

LAPPEENRANTA-LAHTI UNIVERSITY OF TECHNOLOGY LUT
School of Energy Systems
Department of Energy Technology
Master's Thesis

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FUTURE MILL 2040

CARBON NEGATIVITY IN INTEGRATED MILLS

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ABSTRACT

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LUT School of Energy Systems
Master's Degree Programme in Energy Technology

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Future Mill 2040 **Carbon negativity in integrated mills**

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2020

87 pages, 35 pictures, 13 tables and 1 appendix

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Key words: pulp and paper industry, kraft mills, carbon capture, carbon storage, CO₂ utilisation, carbon neutral, carbon negative

The pulp and paper industry accounts for around 6% of global CO₂ emissions. There is a growing pressure to reduce GHG emissions due to global warming. The Finnish forest industries have published a climate roadmap to demonstrate the emission reductions needed, in order to support the ambitious climate neutrality goal of Finland by 2035. Stora Enso is committed to SBTi and are therefore actively looking for solutions for emission reductions in their operations. This work evaluates the possibility of achieving carbon neutrality and even negativity within an integrated pulp and paper mill owned by Stora Enso located in Imatra, Finland. A solution is replacing fossil fuels and implementing carbon capture technologies. Most of carbon emissions from pulp and paper industry are treated as carbon neutral due to their biogenic origin, which means the conventional carbon capture technologies are mainly carbon capture technologies for biomass carbon. The swap from fossil fuels to renewable ones is a step towards carbon neutrality but will not be enough to reach carbon negativity. Therefore, methane and methanol production from captured CO₂ was also studied. Results show that methane or methanol production can be feasible in the future if there is a established market for CO₂ or methanol with low electricity prices. This work provides a roadmap with concepts to reach carbon negativity within an integrated mill by 2040.

TIIVISTELMÄ

LAPPEENRANTA-LAHTI UNIVERSITY OF TECHNOLOGY
LUT School of Energy Systems
Energiatekniikan koulutusohjelma

Max Ek

Tulevaisuuden tehdas 2040 **Hiilinegatiivisuus integroiduissa paperi ja sellutehtaissa**

Diplomityö
2020

87 sivua, 35 kuvaa, 13 taulukkoa ja 1 liite

Työn tarkastajat: Apulaisprofessori, TkT Tero Tynjälä
Professori, TkT Esa Vakkilainen

Työn ohjaajat: DI Marjukka Kujanpää, Stora Enso
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Avainsanat: massa- ja paperiteollisuus, sulfaattisellutehdas, hiilidioksidin talteenotto, hiilidioksidin varastointi, CO₂ hyötykäyttö, hiilineutraali, hiilinegatiivinen

Massa- ja paperiteollisuuden osuus maailman hiilidioksidipäästöistä on noin 6%. Paine vähentää kasvihuonekaasupäästöjä kasvaa ilmaston lämpenemisen seurauksena. Suomen metsäteollisuus on julkaissut ilmastotiekartan osoittaakseen tarvittavat päästövähennykset tukeakseen Suomen kunnianhimoista ilmastoneutraaliustavoitetta vuoteen 2035 mennessä. Stora Enso on sitoutunut SBTi: hen ja etsii siksi aktiivisesti ratkaisuja päästövähennyksiin toiminnassaan. Tässä työssä arvioidaan mahdollisuutta saavuttaa hiilineutraalisuus ja jopa negatiivisuus Stora Enson omistamassa integroidussa sellu- ja paperitehtaassa Imatralla. Yksi ratkaisu on fossiilisten polttoaineiden korvaaminen ja hiilidioksidin talteenottotekniikoiden käyttöönotto. Suurin osa sellu- ja paperiteollisuuden hiilipäästöistä käsitellään hiilineutraaleina niiden biogeenisen alkuperän takia, tämä muuttaa perinteiset hiilidioksidin talteenottotekniikat lähinnä hiilidioksidin talteenotoksi biomassan poltosta. Vaihto fossiilisista polttoaineista uusiutuviin on askel kohti hiilineutraaliutta, mutta se ei riitä hiilinegatiivisuuteen. Tässä työssä arvioidaan myös metaanin sekä metanolin valmistus talteenotetusta hiilidioksidista. Metaanin sekä metanolin valmistus voi olla kannattavaa jos niille sekä hiilidioksidille on kehittyneet markkinat ja edullista sähköä on saatavilla. Tämä työ tarjoaa tiekartan mahdollisen hiilinegatiivisuuden saavuttamiseksi integroidussa tehtaassa vuoteen 2040 mennessä.

ACKNOWLEDGEMENTS

I would like to thank Stora Enso for allowing me to complete my degree while working full-time in their company. Especially Eija Hietavuo who encouraged me to allocate time for my work. I would also like to thank Marjukka Kujanpää who gave me invaluable advice and support during this time. I am also grateful for the inputs of many others in the Stora Enso organisation.

From my university I would like to thank Tero Tynjälä and Esa Vakkilainen who gave valuable input and ideas for my work. Without their extensive knowledge in their field I would have been lost.

Finally, I extend my gratitude to my friends and family who supported me on this journey.

Max Ek

Helsinki, 5.10.2020

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Appendix: TRL levels in Europe

LIST OF ABBREVIATIONS

| | |
|--------|--------------------------------------------------------|
| ADt | Air Dry tonne |
| AGR | Acid Gas Removal |
| BAT | Best Available Technology |
| BECCS | Bioenergy with carbon capture and storage |
| BECCUS | Bioenergy with carbon capture, utilization and storage |
| BFB | Bubbling Fluidized Bed |
| BLG | Black liquor gasification |
| BM | Board machine |
| CAP | Chilled Ammonia Process |
| CC | Carbon Capture |
| CCS | Carbon Capture and Storage |
| CCUS | Carbon capture, utilisation and storage |
| CDP | Carbon Disclosure Program |
| CEPI | Confederation of European Paper Industries |
| CFB | Circulating Fluidized Bed |
| CTMP | Chemi-Thermomechanical Pulp |
| DEA | Diethanolamine |
| DGA | Diglycolamine |
| DIPA | Diisopropylamine |
| DME | Dimethyl ethanol |
| EOR | Enhanced Oil Recovery |
| ETS | Emissions Trading System |
| EU ETS | European Emission Trading System |
| FEED | Front End Engineering Design |
| FGD | Flue-gas Desulfurization |
| GHG | Greenhouse gas |
| HPC | Hot Potassium Carbonate |
| HSS | Heat Stable Salts |
| IEA | International Energy Agency |
| IPR | Inevitable Policy Response |
| LCA | Life Cycle Assessment |
| LPG | Liquefied Petroleum Gas |
| MDEA | Methyl diethanolamine |

| | |
|------|----------------------------------|
| MEA | Ethanolamine |
| MTG | Methanol to Gas |
| NCG | Non-Condensable Gas |
| NG | Natural gas |
| PM | Paper machine |
| RFI | Request for Information |
| SBTi | Science Based Targets initiative |
| SDS | Sustainable Development Scenario |
| UNGC | United Nations Global Compact |
| WRI | World Resource Institute |
| WWF | World Wide Fund for Nature |

1 INTRODUCTION

This thesis is done for Stora Enso Imatra mill integrate as part of a roadmap to aid in the idea of a carbon negative mill by 2040. The reason for choosing Imatra mills integrate is that in the Future mill 2040 project, it was seen that big integrated mills will likely be the most prevalent form of production as they offer many benefits over separate pulp or board mills, namely reduced transport need and increased energy and material efficiency (Stora Enso Oyj, 2019). The Finnish forest industries have also published a climate roadmap to demonstrate the emission reductions that are needed, in order to support the ambitious climate neutrality goal of Finland by 2035 (Finnish Forest Industries, 2020). The aim of the thesis is to evaluate current & emerging technological solutions which reduce CO₂ emissions gradually, leading to a carbon negative mill integrate in 2040. The introduction is followed by looking at current trends and developments affecting the pulp and paper industry, what an integrated mill is and what processes are affecting the emissions from the mill. The empirical part of the thesis provides possible technological solutions which could be implemented in Imatra mills based on its technical blueprint. This will serve as a roadmap for future CO₂ reduction considerations.

1.1 Background

Humanity is suffering from a phenomenon called global warming which is a natural phenomenon that unfortunately without a doubt has been accelerated immensely by human activity. The rise in compounds that trap the heat in our atmosphere has mainly risen because of the industrial revolution starting in the 18th century. This revolution gave mankind seemingly unlimited potential for growth and lead to new innovations that we could have only dreamed of, simultaneously raising the living standards of western countries to a new high. This however was not achieved without a cost. In recent years increasing awareness around global warming has sparked many movements around the world pleading the industrialized world to cut back on the emissions caused to our environment. The total anthropogenic GHG (greenhouse gas) emissions have increased at a higher rate since 1970 onwards. Even though several climate change mitigating policies were already adopted since then the GHG emissions continued to grow at an average rate of 0.4 gigaton carbon dioxide equivalent (GtCO_{2eq}) during 1970-2000 and 1.0 GtCO_{2eq} from 2000-2010 (Edenhofer, et al., 2014) (Marks, 2007). In 2017 total annual GHG emissions reached a record high of 53.5 GtCO_{2eq} this means that in 2030 global GHG emissions need to be 25 % or 55 % lower than in 2017 to achieve a reasonable cost pathway that limits global warming to 1.5 °C and 2 °C respectively (Olhoff, 2018).

This awareness of the strain we continue to put on our planet has led to many policies being developed. One of the most notable, the Paris agreement by the United Nations in where consignees are obligated to reduce their CO₂ emissions to reach the targets of a temperature rise of 2 °C or 1.5 °C. In order to reach those targets Finland must reduce the CO₂_{eq} emissions by 59 – 61% by 2030 and 133 – 152% by 2050 (Rocha, et al., 2016).

Many companies are faced with potential problems trying to tackle the ambitious goals set. As Stora Enso operates globally the company is also committing to reducing its impact on the environment while creating additional value from its products on the market. Stora Enso's plan for action has been approved by the Science Based Targets initiative (SBTi), which is a collaboration between CDP (Carbon Disclosure Program), World Resources Institute (WRI), the World Wide Fund for Nature (WWF) and the United Nations Global Compact (UNGC). This means that Stora Enso's emission reduction plan meets the SBTi criteria set by the initiative and is verified by the SBT team. These ambitious targets will be met by different means, and therefore this Master's thesis will introduce some measures needed to reach a carbon negative mill by 2040 to support the SBT commitments. The carbon negativity will be studied for a Packaging Materials integrated mill located in Imatra, Finland. This work will consist of two separate parts wherein the first will consist of theoretical analysis of available technologies to reduce carbon emissions in existing mills. The second part will then evaluate which solutions fit for the Imatra mills integrate, in terms of reduction potential, time and technical readiness. The introduced solutions will be summarized in a roadmap for step-by-step implementations and reduction amounts.

1.2 Objective

The objective of this thesis is to evaluate future trends and policies affecting the pulp & paper industry and to describe technologies available to reach a carbon neutral or negative integrated pulp & paper mill. Based on the findings of trends & policies and the technologies available, the impacts of implementing them in an integrated pulp & paper mill owned by Stora Enso Oyj located in Imatra. The technologies described will be used to create a roadmap for the mill to reach carbon neutrality or even carbon negativity and help Stora Enso in reaching its ambitious climate goals.

2 MEGATRENDS AND POLICIES AFFECTING PULP & PAPER INDUSTRY

Our world is being shaped by global Megatrends that are the culmination of past choices and actions while being shaped by the present and affecting the future in ways we cannot foresee. They drive changes to the very basic of human needs. Megatrends comprise of global development in macroeconomic forces that affect business, personal lives and culture itself, defining a new world to live in. Megatrends tend to disrupt the current makeup of business-as-usual and create new opportunities. Nevertheless, they can also cause disturbances that expose broken links in society, business and politics. Therefore, changes caused by Megatrends can turn businesses and industries upside down. (Stora Enso Oyj, 2019)

An evaluation by a multi-stakeholder group consisting of operational staff, customer facing staff, consultants in energy, artificial intelligence (AI) and general megatrends as well as universities and institutes concluded which Megatrends would have the biggest possible impacts on Stora Enso Mills. The Megatrends identified are presented in figure 1.

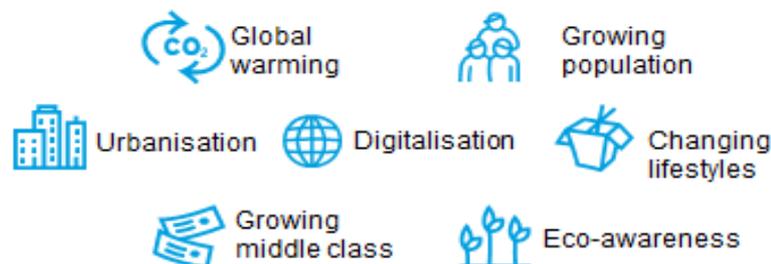


Figure 1: Picture of identified megatrends (Stora Enso Oyj, 2019)

Since megatrends in general are not exhaustive or mutually exclusive, they can contradict or reinforce each other in unforeseen ways. Therefore, it is important to recognize their drivers and importance when looking for future strategic interest (Stora Enso Oyj, 2019).

Digitalization is at the hearth of these megatrends, as it enables the other Megatrends to develop further, therefore it is the single most prevalent driver of the 21st century. Changing the social and political structures around the world together with the synergies of other megatrends like urbanization and the growing middle class and changing lifestyles. (Gillior, 2018)

The pulp and paper industry is not known for its fast paced, agile way of reform so focusing on digitalization and the benefits it provides is crucial going forward.

Policy changes that will affect the pulp & paper industry are driven by aspirations to achieve the goals of the Paris Agreement. The current policies fail to get even close to a 2 °C rise in global temperature let alone the target of well below 1,5 °C as stated in the Paris Agreement. This leads to inevitable changes needed to policies enforced. These possible changes have been outlined by the United Nations in their Inevitable Policy Response report (UNPRI, 2019). The policies listed here are linked to the pulp & paper industry and require fundamental change in the industry to reach the goals of the policies. The policies with implications are described below.

2.1 Carbon pricing

The IPR report takes the route of real-world policies and applied constraints to the model scenario, including explicit carbon pricing. This creates a model where the results are more realistic and non-optimal as opposed to traditional scenario models where, for example, carbon pricing is applied to the results of the model to get optimal pathways (UNPRI, 2019).

The IPR sees applying carbon pricing to processing and combustion of fossil fuels and land use as the areas of highest importance. The carbon pricing scheme is seen as a driver for energy efficiency improvements, coal phase-out, renewables advancement and internal combustion engine phase out. In addition, it drives reforestation to offset CO₂ emissions and bioenergy crop production. The issues with carbon pricing globally are coverage and pricing. Currently less than 5% of carbon pricing is in line with the Paris Agreement and 51% are priced under 10 US\$/tCO_{2e}. Because of its regressivity and other factors carbon pricing is facing strong resistance in some areas, but carbon dividends could be an answer to this problem. (UNPRI, 2019)

Global carbon prices would need to reach 140 US\$/tCO_{2e} in advanced economies by the end of 2040 to be in-line with Paris Agreement temperature targets. Currently the price within the European Emission Trading System (EU ETS) is ~26 US\$/tCO_{2e}. This means that the price of carbon would need to rise by 438 % in 21 years. The current price increase within the EU ETS system is estimated to reach around 44 US\$/tCO_{2eq} in 2024 and then dropping down to ~30 US\$/tCO_{2eq} in 2030. (Andrei, et al., 2019)

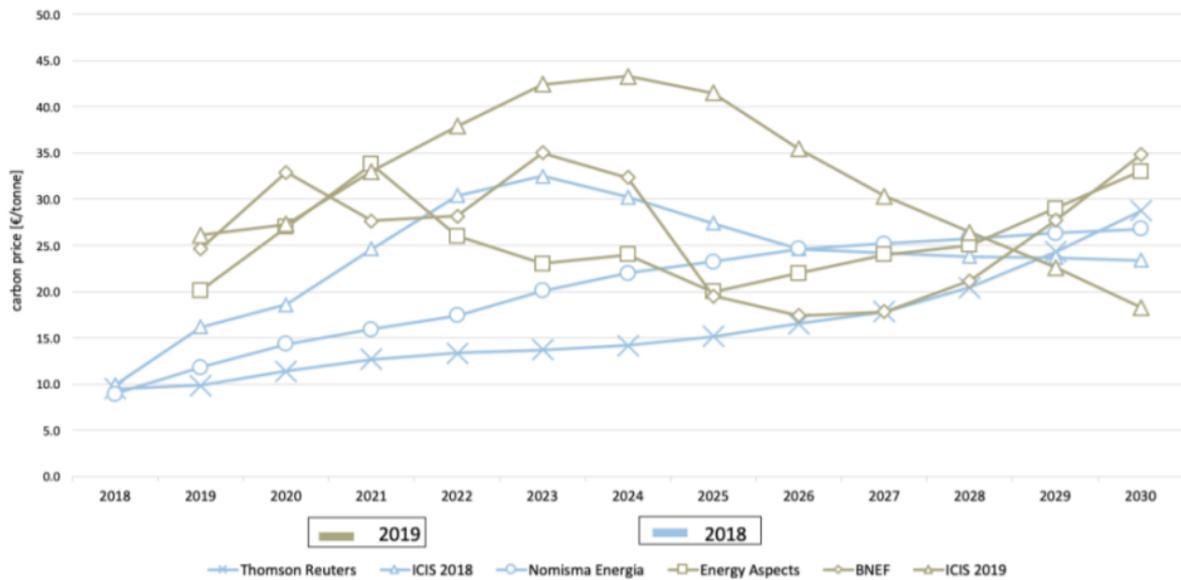


Figure 2: ETS price forecasts (Andrei, et al., 2019).

The increase in carbon price will drive the P&P industry to further their investments into renewable energy sources to phase out the use of fossil fuels, like natural gas that is still in use at mills. Changing market conditions are a key driver for the forecasted price changes over time. The Covid-19 crisis impacted the price of ETS in the spring of 2020 dropping it down to as low as 16 €/tCO₂ from the previous 23 €/tCO₂, but the price has quickly recovered since (Ember climate, 2020).

2.2 CCS & industry decarbonization

Carbon Capture and Storage (CCS) is a technology that removes carbon dioxide (CO₂) from the atmosphere. This technology has the potential to create carbon negative mills for the P&P industry. CCS is currently technically proven but not deployed in commercial scale with a few exceptions in petrol and refining industry. Current forecasts of CCS in power sector do not show significant instalments, except in China, before 2030. The current projects in power and industry sectors under development and operation have a combined capture capacity of 46 MtCO₂ per year, the Sustainable Development Scenario (SDS) target for 2030 is 350 MtCO₂ per year. (UNPRI, 2019)

A large portion (75%) of current CCS capacity is installed in low-cost areas like hydrogen production via steam reforming and biomass fermentation for ethanol production, where CO₂ is available in high concentrations. Technical and economic aspects encompass CCS in energy or carbon intensive industries like iron and steel, as well as P&P. However, unless energy carriers like hydrogen and other energy sources are scaled up quickly, CCS becomes more lucrative with rising carbon prices.

Currently the main issue with large-scale deployment is policies. The US implemented a stimulus programme where the government will provide subsidies for carbon stored or reused if the reduction to emissions can be validated. Asia and the Pacific region are currently the most active regions in CCS, while Europe has seen a slight revival in the field after some North Sea storage options were found to be feasible in cutting the unit cost of stored CO₂. The field requires support to accelerate cost reductions and enhancing or discovering new technologies that can be used. Linking CCS to bioenergy is a proven way to approach negative emissions but innovation is still needed for post-combustion technologies, as retrofitting is much more feasible due to the long lifetime of assets. The storage infrastructure also needs thorough investments and innovations (UNPRI, 2019).

Hydrogen could be a key enabler in the clean energy transition. Currently hydrogen is a marginal energy system option, but its use is dominated within industry applications like oil refining, ammonia production and methanol production. The industrial sectors provide proof that hydrogen can be produced, stored and distributed in large scale. Policy changes are key in tackling the barriers stopping hydrogen applications to be scaled up. Hydrogen is estimated to play a significant role in the medium term (2035-2050) in energy-intensive sectors with adequate policy support. (UNPRI, 2019)

A newly published report by IEA highlights the need for CCUS in the future, “Alongside electrification, hydrogen and sustainable bioenergy, CCUS will need to play a major role. It is the only group of technologies that contributes both to reducing emissions in key sectors directly and to removing CO₂ to balance emissions that cannot be avoided”. (IEA, 2020)

2.3 Zero-carbon power

Although P&P mills are often self-sufficient, or close to it in their energy production they still purchase power from the grid which is produced in varying ways which has its own impacts on greenhouse gas emissions attributed to the mill.

The power sectors capacity to shift to zero-carbon power therefore has an impact on the carbon balance of a P&P mill. Nuclear power is a stable and CO₂ free power source, which has been the backbone for electricity in many countries for a long time. It has seen a considerable capacity reduction in many European countries in recent years. The trend shows nuclear power fading even though it has played a large part in decarbonizing the power sector in advanced economies. Figure 3 shows that even after significant capacity reduction nuclear electricity is the 2nd largest source

of global low carbon electricity generation (International Energy Agency, 2019). A reduction of up to two thirds can happen by 2040, given current level of investments and uncertainty of policy decisions as well as shifting public opinion. The current average age of nuclear installations is around 35 years, which means, new investments need to be planned to avoid shortcomings in low-carbon electricity availability (UNPRI, 2019). According to Finnish Forest Industries, building more nuclear power should play a role in fulfilling a base demand for electricity, complimented by renewable power production. (Finnish Forest Industries, 2010).

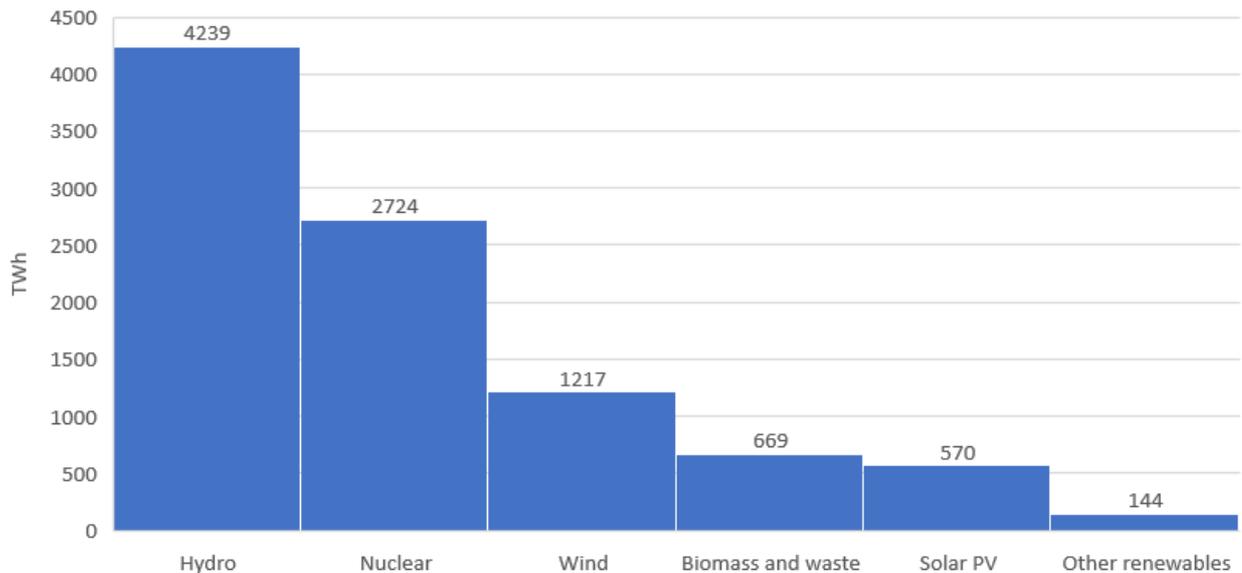


Figure 3: Global low carbon electricity generation by source. (International Energy Agency, 2019)

If the trend of decreasing nuclear based electricity generation continues, the gap needs to be filled by other renewables such as wind and solar, but these sources are not as reliable without intermediate storage. The current growth of renewables is not driven by set directives, instead, declining costs and increased competitiveness with fossil fuels are the driving factors. Renewables share of global electricity generation was at ~26 % in the end of 2018. The net capacity increase on a yearly basis was higher for renewables than for nuclear and fossil fuels combined for the fourth consecutive year. (UNPRI, 2019)

Reaching the goals of the Paris Agreement will require significant increase of the pace of renewables transition. This combined with linked electricity grids across regions and cross sectoral approaches will maximize the effects of decarbonizing the power sector and reaching lowered CO₂ emissions. The renewable power would need to grow by 7% annually between 2018-2030 in order to reach the targets of the SDS (UNPRI, 2019).

