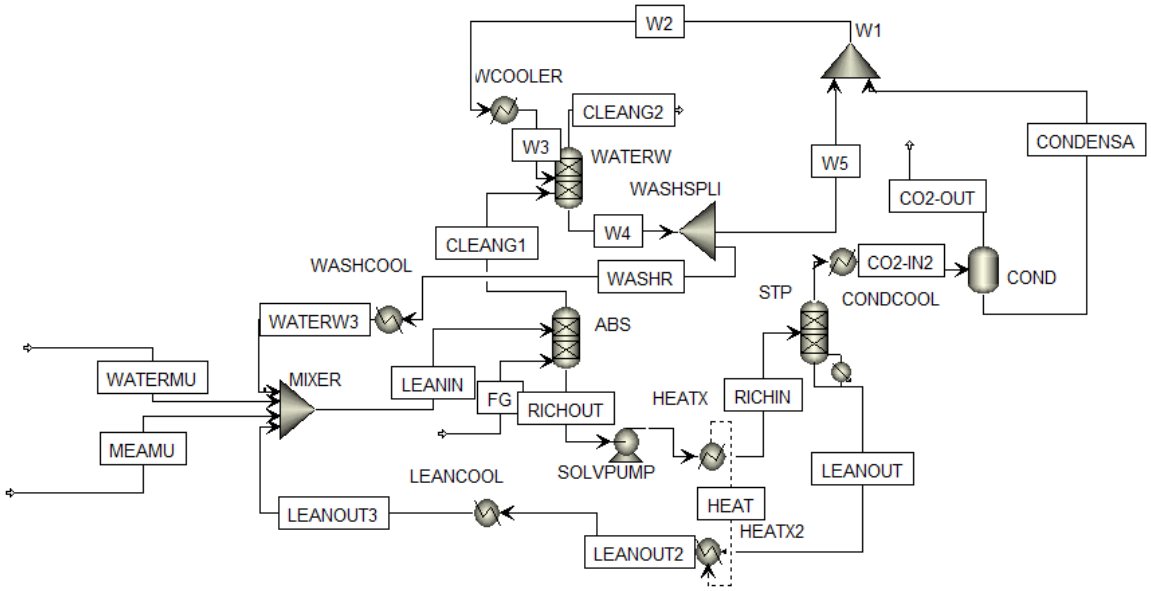

Figure 14. Aspen Plus model flowsheet for chemical absorption with MEA-solvent.

Flue gas is fed into the absorber from the bottom to contact with the solvent, which is fed into the absorber at the top. In the model, the absorber operates at 1.1 bar pressure. According to reaction equations, Flue gas CO_2 is absorbed into the solvent, and cleaned flue gases exit at the top of the absorber. Reactions in the absorber are exothermic, which causes the vaporisation of MEA. Vaporised MEA is washed and recovered in a water wash column to achieve under three ppm-molar concentration in the cleaned flue gas stream before entering the stack. Water wash in this thesis is considered part of the absorber but simulated as a separate unit. The cleaned flue gases exit at the top of the washer unit after washing, and the used wash water exits at the bottom. 15% of the flow exiting from the bottom is recycled back to the water wash unit, while the remaining water is recycled back to the absorber. The CO_2-rich solvent from the absorber is pumped to the stripper through heat exchangers, where the solvent is heated to nearly 110°C before entering the stripper.

The stripper operates at 2 bar pressure. Higher pressure compared to the absorber is done to reduce equipment size and parasitic losses (Warudkar et al., 2013). In the stripper, the solvent is heated using low-pressure steam. Water is removed from the CO_2-rich stream in the

condenser at 45 ℃. Condensate formed is recycled back to the water wash unit by cooling the wash water to 40 ℃.

Lean solvent from the bottom of the stripper is cooled via a heat exchanger and lean solvent cooler to 40 ℃ and mixed with water make-up, MEA-make-up flow, and wash water exiting from the wash section. The make-up mass flow of MEA and water equals the difference between the exiting and inlet flows. MEA and water exit from the stripper and washer unit. On the other hand, water comes into the unit from flue gas.

8.4 Purification and liquefaction

Due to the high purity requirements of CO_2 for both CCS and CCU applications, CO_2- and CO_2-rich streams need deep purification of water and oxygen. In post-combustion capture technologies, CO_2-rich streams after the stripper typically contain 2.8-7.3 mol-% water and 0.1-3 mol-% oxygen, requiring additional purification to match the current recommendations (Abbas et al., 2013a). Water content in liquefied CO_2 should be less than 30 ppm-mol-%, and oxygen should have a lower concentration than ten ppm-mol%.

Several technologies can remove water, including compressing and cooling, adsorption using solid or liquid desiccants, absorption with a deliquescing solid and cooling below the initial dew point (Abbas et al., 2013a). Oxygen can be removed with catalytic oxidations with different oxidants, cryogenic distillation, or chemisorption (Abbas et al., 2013a). This thesis simulation removes water with refrigeration and condensation due to its low investment cost (Abbas et al., 2013a). Oxygen is removed using catalytic oxidation with hydrogen due to simple construction, low investment cost, low operating temperature and the possibility of operating continuously with one reactor (Abbas et al., 2013a). In the simulation, the Peng-Robinson equation of state is used to calculate the properties of streams in Aspen Plus. According to Aliyon et al. (2020), the model has shown good accuracy in CO_2 liquefaction applications. Modelling was based on the study by Aliyon et al. (2020), which proposed a refrigeration cycle using ammonia (NH3) and the liquefaction process suggested by Abbas et al. (2013b). The Aspen model is presented in Figure 15.

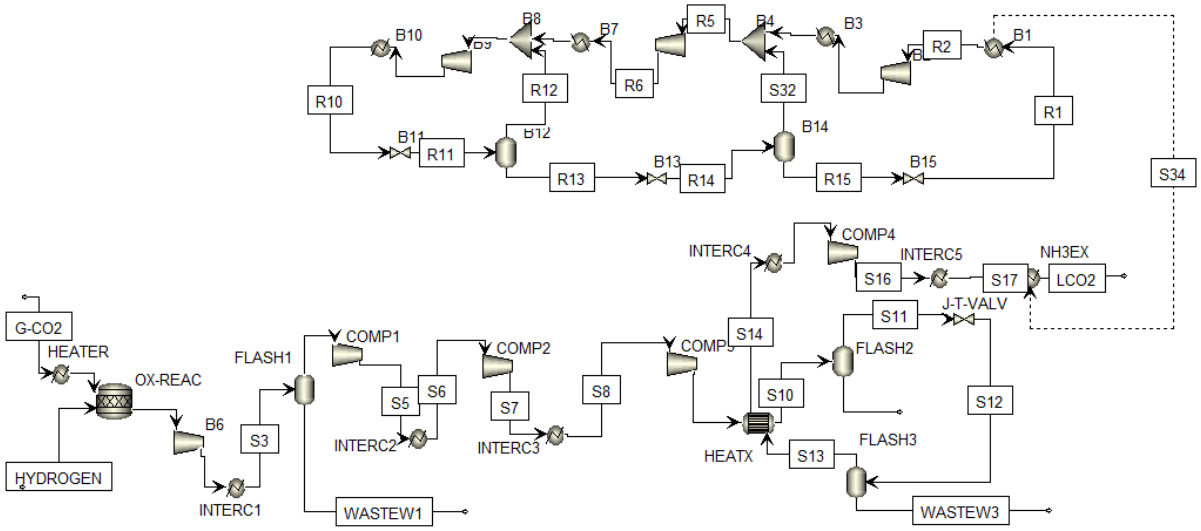

Figure 15. Liquefaction plant Aspen model.

The process starts with heating the gaseous CO_2 stream to 80 °C. In the reactor, 85% of hydrogen oxidises to water with oxygen. The mass flow of the hydrogen stream is controlled to reach an O_2 concentration of less than ten ppm in liquified CO_2. The gaseous stream is compressed and cooled to remove water from the process. Before the Joule-Thompson valve, the stream is pressurised to 41.5 bar, and A Joule-Thompson valve is used to reduce pressure to 9.6 bars, leading to a temperature of -34 °C and water condensation. The condensed water is removed in a flash separator to achieve a water concentration lower than 30 ppm. In the last compressor stage, the CO_2 stream is compressed to 15 bars and cooled to -28 °C in the last heat exchanger to match Northern Lights' requirements for liquefied CO_2.

8.5 Simulation results and discussion

The results and discussion chapter presents simulation results in different part processes and analyses the impacts of carbon capture integration. First, utilities are presented, and steam availability limitations are studied in pulp mill A. After that, simulation results are presented from DCC to the liquefaction plant. At the end of the chapter, utility consumption impacts are analysed and compared between MEA and HPC technologies.

The utilities used in the simulation are electricity, cooling water, and steam. Process values for steam and water flows used in the analysis are presented in Table 13.

Table 13. Utility specs for carbon capture.

Auxiliary flow	T_{in} [°C]	T_{out} [°C]	p_{in} [bar]
Steam for carbon capture	135	134	3
Cooling water	25	45	5

Cooling water return temperatures are chosen based on pulp mills' process values. Steam values suggested by Bahadori (2013) were used for the carbon capture units reboiler. It is recommended that a reboiler operating in the temperature range of 107-127 °C should use saturated steam at a temperature of 135-142 °C, corresponding to a pressure of 3-3.8 bars. The temperature of 135 °C is chosen to prevent excessive skin temperature on the reboiler tubes, which can occur at temperatures exceeding 140 °C. The reboiler operates at the same temperature range in both processes, so the same steam values are used in HPC and MEA processes.

The steam required for the capture unit necessitates modifying the current steam supply network. Pulp mill A uses steam from a steam turbine at various pressure levels. Steam for reboilers could be extracted from all levels but requires lowering pressure and desuperheating. Steam availability and modifications to the steam network have been studied by analysing the mass and energy balances of the back pressure turbine and steam network using IPSEpro process modelling software and pulp mill operating parameters. Results are shown in Figure 16.

