CHALMERS
UNIVERSITY OF TECHNOLOGY

Process Integration of Electric Plasma Calcination in Pulp and Paper Plants

Techno-economic and greenhouse gas emission assessment including CO_2 utilisation options

Master's thesis in Sustainable Energy Systems

Elin Andersson
Arvid Skogström

Department of Space, Earth and Environment
Chalmers University of Technology
Gothenburg, Sweden 2020

Master's thesis 2020

Process Integration of Electric Plasma Calcination in Pulp and Paper Plants

Techno-economic and greenhouse gas emission assessment including CO_2 utilisation options

ELIN ANDERSSON
ARVID SKOGSTRÖM

Department of Space, Earth and Environment
Division of Energy Technology
Chalmers University of Technology
Gothenburg, Sweden 2020

Process Integration of Electric Plasma Calcination in Pulp and Paper Plants
Techno-economic and greenhouse gas emission assessment including CO_2 utilisation options

ELIN ANDERSSON
ARVID SKOGSTRÖM

© ELIN ANDERSSON, ARVID SKOGSTRÖM, 2020.

Supervisors: Holger Wiertzema, Department of Space, Earth and Environment
Elin Svensson, CIT Industriell Energi AB
Examiner: Simon Harvey, Department of Space, Earth and Environment

Master's Thesis 2020
Department of Space, Earth and Environment
Division of Energy Technology
Chalmers University of Technology
SE-412 96 Gothenburg
Telephone +46 31 772 1000

Typeset in LaTeX
Gothenburg, Sweden 2020

Process Integration of Electric Plasma Calcination in Pulp and Paper Plants

ELIN ANDERSSON, ARVID SKOGSTRÖM
Department of Space, Earth and Environment, Chalmers University of Technology

Abstract

The calcination process within the Kraft pulping process is essential since it enables the recovery and reuse of pulp cooking chemicals. Currently, fossil fuels or biomass are used as fuel, making the calcination process one of the main emitters of carbon dioxide. An alternative calcination process using electric gas-plasma technology and steam slaking has been proposed and could provide several benefits compared to the conventional calcination process such as a fuel switch to electricity, as well as the production of a pure stream of carbon dioxide with renewable origin that enables cost-effective further utilisation. However, the integration of the new calcination concept into existing pulp and paper plants has not been investigated in detail.

This project aims to investigate the impact of integrating electric plasma calcination with steam slaking into existing pulp and paper plants by using pinch analysis. Two existing mills, the stand-alone pulp mill Södra Cell Värö and the integrated pulp and paper mill Holmen Iggesund were used as models in case studies to illustrate the proposed assessment methodology. Possible energy savings, emission reductions and increased on-site electricity production were investigated along with an economic assessment comparing the total annualised cost of the conventional calcination concept with the new calcination technology. Furthermore, an inventory of possible CO_2 utilisation options was created while two of the CO_2 utilisation options were investigated more in detail; the production of electro-fuels and the extraction of lignin via the LignoBoost process.

The results show that the implementation of the electric plasma calcination concept would require an input of 23.2 MW of electricity for the Iggesund mill and 37.9 MW for Värö. However, it was found that by heat integration of the new calcination process, it is theoretically possible to achieve hot utility savings of about 10% of the total utility demand together with a possible increase of the on-site electricity production. A fuel shift from biomass to electricity results in a net increase of greenhouse gas emissions with the energy market conditions of today. However, if the biomass saved by the fuel shift is used to replace fuels of fossil origin, there is great potential to achieve significant emission reductions. The investment cost of the new calcination technology was found to be in the same order of magnitude as the investment cost of the conventional lime kiln.

With regard to the CO_2 utilisation investigations, it was found that only a share of the available CO_2 can be utilised due to limitations of available grid capacity for electro-fuel production while the recovery boiler operation is the limiting factor for the LignoBoost concept. The greenhouse gas emissions from the electro-fuels are highly depending on the emissions associated with the electricity production, but provided sustainable electricity production, there is high potential to reduce the greenhouse gas emissions with both CO_2 utilisation options if they were to replace fuels of fossil source. The economic potential for both options was investigated with promising results with production costs in the same order of magnitude as the current market price for similar fuels, however, the uncertainties in the cost estimations and the lignin market should be emphasised.

Keywords: Kraft pulp process, electric plasma calcination, pinch analysis, CO_2 utilisation, techno-economic assessment, greenhouse gas assessment, electro-fuels, LignoBoost

Acknowledgement

We would like to express our great appreciation to our supervisors, Holger Wiertzema and Elin Svensson, for their commitment and guidance throughout this work. Your willingness to share your knowledge and experience with us was not only a key factor for us to complete this work but have also given us valuable experience for future challenges.

A special thanks goes to our examiner Simon Harvey for contributing with his expertise in the field and valuable feedback and insights.