The P&P industry is a big consumer of electricity. Electricity consumption varies between grades, grades like Tissue & Towel and Newsprint require substantial purchases of electricity and fuels while in production of wood-free paper high levels of power self-sufficiency is achieved. The paper industry in EU is purchasing over 54 billion GWh of market electricity annually which means there is great potential for CO₂ reductions after achieving zero-carbon power. (Huotari, 2017)

2.4 Land-use based greenhouse gas removal

Reforestation and afforestation are key components for the P&P industry as it relies fully on one source of raw material: forests. The GHG sequestration potential of afforestation and reforestation is estimated to be 1.1 – 18 GtCO₂/year depending on calculation method (UNPRI, 2019). This is significant when you take into account the pathways that limit global warming to 1.5 °C which estimate the GHG removal to the order of 100-1000 GtCO₂ during the 21st century. Current reforestation and afforestation rates have remained at a relatively constant rate of 18 Mha/year (UNPRI, 2019).

Current estimates for the remaining land available for reforestation & afforestation is roughly 300-1800 Mha. The main determinant for the sequestration potential is land availability, which is highly dependent on biophysical and economic constraints. Other factors are the use of land for food production, biodiversity questions and global warming. (UNPRI, 2019)

In 2019 WRI conducted a survey to evaluate the demand on guidance for accounting carbon removals (natural and technological), land-use, land-use change and bioenergy. The survey showed that 88% of respondents considered land-use and land-use change very important or somewhat important, and that they require more guidance in the areas. Even though over half of the respondents to the survey have relevant links to land-use and land-use change only a few are accounting for them in their GHG inventories. Figure 4 demonstrates the issues listed by the respondents. (Greenhouse Gas Protocol, 2019)

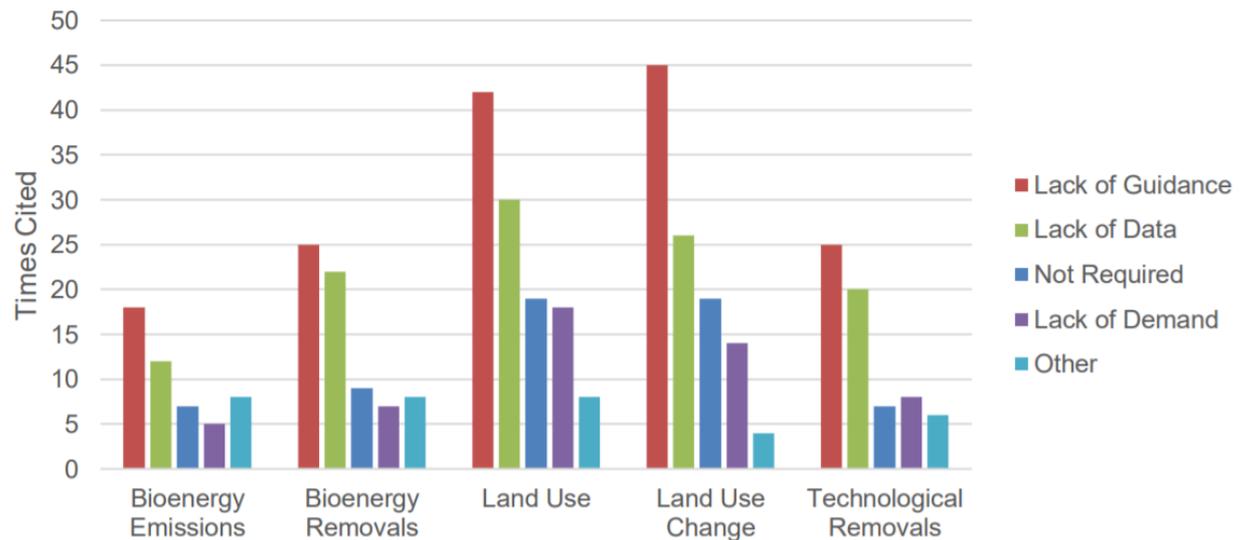


Figure 4: Reasons for not accounting for a category. (Greenhouse Gas Protocol, 2019)

This highlights the need for future guidance and legislation in this field. The WRI has already started the development of a standard for carbon removals and land sector as well as bioenergy guidance, set to be completed by end of 2021 (World Resource Institute, 2020). An increase in accounting for the current lacking areas can be foreseen in the near future. This will further enhance the accuracy of carbon calculations and put more pressure on companies to manage their GHG inventories. The policy forecast is seen as more domestic action and carbon markets that take into consideration land use sectors in an increasing amount during the 2020s. Nations will also promote re/afforestation before the carbon markets begin to take hold in 2030. The international programs like the Bonn challenge will drive some afforestation but will face challenges in tropical regions where food production is competing with forestry (UNPRI, 2019).

2.5 Energy efficiency

Energy efficiency is closely related to emissions and profitability which is why continuous efforts are being made to further improve energy efficiency throughout all sectors. Overall primary energy intensity has decreased by 1.2 % in 2018 (IEA, 2019). Economic growth keeps putting pressure on energy demand, even more so in developing countries. Current major issues in advancing energy efficiency lie in the coverage and stringency of policies globally, currently only about one third of final energy use in industry, transport and buildings is covered by mandatory energy efficiency policies. China is currently leading the development of mandatory standards and regulation accounting for around 60 % of their energy use. (UNPRI, 2019)

Europe is strongly driven by the energy efficiency targets of the EU by things like the amended Energy Efficiency Directive (2018/2002) and the recent development of the European Green Deal programme (European Commission, 2020).

3 BIOMASS USAGE AND CONVERSION PROCESSES

Currently a considerable share of biomass is used in developing countries for residential purposes due to lack of infrastructure or alternatives. In the case of industrialized countries however, biomass is mainly used for industry, electricity and transportation biofuels as new modern methods are implemented. Biomass alone cannot solve the problem of climate change, but it has an opportunity to play a role. Global energy supply in 2017 was 585 EJ of which 81.1 EJ (13.9%) was from renewables of which 70 % is from biomass. The energy share of renewables grew by 1 percent in the past 14 years during which annual average growth was at 2.8 % which is higher than the growth of total energy supply (World Bioenergy Association, 2019). Asia is currently leading the total energy supply globally as well as having the highest renewable energy supply which is growing every year. Traditional biofuels (fuelwood, forestry products, animal and agricultural wastes) accounted for ~64% of biomass energy in 2017 (Ritchie & Roser, 2020). In 2018, 33 % of the pulp and paper sectors final energy demand was covered with fossil fuels (IEA, 2020), thus, it is essential to find new solutions to cover this energy. Biomass is usually a very local feedstock and not abundant enough for large scale users like industries. Industrial users usually need to source biomass from a large area. The properties of unrefined biomass (moisture, low energy density and consistency) make it more expensive and challenging to transport.

3.1 Biomass as energy

Biomass is organic matter, mainly plants, that can be converted to a variety of fuels and energy. Biomass is considered to be a renewable energy source. This thesis will focus almost exclusively on biomass derived from wood as it is the main raw material for a pulp & paper mill. Today biomass use for direct energy still accounts for a big share of the total global available biomass, as demonstrated in the Sankey diagram below (Figure 5). This type of biomass use for energy is very inefficient and occurs mainly in developing countries where refinement is not possible. Another fact is that a considerable share of the biomass classified as material utilization indirectly ends up in energy use through the pulp & paper industry process.

Renewable electricity generation accounted for 23 % of the global electricity generation in 2017. Biomass is the 3rd largest source of renewable electricity generating 493 TWh. Fuelwood is the biggest feedstock of biomass, accounting for 67% of the total. The key sector for biomass development is forestry, as a whole accounting for 87% of the supply of biomass. (World Bioenergy Association, 2019)

Wood Flow Analyse
EU 28
2010

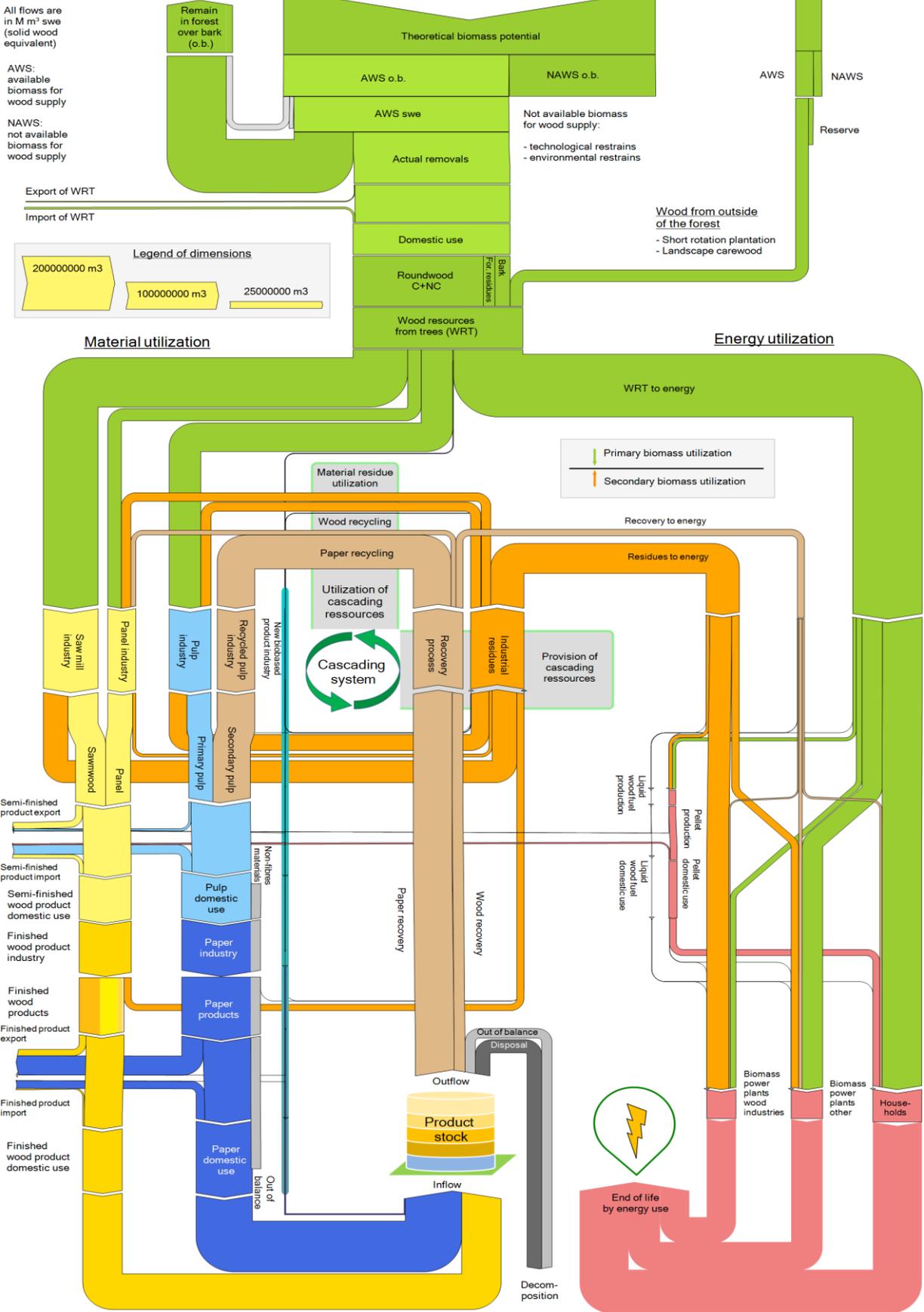


Figure 5: Sankey diagram of wood flows. (Reichenbach, et al., 2016)

The shares of energy carriers in pulp and paper mills are demonstrated in figure 6 below by Confederation of European Paper Industry (CEPI, 2018).

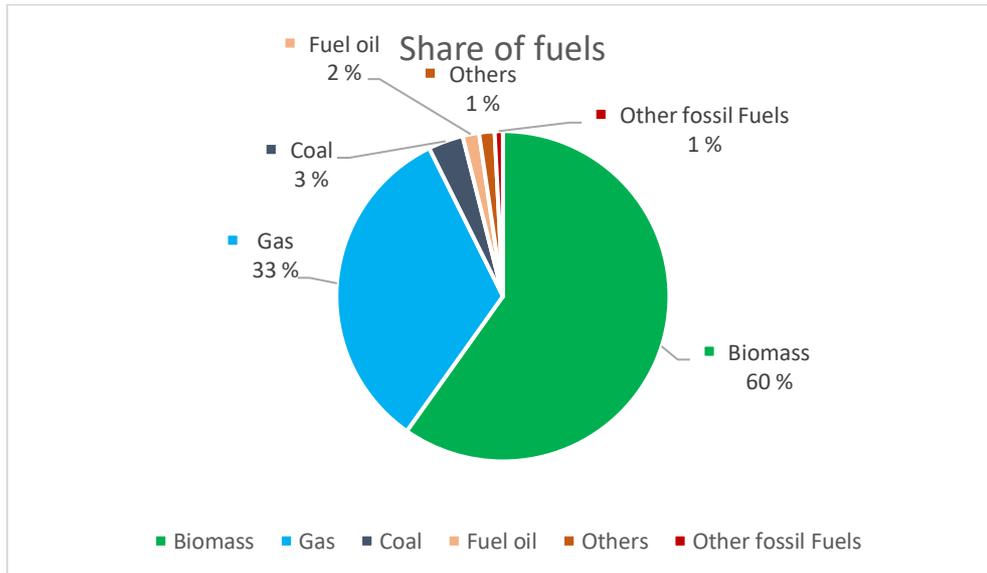


Figure 6: Shares of fuel consumption in 2017 (CEPI, 2018).

Black liquor is the most important biomass fuel in a kraft pulp mill. Burning the black liquor in a recovery boiler and running the generated steam through a back-pressure turbine usually covers the steam and electricity needs of the pulp mill.

There are several sources of biomass in a kraft pulp mill, they are listed in table 1 below.

Table 1: Sources of biomass in kraft pulp mills (Gavrilescu, 2008)

| Type of biomass | Sources of biomass wastes |
|---------------------------------------------|-----------------------------------------------------------------------------|
| Black liquor | Chemical pulp manufacture (kraft pulping) |
| Bark and wood residues | Chemical and semi-chemical pulp processes and mechanical pulp manufacturing |
| Rejects of screening and cleaning processes | Chemical pulp production; recycled paper processing; paper stock preparing |
| Mechanical-chemical sludge | White water treatment and effluent treatment |
| Biological sludge | Biological effluent treatment |
| Deinking sludge | De-inking of recycled paper |
| Mixed sludge | Different sources |

3.2 Biomass conversion processes at kraft pulp mills

There are several different process paths to follow in order to convert biomass into energy. The most notable ones being direct combustion, conversion to liquid biofuels and conversion to biogas (Turkenburg, et al., 2000). However, the ultimate path for choosing the best process depends on a variety of factors like, end use, purity requirements and availability to name a few. The most relevant ones in a pulp & paper mill are direct combustion and gasification. The main biomass energy conversion routes are displayed in figure 7 below.

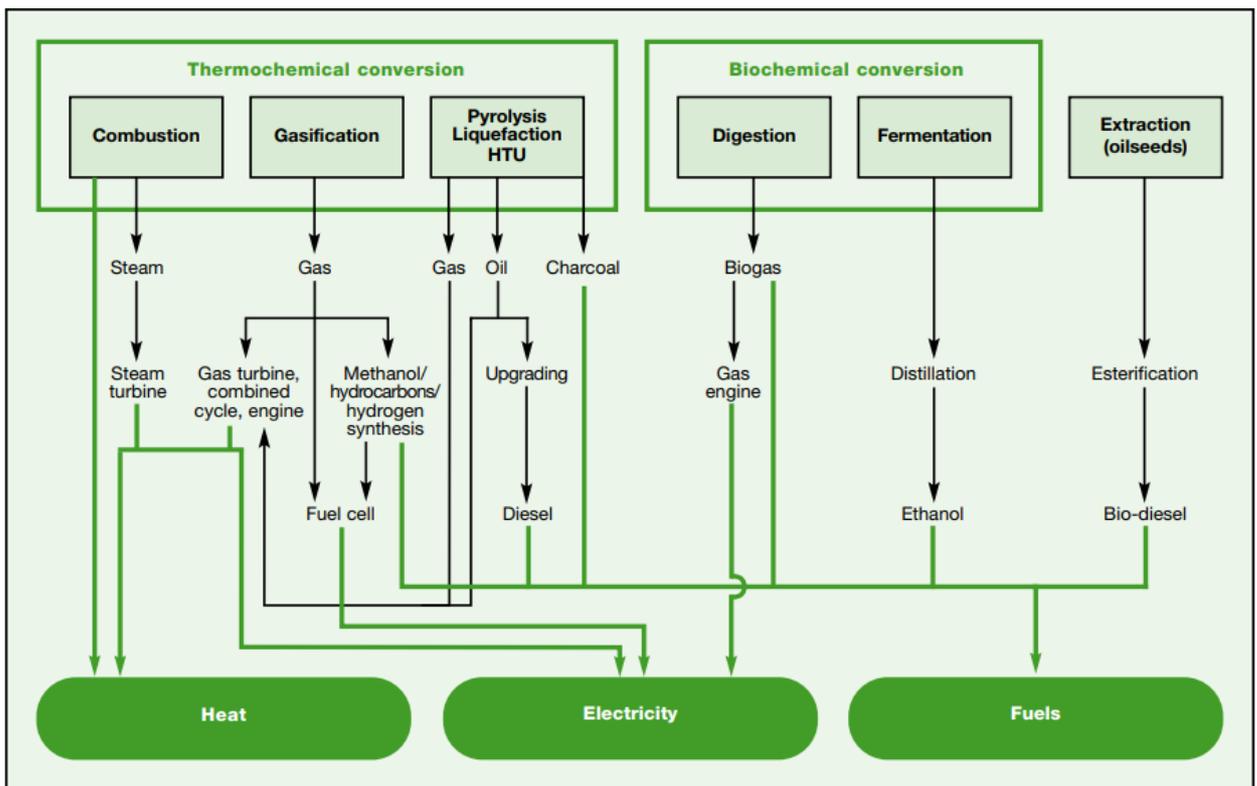


Figure 7: Main biomass conversion routes. (Turkenburg, et al., 2000)

Combustion of biomass is commonly used at pulp and paper mills as a combined heat and power solution. This is usually done with different set-ups of grate firing or fluidized bed concepts. These boilers allow for the recovery of chemicals in kraft pulp mills and generation of heat and electricity. The electrical efficiency of small and medium sized generators (20-50 MWe and 50-80 MWe respectively) is around 20-30%. Finland is a global leader in the development and deployment of Bubbling Fluidized Bed (BFB) and Circulating Fluidized Bed (CFB) boilers. These boilers have a large fuel flexibility, lower investment costs and high efficiency, making them popular in the pulp and paper industry. (Faaij, 2006)

The energy from the biomass fuels recovered through the recovery boiler usually covers more than half of the energy needs of the integrated mill. The CO₂ emitted from biomass combustion is reported separately from the fossil CO₂ accounting for the GHG inventory. The second most important biofuel is the wood waste, like bark and chipping rejects. In the Northern pulp mills, the moisture and ash content of wood waste is high, which leads to a poor heating values and thus typically requiring fossil fuels to be co-fired to enhance the boiler efficiency and stability.

The following chapters 3.2.1 – 3.2.4 describe in more detail the biomass conversion steps and processes needed to make it into a suitable fuel at an integrated pulp and paper mill. The processes are important to understand as some of them require significant equipment and/or changes to current operating processes. The need for utilizing biomass comes from the need to reduce fossil fuel dependency and use.

3.2.1 Biomass pre-treatment

Biomass that can be considered for fuel is usually available only with high water content >40 %. This means that in order to use it for fuel it must be dried to avoid low efficiency due to water evaporation during the conversion to fuel. The biomass must also typically be milled or otherwise converted into suitable particle sizes. The biomass should also be purified from contaminants like sand and metals to reduce equipment malfunctions in the processing equipment. The final moisture content, particle size and numerous other qualities needed are specified by the chosen conversion process. (Kuparinen, 2019)

3.2.2 Torrefaction

The use for torrefied biomass is relevant for pulp mills using solid fuels like coal or wood residues. The use of torrefied biomass has the potential to replace some fossil fuels or increase the efficiency of the bark boiler which means reduced amount auxiliary boiler use typically operated with fossil fuels.

Torrefaction is classified as a thermal pre-treatment method that significantly improves several properties of biomass. Torrefied material has higher energy density, is hydrophobic, fragile and has more homogeneous structure. The advantage of using torrefaction as a pre-treatment technique before gasification is that the biomass is easier to mill, since the fibrous composition of the biomass has been shattered (Isaksson, et al., 2014). This process gives the biomass coal-like properties which allows it to be fired in coal operated lime kilns. The torrefied biomass can be combusted as

the main fuel in a lime kiln using pulverized fuel burners, or as supplementary fuel mixed with lime mud or blown from the burner end in the lime kiln (Kuparinen, 2019).

Torrefaction of biomass is the process of scorching it in high temperatures (200-300 °C) in anaerobic conditions so that water and other compounds are evaporated. During this process the weight of the biomass is reduced by approximately 30 % while losing only 10 % of the energy content. The energy density can be further raised by pressing the torrefied biomass into briquettes or pellets (Schorr, et al., 2012). The residual biomass obtained from pulp mills wood handling is well suited for this process (Kuparinen, 2019).

3.2.3 Gasification

Gasification is the most effective process for hydrogen production from biomass. Gasification can utilize various types of feedstocks based on carbon. Current feedstocks and percentages for syngas production globally are visualized below in figure 8.

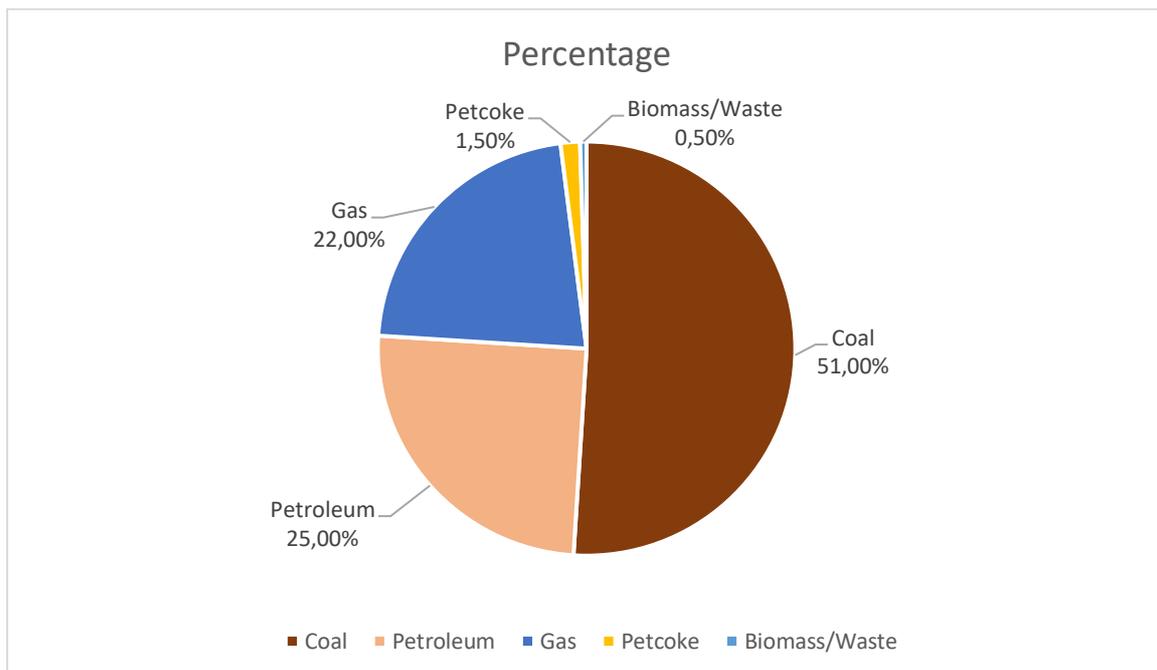


Figure 8: Shares of feedstock for syngas production globally (Anis, et al., 2016).

Biomass gasification is the alteration of carbonaceous substances into a gaseous product or synthesis gas that largely consists of hydrogen (H₂) and carbon monoxide (CO), with lesser amounts of carbon dioxide (CO₂), water (H₂O), methane (CH₄), higher hydrocarbons (C₂p), and nitrogen (N₂) (Anis, et al., 2016). The gasification process is achieved in the presence of a gasifying agent (e.g. air, pure oxygen, steam or mixtures of these components) at elevated temperatures between 500-1400 °C and atmospheric or raised pressures of up to 33 bar.