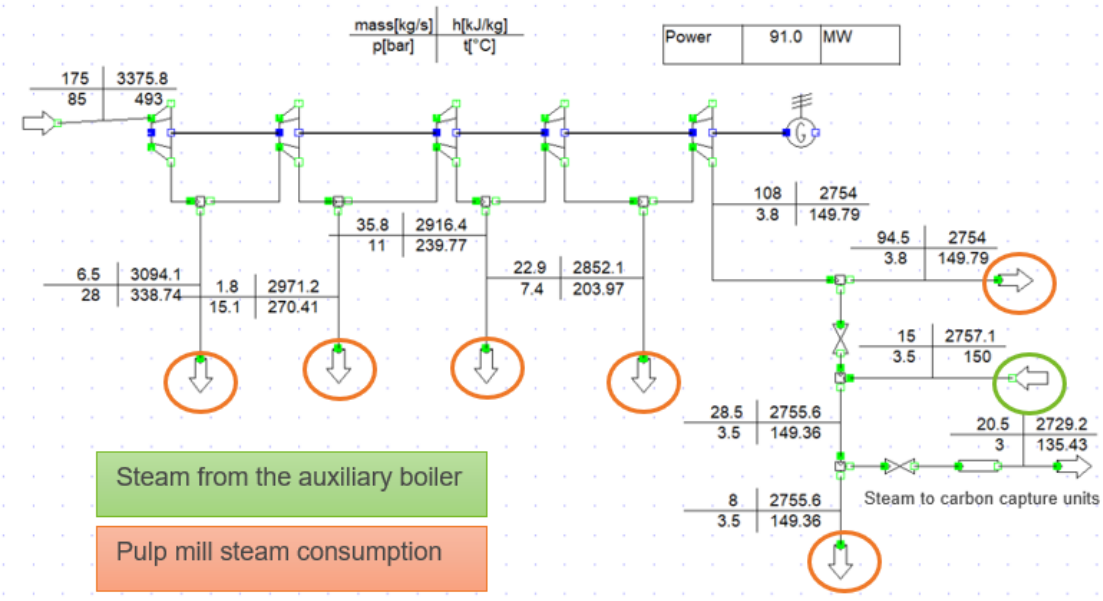

Figure 16. Steam availability at the pulp mill A.

Figure 16 demonstrates the pulp mill's back pressure turbine and steam consumption at various pressure levels. Integrating carbon dioxide capture into the 3.5 bar steam header is the most practical option, as the auxiliary boiler is also connected to it, and integration does not impact back-pressure steam turbine electricity production. An auxiliary boiler can consistently produce a maximum of 15 kg/s of low-pressure steam throughout the year. With this connection, steam availability is 20.5 kg/s if the pulp mill operates at a nominal load. The process parameters for the back pressure turbine are derived from the operational conditions of the pulp mill during its nominal load in the winter season when steam demand at pulp mills reached its peak and did not necessitate additional steam from the auxiliary boilers. It is important to note that the steam production of the pulp mill typically varies based on pulp production and season. Also, steam availability depends on interconnected processes.

The carbon capture simulations start with a DCC. Table 14 presents the properties of flue gas streams before and after the flue gas condenser.

Table 14. Flue gas properties before and after DCC.

Flue gas components	RB FG before DCC	LK FG before DCC	RB FG after DCC	LK FG after DCC
CO_2 [mol-%]	11.4	20.9	13.1	25.3
O_2 [mol-%]	2.6	3.0	3.0	3.7
N_2 [mol-%]	65.6	51.6	77.2	64.4
H_2O [mol-%]	20.4	24.5	6.7	6.6
Mass flow [kg/s]	250	30	224	26
Temperature [°C]	180	290	40	40

Due to water condensation in a DCC, the mass flows of both flue gas streams are reduced, and the molar concentrations of nitrogen, oxygen, and carbon dioxide increase. Process water used for cooling flue gas can be measured by the liquid-gas ratio in mass units. The liquid-gas ratio (L/G) for the recovery boiler DCC was 2.1 kg_{water}/kg_{fg}, and the same ratio for the lime kiln DCC was 2.9 kg_{water}/kg_{fg}. Water consumption for lime kiln flue gases is higher due to the elevated flue gas temperatures, requiring more process water to reach the desired temperature.

The number of parallel absorption units needed to be determined in the carbon capture part for both technologies. This thesis determined the number of parallel units by calculating the

minimum lean solvent rate required for the absorber with theoretical infinity height to achieve a 90% carbon dioxide removal efficiency.

To determine the minimum solvent rate and calculate the diameter, actual flooding velocity and optimal lean solvent need to be selected. The flooding velocity is the highest gas flow rate in a gas-liquid column. Exceeding this rate causes the gas to carry liquid upwards, increasing pressure and reducing separation efficiency. Flooding velocity should not exceeded. Flooding velocity impacts the minimum diameter required for columns. A flooding velocity of 80% of the theoretical value was used in the simulation. A typical design value for packed bed columns is 60-80% (Madeddu et al., 2019).

Loading describes how much CO_2 is absorbed in the solvent. The optimal lean loading for HPC was proposed to be 0.18-0.27 (Gustafsson et al., 2021). In the thesis, 0.21 was used. For MEA, the optimal lean solvent loading varies between 0.2 and 0.4 (Madeddu et al., 2019). In the thesis, lean loading 0.3 was used. Lean solvent loading can be calculated from the following equations for MEA and HPC.

$$L_{\mathrm{MEA}} = \frac{[\mathrm{CO_2}] + [\mathrm{HCO^-}] + [\mathrm{CO_3^{2-}}] + [\mathrm{MEACOO^-}]}{[\mathrm{MEA}] + [\mathrm{MEAH^+}] + [\mathrm{MEACOO^-}]} \tag{22}$$

where L is solvent loading.

$$L_{\mathrm{HPC}} = \frac{[\mathrm{HCO^-}]}{[\mathrm{K^+}]} \tag{23}$$

According to Madeddu et al. (2019), the recommended maximum limit for a single-packed bed-type absorption unit diameter is 12 m. The detailed results are presented in Appendix 6. The results show that only a parallel absorption unit in this case is needed in full carbon capture cases for MEA. The need for parallel units can lead to additional costs due to the higher need for piping and the higher modification needed for the pulp mill. However, this can increase the operational flexibility of the system because it can, for example, increase the minimum capacity of carbon capture at partial load. Using higher pressure in HPC cases enables more compact absorber designs and can lead to lower investment costs required for absorption units.

Carbon capture unit operation is optimised by varying several parameters to achieve low reboiler heat duty per produced CO_2 unit and the targeted capture level and emission levels but with the compact design possible. These parameters include lean flow solvent rate,

reboiler duty, and absorber and stripper unit height. All pumps and compressors were simulated to have 85% and 98% driver efficiency. Table 15 presents the key results of carbon capture units in different cases when a 90% capture rate was achieved.

Table 15. Results of chemical absorption units in different cases.

Variable	HPC RB & LK	HPC RB	HPC LK	MEA RB & LK	MEA RB	MEA LK
Purity [mol-%]	92.5	92.5	92.5	95.0	95.0	95.0
L/G	3.3	3.0	5.1	3.1	3.8	4.5
Lean loading	0.15	0.14	0.16	0.23	0.30	0.19
Rich loading	0.75	0.71	0.81	0.51	0.51	0.51
Reboiler duty [GJ/t_{CO2}]	1.75	1.77	1.67	3.51	3.54	3.47
Electricity demand [GJ/t_{CO2}]	0.91	0.98	0.60	0.04	0.04	0.03
Absorber height [m]	45	40	20	36	58	31
Absorber diameter [m]	7.6	7.1	2.7	8.9	12	4.4
Stripper height [m]	25	20	13	15	30	15
Stripper diameter [m]	7.5	6.8	3.0	5.3	6.8	3.1

The results of Table 15 show that both processes can produce high CO_2 product purities (99.9%-dry) since most of the substances remaining in the product stream are water. From the energy consumption point of view, solvent regeneration in the HPC process requires significantly less energy to regenerate solvent than in the MEA process with similar carbon capture efficiency. Lower energy consumption results mainly from using a steam vapour compressor, which considerably reduces energy consumption in the stripper. However, it should be noted that the reboilers' heat duty does not account for the compressors' electricity consumption.

Electricity consumption is higher for the HPC process, mainly due to the flue gas compressing and using a vapour compressor. In the MEA process, the only electricity consumption comes from the flue gas booster fans in DCC and pumps required in the capture process, which consume significantly less electricity than compressors in HPC. However, total energy consumption is still at a lower level than that of the MEA process.

Energy consumption decreases as the partial pressure of CO_2 increases in flue gases, resulting in the lowest specific energy consumption when carbon capture is integrated into a lime kiln. In the HPC process, higher CO_2 partial pressure in flue gas leads to increased

energy consumption because flue gas CO_2 partial pressure affects rich loading more in HPC than in the MEA process. These results also indicate that combining the flue gas flows from the recovery boiler and lime kiln can give benefits compared to only recovery boiler carbon capture from an energy point of view because combining increases the CO_2 partial pressure.

Operating values and energy consumption are compared to those found in the literature. In the literature for power plant applications, for MEA, the heat duty has typically been between 3-5 GJ/t_{CO2}, with loading values for lean solvent between 0.2-0.4 and rich solvent between 0.4-0.5 (Romeo et al., 2020). The findings imply that the simulated MEA process closely corresponds to the values commonly observed in real-world power plant applications.

The HPC process has less information available due to its lower technological maturity. However, the results are in the same range when comparing reboiler duties with modelled HPC processes found in the literature. Gustafsson et al. (2021) had results of reboiler duty around 2.2 GJ/t_{CO2} with a similar vapour compressor connection and without around 4.4 GJ/t_{CO2}. However, they used a higher L/G ratio and lower absorption pressure, which can explain differences in results. Typically, a higher L/G ratio and lower absorption pressure increase heat duty in the reboiler. Another study by Chuenphan et al. (2022) suggested that the optimal reboiler duty for the HPC process is approximately 2.2 GJ/t_{CO2} without vapour compression, indicating that the achieved results for reboiler duty fall within the appropriate range. In reference studies, lean solvent loadings are typically 0.18-0.27, and rich solvent loadings are around 0.8-0.81 (Gustafsson et al., 2021). These indicate that the results give a good approximation for real operating values.

Carbon capture unit dimensions show that the HPC process generally has a smaller absorber height due to the higher operating pressure in the absorber and the lack of a wash section. However, the required land area for the absorber and stripper is similar in both processes due to small differences in the absorber and stripper diameters.

Due to the fixed capture rate, sensitivity analysis for the capture units was performed using other capture rates. Varying reboiler heat duty and its impacts on carbon capture rates and heat duties per captured CO_2 were simulated for the MEA and HPC processes, and their results are presented in Figures 17 and 18.

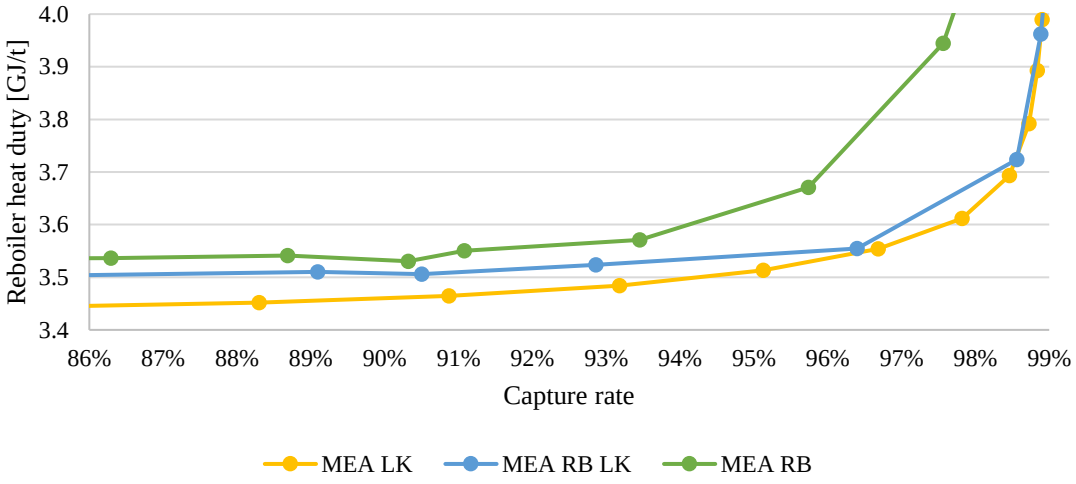

Figure 17. MEA capture rate impact on heat duty.