We would also like to thank the department of energy technology for a warm welcoming and great colleagues who have brighten our days during this project.

Lastly, we want to thank our family and friends for their support, not only during this project, but during our whole period at Chalmers.

Thank you

Elin Andersson & Arvid Skogström, Gothenburg, June 2020

Contents

List of Figures viii

List of Tables x

Abbreviation xi

1 Introduction 1
1.1 Aim . . . 2
1.2 Limitations . . . 3

2 Background 4
2.1 Pulp and Paper Industry . . . 4
2.1.1 The Kraft Pulp Process . . . 5
2.1.2 Existing Technology for Calcination and Cooking Chemical Recovery 5
2.2 Electric Plasma Calcination and Steam Slaking . . . 7
2.2.1 Electric Plasma Technology . . . 8
2.2.2 Steam Slaking . . . 10
2.3 Carbon Capture and Utilisation Options . . . 11
2.3.1 Electro-fuels . . . 11
2.3.1.1 Electrolysis of Water . . . 11
2.3.1.2 Methane Synthesis . . . 12
2.3.1.3 Methanol Synthesis . . . 12
2.3.2 LignoBoost Process . . . 13
2.3.3 Carbon Capture and Storage Processes . . . 14
2.3.4 Carbon Dioxide Utilisation on Site . . . 15

3 Methodology 16
3.1 Steps of the Methodology . . . 16
3.2 Pinch Analysis and Modelling . . . 17

4 Input Data and Assumptions 19
4.1 Investigated Mills . . . 19
4.2 Electric Plasma Calcination . . . 21
4.2.1 Electric Plasma Technology . . . 21
4.2.1.1 GCC Construction . . . 21
4.2.1.2 Scaling of Electric Plasma GCC . . . 22
4.2.1.3 Electricity Demand . . . 22
4.2.2 Steam Cycle Integration . . . 23
4.2.3 Greenhouse Gas Emissions . . . 24
4.2.4 Economic Evaluation . . . 25
4.3 Carbon Capture and Utilisation Processes . . . 26
4.3.1 Electro-fuels . . . 26
4.3.1.1 Production Capacity . . . 26
4.3.1.2 Economic Evaluation . . . 28
4.3.2 LignoBoost . . . 29

5 Results 31

5.1 Integration of Electric Plasma Calcination . . . 31
5.1.1 Electric Plasma Calcination with Steam Slaking . . . 31
5.1.2 Heat Integration of Electric Plasma Calcination . . . 32
5.1.3 Steam Cycle Integration . . . 34
5.1.4 Emission Comparison and Electricity Demand . . . 36
5.1.5 Economic Evaluation . . . 38
5.1.6 Summary . . . 41
5.2 Carbon Dioxide Utilisation . . . 42
5.2.1 Production of Electro-fuels . . . 42
5.2.1.1 Electro-fuel Synthesis . . . 42
5.2.1.2 Greenhouse Gas Emission Comparison . . . 44
5.2.1.3 Heat Integration of Electro-fuels . . . 45
5.2.1.4 Economic Evaluation . . . 47
5.2.2 LignoBoost . . . 49

6 Discussion **51**
6.1 Electric Plasma Calcination . . . 51
6.2 Carbon Dioxide Utilisation . . . 53
6.2.1 Electro-fuels . . . 53
6.2.2 LignoBoost . . . 55
6.2.3 Comparison . . . 55