Gasification is typically feasible for feedstocks with a moisture content of less than 35%. Therefore, pre-treatment of the biomass to remove excess moisture is crucial to avoid substantial energy losses in the overall process. The recommended moisture content for the gasifier is 10-20% (Anis, et al., 2016).

The produced gas blends are intermediate energy carriers that can either be combusted for heat and power generation or they can further be synthesized into fuel for transportation sector. Figure 9 shows an example of a CFB gasifier. The produced gas can also be used as a feedstock to produce high-value chemicals, such as olefins and formaldehyde. (Kajan, et al., 2012)

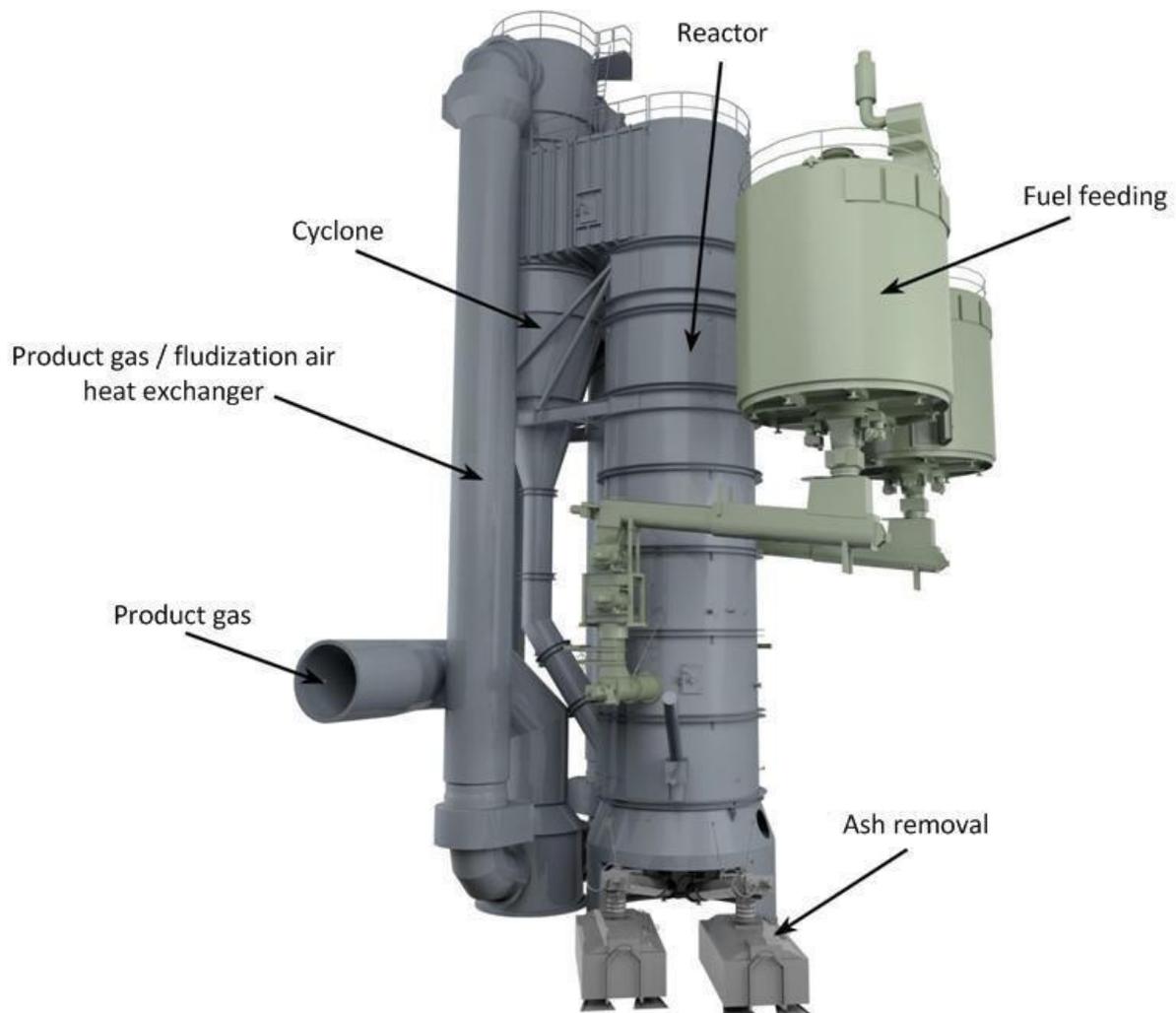


Figure 9: Example of CFB gasifier with components (Bolhar-Nordenkampf & Isaksson, 2016)

The use of gasified biomass in lime kiln operation is well-known. Gasification gas has a lower adiabatic combustion temperature than natural gas thus lowering the hot-end temperature. This means that in order to maintain the production capacity of the lime kiln more fuel is needed. This

leads to higher back-end temperature and thus leads to higher heat losses from the flue gas. (Kuparinen, 2019)

3.2.4 Pulverizing

There are examples of pulverized wood used as fuel for lime kiln operation fuel (Stora Enso, 2015). The use of untreated wood residues in a lime kiln is challenging because of the varying particle size, moisture content and unpredictable flow patterns as well as non-process related contaminants. If the pulverized wood is high in moisture the boiler must be fuelled by fossil fuels to compensate for the moisture content and to allow for better control over the process. Therefore, drying is necessary before pulverization of the fuelwood. Wood has properties that make it more energy intensive to pulverize for instance when compared to coal or torrefied biomass (Kuparinen, 2019).

The species and quality of the fuelwood has effects on the combustion process within the lime kiln, high moisture causes system efficiency loss and increases the flue gas amount. Bark usually contains larger amounts of impurities; these can cause ring formation and corrosion in the kiln as well as having negative effects on the lime quality. This leads to the use of more make-up lime based on the proportion of impurities in the fuel. (Kuparinen, 2019)

As noted by Kuparinen (2019) problems in the renewable fuel system causes breaks in the firing of biofuel. The most common issues being fires in the fuel feeding system and hammer mill clogging's. When flue gases are used for drying the biofuel fires are more common, hence secondary heat is recommended in place of flue gases for drying the biofuel. (Kuparinen, 2019)

4 INTEGRATED MILLS

The term ‘Integrated mill’ is used when describing a manufacturing facility in which all pulp and paper or board is being produced within the same facility area. This may also include converting operations, such as coating or sheeting. In integrated mills, produced pulp is not dried before use, however the integrate might use dried pulp purchased externally. If two or more companies that are not legal units belonging to the same group/corporation work at the same location it is not considered as an integrated mill (CEPI, 2014).

4.1 Pulp & paper manufacturing

Pulp and board manufacturing can be divided into seven main process areas, (1) wood handling; (2) pulping; (3) oxygen delignification (4) bleaching; (5) chemical recovery; (6) pulp drying (mostly in non-integrated mills); (7) board making. Although pulp drying mainly occurs in non-integrated mills, it is included here as some integrated mills also dry pulp for sale. Some mills also operate coating or converting facilities on their premises, however, usually these processes are done at separate facilities. Figure 10 shows the flow of processes in more detail as well as energy generation at the mill.

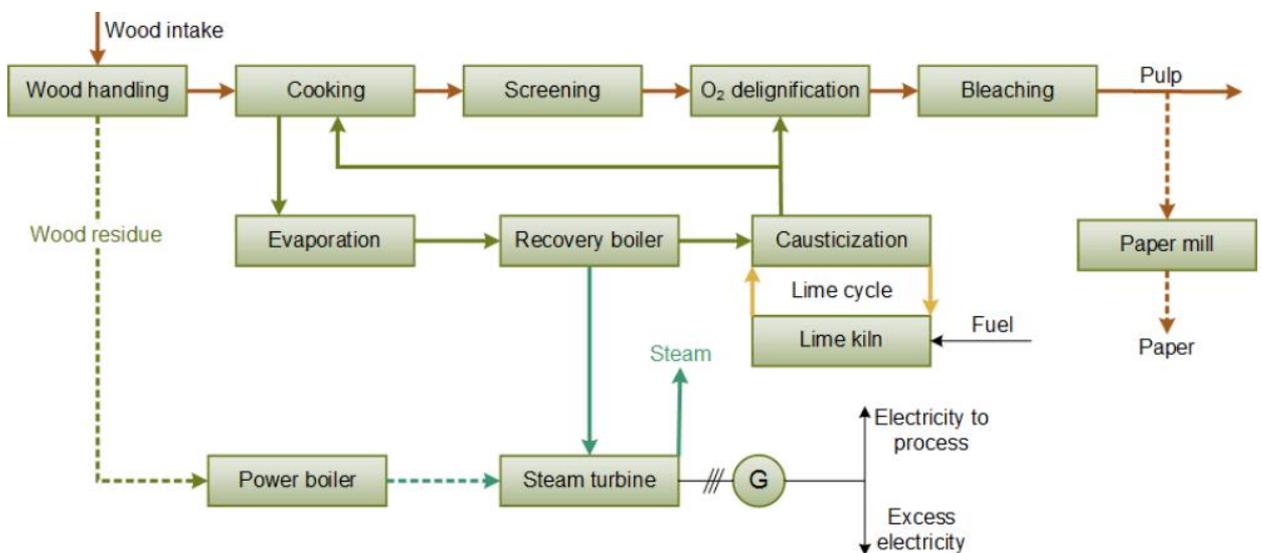


Figure 10. Example of pulp & paper mill operations (Kuparinen, 2019)

Wood is the most common raw material used to manufacture pulp, although other raw materials can be used if non-virgin fibre-based pulp is being produced. The wood usually enters the mill to the woodyard as logs (either with or without bark) or as chips. The woodyard operations are usually a standalone process independent of pulp production process. When logs are being

prepared for pulping, they go through a series of treatments to make them suitable for pulping. (EPA, 2010).

The next step in the process is pulping. Stora Enso manufactures kraft pulp & Chemithermomechanical pulp (CTMP) at Imatra mills. In the chemical pulping process, wood chips are separated into cellulose fibres by removing the lignin that binds the cellulose fibres together. The second type of pulping used at Imatra mills is CTMP. In the CTMP pulping process chips are pre-treated with chemicals before being refined using two rotating disks. This method has a high yield (up to 95%) and CTMP is typically used in the middle layer of a board product to improve its stiffness and allows for lower basis weight (Stora Enso Oyj, 2019).

Oxygen delignification is a direct extension of the lignin removal process following the cooking of wood chips. Oxygen delignification also has a significant effect on the mill's emissions, the emission reduction is directly proportional to kappa reduction, i.e. directly proportional to the pre-bleaching stage lignin removal. Removed lignin is circulated to the chemical recovery from the washing stages. The next step is bleaching which is done to improve the brightness and cleanliness. (Stora Enso Oyj, 2019)

To improve environmental and economic performance chemicals are recovered from the process at kraft pulp mills. The spent cooking chemicals from the pulping phase is called "weak black liquor", it is routed from the brown stock washers to the recovery area at the mill. The process of refining the weak black liquor involves concentrating it by evaporation and reducing inorganic compounds and combusting organic compounds.

After successful reduction of the weak black liquor into black liquor it is sprayed into a recovery boiler where it combusts. This energy is used to create steam needed for many process areas. Mills also typically install a turbine to produce electricity from excess steam. The molten inorganic salts forming at the base of the boiler are referred to as "smelt" which is drawn off and dissolved in a smelt dissolving tank to form "green liquor" (EPA, 2010).

The green liquor is then transported to a causticizing area where it is transformed to Sodium hydroxide (NaOH) by adding lime (calcium oxide [CaO]). The green liquor is cycled through a slaker tank where it reacts with water to form calcium hydroxide ($\text{Ca}(\text{OH})_2$). Lime mud is then calcined in the lime kiln to produce reburned lime that is fed into the slaker tank. The white liquor from the clarifier is recycled back to the digesters at the pulping stage (EPA, 2010).

Pulp drying is only done at non-integrated mills to get the pulp in a transportable format or at integrated mills if production exceeds the board or paper making volume, or if it is being sold. This process involves drying the pulp and then baling and storing it for shipping.

The board making process consists of the pulp being fed onto a conveyer from a head box together with water. During this some additives are also added to the pulp to improve runnability or give the board special characteristics like added density or strength. The conveyer belt leads the pulp through a series of presses to “glue” the sheets together to create the board material. The board is then going through dryers to lower the moisture content and finally proceeds to be wound into storage reels.

4.2 Energy losses and improvement areas

Steam is used in a number of applications which are important for the typical pulp & paper making process. The most significant process steps being cooking, evaporation, bleaching and drying. Over 80 percent of consumed energy by the pulp and paper industry is in boiler fuels while the remaining 20 percent is in other sources like purchased electricity (EPA, 2010). Therefore, energy efficiency improvements to steam systems account for the most significant opportunities for energy savings at the mills. The primary sources of steam in a pulp and paper mill are the recovery boiler/-s and power boiler/-s. As stated earlier the recovery boilers use black liquor as fuel to produce steam for mill process heating applications and recover pulping chemicals, and in most cases co-generation of on-site electricity. The power boilers are usually fired with a wide array of fuels like bark, biofuels, natural gas and heavy fuel oil. (Kramer, et al., 2009)

There is great potential for steam usage savings especially in older mills e.g. Imatra mills that do not have state-of-the-art technology. Many of the possible improvements have very short payback times, < 2 years in some cases. The benefit, however, depends largely on the current process and most of the cheap investments have already been carried out at most mills. The main areas of improvements can be divided into a few distinct categories, steam distribution controls, improved insulation and steam traps (Kramer, et al., 2009).

Steam distribution controls are necessary because steam demand can change abruptly because of operating process halts at steam consuming stages (e.g., paper machines or turbines) or failures in operations (e.g., sheet breakage). Without proper control equipment, this leads to dumping of excess steam or additional fuel consumption at auxiliary boilers. Improved insulation is a major factor in steam energy savings. The steam distribution pipelines are usually long and exposed, so

optimal insulation is essential. The key characteristics of insulation are low thermal conductivity, dimensional stability under temperature changes and resistance to water and combustion. Steam traps using modern thermostatic elements can not only reduce energy use but also improve reliability. This advantage comes from the ability to open them when temperatures are reaching very close to saturated steam (typically within 2 °C). The purging of non-condensable gases with each opening and being open on start-up allowing for fast steam system warm-up. A program for checking the steam traps and having a maintenance schedule and monitoring is essential as well to reduce unwanted losses. (Kramer, et al., 2009)

A reference pulp mill using the Best Available Technology (BAT) has been defined to consume electricity at the rate of 712 kWh/ADt (European Commission, 2015). In addition, the manufacturing of paper or board needs to be taken into account. The energy consumption depends on the age and speed of the machine and grade of paper, the energy consumption is ranging from 510 – 1100 kWh/ADt (Kähkönen, 2019). A modern pulp mill uses around 540 – 800 kWh/ADt, it is worth noting that this does not include the electricity consumption of paper or other product manufacturing, only the pulping process (Penttinen, 2018). At Imatra mills the average electricity consumption is around 1300 kWh/t_{product} which is a good level considering the age of the mill. Currently about half of the consumed electricity is purchased from the grid. This is due to the design of the turbines to produce the needed steam for the mill processes. In the next section there are listed some emerging energy efficiency technologies that would reduce either the demand for steam or electricity or both. These changes to the processes would allow for higher electricity production and therefore better enabling technologies like electrolysis to be operated on site.

4.3 Emerging energy-efficiency technologies

Thinking about what the future will have in store for us is often a daunting task and as Stora Enso and this thesis is focusing on the concept of a carbon negative integrated mill in 2040 it is imperative to look at some future technologies and solutions which would enable cutting substantial energy demand from paper manufacturing as well as reduce emissions. The possibility of rebuilding a whole mill is not a path that we want to pursue unless necessary. CEPI has published a report in 2013 called the Two Team Project (CEPI, 2013), which lists some of the envisioned breakthrough technologies for 2050. The idea is to have the breakthrough technologies ready by 2030 in order to have them implemented to operations by 2050 as this is equal to about two investment cycles in the pulp and paper industry today. The report lists eight different new

breakthroughs or solutions for CO₂ reduction and energy savings, a few of the most promising solutions are described next.

Flash condensing

Flash condensing with steam would allow for almost waterless paper production. This would be done by largely dry fibres being blown into a forming zone with agitated steam and condensed to form a web, using one-thousandth of the water volume used today. The fibres would be combined with the additives and chemicals to a highly turbulent steam. This steam would then carry over to a forming zone, here the condensing and steam expansion would work in combination to allow for the formation of a paper sheet. This would also lower the heating need after the wire as the moisture content would be less than 30 %. Steam forming could also enable massive water savings as steam has a much higher volume per unit. According to the report this concept would reduce fossil CO₂ emissions by about 50 % compared to 2011 baseline and about 20 % savings in primary energy. Compared to today's drying energy demand steam forming would reduce it by at least 50 %. Reduced heating need and the drastically reduced amount of freshwater needed would make an effluent treatment plant obsolete. Commercialization could be seen around 2025 at the earliest. Another technology like the flash condensing would be using superheated steam first, replacing air as a heat carrier at the dryer section. The next step would be to expand the steam to the papermaking process as described above. This would be a step-by-step process to progressively replace air and finally water in papermaking. The expected first commercialization could be seen in 2030. (CEPI, 2013)

Dry pulp for cure forming

Cure forming with dry pulp utilizes cellulose fibres that are treated against shear using bio-based substances and then having the fibres in a highly viscose solution at up to 40 % concentration as opposed to ~2 % concentration currently used. Following the sheet formation, the next step would be cure-forming which occurs after pressing has generated a web consisting of up to 80 % fibres. The sheet would then be cured to a desired end-product. According to the report this could lead to an energy demand reduction of up to 25 % and a 55 % to CO₂ emission reduction. This is due to the removal of water needed to be dried, which also reduces effluent treatment load and removes the need for a coating process. (CEPI, 2013)

Supercritical CO₂

Supercritical CO₂ is a common substance used currently in many applications like decaffeination and textile dyeing. This technology would utilize supercritical CO₂ to dry and remove contaminants. The first application would be to utilize it in the drying section of the paper making process. Lab test already show that a moisture content of 50-60 % can be completely dried with minimal impact on the paper quality and much lower energy demand. The CO₂ could also be obtained through carbon capture technologies installed at the mill, which would make external acquisition obsolete. The substance could also be utilized in recycled paper to remove waxes, stickies and even mineral oils. The estimated energy savings from the implementation are 20 % to the 2011 baseline and fossil CO₂ reduction of up to 45 %. Time to commercialization is estimated to be around 15 years. (CEPI, 2013)

100 % electricity

100 % electricity is a scenario that is driven by the shift away from fossil fuels and replacing flame-based heat applications with electricity instead. When renewable electricity is used the CO₂ would be cut completely. There would also be a possibility to store energy for later use through power-to-x technologies, utilizing electrolysis for hydrogen generation for example. The implementation into pulp and paper making would be stepwise, starting with the drying section utilizing already existing technologies like microwave drying, ultrasonic drying and infrared drying. Later the introduction of power-to-x facilities at the mill would allow for more flexibility in power generation and sale. If the EU meets its target for 2050, which is nearly 100 % CO₂ free electricity it would allow fully decarbonized papermaking with a decrease in energy costs of about 8 %. (CEPI, 2013)

A number of other emerging energy efficiency technologies especially for the pulp and paper industry have been assessed by (Kong, et al., 2016), evaluating potential for energy savings and CO₂ emissions. The technologies listed here are on a TRL of 7+ or semi-commercial status as described in the paper. The technologies are biological pre-treatment, borate auto-causticizing and black liquor gasification concepts.

Biological pre-treatment

Biological pre-treatment is developed mainly for mechanical pulping and therefore not as important for the Imatra mills case, excluding the CTMP plant at Tainionkoski. This is done through modifying the cell wall of fibers and changing its external environment to enhance refining capability without negative impacts on the pulp quality. Fungal pre-treatment has been

implemented on a conventional mechanical pulping line in Wisconsin (U.S) where a 33% reduction in energy costs was reached (Swaney, et al., 2003).

Borate auto-causticizing

Borate auto-causticizing is especially lucrative for kraft pulp mills as it allows higher caustic production and lowering lime demand or even removing lime demand all together. Sodium borate ($\text{Na}_2\text{B}_4\text{O}_7 \cdot 5\text{H}_2\text{O}$) is used to replace lime, each tonne added replaces 10 – 30 times its weight in lime. In kraft pulp mills reducing the lime kiln load leads to significant savings in operating costs, emissions and energy consumption and lime mud disposal cost (Kong, et al., 2016). Figure 11 shows the example process of borate auto-causticizing in a kraft pulp mill. Stora Enso was running a full-scale trial at the now closed Norrsundet kraft pulp mill in Sweden, the trial was a success reducing lime requirement by about 7 % and auto-causticizing level was around 9 – 11 % during the 15 month trial (Kong, et al., 2016). There was even some indication that borates can enhance pulp yield as well (Björk, et al., 2005).

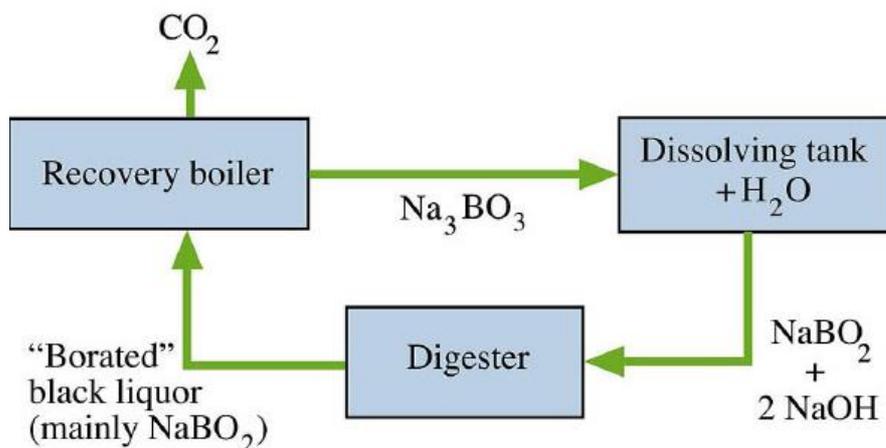


Figure 11: Borate auto-causticizing process example (U.S Department of Energy, 2007)

The main problem with borate currently is that 73% of known global supply is in Turkey and it is almost exclusively produced by Eti Maden IGM for the EU market (CRM alliance, 2019). This can lead to problems with availability and price.

Black liquor gasification

Black liquor gasification (BLG) has a long history of research and development, BLG means pyrolyzing the concentrated black liquor to gas phase and to inorganic phase. This technology can be an alternative to recovery boilers for producing electricity, chemicals and fuels, like synthetic

gas and methanol. BLG can increase the energy recovered from the pulping process by up to 10 % and can increase the electrical output of the mill by up to threefold. The drawbacks are a relatively high investment cost compared to conventional recovery boilers (200 - 400 million vs. 100 – 150 million) and an increased load to lime kilns. The increased load to lime kilns can be counter-acted with auto-causticizing as mentioned earlier. (Kong, et al., 2016)

In addition to the presented technologies there are many different paths currently being pursued with varying degrees of development as seen in table 2 below. From the presented options the black liquor gasification would be the most impactful and needs the biggest investment. It would allow the mill to produce significantly more electricity which could enable more cost-efficient hydrogen production for alternative fuels like methane and methanol. BLG coupled with borate-auto causticizing would also lower the load on the lime kilns but as mentioned before the availability of borate is a risk.