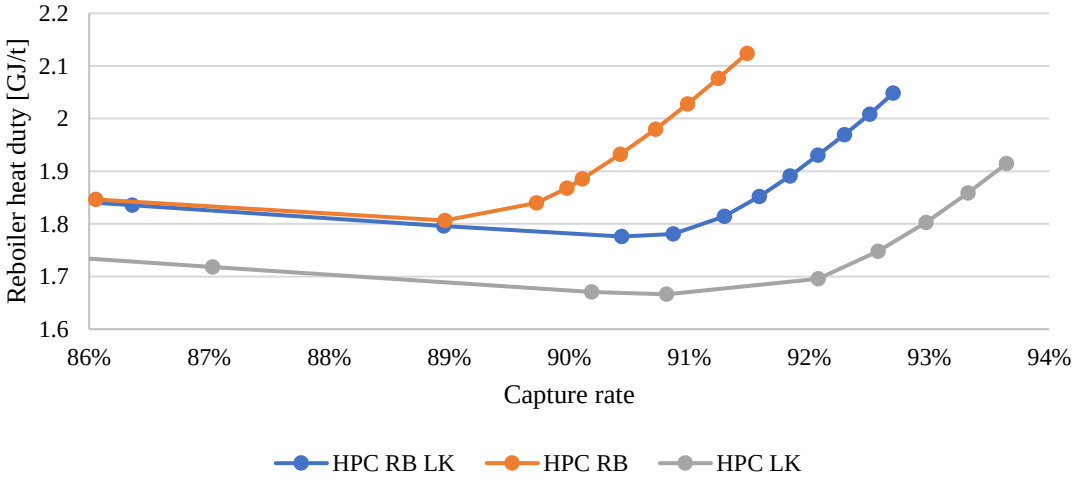

Figure 18. HPC capture rate impact on heat duty.

In the MEA process, the specific energy consumption remains stable until a 90% capture rate. After exceeding this capture rate, specific energy consumption increases in all cases. Specifically, specific energy consumption experiences an exponential increase once the capture rate exceeds 95%. The increase in specific energy consumption is stronger in capture cases with lower carbon dioxide concentrations in flue gas, as regeneration requires more heat than higher CO_2 concentration flue gas cases.

In the HPC process, specific energy consumption decreases until the 88.5-91% capture rate and then increases sharply, clearly showing the optimal point for carbon capture. In the HPC process, specific energy consumption varies more strongly as the recovery rate increases compared to MEA cases. The optimal capture rate also changes slightly as the CO_2 partial pressure in flue gas changes, which did not occur with the MEA cases. In carbon capture cases where CO_2 concentration is higher in flue gas, the optimal recovery rate is also higher, and the increase in energy consumption is more moderate as the recovery rate increases compared to cases with lower CO_2 concentrations in flue gas.

However, from the sensitivity analysis, the 90% recovery rate used for comparability is a reasonable approach for this study. However, when designing a capture unit, setting the design point at optimal specific energy consumption is important to minimise variable costs. From the charts, it can be concluded that the optimal recovery rate for MEA is between 90-95%, depending on the properties of the flue gas in different point sources and carbon capture goals. The optimal carbon capture rate for HPC processes is 88.5-91%, depending on flue gas properties and capture rate goals in different point sources.

After the carbon capture, the liquefaction process is simulated. All compressors are modelled with 85% and 98% driver efficiency in the liquefaction part. Table 16 presents key results from the liquefaction plant.

Table 16. Liquefaction plant results.

Before liquefaction:	RB & LK HPC	RB HPC	LK HPC	RB & LK MEA	RB MEA	LK MEA
CO_2 [mol-%]	92.5	92.5	92.5	95.0	95.0	95.0
O_2 [ppm]	34	33	20	94	119	94
H_2O [mol-%]	7.5	7.5	7.5	4.9	4.9	4.9
After liquefaction:						
CO_2 [mol-%]	99.96	99.96	99.98	99.86	99.82	99.90
O_2 [ppm-mol%]	7.2	6.4	5.6	7.3	9.9	5.9
H_2O [ppm-mol%]	12	28	28	12	29	29
Electricity [kWh/t_{CO2}]	135	135	135	122	122	122
Hydrogen [kg/h]	0.5	0.5	0.1	1.6	1.8	0.3
Total capture rate [%]	87.7	87.6	87.0	87.8	87.8	87.1
Captured CO_2 [kt/a]	1450	1190	250	1450	1190	250

The results show that MEA has higher oxygen process purification requirements but lower needs for water removal. This indicates that more water vapour is released during regeneration in the HPC process. However, the selected stripper condenser temperature in simulation can slightly impact the water content before liquefaction. Rich solvent flashing during carbon capture in HPC explains the reduced need for oxygen removal. However, even if hydrogen consumption is higher in the MEA process, it is very small compared to handled CO_2 amounts.

Both technologies can produce high-purity liquified CO_2 after purification and dehydration, fulfilling the purification criteria with the correct pressure and temperature recommended by Northern Lights (Northern Lights, 2024). Also, CO_2 is pure enough to be used in CCU applications when food-grade is used as a reference quality.

There is also a clear trend of higher compressor electricity demand with lower CO_2 concentration streams after the carbon capture unit due to lower product density. However, compressor electricity demand differences are small and do not significantly impact the electricity consumption of the entire carbon capture chain. Compressor electricity demand corresponds to typical chemical absorption liquefaction-plants electricity demands, typically 100-140 kWh_e/t_{CO2} for post-combustion applications, depending on impurity levels and requirements (Abbas et al., 2013b). From total carbon capture rates, it can be seen that small carbon dioxide losses happen during the other processes, even if the carbon capture rates in the carbon capture unit were 90%. Small losses mainly occur in DCC, where small amounts of CO_2 are dissolved into condensate.

Figure 19 illustrates the absolute energy requirements for electricity, heat, and cooling in power units. Meanwhile, Figure 20 displays the specific energy consumption per liquefied CO_2 produced for all cases, from DCC to the liquefaction plant.

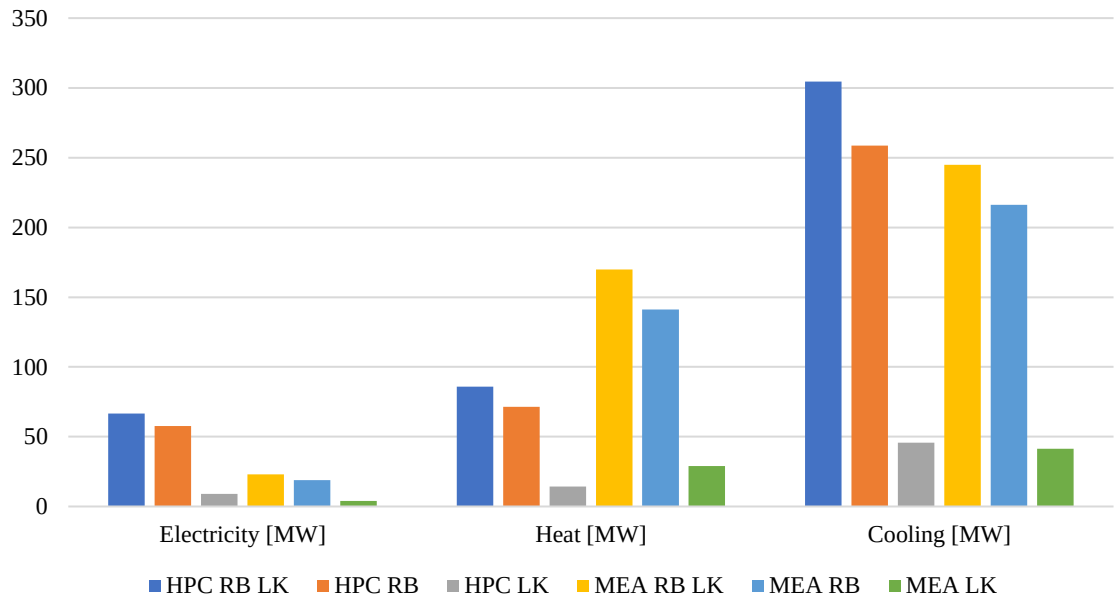

Figure 19. Power, heat and cooling demand different carbon capture cases.

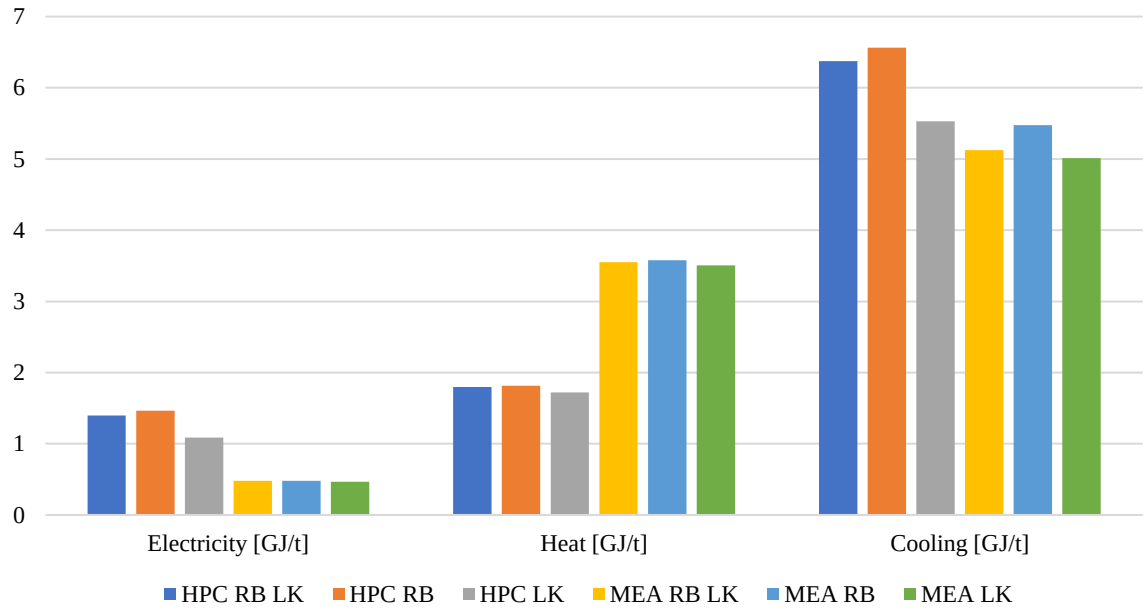

Figure 20. Total energy demands in different carbon capture cases.

The figure demonstrates that the pulp mill will shift from a net electricity producer to a net consumer regardless of the technology when carbon capture is integrated into a recovery boiler. Figures 19 and 20 show that the HPC process has a higher impact on electricity consumption and requires more electricity from the grid due to the compression needs of the

capture unit. However, for the same reasons, heat consumption is smaller for the HPC process, making it an interesting option due to the limited excess steam at a pulp mill.

The highest heat demands in both processes occur in the reboiler of the strippers, and energy consumption savings in reboiler duty can lead to significant energy and variable cost savings in the process. Cooling demands are higher for the HPC process because compressed flue gas needs to be cooled to absorption temperatures, resulting in higher cooling demands than in the amine process. However, the HPC process offers the potential for heat utilisation at higher temperatures, enabling utilisation, for example, in district heat production or pulp mills' hot or warm water heating needs.

Recoverable heat can be approximated from simulated coolers in the Aspen model. In this thesis, only streams cooled to 50°C or higher are considered when analysing the amount of recoverable heat from the stream coolers. Recoverable heat from all capture part processes is assessed, and the results are shown in Table 17.

Table 17. Recoverable heat at the pulp mill.

Heat source	HPC RB & LK	HPC RB	HPC LK	MEA RB & LK	MEA RB	MEA LK
DCC [MW]	44	36	7	44	36	7
Capture unit [MW]	122	102	17	0	0	0
Liquefaction [MW]	5	4	1	5	4	1

In the simulated MEA process, recoverable heat can only be utilised from DCC condensate streams. However, if heat utilisation is integrated into the absorber or stripper, higher value heat utilisation can be possible, but this was not analysed in this thesis. The HPC process has more potential for utilisable heat sources. In addition to DCC, most heat can be utilised from flue gas coolers before the absorber, where high-temperature heat (>80 °C) is available and from the lean solvent cooler, where moderate-level heat is available (60-80 °C). Also, added potential can be if heat utilisation units are integrated into the absorber or stripper. A simulated liquefaction plant has low potential to be considered utilised at pulp mills in all cases due to the small amounts of heat available from compressor intercoolers to be utilised.

When all the heat from the high-temperature sources can be utilised, less cooling water will be needed compared to when cooling demand is fulfilled by using cooling water. However, using all waste heat is not necessarily techno-economical and should be assessed separately

for heat sources. This thesis assumes that all valuable heat can be used at pulp mill processes in district heating or process heating. The assumption is made to calculate utility consumption. Increases in utility consumption of electricity, steam, and water in different cases are presented in Figure 21.

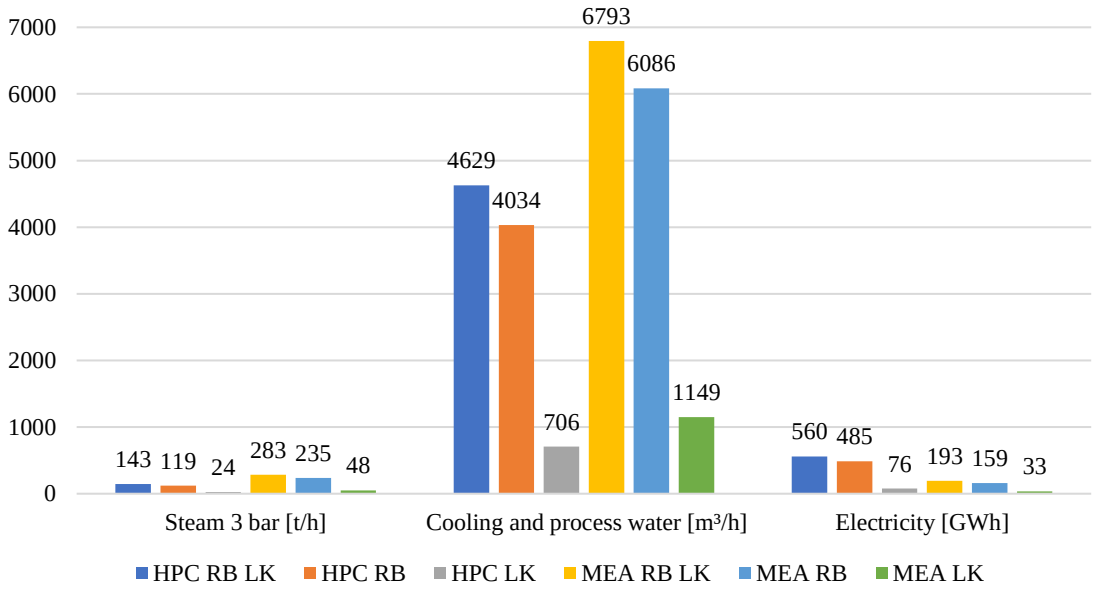

Figure 21. Utility consumption increases at pulp mill A after carbon capture is integrated.

If the full carbon capture potential is utilised, the carbon capture process necessitates more steam availability when capturing carbon dioxide from the recovery boiler. Without additional investment in an auxiliary steam boiler or increased steam savings, the current steam availability (20.5 kg/s or 73.8 t/h) will limit maximal utilisation in the recovery boiler or lime kiln cases, as combining flue gas streams is no longer practical. Maximal annual CO_2 capture can be achieved when carbon capture is integrated into a recovery boiler. The maximum yearly captured CO_2 with 90% capture efficiency, if not invested in a new auxiliary boiler, is presented in Figure 22. Captured yearly CO_2 is calculated from MEA and HPC reboiler heat duties and steam heat value.

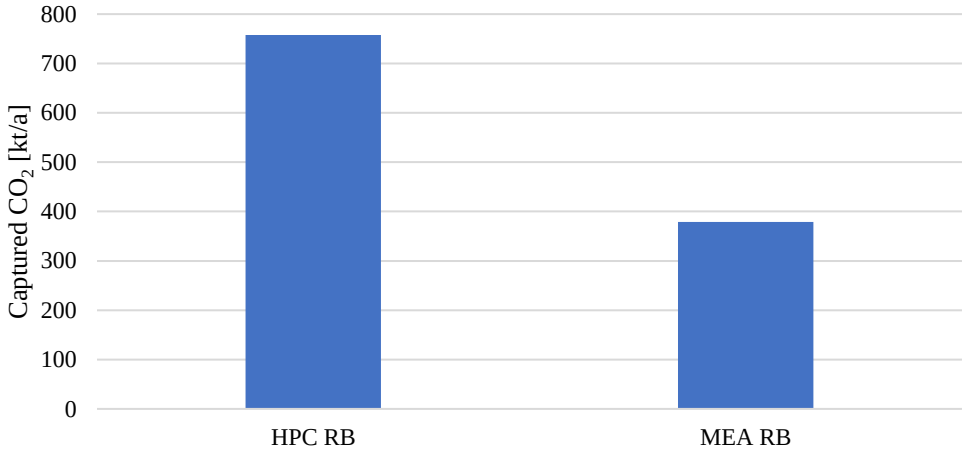

Figure 22. Maximal annual captured CO_2 if not invested in a new auxiliary boiler.

Suppose a new auxiliary boiler is not installed. Only 46% of the pulp mills' full carbon capture potential can be achieved using the HPC process, and 23% using the MEA process when the recovery boiler and lime kiln are considered point sources. If full carbon capture potential is utilised in a pulp mill, in the MEA recovery boiler cases, a steam boiler with almost double capacity is needed compared to the HPC process with maximal steam use. However, According to Navedkhan et al. (2022), the HPC process is commonly integrated with a steam generation unit, utilising heat from the capture unit to reduce low-pressure steam consumption; therefore, steam consumption can be significantly lower.

When integrating carbon capture, cooling water demand will increase significantly, potentially requiring adjustments to the current system or establishing a separate circulation system. In the lime kiln case, integration leads to a 9% increase in water consumption with HPC and 14% with MEA. Similarly, there was a 50% increase with HPC and 76% with MEA for recovery boiler cases. For the combined cases, there was a 58% increase with HPC and an 85% increase with MEA. In the HPC process, water consumption may increase if all the recoverable heat is not effectively utilised. Also, water consumption can be higher if the return temperature of the cooling water needs to be lower than selected for analysis.

Increased power consumption will lead to a significant need for electricity from the grid, posing a potential risk for electricity price changes. Net electricity production is around 150 GWh without carbon capture, which will change to net negative when recovery boilers' carbon dioxide is captured. In lime kiln cases, net electricity production remains positive.

9 Techno-economic analysis of carbon capture in kraft pulp mill

Economic analysis is conducted for the HPC and MEA carbon capture process in selected cases, considering that the full potential in each case is utilised, including the liquefaction in the analysis scope. Transport and possible CO_2 storage or utilisation are excluded from the analysis. Also, economic analysis does not consider cost savings and profits from waste heat utilisation in the carbon capture process. Cost analysis also excludes additional costs due to the need for parallel units in the MEA full carbon capture case. In this analysis, the cost of carbon capture is determined for both technologies based on utility consumptions in simulation and investment cost data based on literature by scaling investment cost to the right scale. Scaling is done by following the equation.

$$\frac{I}{I_0} = \left(\frac{S}{S_0}\right)^n \tag{24}$$

where I is the scaled investment cost (€), I_0 is the reference investment cost (€), S is the target capacity (t/h), S_0 is the reference capacity (t/h), and n is the scaling factor.

The carbon capture unit and CO_2 liquefaction plant were scaled using a scaling factor of 0.7. According to Sinnott & Towler (2020), a scaling factor of 0.7 is commonly used to scale capital costs in petrochemical applications.

The cost data is adjusted to match the 2024 Q1 prices using the chemical engineering plant cost index (CEPCI). This is accomplished by multiplying the cost component by the ratio of the 2024 Q1 CEPCI to the CEPCI of the reference year. The cost index in 2024 Q1 was 800.7 (CEPCI, 2024). The Euro currency is assumed to be Euro =1.1 U.S. Dollar and Euro = 0.83 British pound. The cost data information and data references are presented in Appendix 7. In this thesis, the investment costs of the capture unit also include a DCC and its installation.

The economic analysis assumes that the HPC process has a steam regeneration unit that generates low-pressure steam by utilising available heat from the capture process, which reduces steam consumption so that it needs only 40% of the reported maximum steam consumption. When this assumption is used, an additional auxiliary boiler is unnecessary for HPC processes. In the MEA process, an additional boiler is needed in the full carbon capture

and recovery boiler cases. Additional boiler cost estimations are made using the equation proposed by Sinnott & Towler (2020).

$$I_{\mathrm{b}} = a + b \cdot S^{n} \tag{25}$$

where I_{b} is the purchased cost of the boiler ($), a and b are cost coefficients.