Table 2: Promising biorefinery chains under development, adopted from (Kong, et al., 2016)

| | Biorefinery chain name | Technology challenges | Development status | Preliminary cost estimates demonstration plant / first commercial plant (million €)* |
|------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------|---------------------------|---------------------------------------------------------------------------------------------|
| <i>Thermochemical paths</i> | | | | |
| 1 | Synthetic liquid fuels and/or hydrocarbons (e.g. gasoline, naphtha, kerosene or diesel fuel) and blending components through gasification | Feeders, gas cleaning, catalysts | Demonstration stage | 40- 100/ 400- 1000 |
| 2 | Bio-methane and other bio-synthetic gaseous fuels through gasification | Feeders, gas cleaning, catalysts | Pilot stage | 20 – 40 / 200 - 300 |
| 3 | High efficiency heat & power generation through thermochemical conversion | Bio-feedstock compatible materials, high share of power generation | Demonstration stage | 20 - 40/ 150 - 250 |
| 4 | Intermediate bioenergy carriers through techniques such as pyrolysis and torrefaction | Handling/stability of biooil, materials, specifications of intermediates | Demonstration stage | 20 - 40/ 50 - 100 |
| <i>Biochemical paths</i> | | | | |
| 5 | Ethanol and higher alcohols from lignocellulosic feedstock through chemical and biological processes | Preparation of feedstock and enzymes | Demonstration stage | 20 - 40/ 100 - 200 |
| 6 | Hydrocarbons (e.g. diesel and jet fuel) through biological and/or chemical synthesis from biomass containing carbohydrates | Microorganisms, catalyst performance, bioprocessing | Demonstration stage | 5 - 20/ 50 - 100 |
| 7 | Bioenergy carriers produced by microorganisms (algae, bacteria) from CO ₂ and sunlight | Selection of strains, process design, water treatment, scale up | Pilot stage | 20 – 40 / 100 - 300 |
| *Because demonstration and first commercial plants are first of a kind the costs are estimates | | | | |

5 EMISSION SOURCES AND REDUCTION POTENTIAL

The emission sources at pulp & paper mills can be divided into two categories, namely, (1) combustion of on-site fuels; and (2) non-energy related emissions. If any excess electricity or heat is bought and used by the mill the emissions from the production of those are also accounted to the mill. The GHG emissions at a pulp & paper mill largely consists of CO₂ and smaller amounts of methane (CH₄) and nitrous oxide (N₂O). (EPA, 2010)

This thesis is focused on the emissions occurring within the mill gates, therefore emissions such as transport of raw materials to the mill or transports of the finished products is excluded. The offsetting of emissions by green electricity generation at the mill elsewhere than when used directly in processes at the mill is also excluded. The emission reduction of any other side-products like sludges for agriculture, lime or green liquor dregs, and any chemicals is also excluded as they are not used within the boundaries of the mill.

Many definitions for carbon neutrality and negativity exist today but they often aim to convey the same things and scope. The definitions chosen for this work are presented in table 3 below.

Table 3: Carbon neutral and carbon negative definitions (World resource institute, 2015)

| | |
|-----------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Carbon neutral | Carbon neutrality means annual zero net anthropogenic (human caused or influenced) CO ₂ emissions by a certain date meaning every ton of anthropogenic CO ₂ emitted is compensated with an equivalent amount of CO ₂ removed (e.g. via carbon sequestration). |
| Carbon negative | The measures for the scoped activity go beyond neutrality removing carbon from the atmosphere with additional carbon sequestration like carbon capture or reforestation. |

The pursuit of achieving carbon neutrality for this work is only within the boundaries of operations at the mill. Any activities outside of the mill's premises like wood harvesting, transport to the mill or converting operations outside of the mill area are excluded in this thesis. The reason why converting is outside of the scope along with wood procurement is that the mill cannot influence the emissions related to it.

Kraft pulp mills are emitting significant amounts of biogenic CO₂ during operation but as it is not part of the carbon emissions trading there is currently no cost for it. Biogenic carbon is seen as

carbon neutral as the consensus is that trees growing in sustainably managed forests absorb the amount that is released from the harvested trees (World Business Council for Sustainable Development, 2015). Hence achieving carbon neutrality in this regard is rather straightforward, stopping utilization of fossil fuels at the operations. This often means finding alternative fuels. Described next are some steps to reach carbon neutrality.

5.1 Combustion of on-site fuels

Pulp mills utilize the black liquor from the kraft pulping process to cover a large part of their energy needs. This allows mills to use much lower amounts of fossil fuels like natural gas and heavy fuel oil. However, according to CEPI's statistics of the European pulp and paper industry natural gas still represents some 33% of fuels used at mills while coal is at 3% in 2017 (CEPI, 2018). The natural gas usage is mainly from lime kiln operation and auxiliary boilers and/or during maintenance and start-up of biomass boilers. Figure 12 shows the main sources of CO₂ from combustion in a kraft pulp mill.

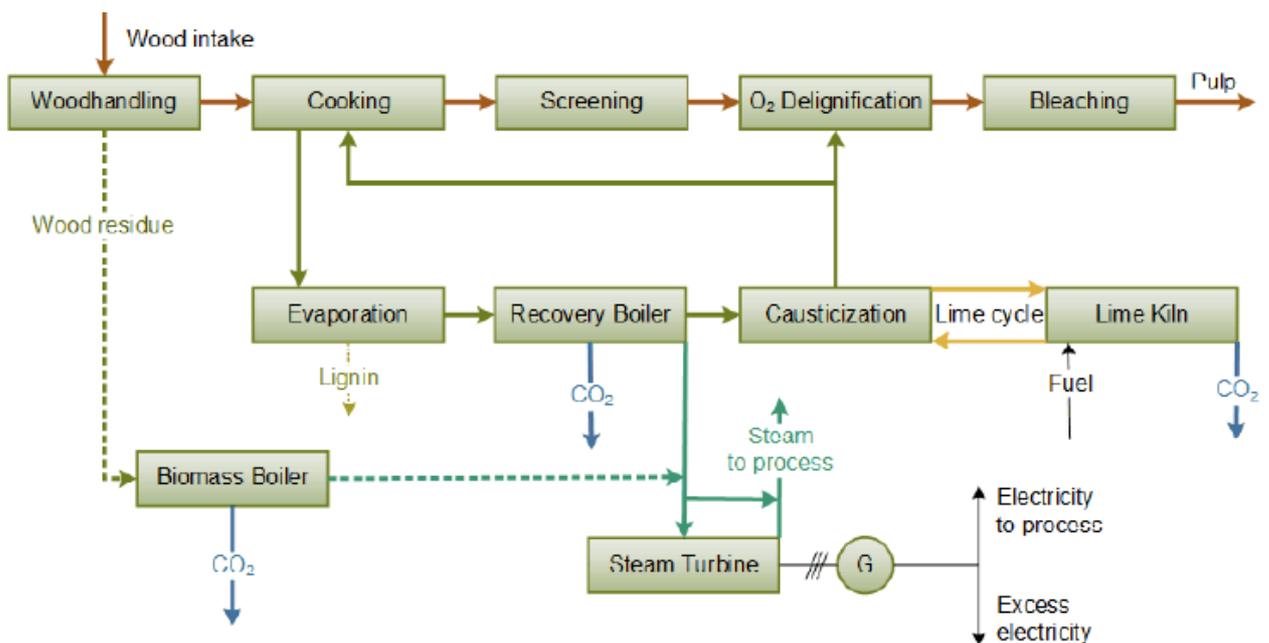


Figure 12: Primary sources of CO₂ from combustion in a kraft pulp mill. (Kuparinen, 2019)

5.2 Non-energy related emissions

The overwhelming amount of GHG emissions occurring at a kraft pulp mill are originating from biomass combustion for energy use. However, there are a few sources of non-energy related emissions, such as, CH₄ emissions from the effluent treatment plant and CO₂ from lime kiln

chemical reactions (EPA, 2010). A few smaller CO₂ sources can also be identified, like, non-condensable gas (NCG) destruction but they are magnitudes lower than the biogenic CO₂ streams. Some newer mills have a bio sludge treatment into biogas which subsequently releases CO₂, but these are currently novel solutions that might become more prominent in the future, hence they are not impactful (Kuparinen, 2019).

5.3 Renewable fuels for lime kiln operation

In kraft pulping most mills are already self-sufficient and most of the energy used is biomass based. The lime kiln of a modern pulp mill or integrate is typically one of the last sources of fossil CO₂ emissions during normal operation of the mill. The lime kiln is used to reduce lime mud, which is mostly composed of calcium carbonate (CaCO₃), into burnt lime, calcium oxide (CaO) and CO₂. This is typically done by combusting fossil fuels. (Kuparinen, 2019)

Using alternative fuels in the lime kiln operations would significantly reduce the remaining fossil fuel usage at a mill. If the fossil fuels could be replaced with existing side streams from the mill operations, it would make investments into new solutions more lucrative and enhance the self-sufficiency of the mill. The main sources of fossil CO₂ emissions at kraft pulp mills are listed below in table 4.

Table 4: Example of emission sources at Imatra mills

| Sources: | Percentage of total fossil emissions |
|------------------------|--------------------------------------|
| Lime kilns | 45 % |
| Auxiliary boilers | 33 % |
| Recovery boilers | 8 % |
| Board & paper machines | 9 % |
| Bark boiler | 2 % |
| Coating plant | 2 % |

From Table 4 we can see that the most impactful areas for de-carbonisation are lime kiln and auxiliary boiler fuels. There are several options for replacing the natural gas that is currently being used at Imatra mills. The easiest way to cut the fossil emissions would be to swap to biogas. Biogas with the same chemical composition as natural gas is continuously being fed into the NG network. A customer can buy biogas in the form of certificates which are managed by a certificate system. The problem however is that the demand for gas by the mill is too great, which leads to there not being enough biogas available for the switch, at least in the current marketplace. A partial swap

would be possible, but it implies additional cost for fuel and is not pursued at this time. As (Mäkelä, 2019) mentions in his thesis the new Baltic Connector that is set to open in 2020 will open up more biogas opportunities and price competition from Europe. This means biogas could be used as backup fuel in the future to allow for renewable fuel operation of the lime kiln and auxiliary boilers.

Lime kilns are usually long rotating cylindrical kilns where lime mud is fed in from the feed end from which it slowly flows through the rotating cylinder placed in a slight incline. There it passes the drying and heating zones in order to reach the burning zone. The temperature must reach over 800 °C in a gas atmosphere containing 20% CO₂ for the reaction to take place in the lime mud (Kuparinen, 2019). The external heat needed is supplied by flames at the hot end from the combustion of fossil fuels, typically natural gas or heavy fuel oil.

There is significant trials and operation with alternative fuels fired in co-combustion in lime kiln operation, but the fossil fuel is rarely replaced completely (Francey, et al., 2011). The fuels that are co-combusted are for example, methanol, tall oil, tall oil pitch and hydrogen. There are numerous side streams occurring at a pulp mill, especially northern pulp mills where the wood is usually de-barked on site instead of in the forest like in eucalyptus plantations.

Renewable liquid biofuels are produced at the mill as side streams, mainly methanol and turpentine, they can be used to cover the role of backup fuel for the lime kiln. The methanol is impure when it comes from the distillery so special attention must be paid to the handling of impurities (Mäkelä, 2019). At the refinement of tall oil an estimate of 20 – 30% of the amount of tall oil comprises of tall oil pitch, however, the amount of tall oil pitch generated from the tall oil refinement of one mill is not enough to cover the whole energy demand of the lime kiln/-s. Moreover, the tall oil generated is usually sold to a separate facility for refinement from where the tall oil pitch can be bought back (Niemeläinen, 2018). In his thesis Niemeläinen, M has studied the possibility of tall oil depitching on-site at a kraft pulp mill. The findings showed that it would indeed be feasible with a short payback period and good applicability to a pulp mill. The biggest drawback from a technical perspective was that not enough is generated to cover the whole energy demand of the lime kilns; thus, lime kilns would require additional feedstock. (Niemeläinen, 2018)

Lime kilns in Finland have typically used mostly either liquid or gaseous fuels, because these types have better combustion properties and are easier to handle than solid fuels. When burning solid woody fuels, one needs to consider the required steps in fuel preparation like, handling, impurities

and higher moisture content. Fuel properties, investment costs and payback time typically state if a solid fuel can be used as pulverized fuel or if it should be gasified.

The gasification process of biofuels at a kraft mill is an established process. In the 1980s the main incentive for building gasifiers for wood was to replace oil use in lime kiln due to the rise in price (Hiltunen, 2009). The process mainly involves the gasification of bark from the de-barking station at the mill, sometimes bark can be bought externally to the mill site. Most gasifiers currently in use are CFB type gasifiers. The benefit of CFB gasifiers compared to fixed bed gasifiers is that they are more forgiving in fuel particle size. The gasification of biofuels offers more lax restrictions on particle size than with pulverizing therefore a crusher at the intake is often enough to turn oversized particles into suitable sizes. A crusher with a particle size allowance of 45 mm is usually enough (Mäkelä, 2019). The heating value of biogas produced in a CFB gasifier is typically within 3 – 7 MJ/kg if air is used as the gasification agent (Vakkilainen & Kivistö, 2008). The removal of lignin from the black liquor at the evaporation stage is also a possibility but as lignin is starting to look promising for lignin based products, like carbon fibres and moisture barriers it is more likely that in the future it is considered as a higher value side stream and will be utilized elsewhere.

In his Master's thesis Mäkelä (2019) focused on three solid biofuels for lime kiln operation. Namely, saw dust, bark and lignin. When considering a fuel switch one of the main considerations is the fuel demand, which usually creates thresholds for the different options considered. The fuel demand is based on the lime kiln burner power demand. (Mäkelä, 2019) Figure 13 shows the needed treatment for wood before lime kiln firing.



Figure 13: Wood treatment before lime kiln firing. (Valmet Oy, 2018)

Drying of the fuel aims to reduce the moisture content from a typical 30 – 55% (wet basis) to around 5 – 8% moisture. The lower moisture increases the efficiency of the kiln operation and capacity. Many types of dryers can be used like rotary, pneumatic or belt dryers. Dryers are chosen based on the characteristics of each individual site layout, heat sources, fuel properties and

environmental issues. Belt dryers are most commonly used because of their robust properties like secondary heat utilization and long residual time. (Valmet Oy, 2018)

Grinding is done in order to reduce the particle size to a homogenous size in order to enhance combustion properties and enable stable fuel quality. Wood structure is arranged in a matrix of cellulose fibres within a matrix of lignin. Wood is anisotropic due to the fibres in the wood and the longitudinal tensile strength is higher than the radial strength. As the material enters the grinder it is coming in random orientations thus high impact forces must be used for grinding to take place (Valmet Oy, 2018).

Once the fuel is dried and ground to a suitable particle size it can be combusted with a burner suitable for pulverized fuel. The main considerations for firing pulverized fuel is the heightened need for particles and air to mix properly & moisture content of the fuel and the burner must be designed in a way that allows for optimal flame output. It also must have the ability to co-fire backup fuels like natural gas or oil.

5.4 Additional measures

Fuel oil is often used for transport vehicles and for auxiliary boilers and during start-up of certain boilers. Heavy fuel oil usage comes mainly from boiler start-up or auxiliary boilers/equipment. Light fuel oil is often used in transports inside the mill area, e.g. trucks, trains and forklifts moving goods around. At Imatra mills there is some 30 fuelling stations for logistics operations to use. Fuel oil stands for about 1% of the fossil emissions currently generated at the mill and therefore the impact on the overall emissions is minimal and not considered in more detail. However, solutions for swapping out fuel oil usage in vehicles include electric vehicles or switching to renewable alternatives. These solutions are merely based on additional cost vs. benefit and do not require significant investments into actual mill infrastructure.

The utilization of side streams occurring at a kraft pulp mill has seen increased interest in order to capture the full potential of a mill. Table 5 shows the typical side streams occurring at a kraft pulp mill. Some of the side streams can be utilized for energy generation or otherwise reduce the energy intensity at the mill, thus reducing emissions.

Table 5: Side stream generation in a kraft pulp mill (Hassan, et al., 2018)

| Type of side stream | Side stream generation stages |
|------------------------|-------------------------------------------------------------------|
| Black liquor | Kraft pulping process |
| Bark | Debarking, peeling of wood logs |
| Biosludge | Biological wastewater treatment |
| Green liquor and dregs | Causticizing process, chemical recovery |
| Dry wood residue | Grading and sorting of wood logs |
| Tall oil | Kraft pulping process, burning and evaporation |
| Lime mud | Recovery and causticizing processes |
| Fly wood ash | Burning of fuel for energy production |
| Primary sludge | Primary wastewater treatment |
| Rejects | Processing of raw material, recovery from screening, causticizing |
| Bottom wood ash | Burning of fuel for energy production in boiler |
| Turpentine | Kraft pulping process, burning and evaporation |
| Screenings | Fiber lining |

Biosludge can be converted to biogas at the rate of approximately 10-15 kWh/ADt (Metsä Group, 2016) and bark, tall oil and turpentine can be utilized on site for energy or further refined to more valuable products. The remaining side streams are of small impact when considering the carbon balance at the pulp mill, therefore they are not described in more detail.

Biochar is a material rich in carbon which is synthesised through pyrolysis of biomass in an environment with limited oxygen. It has been introduced to the agricultural and forest industries for its positive characteristics towards soil function, lowering water demand and fertilizer demand to name a few. (Mohammadi, et al., 2019) During pyrolysis product gas is also formed which could be used for energy production at the mill, replacing some fossil fuels use. However, the end use of the biochar is outside the mills operating premise therefore its benefits and production are not considered further in this work.

6 CARBON CAPTURE AND STORAGE OR UTILISATION

Currently the only option to reach carbon negativity within an integrated pulp and paper mill is to fit in some sort of carbon capture system. Paper products have a relatively short lifetime and therefore the carbon stored in them is often quickly released <5 years, which means they are not effective carbon sinks (CEPI, 2017). In addition, the scope of this work is to evaluate the possibility of a carbon negative integrated mill within the gates of the mill. CO₂ is a significant side-stream from the mill and is currently one of the least utilized due to the lack of business cases for it in the current policy and economic landscape. The captured CO₂ can either be stored permanently or utilized in processes or as a raw material for fuels like methane and methanol.

6.1 Carbon capture

Carbon negativity can be achieved in a pulp & paper mill when CCS technology is installed to capture the fossil and biogenic CO₂ originating from the process areas around the mill in question. Fossil CO₂ capture, transport and storage installations have been part of the EU ETS since 2013. “Capture and storage of CO₂ from combustion of biomass has not yet been incentivised through the ETS. However, it is expected that inclusion of the concept of carbon negatives will be required to meet the stringent long-term emission reductions proposed by for instance the EU. (Jönsson, et al., 2013)” An assumption can be made that in the future biogenic CO₂ is likely to be included in the ETS scheme in some way.

Carbon capture is generally split into two categories, namely, fossil and biogenic carbon capture. In this thesis the focus will be on biogenic CC as the majority of emissions in a pulp mill are of biogenic origin. Furthermore, even though current mills utilize fossil fuels in the lime kiln, there is ongoing efforts to replace or reduce the use of fossil fuels significantly. This furthers the share of biogenic CO₂ emissions from a pulp & paper mill.

Bio-CCS refers to the capture and storage of CO₂ from a biogenic origin, capturing CO₂ from an industrial process utilizing biomass as raw material. Because of the nature of biomass binding carbon dioxide during photosynthesis, carbon captured from biomass fired installations would lead to negative emissions on a life cycle basis, which results in CO₂ removed from the atmosphere. (Arasto, 2015)

The different CO₂ removal technologies are often already available in industrial processes but are not yet mature or there are only small-scale demonstrations available. The different technologies and types with TRLs are presented below in figure 14.

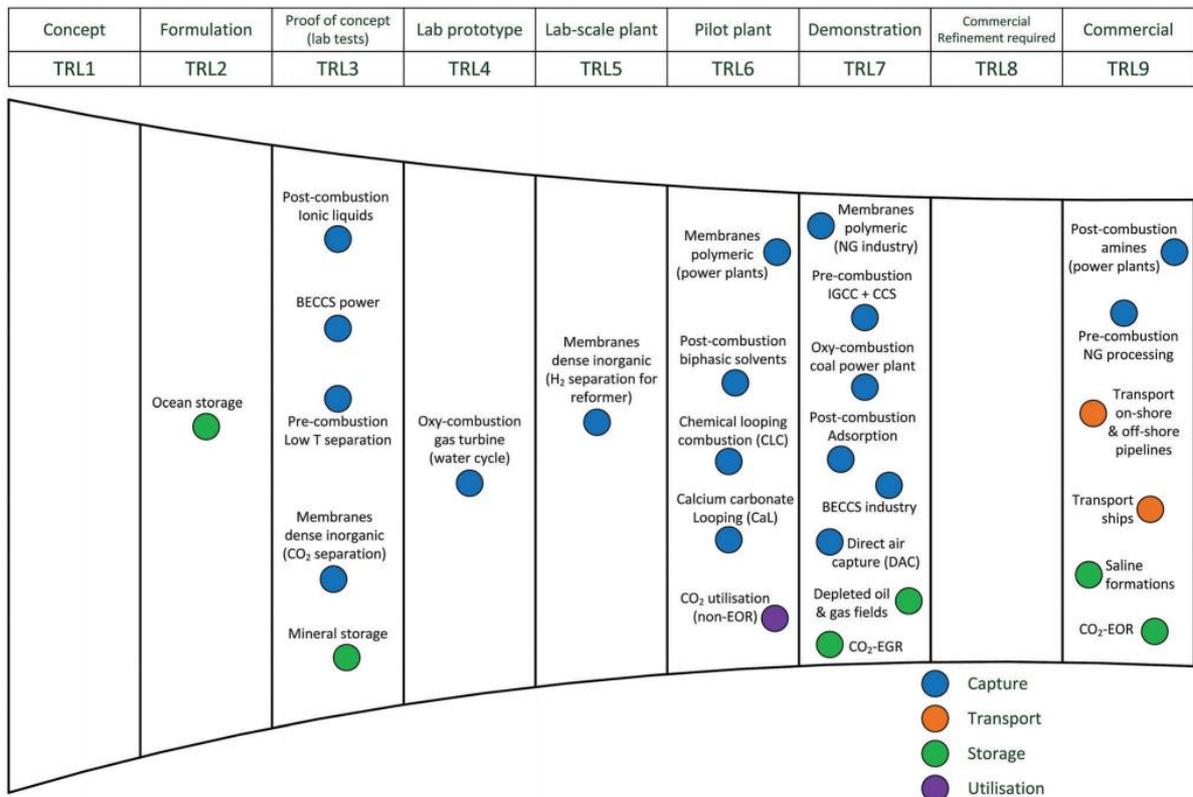


Figure 14: Current development progress of carbon capture, storage and utilisation technologies in terms of technology readiness level. CO₂ utilisation (non-EOR) reflects a wide range of technologies, most of which have been demonstrated conceptually at lab scale. (Bui, et al., 2018)

6.1.1 Methods for carbon capture

In order to understand the development of different technologies for carbon dioxide capture it is important to know that capturing carbon dioxide has been done in large scale for over 60 years. It was developed for the chemical process industry and the oil and gas industry and the first patent of amines for carbon dioxide absorption was taken 1930.

The flue gas carbon dioxide capture technologies derive from three major areas: Ammonia production and Acid Gas Removal (AGR), as well as flue-gas desulfurization (FGD). AGR mainly removes hydrogen sulphide and carbon dioxide from natural gas. It is also called gas sweetening (PetroWiki, 2015). These acid gases need to be removed from natural gas in order to comply with gas quality regulations to minimize environmental impact and ensure gas pipeline transport integrity. Hydrogen sulphide is also extremely toxic to humans and the environment. (BluCarbon Solutions, 2020)

There are 3 main methods currently available for carbon capture presented in figure 15 and they are categorized as follows, Pre-combustion-, oxy-fuel combustion- and post-combustion carbon capture (IPCC, 2005).

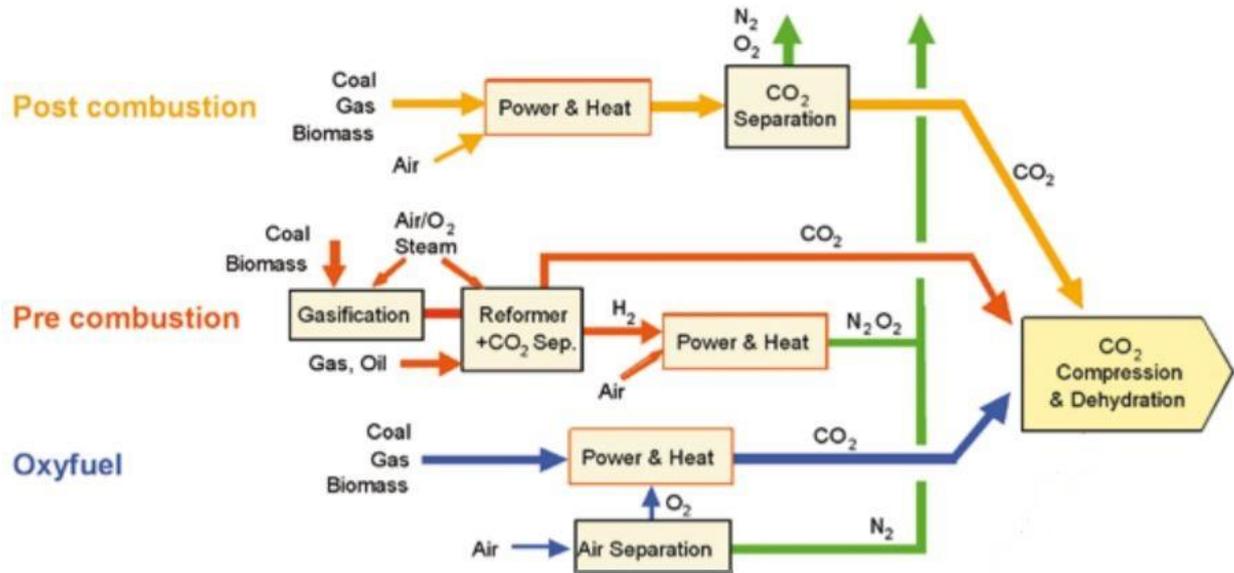


Figure 15: Capture processes for industrial CO₂ emissions (IPCC, 2005).