The cost constant a is $110 000, and b is 45. The scaling factor used was 0.9 (Sinnott & Towler, 2020). The installation cost was assumed to be 300% of the equipment purchase cost. The cost of carbon capture in this thesis is determined by the levelized cost of carbon capture (LCOC). LCOC is calculated using the following equation:

$$\mathrm{LCOC} = \frac{K_{\mathrm{cap}} + K_{\mathrm{O\&M,fixed}} + K_{\mathrm{variable}}}{q_{\mathrm{m,LCO_2}}} \tag{26}$$

where, LCOC is the levelized cost of carbon capture (€/t_{CO2}), K_{cap} is annual capital costs with fixed depreciation (€/a), $K_{\mathrm{O\&M,\ fixed}}$ is the sum of annual fixed operation and maintenance costs (€/a), and K_{variable} is the sum of annual variable costs (€/a) and $q_{\mathrm{m,LCO2}}$ is produced liquified CO_2 annually (t_{CO2}/a).

Capital, operational and management costs and variable costs are presented in detail in Appendix 8. Annual capital expenditures (CAPEX) consist of annual capital charges on total plant costs, which include the investment costs of capture and liquefaction plants, modifications and changes made to the pulp mill. The cost of changes to the pulp mill includes, for instance, auxiliary boilers, ducting and piping. A 10% contingency is also added to these costs. CAPEX includes other capital costs, including spare parts, start-up and commissioning costs, owners' costs, working capital and annual interest during construction. An interest rate of 8% and a lifetime of 25 years was assumed for capital recovery factor calculation.

Operational expenditures (OPEX) include fixed operation and maintenance costs and variable operational costs. Fixed costs include maintenance and labour costs for capture and liquefaction plants. Maintenance costs were assumed to be 3% of the total plant costs, and the additional labour needed was estimated to be 20 people. Indirect labour costs are added and assumed to be 40% of labour costs, including health care insurance, pensions, and other insurances. Variable operational costs include utility, chemicals, process and cooling water consumption. Chemical consumption of MEA is assumed to be a make-up flow in simulation, added by its degradation rate of 1.6 kg/t_{CO2} (Chuenphan et al., 2022). The

consumption of K_2CO_3 is assumed to be 10% of the MEA degradation rate. A study by Kumar et al. (2023) found that chemical make-up flow is around 10% smaller in the HPC process. Process water equals the water inlet flows used in DCC and make-up flows. Also, the wastewater and MEA sludge disposal costs are included in variable costs. MEA sludge disposal amounts are assumed to be the same as the degradation rate. The LCOC, CAPEX and OPEX distribution are presented in Figure 23.

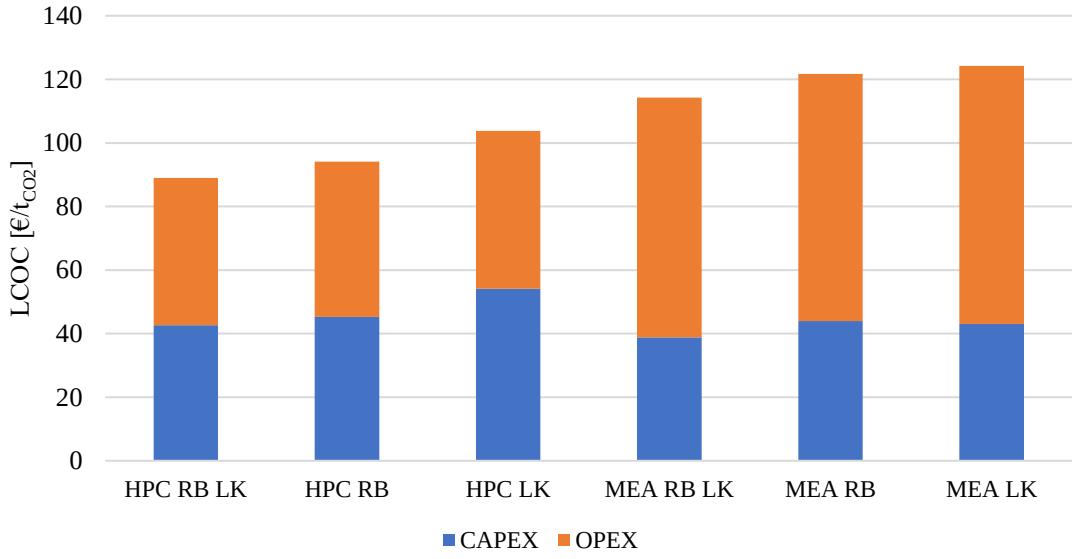

Figure 23. Cost of carbon capture, including liquefaction.

Figure 23 demonstrates that the HPC process is the most cost-effective option across all examined cases. Costs are generally higher than those of previous techno-economic studies, mostly due to general cost increases. Cost indexes have been increased by around 40%, for example, from a techno-economic study by Onarheim et al. (2017b), where costs for similar cases with MEA technology were 62-83 €/t_{CO2}. The unit cost of carbon dioxide capture consistently remains at its lowest in the combined flue gas cases, where the scale advantage significantly influences the profitability of carbon capture. In addition, results show that investing in one high-capacity absorption unit for a recovery boiler and lime kiln flue gases has a lower total CAPEX than investing in those separately with two different capacity absorption units.

In the MEA process, there is no significant difference between the recovery boiler and only lime kiln cases. The difference is low because recovery boiler cases need additional

investments in the auxiliary boiler to produce extra steam to meet reboiler steam needs. Also, the cost difference between a full capture case and only recovery boiler cases can be smaller due to additional costs due to parallel units in a full carbon capture case.

Cost distribution between CAPEX and OPEX costs are more evenly distributed in the HPC process, with capital costs accounting for a greater share of the total costs than in the MEA process. This allocation of higher capital costs in the HPC process is likely attributed to the increased requirement for flue gas pretreatment, alongside greater investments in heat recovery and steam recirculation systems. Higher variable costs in the MEA process are due to higher steam demand in reboiler and higher chemical and disposal costs. Lowering the steam consumption even slightly in amine processes can reduce overall costs significantly.

Carbon capture costs for both processes can be reduced if carbon dioxide is not liquefied but pressurised to make it suitable for pipeline transport (Onarheim et al., 2017b; Teir et al., 2011). However, Finland currently has no pipeline transport network for carbon dioxide, so liquefaction is mandatory. The process cost structure in CAPEX and OPEX exhibits some variances between technologies. Figure 24 presents a detailed CAPEX cost distribution.

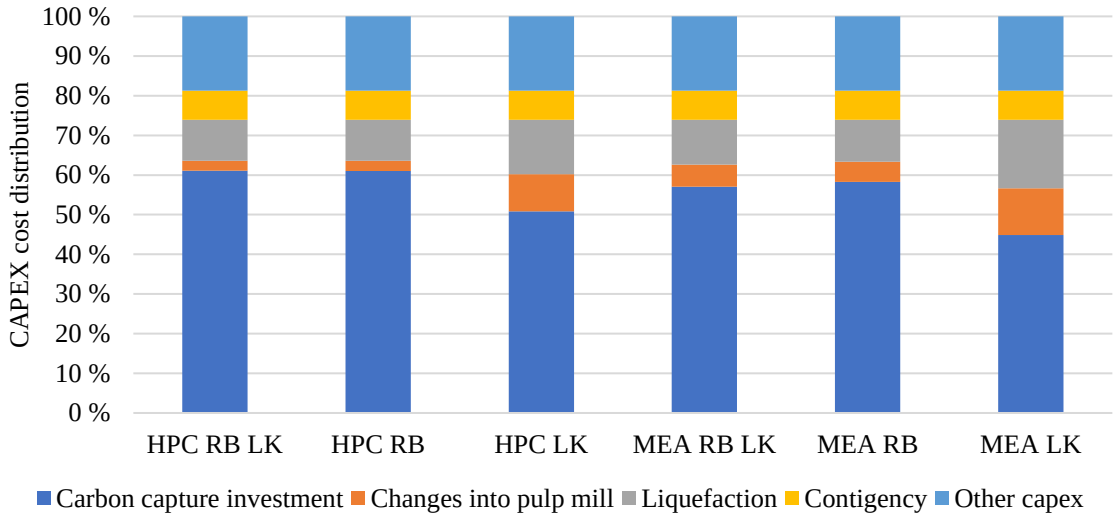

Figure 24. CAPEX cost structure in MEA and HPC.

Capex cost distribution is similar between technologies. In both technologies, costs primarily consist of the investment costs of carbon capture units. The investment cost of a carbon capture unit mainly determines CAPEX and a low investment cost is crucial for reducing

project overall expenses. Other important costs come from liquefaction plant investment and other capital costs. OPEX cost distributions are presented in Figure 25.

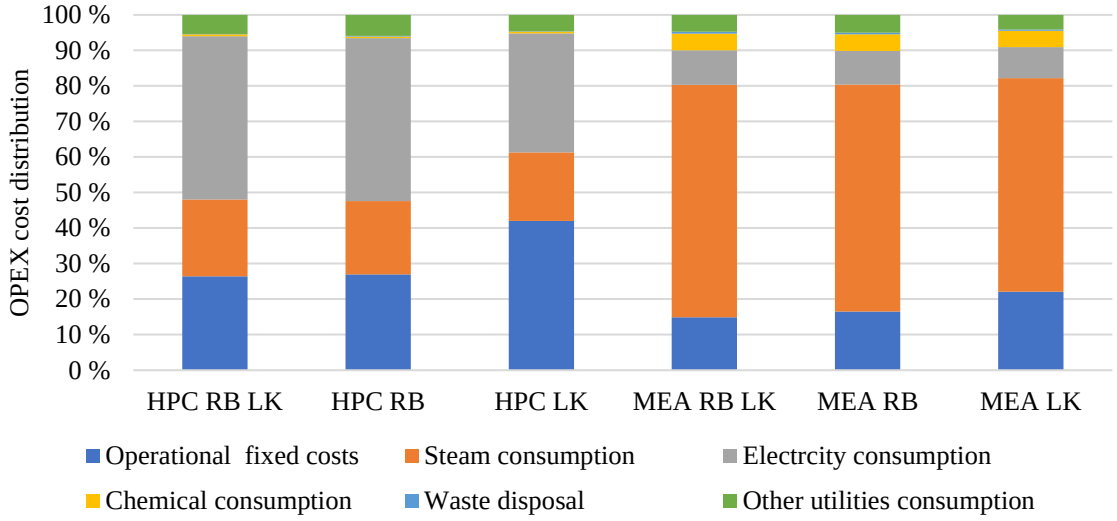

Figure 25. OPEX cost structure in MEA and HPC

In OPEX, cost distributions are more diverse. In both processes, energy consumption dominates operational costs but is structured differently. In HPC, costs are dominated by electricity consumption, while in MEA, energy costs are dominated by steam consumption. In addition, chemical costs in MEA are significantly higher than in HPC, impacting operational cost distribution variance between technologies.

Given the uncertainties in investment costs and the potential variability of the most important utility consumptions, a sensitivity analysis is conducted. This analysis involves varying the investment cost of both processes by ±30% and adjusting the cost of electricity and steam for both processes since they had the highest impacts on CAPEX and OPEX. The goal is to establish a cost range for carbon capture with reasonable accuracy. There are uncertainties about the investment cost of the HPC process, particularly due to its lower technological maturity. Electricity prices fluctuate yearly, and steam costs vary depending on the production method. The sensitivity analysis for MEA processes is presented in Figure 26.

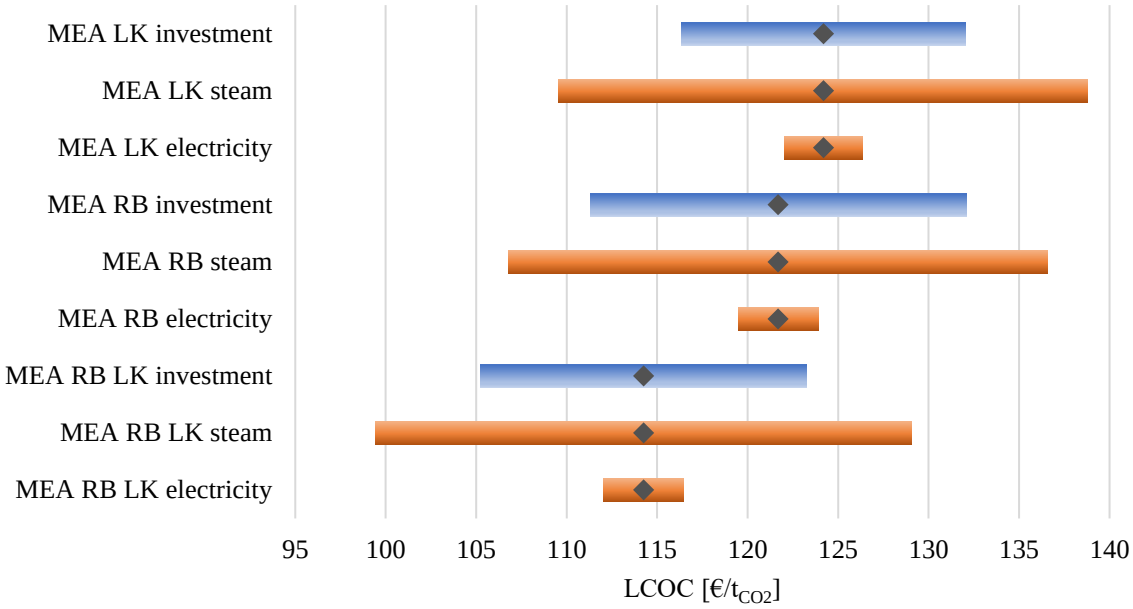

Figure 26. MEA sensitivity analysis: Orange is used for OPEX costs, blue is used for CAPEX costs, and grey mark is used for the base case.

In the MEA process, the most significant cost impact is the price and consumption of steam, which contributes the most to variable costs. However, there is considerable potential for cost savings in the MEA process. Notably, significant cost reductions can be achieved when low-cost steam is available, or steam consumption in the reboiler is slightly reduced when selecting a carbon capture process based on amine solvents. Steam savings can be achieved if more advanced amine solvents are used with lower specific energy consumption. Another crucial factor that significantly influences the cost of carbon capture is the investment cost. When an amine carbon capture process is selected, these factors have the most significant impact on final costs. After sensitivity analysis, the cost of carbon capture for the recovery boiler and lime kiln case is between 100-130 €/t_{CO2}; only the recovery boiler case is between 108-137 €/t_{CO2} and only the lime kiln case between 111-140 €/t_{CO2}, primarily depending on steam and investment cost. The same analysis is also conducted for the HPC process and is presented in Figure 27.

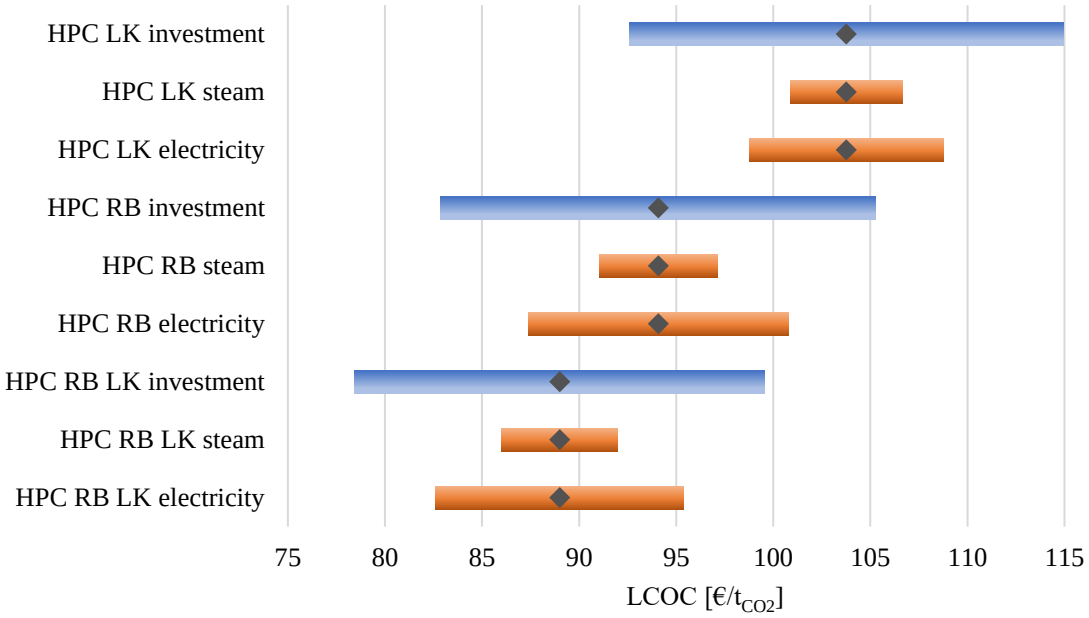

Figure 27. HPC sensitivity analysis: Orange is used for OPEX costs, blue is used for CAPEX costs, and grey mark is used for the base case.

While investment costs have the greatest impact on costs and uncertainty in the HPC process, they do not overshadow its competitive cost-effectiveness. Even when investment costs are 30% higher, the HPC process remains competitive compared to the MEA process. Varying energy costs impact the total costs of the HPC process less than MEA due to a lower share of variable costs on the total costs of carbon capture. In the HPC process, electricity costs impact total costs more than steam costs since regeneration is based more on electricity than utilising low-pressure steam. The HPC process may offer greater advantages for integration into pulp mills, particularly in Nordic countries where the cost of electricity is low compared to other European countries (Eurostat, 2024). From the analysis, the unit cost of carbon capture for carbon capture from the recovery boiler and the lime kiln is between 79-100 €/t_{CO2}. For the recovery boiler case, costs are between 84-106 €/t_{CO2}, and for only the lime kiln case, 94-116 €/t_{CO2}.

10 Conclusions

The first objective of the thesis was to study potential carbon capture technologies and their integration into Finnish kraft pulp mills. The study identified numerous technological options for different carbon dioxide sources in pulp mills, with most technologies still developing, and therefore, limiting the integration possibilities to only a few options.

Based on the criteria defined in this thesis, it was determined that two technology options are currently feasible for integration into a pulp mill due to their high maturity (TRL 8-9). These technologies are HPC using potassium carbonate-based solvents and chemical absorption using amine solvents. Of these two, HPC is the most promising technology for integration into a pulp mill. HPC outperforms amine-based technology in terms of costs, energy balance, and environmental impacts. However, it is worth noticing that amine-based technology remains a strong option for carbon capture as the only commercially available system among the examined technologies.

The potential for developing technologies to reach a commercial level in the future was assessed. The analysis indicated that in the future, calcium looping, adsorption technologies, chemical absorption based on potassium carbonate or aqueous ammonia solvents, and MCFC show potential for widespread. From these, the most promising technologies for integration into pulp mills are MCFC and calcium looping due to their potential to reduce capture costs compared to amines and have a minimal impact on pulp mill's electricity consumption and steam availability. However, adsorption and potassium carbonate-based technologies also demonstrate high potential for integration due to minor differences in analysis. There are uncertainties about the large-scale performance of these technologies under the flue gas conditions of a pulp mill because of low technological maturity.

It is essential to update the results within 5-10 years to identify possible changes in promising emerging technologies and determine whether other integration options are available besides HPC or amine technologies. This is important because thesis results indicate that especially capture costs could be reduced in the future compared to current integration options.

One objective of the thesis was to determine the impacts of carbon capture integration. The thesis analysed the impacts of integrating carbon capture into a Finnish pulp mill using two

chemical absorption technologies: HPC based on potassium carbonate solvent and an amine-based carbon capture process with an MEA solvent. The integration was examined for three carbon capture cases: flue gas from the lime kiln and recovery boiler (RB & LK), only from the recovery boiler (RB), and only from the lime kiln (LK). The analysis of integration demonstrated that both technologies could be successfully integrated into pulp mill point sources with high separation efficiency. Additionally, both technologies can produce high-purity CO_2. However, the findings of this thesis indicate that MEA achieves higher CO_2 separation efficiencies with a reasonable increase in energy consumption. Specifically, MEA can capture up to 95% of CO_2 with a slight specific energy consumption increase, while the HPC process exhibits an optimal range between 88.5-91%.

The analysis revealed that in MEA capture technology, where absorption occurs at low pressure, installing an additional parallel absorption unit might be necessary when capturing carbon dioxide from the recovery boiler and lime kiln with a single absorption unit. This parallel unit could lead to higher retrofit costs than with the HPC process, which does not require a parallel system due to higher absorption pressure. In pulp mills with larger recovery boilers than in pulp mill A, a parallel unit might also be needed in cases where CO_2 is captured only from the recovery boiler with MEA. However, these modular solutions can enhance operational flexibility by enabling running capture units with lower total loads.

The analysis found that the highest impacts of a carbon capture system on the pulp mill are increased steam, electricity, and water consumption. Consumption increases lead to possible modifications to the water circulation system and steam network and a need for steam savings or investment into a new boiler when carbon capture is integrated into the recovery boiler. In addition, in the case of recovery boiler, integration leads to net-negative electricity production in pulp mills.

The MEA process has a greater impact on water consumption than HPC. However, an analysis needs to be conducted about effectively utilising heat in the pulp mill. HPC's cooling water consumption will significantly increase if valuable heat is not utilised effectively.