As processes and technologies of capturing carbon dioxide continue to be developed over time, information around these technologies might change. The various existing manufacturers of the numerous capture technologies from AGR are eager to sell their own existing solutions in the new-found industry area of carbon dioxide capture and storage.

The first defining properties for the choice of carbon dioxide capture technology are based on the carbon dioxide concentration before and after the gas treatment. Concentrations of carbon dioxide in natural gas are typically much higher than those found in flue gases resulting from a combustion process. High carbon dioxide concentrations simplify separation.

When considering only the carbon flows within an integrated mill (figure 12), the carbon balance and net captured CO₂ emissions ($CO_{2,net,capt}$) can be evaluated from the following formula

$$CO_{2,net,capt} = (CO_{2,f} + CO_{2,bio}) \times \eta_{CC} \times \eta_{CS} \quad (6.1)$$

Where $CO_{2,f}$ and $CO_{2,bio}$ refer to the total amount of fossil CO₂ and biogenic CO₂ generated at the mill. Where η_{CC} refers to the efficiency of the carbon capture process. If the carbon capture is fitted only for a specific source at the mill, e.g. the recovery boiler, one must multiply the share of emissions originating from that source by the efficiency of the carbon capture technology. For

example, post-combustion carbon capture has an efficiency of 90 % and the recovery boilers share of the total CO₂ emissions is 85 % it follows that the final η_{CC} for the recovery boiler would be ~77 %. The η_{CS} refers to the carbon storage efficiency. The captured carbon or at least a part of it must be permanently removed from the atmosphere if negative emissions is to be achieved. The efficiency of the carbon storage is expressed as 100 % if all the captured carbon is stored away without any additional emissions occurring from the steps leading and including the final storage.

6.1.2 Post combustion carbon capture

Post combustion was chosen as the go-to technology as it is the easiest to retrofit into an existing mill. Currently there are four main technologies for carbon dioxide capture in a post-combustion environment. These technologies are based on, (1) absorption with chemical and physical solvents, (2) cryogenic separation, (3) adsorption, (4) membrane separation. This thesis focuses on post combustion chemical solvent absorption because of its long commercial use starting in the 1950s for both the chemical process industry as well as the oil and gas industry. Its working principle is that a chemical solvent is reacting with carbon dioxide in the flue gas in order to separate CO₂ as presented in figure 16.

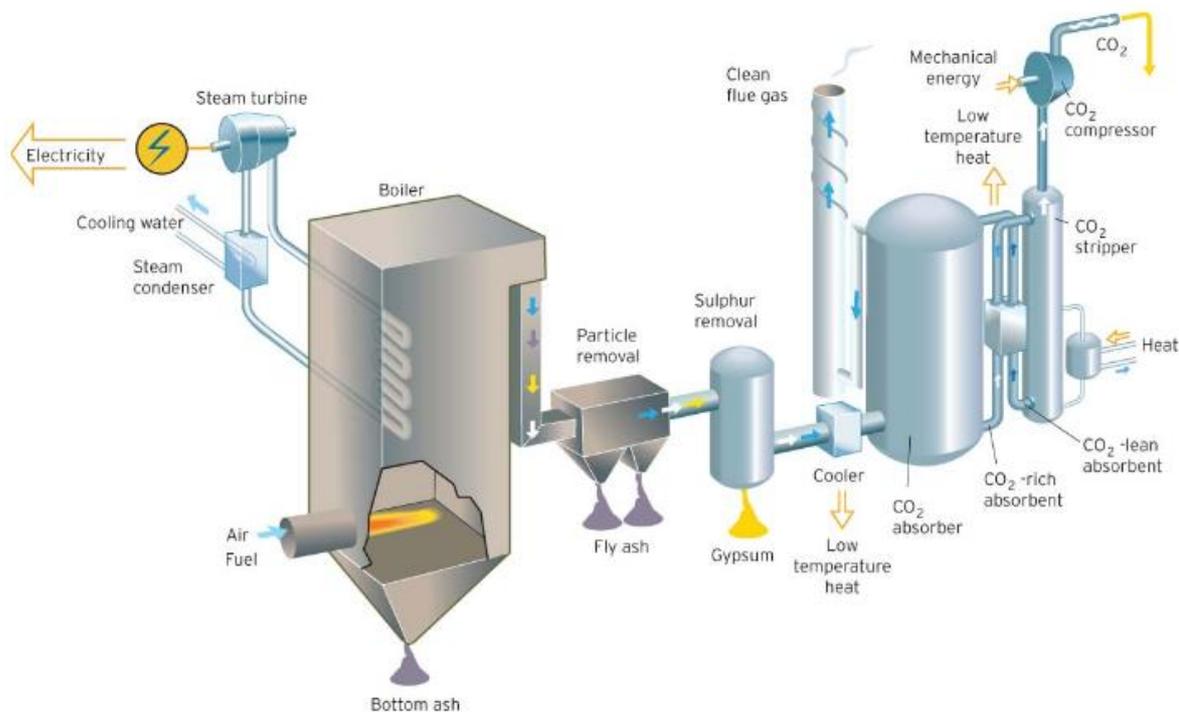


Figure 16: Post combustion capture system example (Arshad, 2009)

Amine based technology is a mature concept and used in the chemical industry over 60 years for AGR. The early demonstration CCS plants and in the newly few erected full-scale plants use amines in different forms. There is a large array of different amines Ethanolamine (MEA),

Diglycolamine (DGA), Diethanolamine (DEA), Diisopropylamine (DIPA), Methyl diethanolamine (MDEA), amine mixtures and hindered amines within three forms: primary, secondary, and tertiary, each of them has advantage and disadvantages. There are also amino acids being used. (BluCarbon Solutions, 2020)

The Hot Potassium Carbonate (HPC) technology is very mature with over 600 installations globally in the chemical industry installed over a 60-year time frame. The technology is used in various applications such as Ammonia plants, AGR from natural gas, as well as in other types of chemical processes such as Ethylene Oxide processing. These chemical processes are normally pressurized with pressure level varies depending on application and presence of catalysts. (BluCarbon Solutions, 2020)

HPC is an inorganic absorption solvent that shows to be selective and does not react with oxygen in the flue gas from a power plant. There are reactions in the flue gas with sulphur oxides and nitrous oxides, where Heat Stable Salts (HSS) are formed. The volume of HPC that reacts to form HSS needs to be replaced. If the flue gas has already installed an FGD there is only a limited amount of Sulphur oxides. The reaction with nitrous oxides is positive since the HSS that are formed are non-toxic and the result is less nitrous oxides in the atmosphere. (Smith, et al., 2016)

Chilled Ammonia Process (CAP) is a post combustion carbon dioxide capture technology that produces a high purity carbon dioxide product stream. The CAP was developed solely for the purpose of CCS by Alstom since 2006. Now BHGE has bought all rights from Alstom. Several pilot and validation facilities using the Chilled Ammonia Process with increasing capacity per evolution step have been built and tested and together operated for over 22.000 hours. The CAP plants treated combustion flue gases from both power and industrial boilers using several different fuels. (BluCarbon Solutions, 2020)

CO₂ is captured in an absorber and then cycled through a regeneration tower to drive off a concentrated stream of carbon dioxide. Traditional water washing steps is employed to recover ammonia and avoiding it in the stack. The reaction mechanism of carbon-dioxide absorption with ammoniated solution has been reviewed in the scientific community for many years. (BluCarbon Solutions, 2020) Figure 17 presents the process diagram for CAP to use for CO₂ capture.

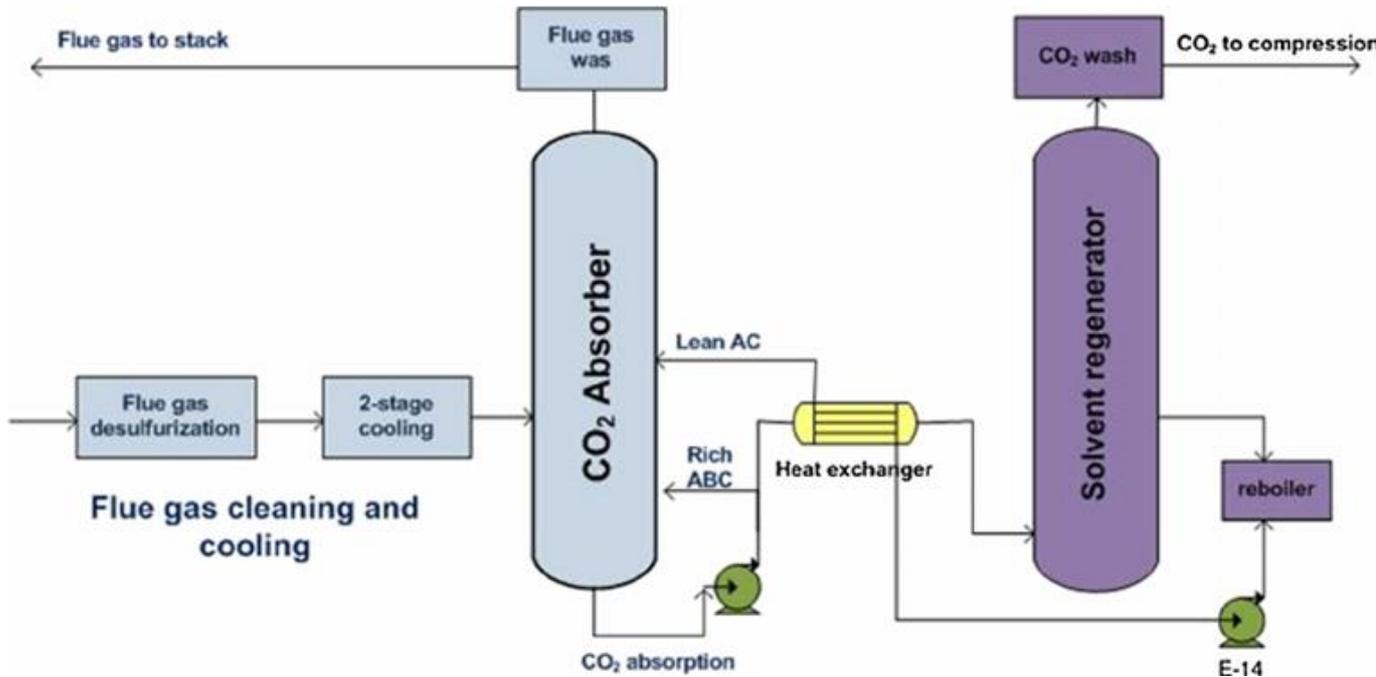


Figure 17: Chilled ammonia process diagram for CO₂ capture simplified. (Rackley, 2010)

The release of carbon dioxide in the desorber is done under pressure. The CAP technology therefore delivers carbon dioxide at a pressure of around 20 bar. This gives an advantage as the carbon dioxide later needs to be transported which is done in liquid phase after compression. CAP technology uses electricity to the refrigeration for the absorber, the compressor for the pressure in the desorber as well as drive pumps. CAP has a high regeneration rate. The only reactions are with impurities in the flue gas such as SO₂ and NO_x where HSS are formed. The volume of ammonia that reacts to form HSS needs to be replaced. Ammonia is however a low-cost solvent which means low Opex cost makeup of the solvent. The quantities of HSS are small and the HSS is harmless. The waste cost is therefore low. (BluCarbon Solutions, 2020)

6.1.3 Transport & storage

Transport of captured carbon dioxide is an established concept which is done in three different ways, pipeline, ship transport or by tank transport (truck or train). Chosen transportation will depend on the volume of the transported carbon dioxide, the storage site and the distance of the transport. The driving factor behind the choice of transport is the cost. Pipeline transport of gases,

including carbon dioxide is a well-established industry. Currently there is more than 550.000 km of natural gas pipeline and 6,500 km of CO₂ pipelines in the US for transport, though most of the CO₂ pipelines are linked to enhanced oil recovery (EOR) operations in the United States there is also a number of new pipelines under development or associated with CO₂ storage. (International Energy Agency, 2013) In Finland there is about 1200 km of natural gas pipeline (Gasum Oy, 2020). Figure 18 illustrates the carbon dioxide transport options after capture.

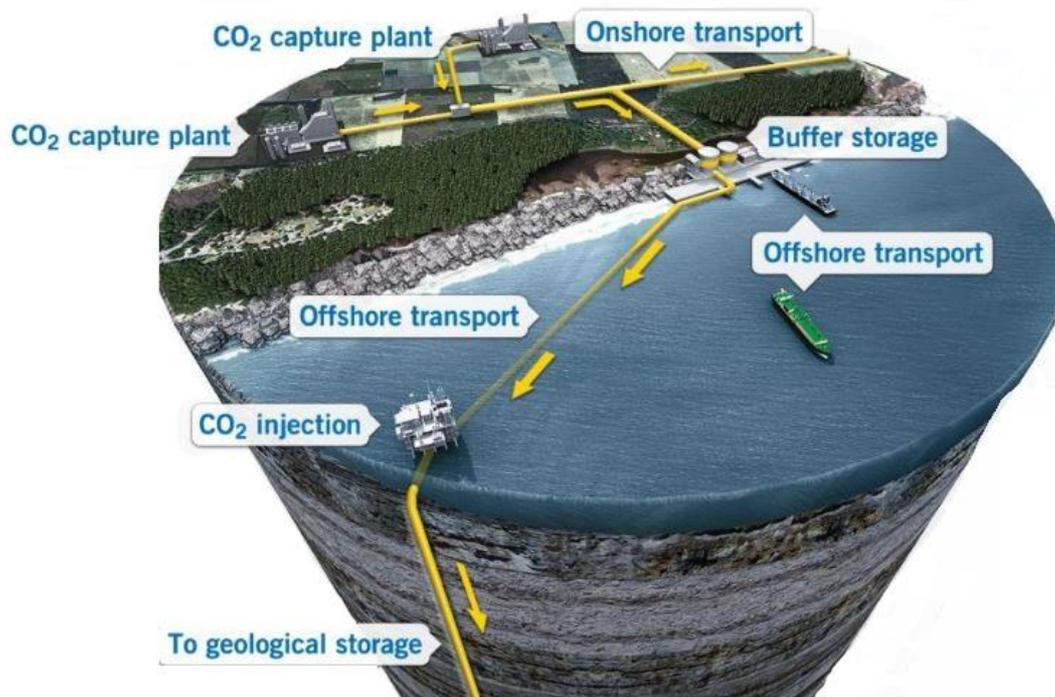


Figure 18: Carbon dioxide transport options illustrated. (Global CCS institute, 2018)

To enable efficient transport of carbon dioxide it must be conditioned for transport. The requirements for transport vary between different methods, hence each case must be considered separately. There are specific guidelines and recommendations drawn from EOR activities and literature. The CO₂ concentration requirement is varying from 95.5 % for pipeline transport to 99.5 % for ship transport respectively (VTT, 2009). Water mixed into the captured CO₂ will cause erosion, ice and hydrate formation and an increase in minimum mixing pressure in EOR utilization. In a saline deposit or in an oil deposit the oxide of sulfur and nitrogen as well as hydrogen sulfide also increase the mixing pressure of carbon dioxide and they are harmful to health and cause a change in acidity. Hydrogen (H₂), nitrogen (N₂) and argon (Ar), oxygen (O₂), carbon monoxide (CO) and methane (CH₄) must be removed from CO₂ for shipping to avoid the formation of hydrates at lower temperatures. During pipeline transport these substances should not pose an

operational threat or environmental threat but one might want to remove them to reduce the gas flow and work needed from the compression. The afore mentioned substances increase the needed compression work in a multitude of ways. Firstly, impurities raise the pressure needed for supercritical and liquid state of the CO₂. Second, the compression work needed to raise the pressure of the gas and finally, impurities, especially hydrogen, causes a higher pressure needed for storage due to reduced mixing pressure. (VTT, 2009)

Pipeline transport requires the carbon dioxide to be compressed to a pressure matching the outlet pressure and consider pressure losses along the pipeline, which typically means the pressure must be above ~55-80 bar including a safety margin subject to the ambient temperature along the entire pipeline. Transporting by ship requires the carbon dioxide to be liquefied at a pressure around 7-20 bar and a temperature of – 55° to -30°C to avoid the risk of dry ice formation.

There are no modern carbon dioxide ships with the right size on the market. The only six existing multi-gas ships constructed by Skaugen in Norway are too small, too old and consumes too much fuel. There is an opportunity to redesign, rebuild and reclassify Liquefied Petroleum Gas (LPG) ships but the initial dialogue with shippers have indicated that that reconstruction will be quite costly and building new purpose-built ships would be more cost efficient. (BluCarbon Solutions, 2020)

The construction of new purpose-built carbon dioxide ships will require 12 months for the Front End Engineering Design (FEED) period and 24 months for delivery. The indication from shippers by the Request for information (RFI) is that a 14000 m³ ship would cost around 40 million EUR and a 7000 m³ ship would cost around 25 million EUR. Detailed cost estimates should be initiated with ship builders in the next steps enabling evaluation the most cost efficient shipping scenario for Stora Enso. (BluCarbon Solutions, 2020)

According to shippers it may be advantageous to construct multi-gas ships that can transport both carbon dioxide and LPG. These ships can be more flexible and find alternative transports on return leg or other times when needed. That lowers the risk for the shippers even though the ship becomes marginally more expensive. The ships will normally have a lifetime of 25 – 30 years. There are technology suppliers, shipowners and shipbuilders contacted in the study who are willing to execute different scenarios of transporting the carbon dioxide, including buying the carbon dioxide, Build Own and Operate (BOO) ships, Build Own and Transfer (BOT) ships. (BluCarbon Solutions, 2020)

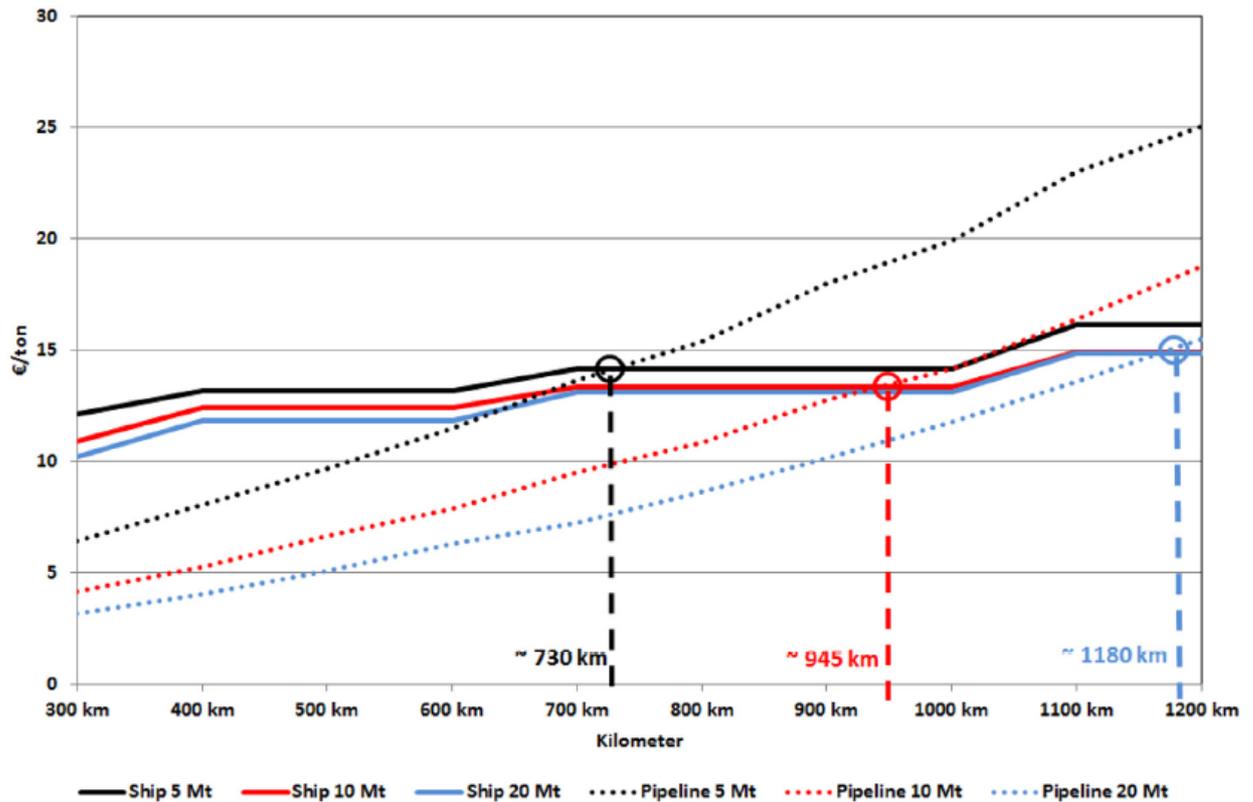


Figure 19: Comparison of cost of transporting CO₂ with ship or pipeline (Kjärstad, et al., 2016)

The cost of transport will depend on the choice of logistics. It can be observed from the figure 19 above that pipeline transport suffers significantly more diminishing returns with increased distances compared to shipping. A pipeline connection to Helsinki, Kotka or Hamina and thereafter ship transport to Faludden is likely to be the most cost-effective way to transport the CO₂. This is based on calculations from (Kjärstad, et al., 2016) where a distance of around 1000 km costs 14 – 22 €/ton for volumes between 1 – 20 Mt/a.

To create a carbon sink, the carbon dioxide needs to be stored permanently. The North Sea has a storage capacity of 70 billion tonnes of carbon dioxide (Norwegian Petroleum Directorate, 2019). The total emissions of carbon dioxide in Europe is 4.4 billion tonnes per year (European Environment Agency, 2019). This means that if 10% of the European emissions could be captured, it would equal 440 plants in Europe each capturing in average 1 million tonnes. Then the North Sea alone could store carbon dioxide for the European emitters for 160 years of production. During that time technologies are likely to have developed to avoid emissions. The storage capacity itself is therefore not a limiting factor. The limiting factors are instead the financing of the project executions of the various storage sites including drilling of injection the holes, injection and monitoring responsibilities and costs.

The two most prominent alternatives for geological storage are saline aquifers and depleted oil/gas fields. The advantage of saline aquifers is that they often are on a deeper level, separated from the oil/gas fields and are more extensive in size. The carbon dioxide plume (the volume and space taken by carbon dioxide in the reservoir) of carbon dioxide will extend as the volume increases and it is important to monitor the extension of the plume (CO₂ GeoNet, 2007). The carbon dioxide will eventually over time react with the rock matrix and form new minerals. The disadvantage is that there is no existing equipment infrastructure in place and limited geological data. Every equipment and study will be green-field, and this is expensive and takes time. Figure 20 shows the storage options for depleted oil/gas fields, deep aquifers and coal seam storage.