Results showed that HPC is a more energy-efficient solution, with lower impacts on steam consumption than MEA. Lower steam consumption in HPC enables a higher maximal capture rate from considered point sources compared to MEA without investment in an additional auxiliary boiler. However, the maximum capture potential in both processes

without boiler investment is far from the full potential of carbon capture in pulp mills. Due to these reasons, steam savings are crucial in pulp mills. Steam savings can be achieved, for example, by reducing hot water, warm water or district heating steam needs by utilising valuable heat from the capture unit. These heat utilisation possibilities and their suitability should be considered separately. Also, investing in an auxiliary boiler should be considered when steam availability limits carbon capture. Even if HPC has a lower steam consumption, it has much higher electricity consumption than MEA. Higher electricity use in HPC increases the risk of exposure to electricity price changes.

Integrating carbon capture into a recovery boiler in an assessed pulp mill enables large-scale BECCS or both BECCS and BECCU since the captured biogenic CO_2 amounts are over 1 Mt CO_2. When integrating carbon capture only to the lime kiln, captured CO_2 in the assessed pulp mill is more suitable for BECCU since the captured amounts match the typical needs of CCU applications. Furthermore, in all evaluated scenarios, even when captured CO_2 is utilised in the pulp mill, there remains a sufficient amount of CO_2 for use in other applications. These findings underscore the significance of carbon capture in Finnish pulp mills for facilitating a hydrogen economy or achieving the potential for negative emissions in Finland.

The last objective of the thesis was to determine the costs of carbon capture for MEA and HPC technologies. Generally, the techno-economic analysis indicates that capturing CO_2 at a larger scale reduces the cost per unit of CO_2 for both technologies. Capture scale benefits hold even in cases where an additional auxiliary boiler is required for MEA. Additionally, the analysis suggests that capturing CO_2 from a single, larger capacity absorption unit in a pulp mill results in lower CAPEX than capturing CO_2 from multiple capture units with different capacities. Cost analysis also showed that energy consumption has the highest impact on operational costs, and energy consumption minimisation is the most important factor for reducing capture costs with chemical absorption technologies. Another important thing that could reduce the cost of capturing carbon dioxide is implementing a CO_2 pipeline network in Finland. Building a CO_2 pipeline network will reduce the capture unit owner's capture costs by opting for CO_2 compression and purification rather than the more expensive CO_2 liquefaction.

The techno-economic analysis of the compared technologies showed that HPC had the lowest cost in every case. However, the cost difference between HPC and amine-based

carbon capture can be smaller if more advanced amine solvents can reduce steam consumption compared to MEA and if HPC has a higher investment cost. Higher investment costs are possible since HPC still has a few techno-economic analyses in the literature, leading to increased uncertainties regarding estimated CAPEX for capture units. Obtaining quotes from technology providers is recommended to improve the accuracy of CAPEX estimations for these technologies. However, even if the investment cost of HPC increases by 30%, it is still a more cost-effective solution due to lower energy consumption. In addition, conducting a techno-economic analysis to determine the potential cost savings from utilising the valuable heat produced by capture units in pulp mills is crucial. This analysis will help to understand how much heat utilisation can reduce the capture costs. If the utilisation of secondary heat is profitable, the cost gap between HPC and MEA will also widen because HPC produces more valuable heat than MEA, and the quantity of this heat is greater.

The thesis results indicate that no definitive technology for carbon capture has been identified as the winner. However, in the future, we may see technologies emerge commercially that challenge amine-based chemical absorption. According to this study, the HPC process could be one of these potential technologies. Calcium looping and MCFC also present promising future integration alternatives.

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Appendix 1. Comparison of chemical absorption solvents (IEAGHG, 2019; Linjala & Kajolinna, 2023; Rackley, 2017; Rezaei et al., 2023).

Technology	Advantages	Disadvantages	TRL
Amine-based systems	Commercialised technology. Low capital investment. A lot of different solvents.	Low amine concentrations to resist corrosion also limit loading capacity. Hazardous degradation products. Low tolerance of SO_x, NO_x, and O_2 in flue gas. Solvent loss due to mist formation High regeneration energy demand 2.5-3.8 GJ/t_{CO2}	6-9
Phase change solvents	Higher solvent loading capacity regeneration at higher pressure. Lower temperature reduces energy requirement (2.2-2.4 GJ/t_{CO2}).	Typically, higher loaded solvent viscosity than amines, including higher process pressure drops.	5-6
Aqueous ammonia	Low regeneration energy requirement (2.4-3 GJ/t_{CO2} (Liu et al., 2020, p. 283). Lack of solvent degradation during the absorption and regeneration cycle. Ammonia can remove NO_x and SO_x from flue gas streams. Tolerant for NO_x and SO_x Low solvent cost Sellable ammonium sulfate and ammonium nitrate fertiliser by-products	High solvent volatility requires reduced operating temperature. High solvent loss during regeneration at high temperatures.	6-7
Potassium carbonate-based	Higher solvent loading capacity than amine-based. Lower corrosivity compared to amine-based. Tolerant for NO_x and SO_x Lower reboiler heat duty with promoters 2-3.2 GJ/t_{CO2} (Borhani et al., 2015, p. 158). 0.7-1.5 GJ_e/t_{CO2} (GCI, 2023). Regeneration is possible at low temperatures, 75-85 ℃ It can regenerate using only electricity (Linjala & Kajolinna, 2023).	Without promoters, the reaction rate is slow, and the reboiler heat duty is high (5.3 GJ/t_{CO2}) (Borhani et al., 2015, p. 158). High cost of promoters. System fouling due to reaction product precipitation.	7-8
Water-lean solvents	Lower regeneration energy demand compared to MEA (2-2.8 GJ/t_{CO2}) due to lower regeneration temperatures < 100 ℃ (Heldebrant et al., 2017). Faster absorption compared to MEA (Heldebrant et al., 2017).	Requires the addition, such as mechanical filtration, solution reclamation, or antifoam additives due to foaming (Heldebrant et al., 2017) Not tolerant of water (Heldebrant et al., 2017). The absence of water increases heat duty (Heldebrant et al., 2017). Higher cost of solvents compared to amines (Heldebrant et al., 2017).	5-6
Chilled ammonia	Low regeneration energy requirement (2.5-3.2 GJ/t_{CO2} Valenti et al., 2012, p. 80) Lack of solvent degradation during the absorption and regeneration cycle Tolerant for NO_x and SO_x Low solvent cost Sellable ammonium sulfate fertiliser by-product	Near-freezing operating conditions are required to limit solvent losses Potential for fouling due to ammonium bicarbonate solids.	6-7

Appendix 2. Comparison of different adsorbents (Ben-Mansour et al., 2016; Zhao et al., 2021).

Sorbents		Advantage	Disadvantage
Metal-organic frameworks	Physical adsorbent	High capacity (5-8 mol/kg). Large surface area (1800-4700 m^2/g). Possibility of tuning pore size. High selectivity CO_2/N_2 (60-700).	Expensive. Low performance at low CO_2 partial pressure. Sensitive to moisture.
Zeolites		Large micropores/mesopores. Moderate CO_2 adsorption at ambient conditions (3-5 mol/kg). Low production cost.	Reduced capacity under moisture conditions. High regeneration energy consumption. Low CO_2/N_2 selectivity (4-30).
Carbon-based		High conductivity. High thermal and chemical stabilities. It is lightweight, with high surface areas and large pore volumes. Low energy regeneration consumption. Reasonable cost	Low adsorption and desorption temperatures (25-30 °C). Low CO_2 capacity compared to Zeolites or MOFs (2-3 mol_{CO2}/kg)
Amine supported adsorbents	Chemical adsorbent	More suitable for ambient pressure. CO_2 capacity increases in the presence of moisture where CO_2/N_2 ratio increases High selectivity Tolerant to moisture. Moderate capacity (2-5 mol/kg)	Expensive sorbents. Higher regeneration energy requirement compared to physical adsorbents. Low reaction rates compared to physical adsorbents.

Appendix 3. Comparison of different membrane types (Hou et al., 2022; Bazhenov & Lyubimova, 2016; Scholes, 2023; Vasudevan et al., 2016; IEAGHG, 2019; Rezaei et al., 2023).

Membrane type	Advantage	Disadvantage	TRL
Thin-film composite membranes	High permeance	Low selectivity CO_2/N_2 (10-55)	6-7
	Use less material compared to traditional asymmetric membranes (1-2 g/m^2 vs. 50 g/m^2)	Material stabilities	
	Commercialised material options available	Sensitive for impurities	
	Low energy consumption 0.5-1.2 GJ_e/t_{CO2}	Low capture rate with a single membrane module (60-80%).	
fixed-site carriers	High CO_2/N_2 selectivity (generally 70-450 and up to 1500).	Material stabilities	5-6
	Low energy consumption 0.5-1.2 GJ_e/t_{CO2}	Sensitive for impurities	
	many materials are available	Low capture rate with a single membrane module (60-80%).	
Membrane contactors	Suitable for low CO_2 concentrations (<10% vol)	Additional mass transfer resistance.	5-6
	Selectivity is not dependent on membrane material.	Physical ageing of material.	
	Smaller and lighter dimensions than absorption.	Process sensitivity to impurities.	
	Many solvents are available.	Limited temperature and pressure range.	
	Lower electrical energy consumption than other membrane types.	Higher energy demand compared to gas separation membranes ~2.9 GJ/t_{CO2}	

Appendix 4. Carbon capture technologies will be widespread in the future.

Technology	Other technology applications	Size of existing market	Availability and maturity of technology components	Development needs	TRL	Retrofit easiness	Cost reduction potential	Score
CLOU	3, Applications in chemical industries and oxygen production	2, There is a sizeable market for fluidised bed systems in the oil & and gas industry and power sector	3, key components are commercially available.	1, the ability to produce power is yet to be demonstrated.	2, TRL 6	1, Needs a boiler replacement, not a retrofit solution	3, -40%	15
Amine-based	3, can be widely applied in other applications	3, used in methanol production and commercial CO_2 capture	3, Commercialized technology	3, Successful operation on a large scale demonstrated in a commercial power plant	3, TRL 6-9	3, End-of-pipe technology	1, 0%	19
Phase change solvents	3, liquid-liquid extraction process and amine-based absorption	1, Non-existent	3, Similar components used in amine-based absorption	2, Emission and degradation rates should be determined. Solvent volatility problems should be solved.	2, TRL 5-6	3, End-of-pipe technology	1, 0%	15
Aqueous ammonia	3, Some components have been commercialised in gas treatment also a similar process to amine-based absorption	2, small-scale applications based on the alkali process.	3, Similar components used in amine-based absorption	2, Needs, pilot-scale evaluation of complete process	2, TRL 6-7	3, End-of-pipe technology	1, +25%	16
Potassium carbonate-based	3, Some components have been commercialised in gas treatment also a similar process to amine-based absorption	2 small-scale applications based on the alkali process.	3, Similar components used in amine-based absorption	3, Low, large-scale piloting has been done successfully.	3, TRL 7-8	3, End-of-pipe technology	2, -12.5%	19
Water-lean solvents	2, Similar process to amine-based absorption	1, Non-existent	3, Similar components used in amine-based chemical absorption	2, solvent stability issues need to be solved.	2, TRL 5-6	3, End-of-pipe technology	2, -12.5%	15
Chilled ammonia	3, Similar process compared to amine-based absorption	1, Non-existent	3, Similar components used in amine-based chemical absorption and chilling technology are commercially available	1. Challenges include dealing with precipitation in a process designed for liquid absorbents.	2, TRL 6-7	3, End-of-pipe technology	1, +65%	14
PSA/VSA	3, PSA/VPSA used commercially for carbon capture in hydrogen production	3, Solid sorbents used in many applications, hydrogen separation, air separation and CO_2 removal from biogas	3, VPSA systems used commercially in natural gas processing	2, electrical energy requirements must be decreased, and scalability challenges should be solved.	2, TRL 6	3, End-of-pipe technology	2, -25%	18
TSA/TVSA	2, Sorbents used commercially for CO_2 removal from air	2, Solid sorbents are used for VOC removal	3, the technology uses commercially available sorbents and air preheaters	2, some TSA/TVSA processes reported having problems with leakage of sealing valves under vacuum conditions and sorbent stabilities in long-term required to confirm	2, TRL 5-7	3, End-of-pipe technology	2, -25%	16
Gas separation membranes	3, used commercially in other gas-treatment applications and reverse osmosis	2, Used especially in natural gas treatment on a small scale	2, other components than membrane modules are commercially available.	1, No experience in integrating several modules in flue gas streams with vacuum	2, TRL 5-7	2, End-of-pipe technology but requires modifications to flue gas paths	1, 0%	13
Membrane contactors	3. Membrane contactors are used in degassing liquids and gas transfer.	1, very small sub-market	2, other components than membrane modules are commercially available.	1, No experience in integrating several modules in flue gas streams with vacuum	2, TRL 5-6	2, Moderate, end-of-pipe technology but requires modifications to flow paths. Not so many units are required compared to other membrane types	1, +12.5%	12
MCFC	3, Modular units for power generation up to 60 MW_e	2, Niche markets, not competitive in large-scale power generation	3, Molten carbonate fuel cells are commercially available for power generation without carbon capture. Also, cryogenic separation is commercialised	2, the impact of flue gas impurities needs to be studied.	1, TRL 4-6	2.5, End-of-pipe technology but needs access to gaseous fuel network	3, -50%	16.5
Calcium looping	3, Calcination process used in the cement and forest industry	2, Cement and forest industry use caustising process	3, key components are commercially available.	2, Needs demonstration of energy saving and cost reduction modifications	2, TRL 6-7	2.5, End-of-pipe technology but needs new logistical solutions	3, -32.5%	17.5
Oxy-fuel combustion	3, potentially combustion system market	1, Non-existent	3, key components are commercially available.	1, Ongoing work developing further efficiency aspects, such as pressurised removal of SOx and NOx components	2, TRL 7	1.5, Needs modifications to the auxiliary boiler or new boiler	1, +20%	12.5

Appendix 5. The most promising technologies to be integrated into pulp mills in the future.

Technology	Operational flexibility	Emissions and waste streams	Cost reduction potential	Energy balance impacts to Pulp mill	Retrofit easiness	Capture and purities that can achieved	Score
Amine-based	2, Moderate operational flexibility possibilities	1, Produces toxic emissions and waste streams	1, Reference	1, Reference	3, end-of-pipe technology	3, capture rates > 90% and purities > 95%	11
Aqueous ammonia	2, Moderate operational flexibility possibilities	1, Produces toxic emissions	1, +25%	2, Reduces steam consumption compared to amines but still has significant impacts on steam availability	3, end-of-pipe technology	3, capture rates > 90% and purities > 95%	12
Potassium carbonate-based	2, Moderate operational flexibility possibilities	3, No additional emissions	2, -12.5%	2. It reduces steam consumption compared to amines but impacts steam availability. If the regeneration of solvent is done by electricity, then there is no impact on pulp mill processes	3, end-of-pipe technology	3, capture rates > 90% and purities > 95%	15
PSA/VSA	3, High operational flexibility	2, No additional emissions but adsorbents generate waste when they are changed	3, -25%	2, Not impacting on steam availability but have high electrical energy consumption compared to other technologies consuming only electricity	3, end-of-pipe technology	3, capture rates > 90% and purities > 95%	16
TSA/TVSA	3, High operational flexibility	2, No additional emissions but adsorbents generate waste when they are changed	3, -25%	2, Can reduce impacts on steam availability, but energy savings are on a moderate level compared to amines	3, end-of-pipe technology	3, capture rates > 90% and purities > 95%	16
MCFC	3, High operational flexibility	3, No emissions	3, -50%	3, Low impacts on steam availability and electricity consumption	2.5, end-of-pipe technology but requires access to gaseous fuel network	3, capture rates > 90% and purities > 95%	17.5
Calcium looping	3, High operational flexibility	3, Decrease sulphur emissions	3, -32.5%	3, Low impacts on steam availability and electricity consumption	2.5, end-of-pipe technology but requires a new logistical solution in the pulp mill	3, capture rates > 90% and purities > 95%	17.5

Appendix 6. Minimum solvent flow rate and minimum diameter of absorption unit.

Absorption case	Lean solvent mass flow [kg/s]	Flue gas mass flow [kg/s]	Flue gas CO_2 [kmol/h]	Clean gas CO_2 [kmol/h]	L/G [kg/kg]	Removal efficiency [%]	Calculated diameter [m]
MEA RB & LK	967	250	4344	596	3.9	86.3%	12.2
	1011	250	4344	456	4.0	89.5%	12.3
	1056	250	4344	327	4.2	92.5%	12.4
MEA RB	789	224	3573	537	3.5	85.0%	11.4
	833	224	3573	393	3.7	89.0%	11.5
	878	224	3573	262	3.9	92.7%	11.6
MEA LK	175	26	750	92	6.7	87.7%	4.3
	180	26	750	78	6.9	89.6%	4.4
	185	26	750	64	7.1	91.5%	4.4
HPC RB & LK	860	250	4324	729	3.4	83.1%	7.6
	920	250	4324	479	3.7	88.9%	7.7
	980	250	4324	232	3.9	94.6%	7.8
HPC RB	700	224	3573	710	3.1	80.1%	7.1
	750	224	3573	505	3.3	85.9%	7.2
	800	224	3573	303	3.6	91.5%	7.3
HPC LK	135	26	750	115	5.2	84.7%	2.6
	140	26	750	91	5.4	87.9%	2.6
	145	26	750	67	5.6	91.1%	2.7

Appendix 7. Cost data for techno-economic analysis.

Cost type	Cost	Comment	Reference
Investment costs:			
HPC RB LK & HPC RB	200 M €	2019 CEPCI and reference capacity 800 000 tCO_2/a	Levihn et al. (2019)
HPC LK	78 M £	2022 CEPCI and reference capacity 350 000 tCO_2/a	AECOM (2022)
MEA RB LK	239 M €	2015 CEPCI and the same reference capacity	Onarheim et al. (2017b)
MEA RB	228 M €	2015 CEPCI and the same reference capacity	Onarheim et al. (2017b)
MEA LK	36 M €	2015 CEPCI and the same reference capacity	Onarheim et al. (2017b)
Liquefaction plant	113 M $	2013 CEPCI and reference capacity 515 tCO_2/h	Abbas et al. (2013b)
Modifications to pulp mill:			
RB & LK	12 M €	2015 CEPCI and the same reference capacity	Onarheim et al. (2017b)
RB	11 M €	2015 CEPCI and the same reference capacity	Onarheim et al. (2017b)
LK	10 M €	2015 CEPCI and the same reference capacity	Onarheim et al. (2017b)
Other capex:			
Spare parts	1%	1% of total plant costs	1%, Onarheim et al. (2017b)
Start-up and commissioning cost	2%	3% of total plant costs	3%, Onarheim et al. (2017b)
Owner's cost	6%	7% of total plant costs	8%, Onarheim et al. (2017b)
Working capital	6%	6% of total plant costs	8%, Onarheim et al. (2017b)
Interest during construction	8%	8% of total plant costs	8%, Onarheim et al. (2017b)
Fixed operational & maintenance costs:			
Maintenance costs	3%	3% of total plant costs	Onarheim et al. (2017b)
Labor cost	60 000 €/a	1 person annually, assumed that 20 people are needed	Onarheim et al. (2017b)
Indirect labor costs	24 000 €/a	1 person annually, assumed that 20 people are needed	Onarheim et al. (2017b)
Variable operational & maintenance costs:			
K_2CO_3 solvent cost	1300 €/t	Same as in reference	Chuenphan et al. (2022)
MEA solvent cost	2200 €/t	Same as in reference	Chuenphan et al. (2022)
Steam cost	30 €/t	Assumed	
Electricity cost	55 €/MWh	~53 €/MWh, last 3-year average spot price excluding the year 2022	Nord Pool (2024)
Cooling water	0.05 €/m^3	Same as in reference	Onarheim et al. (2017b)
Process water	0.1 €/m^3	Same as in reference	Onarheim et al. (2017b)
Wastewater effluent discharge	0.09 €/m^3	Same as in reference	Onarheim et al. (2017b)
MEA sludge disposal	190 €/t	1.6 kg/tCO_2 degradation rate (Chuenphan et al. 2022)	Onarheim et al. (2017b)
Hydrogen cost	30 €/kg	25 $/kg (Abbas et al., 2013b)	Abbas et al. (2013b)

Appendix 8. Capital expenditure and operational expenditures.

CAPEX:	HPC RB LK	HPC RB	HPC LK	MEA RB LK	MEA RB	MEA LK
Investment to CO_2 unit [M €]	402	351	73	342	325	52
Changes to pulp mill [M €]	17	15	14	33	28	14
Investment to liquefaction unit [M €]	68	59	20	68	59	20
Contingency, 10% [M €]	49	43	11	44	41	8
Total plant cost [M €]	535	468	118	487	454	93
Other capex [M €]	123	108	27	112	104	21
Total capital requirement [M €]	658	575	145	599	559	115
Annual capital requirement, 8% interest rate, 25 years [M €]	62	54	14	56	52	11
OPEX:						
Fixed costs:						
Direct labour costs [M €]	1.2	1.2	1.2	1.2	1.2	1.2
Indirect labour costs [M €]	0.48	0.48	0.48	0.48	0.48	0.48
Maintenance, 3% of TPC [M €]	16	14	4	15	14	3
Total fixed costs [M €]	18	16	5	16	15	4
Variable costs:						
Chemicals [M €]	0.31	0.25	0.05	5.2	4.3	0.91
Utilities [M €]	49	42	7	87	73	15
Waste and disposal [M €]	0.11	0.09	0.01	0.53	0.44	0.09
Total variable costs [M €]	49	42	7	93	77	16
Total OPEX [M €]	67	58	12	109	93	20
Total annual costs [M €]	129	112	26	165	145	31
LCOC [€/t_{CO2}]	89	94	104	114	122	124