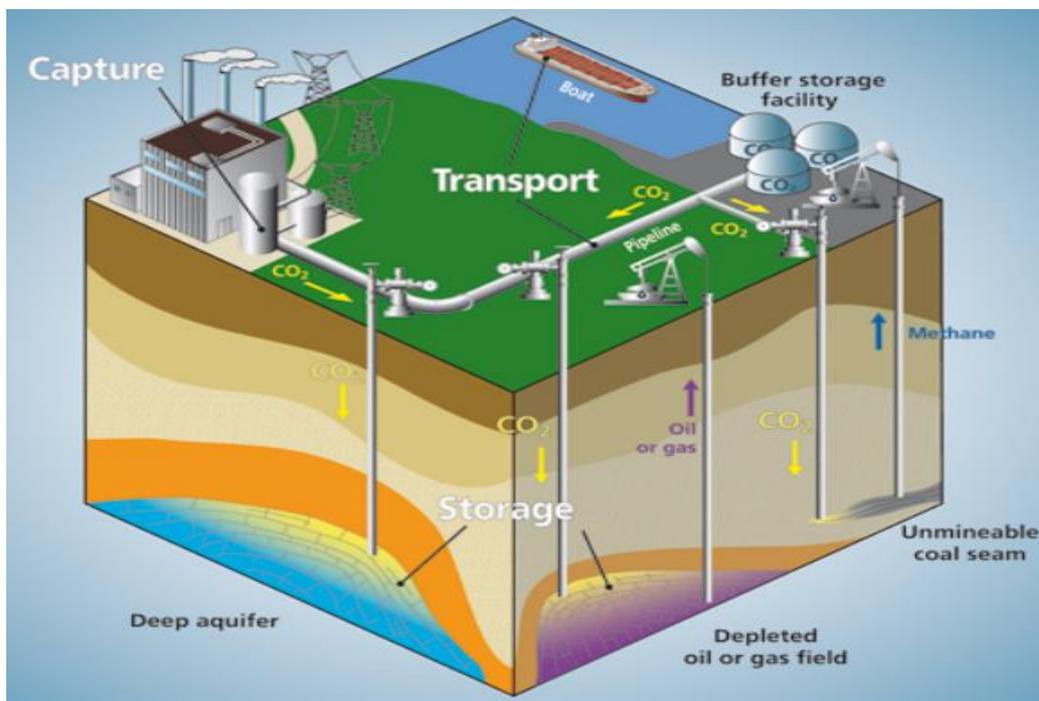


Figure 20: Depleted oil/gas field, deep aquifer and coal seam storage. (BRGM, 2013)

One advantage of the depleted oil/gas fields is that there exist extensive data of the geology of the reservoir and matrix around. There are also physical equipment infrastructures to be re-used such as holes, possibly platforms, equipment and pipelines. The owner of the field has good information about the base of the geology and capacity. The disadvantages are that there is a limited capacity in fields and there can exist leaking injection and production holes that need to be plugged and monitored (Loizzo, et al., 2010).

The large established oil companies are the organizations that will execute the storage work as they know the areas and have the skills and equipment. Figure 21 is a map over the North Sea and the capacity for CO₂ storage of 40 years of emissions from large point sources.

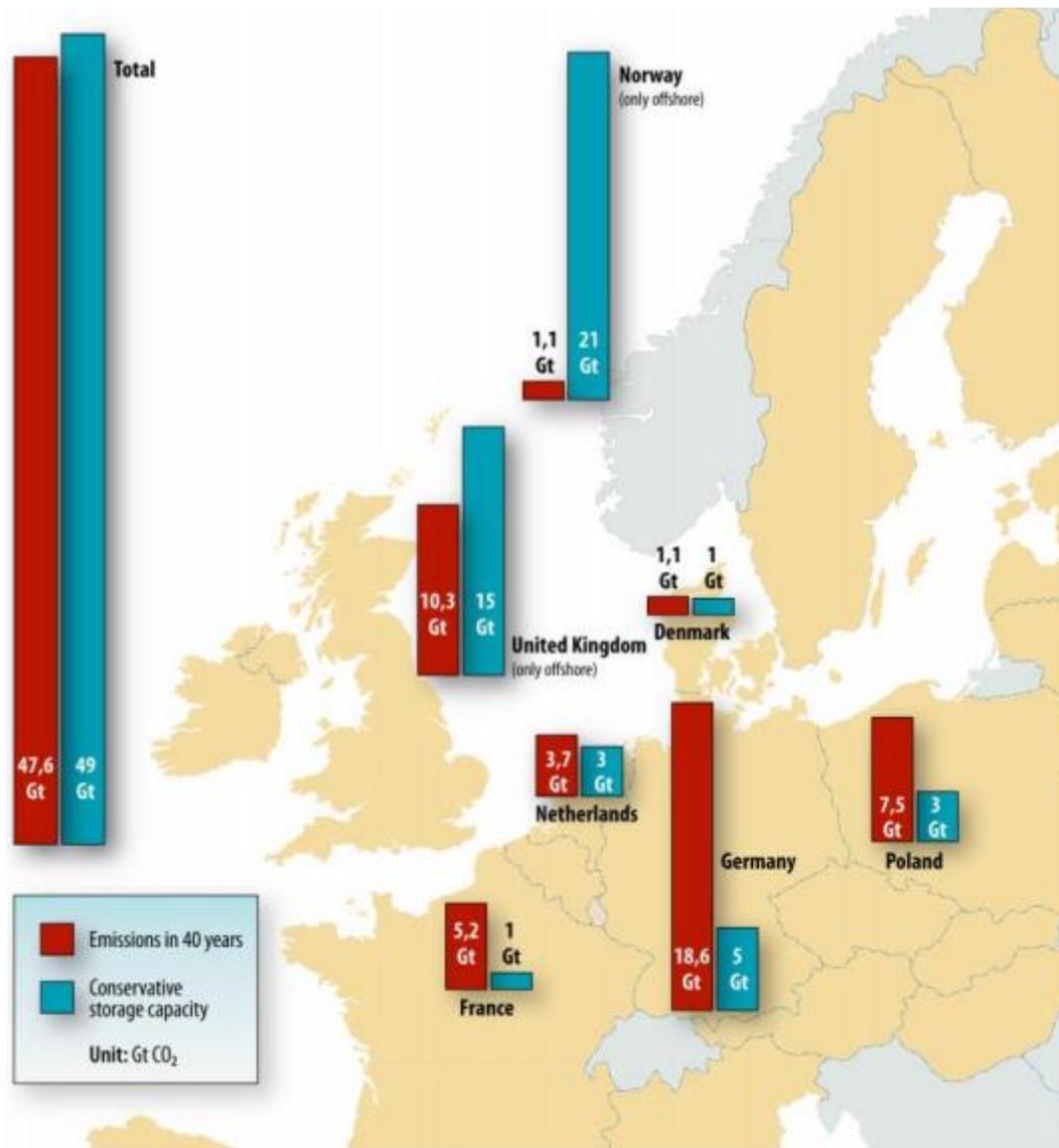


Figure 21: Overview of conservative capacity estimates for CO₂ storage compared with 40 years emissions of large point sources. (Höller & Viebahn, 2011)

6.2 CO₂ utilization possibilities on-site

The options for utilizing CO₂ on-site depend on specific details for a mill, like processes used and pulp raw material. A few concepts for utilization are listed next, lignin extraction, precipitated calcium carbonate (PCC) and tall oil manufacturing.

The CO₂ use for lignin extraction is the most significant one as it is used for acidifying a fraction of the black liquor which typically uses sulphuric acid. CO₂ use is around 150 – 250 kg per ton of separated lignin (Tomani, 2011).

Crude tall oil (CTO) is only an option for softwood mills as with hardwood and eucalyptus mills the levels of tall oil is so low that it is rarely worth extracting. CO₂ can be used as a replacement

for sulphuric acid for the acidulation of the soap to CTO from the weak black liquor, however CO₂ is a weaker acid which means only up to about 50 % can be replaced leading to a usage of about 4 – 6 kg/ADt. (Kuparinen, 2019)

PCC is utilized as a paper filler and for coatings to smoothen and whiten the papers surface (Nanri, et al., 2008). When the mill is an integrated pulp and paper mill CO₂ can be used for production of PCC by blowing the lime kiln flue gases through a solution of calcium hydroxide. The production is usually scaled to meet the local demand at the mill, an estimated CO₂ use could be 20 kg/ADt which corresponds to a PCC production of 33 kgCaCO₃/ADt. (Kuparinen, 2019) Imatra mills used to have PCC production but has since 2019 terminated the production.

6.2.1 Methane/Methanol production

Converting the CO₂ emissions originating at the mill operations into methane or methanol provides a possibility to drastically reduce a mills emissions and even reach negative emission status due to the biogenic nature of most CO₂ released provided that some or all of the produced fuel is replacing fossil fuels or sold. A recent study done by Laaksonen, et al. shows that the CO₂ emissions from the 10 largest pulp mills in Finland would provide sufficient CO₂ to cover all of Finland's energy need in transport with a energy surplus of over 60% (Laaksonen, et al., 2020).

Methane production

Methane is the second cleanest fuel to combust after hydrogen, this is because it contains only one carbon atom (CH₄). Methane also has other desirable properties like ignition in air is around 5 – 15 % while hydrogen has around 5 – 75 %, which means methane is much safer in an accident environment. The heating value of pure methane is 50 MJ/kg. (Petäistö & Snellman, 2011) Figure 22 shows an example of power-to-gas to enable sustainable chemical and fuel production.

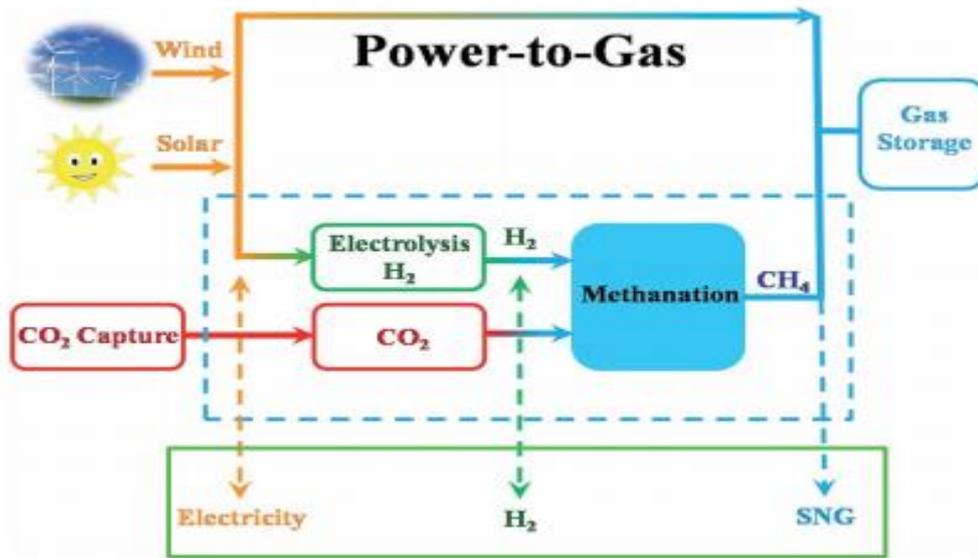


Figure 22: Example of power-to-gas enabling sustainable chemical and fuel production. (Wenhui, et al., 2018)

Methane can be produced by reacting CO_2 and H_2 under moderate heat and pressure as discovered and named after Paul Sabatier in 1912. The Sabatier-reaction can be used as a carbon dioxide removal especially suited for capturing CO_2 from fossil origin. The production of methane has even greater CO_2 reduction potential if the hydrogen is produced through renewable energy, like solar or wind powered electrolysis. The process is scalable and different catalysts have been studied, like the original nickel dust that has since been replaced by aluminium dust which provides higher efficiency.

The Sabatier-reaction is as follows:



The Sabatier reaction is one of the more established processes for utilizing carbon captured from an industrial asset. A new method using ruthenium (RU) as catalyst in an electric field lowered the temperature required for the formation of methane below 473 K, currently temperatures range from 523-673 K. Low operation temperatures are favourable to improve the CO_2 conversion because the reverse reaction of steam reforming and side reactions are suppressed at these low temperatures. (Kensei, et al., 2020)

Methanol production

Methanol is currently widely used in the chemical industry as a fuel or reactant and as a starting material for further conversion, globally almost one-third of methanol produced is used for formaldehyde production (Pérez-Fortes, et al., 2016). Figure 23 provides an example of the layout for a methanol synthesis plant with purification.

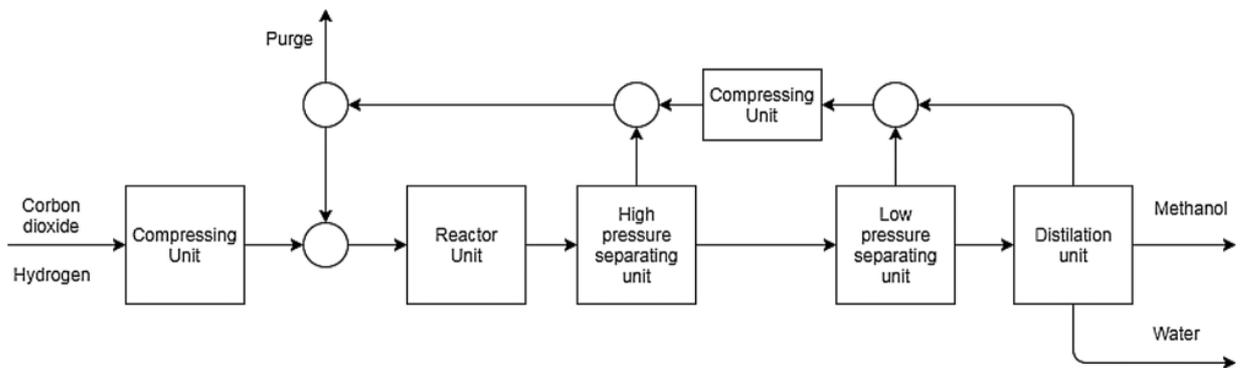


Figure 23: example layout of a methanol synthesis and purification plant (Szima & Cormos, 2018)

Converting CO₂ is a promising idea for power-to-x solutions. It may be used as a precursor for more complex transportation fuels or as an intermediate energy storage and in a large array of industrial chemicals. The current conventional way to produce methanol is typically through syngas derived from gasified coal or natural gas (Ott, et al., 2012). When synthesizing methanol from syngas it usually means having CO and CO₂ as well as many other light and heavy coproducts. Much of the energy and costs for conventional methanol plants come from the treated and separated from the final methanol product. Using pure streams of CO₂ and H₂ makes the chemistry much simpler along with the reaction products. When conventional methanol plants operate the reaction is mainly occurring as CO is reacting with H₂ which is significantly more exothermic. This means conventional plants require more cooling and are often limited to boiling water reactors, which are expensive and complex equipment. The methanation from pure streams is less exothermic and therefore enables the use of tube-cooled reactors which are cheaper, more efficient and easier to operate. (Marlin, et al., 2018)

Methanation with pure CO₂ and H₂ streams occurs through the following reaction:



The unreacted CO and H₂ can be recycled into the process to allow for maximum methanol formation. Methanol is in liquid form in its standard state which makes it ideal to transport without the need of special equipment.

A disadvantage of methanol is the fact that it is toxic towards humans, therefore, it would be preferable to further convert it to dimethyl ether (DME) which is obtained from dehydration of methanol and is less toxic and has a higher energy content (Szima & Cormos, 2018). The reactants for the DME production process can be produced from renewable sources like, biomass and solar or wind, this makes DME a liquid energy storage which is easily dispensable, has a high energy content and is easily applicable. Currently many trial plants of varying size are running or being built, for example, TOYO has developed a catalyst and technology for indirect DME production (TOYO, 2006) and ExxonMobil developing methanol to gasoline (MTG) solutions in six plants currently operating with DME as an intermediate leading to the formation of light olefins (ExxonMobil, 2014).

7 CASE: IMATRA MILLS

Imatra Mills in southern Finland consists of two production units – Kaukopää and Tainionkoski – that produce chemical pulp and consumer board. Imatra Mills is one of the largest Consumer Board mills in the world, with over 90% of its produce exported to Europe and Southeast Asia. Annual capacity consists of 1.155.000 tonnes consumer board, 825.000 tonnes of pulp from Kaukopää, 195.000 tonnes of pulp from Tainionkoski and 285.000 tonnes of plastic coating. Imatra mills is an old integrate and therefore many of the concepts would require significant investments into new equipment and processes to get to the level of a modern mill. However, within 20 years it is assumed that a number of larger investments will be directed to the mill.

The goal is to reach carbon negative operations by the year 2040 within the Imatra mill integrate, the steps described in section 7.3 technological and economical evaluation of outlined concepts aim to reach this goal and are dimensioned accordingly.

The evaluation of heat and electricity balance changes was calculated using excel where inputs of mill operational data are fed in. The heat and electricity demand of different CO₂ capture set-ups are taken from a report by BluCarbon Solutions.

7.1 Mill description

Assumed initial values for the mill are based on information from the master excel for LCA calculations gathered in 2019 containing the data for Imatra Mills integrate. Figure 24 shows the simplified principle of energy allocation used in the master excel.

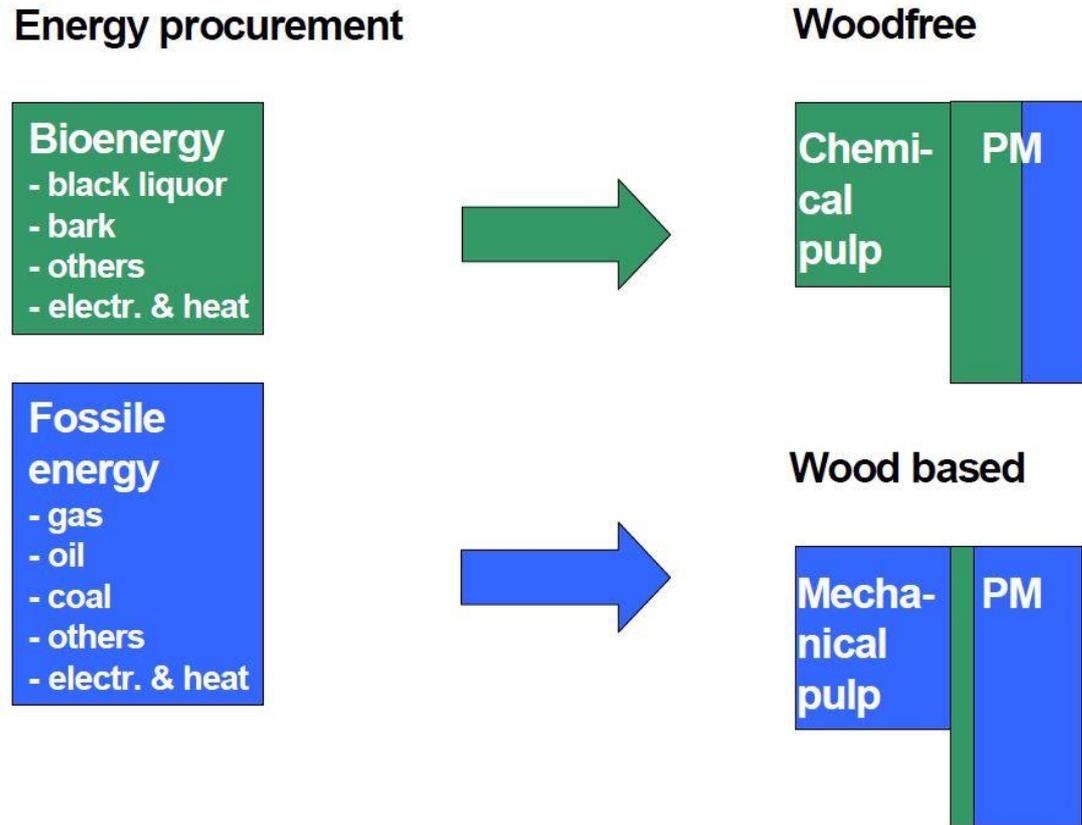


Figure 24: Simplified energy allocation for produce (Stora Enso Oyj, 2019)

Table 6 presents some key parameters for the Imatra mills integrate that were used as a basis for calculations.

Table 6: Data for Imatra mills integrate calculations

| Pulp and paper mill | | |
|---------------------------------|------------|-------|
| Operating days | d | 355 |
| Availability | % | 95 |
| Load factor | % | 100 |
| FLH | h | 8094 |
| Electricity | | |
| Electricity production | % of total | 51 |
| Electricity purchase | % of total | 49 |
| Steam | | |
| Steam to process | % | 83 |
| Steam to electricity production | % | 17 |
| CO2 sources | | |
| Power Boilers | kg/s | 18.05 |
| Recovery boiler | kg/s | 53,33 |
| Lime kiln | kg/s | 2.61 |
| CO2 sources in TCO2/year | | |
| Power Boilers | MTCO2/year | 0.55 |
| Recovery boilers | MTCO2/year | 1.64 |
| Lime kilns | MTCO2/year | 0.08 |
| Total | MTCO2/year | 2.27 |

The values for heat consumption and electricity consumption for carbon capture are currently taken from a report from (BluCarbon Solutions, 2020) investigating a Swedish pulp mill implementing CCS. Cost analysis is on a high level and will have great variation depending on mill set-up and location. The data is also classified as confidential and therefore not part of the public version of the thesis.

7.2 Basis for mill calculations

The mill mass and energy balances are taken from a master excel used for LCA calculations of Imatra mills products in 2019, which contains aggregated data from raw material consumptions and process measurements. Some data is also from a report by BluCarbon Solutions (BluCarbon Solutions, 2020). The data is generated annually for Imatra Mills, it is not presented in the work as there is business critical information included.

7.2.1 Processes at the mill

Imatra mills is a large integrated pulp and paper mill, figure 25 shows the shares of biogenic and fossil CO₂ emissions occurring from the mill. The consumption data of steam and electricity is from 2019 gate-to-gate inventory data for the mill, which is annually updated. It consists of detailed energy and material balances for the mill and is the basis for all the information and calculations done in this thesis.

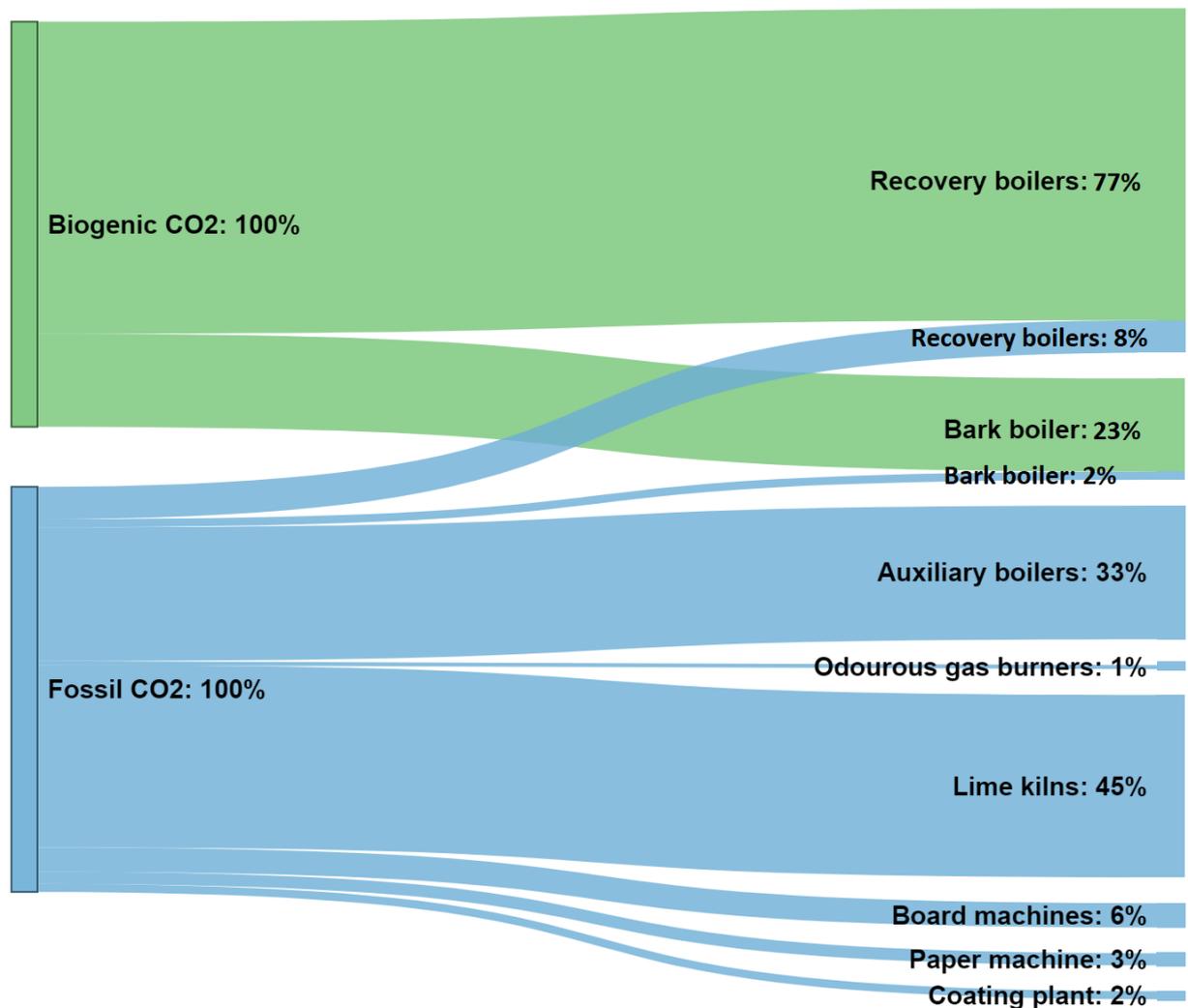


Figure 25: CO₂ flows at Imatra Mills (Stora Enso Oyj, 2019)

The recovery boilers and bark boilers have 8 % and 2 % fossil CO₂ fraction because natural gas and fuel oil is used during start-up and during process disturbances. The lime kiln is the single largest source of fossil CO₂ because it is utilizing natural gas as fuel, still the fossil emissions only account for about 10 % of the total CO₂ emissions occurring at the mill.

The steam consumption at Imatra mills is mainly split between the evaporators, board- and paper machines and fiberlines as figure 26 demonstrates. It is therefore important to evaluate possibilities of enhancing the evaporation and board- and paper machine processes.

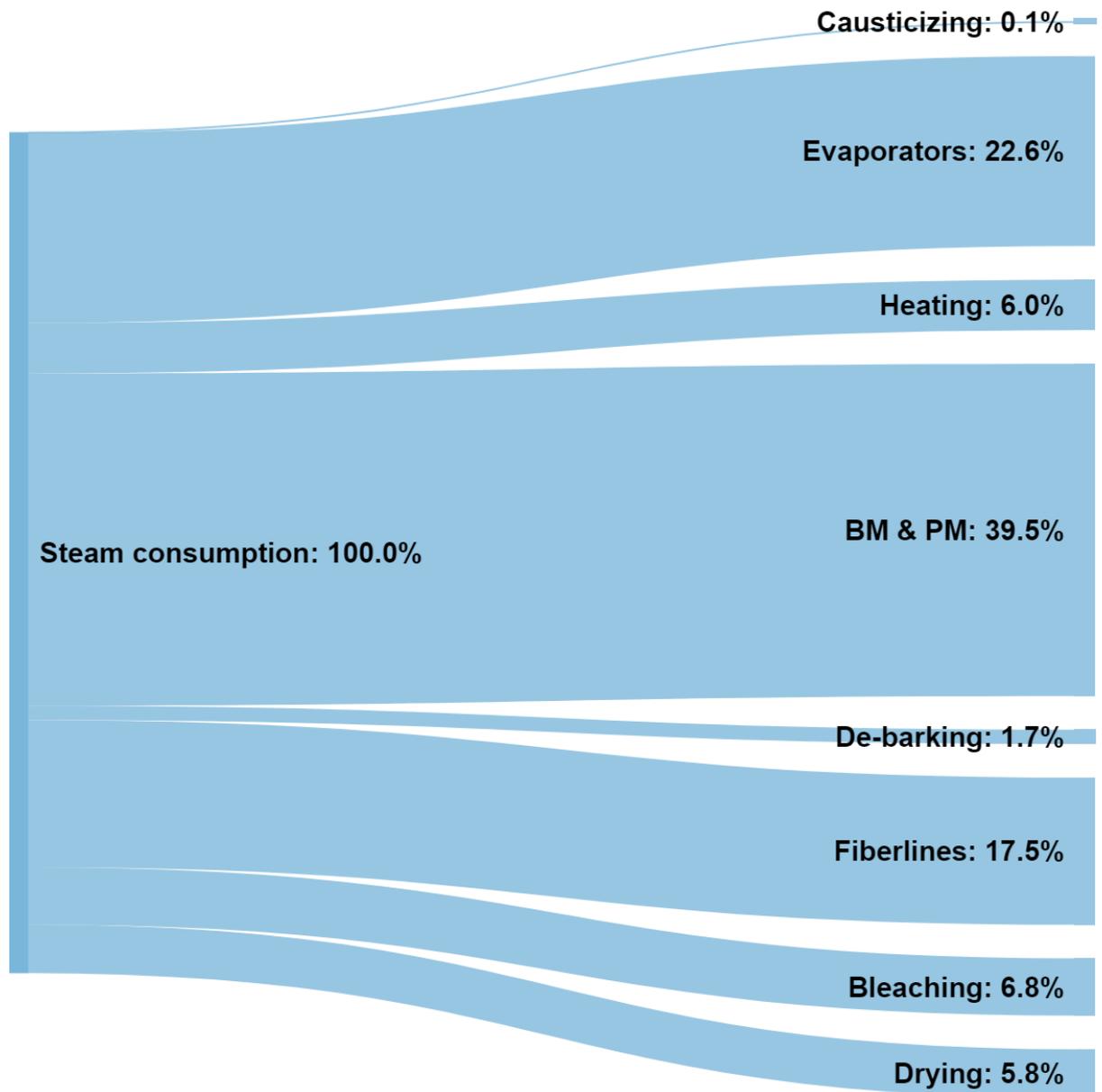


Figure 26: Steam consumption in processes (Stora Enso Oyj, 2019)

The electricity consumption of the processes at the mill is displayed in figure 27, it is to be noted that Imatra mills is an old mill and therefore the energy efficiency is not state of the art, and the paper and board production uses quite a significant share of the electricity approximately 35 %.

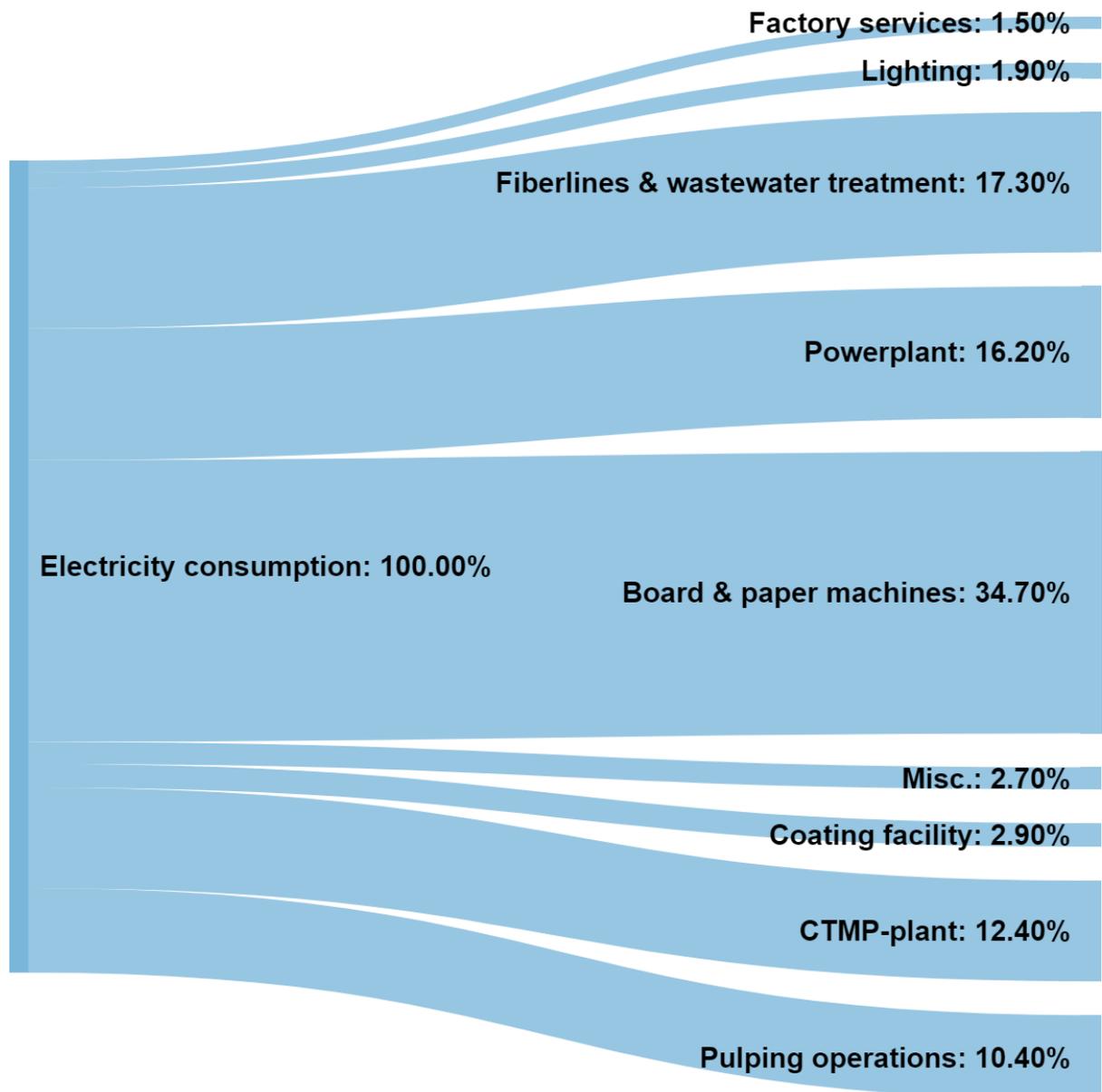


Figure 27: Electricity consumption in processes (Stora Enso Oyj, 2019)

7.3 Technological and economical evaluation of outlined concepts

In this chapter the outlined concepts are evaluated from technological and economical aspects. The variables are indicative and in order to have a more detailed evaluation a much more detailed and site-specific study must be done. However, the evaluation should give an indication of what is currently feasible and what is not.

7.3.1 Increasing energy production/reducing energy consumption

In this section some of the possible ways to increase the energy production capacity or reducing the energy consumption are described. Some of the solutions are easier and cheaper while others would require significant investments.

Increasing the operating parameters for the recovery boilers to 100 bar and 510 °C as described as BAT (European Commission, 2015) would increase the electricity production in the turbines by increasing the quality of the steam. The main limiting factor is the corrosion of the recovery boilers heat transfer surfaces which means that the parameters cannot be raised indefinitely (Salmenoja, 2013). At Imatra mills there is an idea to increase the capacity of recovery boiler 5 by 26 % in 2020 which should positively affect the mills energy balance. Another measure is to increase the dry solids content in the black liquor closer to a modern 85 % but this would require additional or new evaporators to be installed.

The use of excess steam in a low pressure turbine would increase the electricity production but is only feasible if the steam production exceeds the demand (European Commission, 2015). This is not the case throughout the year at Imatra mills, during winter back-up boilers must be used to generate steam. In the summer however excess steam is available, and a more detailed cost vs. benefit analysis would have to be made.

With increased steam production, the drying of bark used at the bark boiler could be implemented to raise the efficiency of the bark boiler. Usually the bark has just been ripped and pressed at the de-barking station but requirements to increase the efficiency has re-introduced the drying process to increase the heating value and combustion properties of the bark. (Holmberg & Ramm-Schmidt, 2017)

Typically, one can find steam saving potential in the range of 5 – 10 % and in electricity consumption around 2 – 5 %. These savings usually come from process optimisation like valve and motor operation and new ways of working and can be achieved without significant investments to new equipment. (Kyllönen, 2020)

Implementing the concepts from section 4.3 Emerging energy-efficiency technologies is not considered in this thesis as the concepts are still in development phase and a much more detailed evaluation on the effects on the processes would be needed for each technology.

7.3.2 Fossil fuel swap in lime kiln

The swap from fossil fuels to renewable alternatives at Imatra mill has been evaluated earlier (Mäkelä, 2019). The evaluation was done for two options, upgrading the old lime kilns to use bio-based fuels and the second option was to build a new lime kiln all together and integrate it to the old causticizing plant or upgrading the causticizing plant as well. The evaluation found that currently only solid biofuels would be an option to consider in replacing the fossil fuels currently in use, namely, natural gas and heavy fuel oil. The recovery boilers at Imatra mills are not a bottleneck for pulp production and the integrate is already relying on some purchased fuels. This excludes lignin separation as it would reduce the output from the recovery boilers even further. Additionally, reducing recovery boiler energy output would mean a bigger load to the bark boiler and auxiliary boilers. When gasifying fuel the homogeneity of the feedstock is important so the bark from the debarking station should be used for gasification. Using the bark for gasification means the bark boiler would need to be supplied with external fuel like wood residue from forestry or other residues that cannot be utilized for pulp or timber industry.

Sawdust is being separated at the hardwood pulp production which equals around 15 000 tonnes a year, this is not enough even for the smaller lime kiln that would require some 60 000 tonnes a year. Additional saw dust could be purchased from the close by Stora Enso Honkalahti sawmill but relying on a single raw material and potential source is too great of a risk. Saw dust is better utilised for refinement as raw material for pulp and pellets.

Poor availability and bad synergy for other burners like auxiliary boilers means that pulverised fuels are not to be considered. Natural gas will most likely remain as an important backup fuel and certificates for biogas could be purchased in the future, allowing for fossil free production of lime. Some other options like methanol which is currently combusted at the odorous gas boilers where sodium bisulphite is produced from the scrubbers for the CTMP plant or tar oil and turpentine could also be considered but they are most likely to become commercial products for refinement or selling, therefore it is not foreseen to utilize them.

The main concern when considering the switch is linked to the capacity of the lime kiln. With pulverised fuels or lignin, the exhaust gas flows and energy consumption are comparable to that of natural gas. However, the use of product gas increases the exhaust gas flows causing a limited capacity in the lime kiln. Thus, using the old kilns means accepting reduced capacity if the kilns turn out to be a bottleneck. Therefore, the best solution would be to invest in a new lime kiln with a capacity of 1 200 tCaO/d if the pulping capacity allowed by the recovery boilers is to be fully

utilized in the near future. Dimensioning for the new lime kiln would lead to an 8.3 MW energy demand for drying and a fuel consumption of about 630 GWh/a which requires a significant amount of bark. The increase of solids for the black liquor from current ~70 % to a modern ~80 % is desired to guarantee competitive energy efficiency and fuel availability. (Mäkelä, 2019) Figure 28 shows an example for gasification of biomass for use as fuel in a lime kiln.

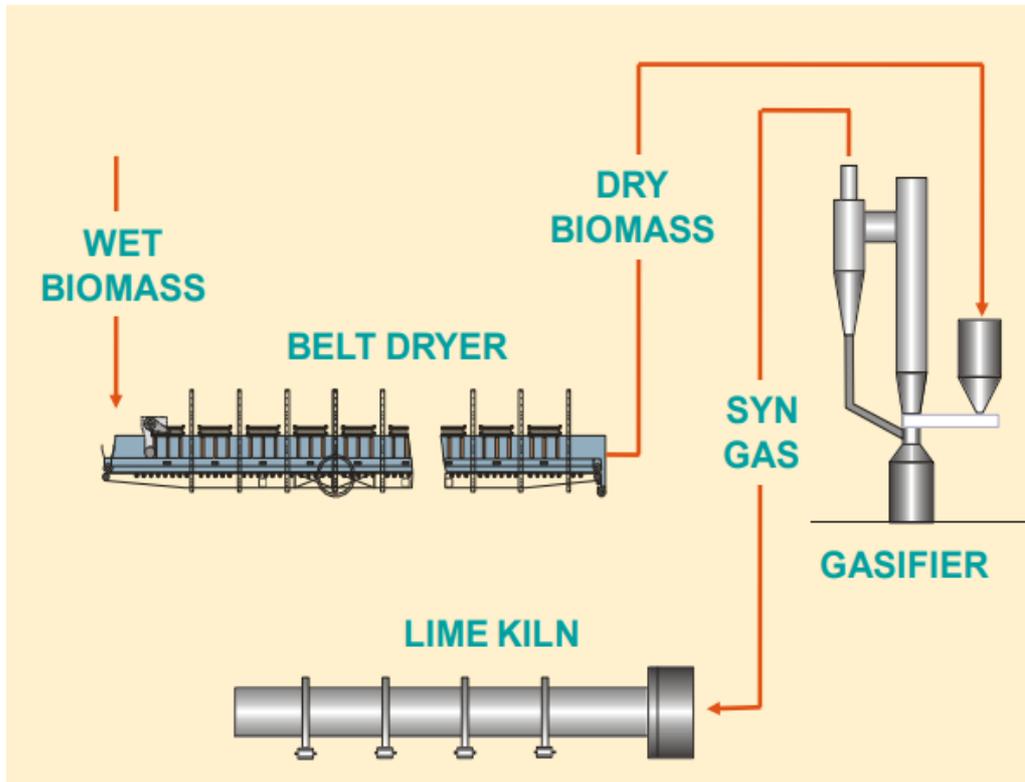


Figure 28: Lime kiln gasification example (Metso, 2013).

An important consideration to take into account when deciding investments for the fuel swap of the lime kilns is the environmental targets set by the EU, Finnish government and Stora Enso, meaning that the payback time cannot be the only consideration for the future. Investment cost for the new plant is in the range of 20-100 m€ based on other similar projects done by (Metso, 2013) & (Metsä Fibre Oy, 2013) The swap to biogas would incur an additional cost of about ~8 M€/a assuming NG price of 30 €/MWh with current prices of biogas being around 25 % higher (Gasum Oy, 2020), excluding the sale of any ETS credits available. Switching to biogas for the lime kiln reduces the fossil CO₂ emissions to 0, however integration for power boilers and recovery boilers needs to be assessed separately.

7.3.3 Carbon capture and storage

Stora Enso has conducted a feasibility study with BluCarbon Solutions consultancy, where the aim was to evaluate the implementation of CCS in two of our mills located in Sweden, Skutskär pulp mill and Skoghall pulp and paper mill. Skutskär is in eastern Sweden, primarily producing fluff pulp and pulp for liquid packaging and speciality paper, the capacity being 540 000 tonnes/a. Skoghall mill is in western Sweden and is producing liquid packaging board and packaging for dry food. Skoghall mill has annual capacities of 875 000 tonnes of board, 657 000 tonnes of pulp and 120 000 tonnes of polymer coating. The feasibility study looked at the possibility of capturing 1 million tonnes of CO₂ annually only from Skutskär. This is because Skoghall is located inland with difficult logistics. The findings are applicable for Imatra mills as well. This is because Skutskär and Imatra fiberline 3 are of similar size. The results are still indicative and worth considering for Imatra mills as well when the goal is to reach carbon negativity within the next 20 years.

The carbon capture plant was designed to capture the flue gases as a combined flow from the lime kilns and recovery boiler, resulting in a flow of 468 000 Nm³/h and with a CO₂ content of 19 % (Stora Enso Oyj, 2019). The study only looked at post-combustion capture as it was the only feasible route without major rebuild and process changes to the current set-up of the mill. The flue gas temperature of 192 °C must be cooled down to fit the requirements of the carbon capture plant. The cooling need for the flue gases is found in the “Flue gas” column of table 7 below, as well as the cooling need for the capture plant and liquefaction plant. The cooling could be done by water pumped from the nearby lake.

Table 7: Energy demand for mixed flue gas of recovery boiler and lime kiln (BluCarbon Solutions, 2020).

| Technology | kWh/t CO ₂ | | | Total kWh/t CO ₂ |
|-------------------|-----------------------|---------|------|-----------------------------|
| | Flue gas | Capture | Liq. | |
| Amine | 847 | 660 | 250 | 1757 |
| HPC electric | 855 | 42 | 250 | 1148 |
| HPC steam capture | 962 | 831 | 250 | 2043 |
| CAP | 843 | 659 | 0 | 1503 |
| PSA/Cryocap | 879 | 524 | 0 | 1403 |

The study found a positive scaling effect noticed as capture cost reduction with increased volumes of flue gases and increased CO₂ concentration. This means that implementing a full-scale CCS should be designed around the mixed flue gas flow rather than just the lime kiln or recovery boiler. Table 8 shows the steam, electricity and cooling needs for each technology investigated.

Table 8: Steam, electricity and cooling needs for the technologies (BluCarbon Solutions, 2020).

| Technology | Steam | Electricity | Cooling | Unit |
|-------------------|-------|-------------|---------|------|
| Amine | 102 | 43 | 215 | MW |
| HPC electric | 0 | 71 | 128 | MW |
| HPC steam capture | 75 | 70 | 228 | MW |
| CAP | 94 | 44 | 215 | MW |
| PSA/Cryocap | 0 | 68 | 192 | MW |

The estimated size for the intermediate storage on the mill site is 15.000 m³ CO₂. The intermediate storage is needed in order to optimize the transport of the CO₂ and to protect against any possible disturbances in the transportation chain. The tanks would be spherical tanks with each having a volume of 7500 m³ being 23 meters in height and require an area of approximately 3700 m². The area for the capture and liquefaction plant would be around 10.000 m² optimally located close to the recovery boiler and lime kiln, a pipeline connecting to the buffer storage would need to be built as well. The cost for transport is ranging from 10 – 20 €/t CO₂ obtained from a request for information from shippers of CO₂. A cost of around 30 €/t CO₂ was used as a more conservative estimate (Johnsson & Kjärstad, 2019).

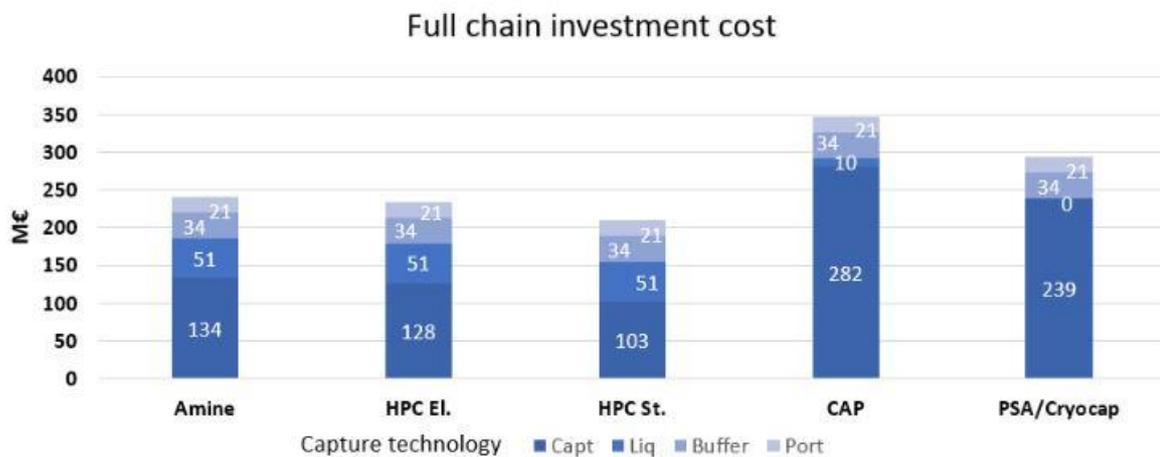


Figure 29: Investment cost for mixed flue gases with different technologies. (BluCarbon Solutions, 2020)

Figure 29 describes the estimated full chain investment cost for the different technologies concerning CO₂ capture, liquefaction, buffer plant and port systems for the mixed flue gases of recovery boiler and lime kiln. The range is 203 – 347 m€.

Figure 30 shows the total full chain cost for 20 years of operation for mixed flue gases where transport and storage costs are assumed as 30 €/t CO₂ and 22 €/t CO₂, respectively. It is unclear if the storage costs include monitoring costs as well. A report by IPCC estimates storage costs to be

6 – 31 \$/t CO₂ for pipeline dispersion and 14 – 15 \$/t CO₂ for platform or ship dispersion, hence a value of 22 €/t CO₂ was chosen (IPCC, 2018). This means that an investment of around 300 m€ combined with yearly costs of around 100 m€ is required to capture 1 Mt of CO₂/a.

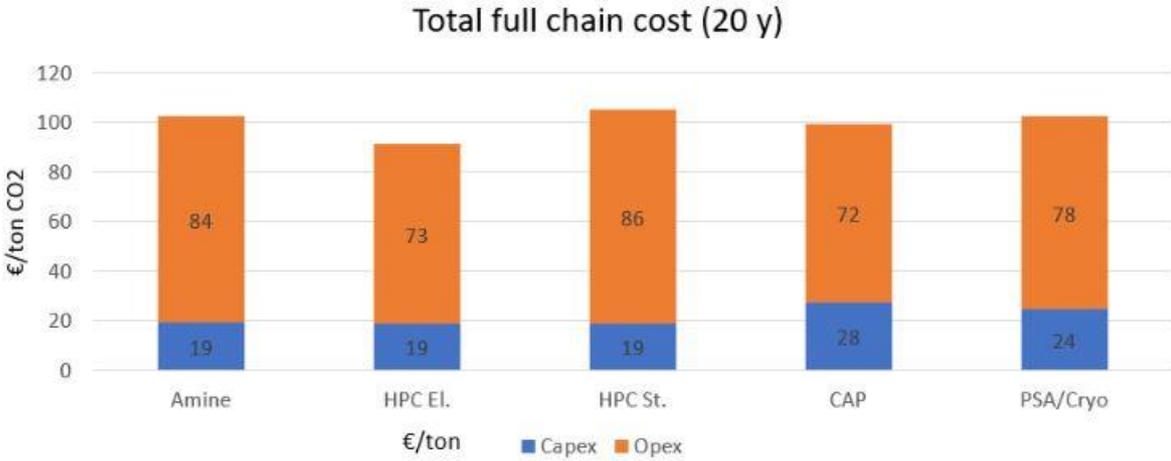


Figure 30: Full chain cost of BECCS for mixed flue gases. (BluCarbon Solutions, 2020)

Figure 31 shows the Opex cost of CC assuming Amine capture to be the chosen technology, and assuming biogenic CO₂ is included in the EU ETS system in a way of which one could get compensation for storing CO₂ permanently. Compensation is assumed to be 50 % of ETS CO₂ price (40 €/tCO₂) in 2030 and 100 % in 2040 (50 €/tCO₂).

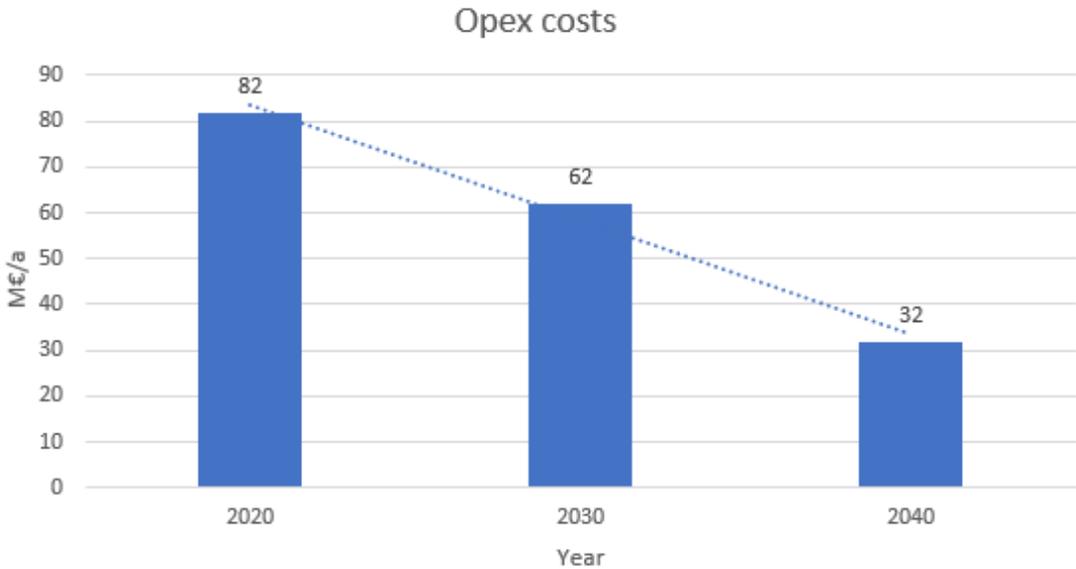


Figure 31: Opex of amine based CC from 2020-2040

This shows that CCS will not be economically feasible with the current assumptions even in 2040. A large share of the cost of BECCS is the transport and storage (52 €/t CO₂) which means that the

actual captured amount has a smaller impact on the final cost, this is why methane or methanol production is evaluated as an option.

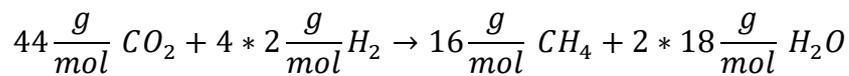
7.3.4 Methane/Methanol production

The calculations are made in excel based on equations number 6.2.1 and 6.2.2. Table 9 shows the molar masses of reactants and products. Molar masses for carbon, hydrogen and oxygen are 12, 1 and 16 g/mol respectively.

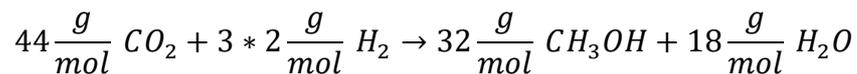
Table 9: Elements with molar masses

| Element | Molar mass (g/mol) |
|--------------------|--------------------|
| H ₂ | 2 |
| CO ₂ | 44 |
| H ₂ O | 18 |
| CH ₄ | 16 |
| CH ₃ OH | 32 |

Methane synthesis $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$, the proportion for molar masses is:



Methanol synthesis $CO_2 + 3H_2 \rightarrow CH_3OH + H_2O$, the proportion of molar masses is:



Based on these proportions the methane or methanol production and hydrogen demand for methane production was calculated based on available CO₂ presented in table 10. The ratios are in-line with calculations for a methanol plant (Kourkoumpas, et al., 2016).

Table 10: Masses of substances for methane and methanol production

| Substance | Mass (t) |
|---------------------------|----------|
| CO ₂ | 190 000 |
| H ₂ (methane) | 34 545 |
| H ₂ (methanol) | 25 909 |
| CH ₄ | 69 091 |
| CH ₃ OH | 138 182 |

The calculations are based on capturing approximately 1 Mt of CO₂/a, which corresponds the capture amount in the studied carbon capture installation. Purified CO₂ is obtained as a result of carbon capture from the flue gases at the mill. The methane or methanol production are dimensioned so that the produced product covers the current fossil fuel demand. In addition, it is assumed that the remaining captured CO₂ will be sold for further use and the fossil CO₂ credits are sold to the market and the oxygen from the electrolysis is used for bleaching of pulp, the produced amount of oxygen is enough to cover the whole demand at the mill (Ragnar & Ala-Kaila, 2004). The excess oxygen is sold to market. The ETS credits allocated for the mill are assumed to remain the same. There are 3 scenarios for both methane and methanol solutions with varying parameters for the calculations, they are presented in table 11 below. The oxygen price is estimated by (Hurskainen, 2017) and liquid CO₂ by (Mikunda, et al., 2015) (BOC Limited, 2020). Grid electricity cost is assumed to decline over 20 years as higher energy efficiency is reached in all sectors and capacity will grow. The liquid CO₂ price is also expected to increase as the demand for bioCO₂ will increase to provide 100 % renewable fuel production in the future.

Table 11: Different parameters used to evaluate economic aspects of methane and methanol production

| Parameters | 2020 | 2030 | 2040 |
|-----------------------------|------------------------|------------------------|------------------------|
| Grid electricity | 30 €/MWh | 25 €/MWh | 20 €/MWh |
| ETS price | 25 €/t CO ₂ | 40 €/t CO ₂ | 50 €/t CO ₂ |
| Oxygen sold | 48 €/t | 48 €/t | 48 €/t |
| Liquid CO ₂ sold | 60 €/t | 70 €/t | 75 €/t |

Capex costs are based on current price estimates and might change drastically if CC or electrolysis becomes widely implemented during 2030-2040. Table 12 shows the specifications for the electrolysis plant for both methane and methanol production. The price for the cost of the electrolysis plant needed was calculated using data from works by (Saba, et al., 2018), (Petäistö & Snellman, 2011), (de Jong, 2018) and (Hydrogenics, 2019) The price of freshwater for the electrolysis is inconsequential as it typically is 1-2 % of other costs, and the process produces water as a by-product which can be looped back into the process.

Table 12: Electrolysis plant data

| Electrolysis plant data | Values | |
|------------------------------------|--------------------------------|---------------------|
| | Methane production | Methanol production |
| H ₂ production | 34 545 t | 25 909 t |
| Oxygen produced | 276 363 t | 207 273 t |
| Efficiency of electrolyser, η | 80 % | |
| Power demand | 178 MW | 133 MW |
| Electrolyser type | Proton exchange membrane (PEM) | |
| Investment cost | 500 €/kW | |
| Area needed | 285 m ² | 213 m ² |

CC and liquefaction have a lower investment and Opex cost due to the fact that a transport hub and transport and storage is not needed for these cases. Table 13 shows the Capex and Opex assumptions used for the methane and methanol production calculations for 2020. In 2030 and 2040 the electrolysis Opex costs for methane production are 36 M€/a and 28.9 M€/a respectively. For methanol production the Opex costs in 2030 and 2040 are 21.6 M€/a and 16.2 M€/a respectively.

Table 13: Capex and Opex for equipment and operation

| Equipment & operation | Capex (M€) | | Opex (M€/a) | |
|----------------------------------|-----------------------------|-------------------------------|------------------------------|--------------------------------|
| Carbon capture & liquefaction | 219 | | 30 | |
| Methane/methanol synthesis plant | 180 | | 5.4 | |
| Electrolysers | 89 (CH ₄) | 67 (CH ₃ OH) | 43.2 (CH ₄) | 32.4 (CH ₃ OH) |
| Total | 488 (CH₄) | 466 (CH₃OH) | 78.6 (CH₄) | 67.8 (CH₃OH) |

In the calculations it is assumed that all electricity needed for electrolysis is bought from the grid and is renewably produced. Assuming that the ETS credits, excess oxygen and liquefied CO₂ is sold at respective prices in table 11 the revenue from those in methane production is presented in table 14.

Table 14: Revenue streams in methane production

| Revenue streams | Methane, M€/a | | | Methanol, M€/a | | |
|------------------------|---------------|-------------|-------------|----------------|-------------|--------------|
| | 2020 | 2030 | 2040 | 2020 | 2030 | 2040 |
| ETS credits | 6,3 | 10 | 12.5 | 6.3 | 10 | 12.5 |
| Liquid CO ₂ | 48.6 | 52.5 | 56.3 | 45 | 56.7 | 60.75 |
| Oxygen | 11.6 | 11.6 | 11.6 | 8.2 | 8.2 | 8.2 |
| Total | 66.5 | 74.1 | 80.4 | 59.5 | 74.9 | 81.45 |

This means that for the case of methane production the costs of operation from 2020 – 2040 will steadily decrease and be profitable in 2030 as presented in figure 32.

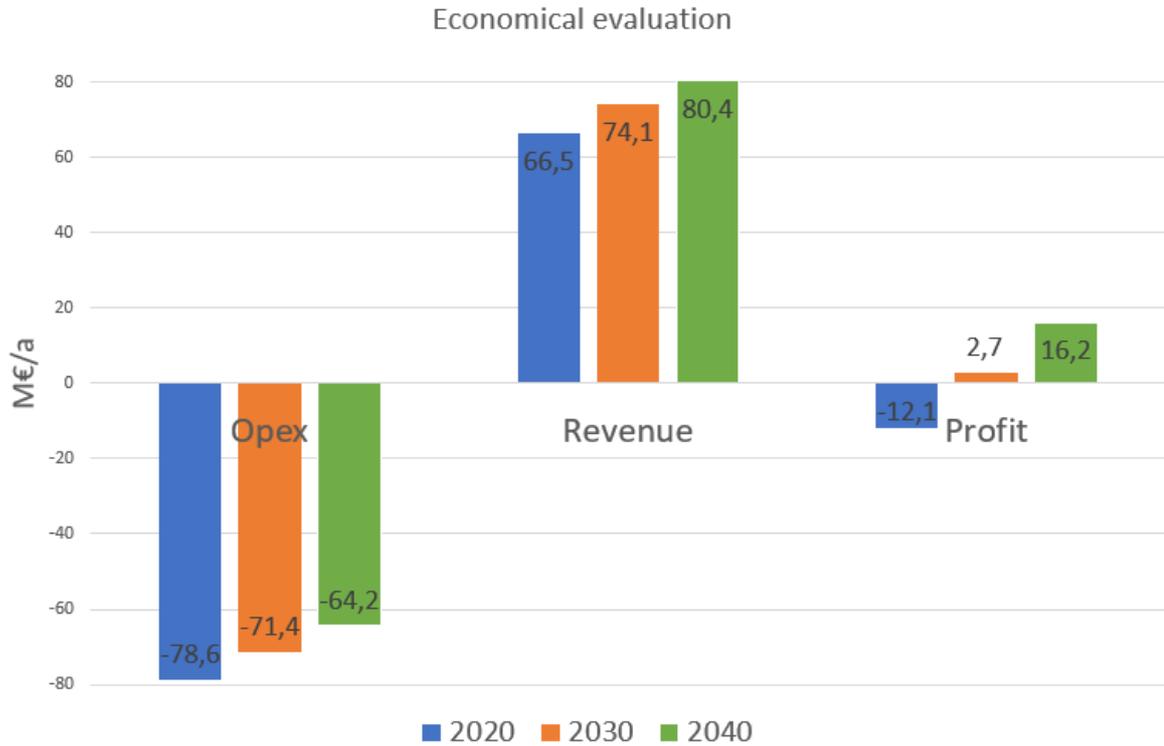


Figure 32: Economic analysis of methane production

Figure 33 shows the economic evaluation of methanol production from 2020 – 2040.

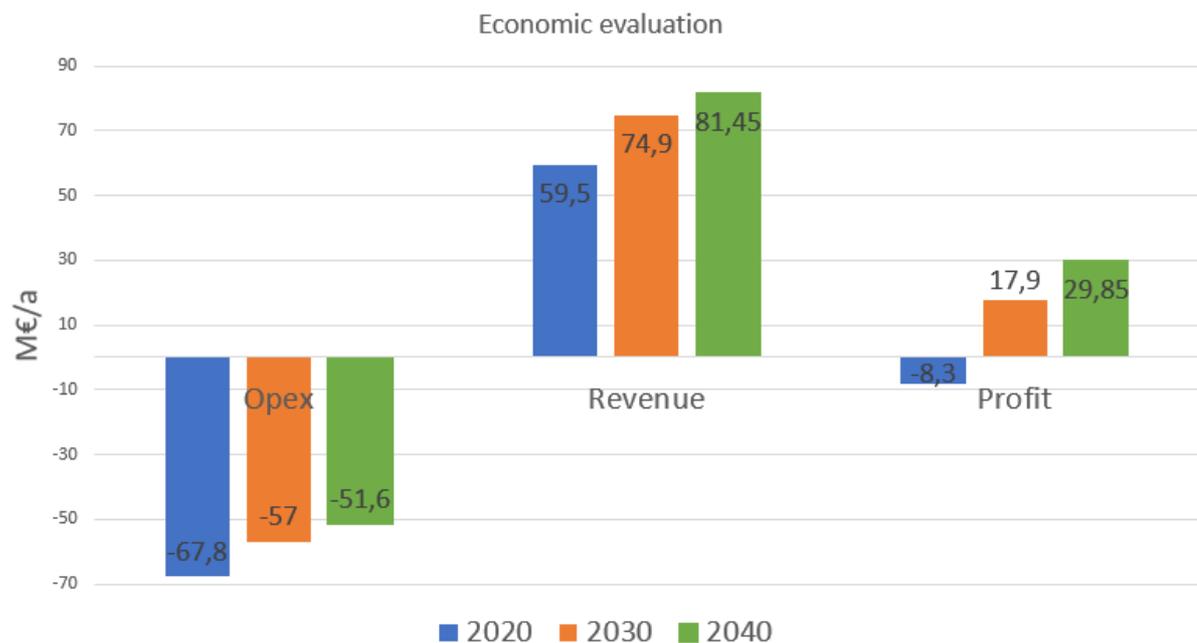


Figure 33: Economic analysis of methanol production

The profitability of the methanol synthesis plant could be assumed to increase if methanol prices continue to rise and grid electricity prices decrease, thus it becomes feasible to produce more methanol from the captured CO₂ even with increased demand of electrolysis.

The optimal way to increase the profitability of the installations is for the mill to produce excess electricity for electrolysis, as in the current scenarios all electricity is supplied from the grid. Figure 34 shows the correlation of electrolysis cost versus the amount of electricity supplied by the mill.

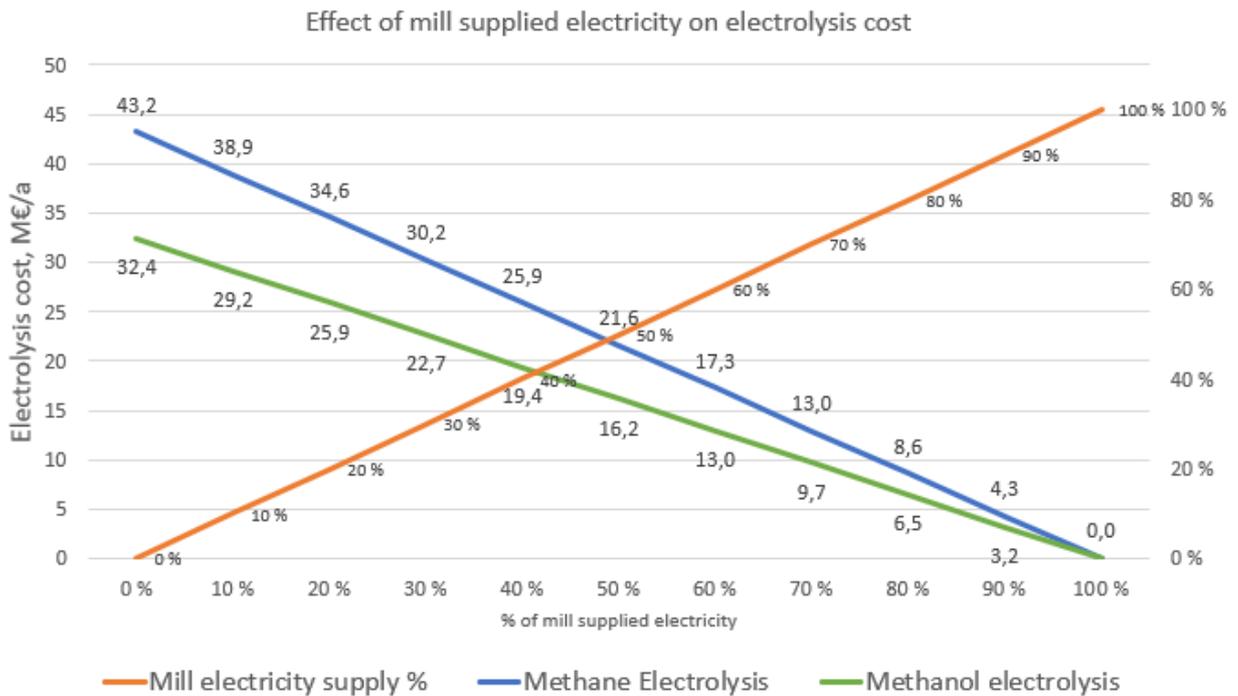


Figure 34: Effect of % of electricity supplied by the mill on electrolysis cost in 2020

The challenge with an integrated pulp and paper mill compared to a stand-alone pulp mill is that a large part of the energy produced goes into the paper making process which dramatically lowers the availability of energy for utilisation. However, assuming new paper making processes are implemented as mentioned in chapter 4.3 the possibilities of excess energy use might become more favourable. The cheaper the grid electricity price, the smaller the marginal saving will be from using own electricity produced at the mill.

The studied solutions show that it is possible to reach carbon neutrality and negativity within an integrated pulp and paper mill. However, investment costs are high and modifications to existing processes and equipment are likely. The progression from the current situation of 2.23 MtCO₂/a emitted, to 2040 where the mill would be carbon negative with zero fossil fuel use and capturing 1 MtCO₂/a is described next. In 2025 the fossil fuel use would be halved by efficiency

improvements and fossil fuel replacement in the lime kiln. In 2030 the remaining fossil fuel use is replaced by biofuels in the auxiliary boilers and construction of methane or methanol production plant begins. In 2035 the methane or methanol plant construction is completed together with carbon capture and the captured amount is equal to the energy demand of fossil fuels before. In another scenario implementing only carbon capture with transport and storage the carbon capture plant is expected to take 5 years to operate at full capacity so in 2040 the net captured amount is 1 MtCO₂/a. If the methane or methanol plant is built it is assumed that by 2040 190 000t of CO₂ is consumed in the methane or methanol production process, thus leaving 810 000t of CO₂ to be sold to the market outside the mills gates which makes the mill still carbon negative. The concepts with their emission reduction estimations are visualised in figure 35.

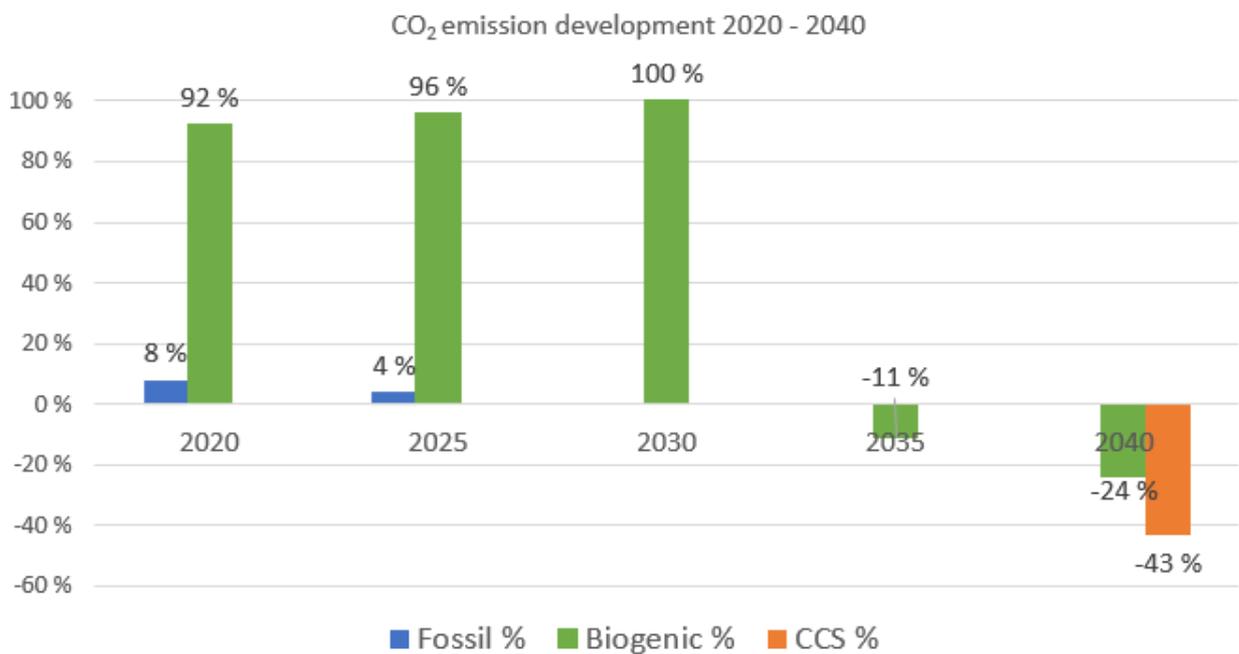


Figure 35: Emission development from 2020 – 2040

It is clear that CCS offers a higher removal of CO₂ but also at a higher cost and much smaller potential for revenue compared to methanol production for example.

8 CONCLUSIONS

The pulp and paper industry is in a unique position to combat climate change due to the renewability of their most important raw material, wood. The opportunity to contribute to a fossil free future should not be overlooked. The strategies and goals should support business but also national and international environmental goals. Policies and megatrends are driving the change and business as usual is no longer an option. It is clear that GHG emission reductions are a necessity for future business, the road is long and will require substantial investments.

This thesis looked at the possibilities to reduce CO₂ emissions in an integrated P&P mill. The options to reach carbon neutrality are easier and cheaper to implement than the options for carbon negativity because of the established technology and knowledge. Like switching the fossil fuel use in the lime kiln, however, this does not solve the fossil fuel use in the recovery boilers and power boilers. Therefore, in order to reach carbon negativity, carbon capture and methane/methanol production was studied.

The results show that carbon capture and storage will never offer possibilities for profit with current assumptions. This is due to the high costs of transport and storage. In a future where the compensation for storage is greatly increased it might allow for economic feasibility. The current benefit of it is that it provides a company only the opportunity to reach their reduction targets. For methane and methanol production the outlook is better with the current assumptions. The profitability depends largely on the price of electricity and available supply from the mill. As noted, a stand-alone pulp mill generates much more excess electricity than an integrated mill. Therefore, both carbon capture and methane or methanol production is more feasible in such a mill. However, it was shown that even an integrated mill can reach economic feasibility in the future with the assumptions made for electricity and CO₂ price. The calculations here are a rough estimate for evaluation and it is impossible to predict what the future will look like in terms of costs and product availability.

In any case if one wants to reach carbon negativity within an integrated pulp and paper mill, the investments needed are substantial and over the course of many years.

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APPENDIX

TRL levels in EU

| | |
|-------|-----------------------------------------------------------------------------------------------------------------------------------|
| TRL 1 | Basic principles observed |
| TRL 2 | Technology concept formulated |
| TRL 3 | Experimental proof of concept |
| TRL 4 | Technology validated in lab |
| TRL 5 | Technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies) |
| TRL 6 | Technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies) |
| TRL 7 | System prototype demonstration in operational environment |
| TRL 8 | System complete and qualified |
| TRL 9 | Actual system proven in operational environment (competitive manufacturing in the case of key enabling technologies; or in space) |