7 Conclusions and Future Work **57**

References **59**

Appendix A Electricity Demand of Electric Plasma Concept **I**

Appendix B Stream Data **II**
B.1 Värö . . . II
B.2 Iggesund . . . IV

Appendix C Heat of Reaction **VI**

Appendix D Physical Properties **VIII**

Appendix E Economic Data **IX**

Appendix F LignoBoost **X**

Appendix G Steam Cycle Integration **XII**

Appendix H Heat Integration of Electro-methanol **XIII**

List of Figures

1 Process overview of a chemical pulp plant. 4
2 Visualisation of the cooking chemical recovery process. 5
3 Visualisation of the existing calcination and causticising processes. 6
4 Illustration of a lime kiln with temperatures for the lime in each zone. . . . 7
5 Simplified process schematic of causticising plant with electric plasma calcination and steam slaking. 8
6 Detailed process schematic of electric plasma calcination with steam slaking. Black lines represents mass flows and red lines represents heat flows. . 9
7 Simplified process schematic of electro-fuel process. 11
8 Simplified process schematic of LignoBoost process. 13
9 Flow sheet of work process. 16
10 Grand composite curve for Iggesund pulp and paper mill. 20
11 Grand composite curve for Värö pulp mill. 21
12 Grand composite curve of electric plasma calcination process based on a flow rate of one kilogram wet lime mud per second. 31
13 Visualisation of the identified process streams connected to the conventional calcination process at Iggesund by a split-GCC. Calcination process in red and background pulp and paper process in blue. 32
14 Visualisation of the identified process streams connected to the conventional calcination process at Värö by a split-GCC. Calcination process in red and background pulp process in blue. 33
15 Visualisation of heat integration possibilities of electric plasma calcination at Iggesund mill by a split-GCC. Electric plasma calcination process as foreground (red line) and remaining pulp and paper process as background (blue line). 33
16 Visualisation of heat integration possibilities of electric plasma calcination at Värö mill by a split-GCC. Electric plasma calcination process as foreground (red line) and remaining pulp process as background (blue line). . . 34
17 Integrated steam cycles (blue dashed lines) for Iggesund (left) and Värö (right) for the original mills (red lines). 35
18 Integrated steam cycles (blue dashed lines) for Iggesund (left) and Värö (right) when the electric plasma calcination and steam slaking process is completely integrated with the rest of the mill (red lines). 35
19 Integrated steam cycles (blue dashed lines) for Iggesund (left) and Värö (right) when the electric plasma calcination and steam slaking process is integrated with the rest of the mill via the steam system. The red line represents the pulp mill process with the new electric plasma calcination process represented by its potential steam generation. 36
20 Sensitivity analysis of the total annualised cost with respect to electricity price for Iggesund (left) and Värö (right). 40
21 Sensitivity analysis of the total annualised cost with respect to the interest rate for Iggesund (left) and Värö (right). 40
22 Visualisation of heat integration possibilities of electro-methane synthesis at Iggesund mill by a split-GCC. 45
23 Visualisation of heat integration possibilities of electro-methane synthesis at Värö mill by a split-GCC. 45

24 Visualisation of heat integration possibilities of electro-methane synthesis with an electrolyser temperature of 50 °C at Värö mill by a split-GCC. . . 47

List of Tables

1 Plant specifications for the two investigated mills. 20
2 Assumptions related to the GCC construction of electric plasma calcination with steam slaking. 22
3 Scaling factors for investigated mills. 22
4 Specific power demand of the electric plasma calcination concept. 23
5 Specific emissions of power sources for the reburning unit. 25
6 Economic parameters for the calcination process. 26
7 Electro-fuel synthesis process specifications. 27
8 Fundamental assumptions for economic calculations. 28
9 Equipment cost assumptions for electrolysis of water, year 2019. 28
10 Equipment cost assumptions for electro-fuel synthesis reactor, year 2019. . 29
11 Assumptions for lignin production calculations. 29
12 Stream data for electric plasma calcination based on a flow rate of one kilogram wet lime mud per second. 32
13 Theoretical utility savings by integration of electric plasma calcination. . . 34
14 Emissions from the different fuels that a lime kiln can use. Iggesund currently uses tall oil pitch and Värö uses wood in the lime kiln. 37
15 Electricity demand of electric plasma calcination concept for the two investigated mills. 37
16 Emissions from electricity used in the electric plasma calcination with two different system boundaries. 37
17 Costs for investment, installation and operation of the electric plasma calcination process. 39
18 Costs for investment, installation and operation of a lime kiln. 39
19 Summary of results in terms of utilities, emissions and economics of integration of electric plasma calcination with steam slaking. 41
20 Theoretical maximum production of electro-fuels at the two investigated mills. 43
21 Available electric power capacity for electro-fuel production. 43
22 Maximum capacity for electro-fuel production at the two investigated mills. 44
23 Emission comparison of combustion of electro-fuels and equivalent fossil fuels. 44
24 Stream data for electro-methane synthesis. 46
25 Theoretical utility savings by heat integration of electro-methane synthesis. 46
26 Theoretical utility savings by heat integration of electro-methane synthesis with an electrolyser temperature of 50 °C at Värö mill. 47
27 Production cost calculations for electro-methane and electro-methanol production. 48
28 Theoretical maximum lignin production and corresponding load reduction of recovery boiler at maximum lignin extraction rate. 49
29 Maximum allowed load reduction of recovery boiler to ensure hot utility supply. 49
30 Summary of theoretical maximum lignin production and corresponding economic assessment, applies for year 2019. 50

Abbreviations

ADt - Air-dried tonne, pulp with 90 % dry matter

CAPEX - Capital expenses

CEPCI - Chemical Engineering Plant Cost Index

CCS - Carbon capture and storage

CCU - Carbon capture and utilisation

GCC - Grand composite curve

LHV - Lower heating value

OPEX - Operating expense

TOP - Tall oil pitch

1 Introduction

With increasing greenhouse gas emissions and anthropogenic global warming, the mitigation of climate change has become a top priority on the political agenda. The European Union has released directives for the member states to reduce their emissions of greenhouse gases and to improve their energy use by increased energy efficiency and sustainable electricity production [1]. The Swedish Government set the target for Sweden to have a net zero greenhouse gas emissions to the atmosphere by 2045 and negative emissions thereafter [2].

In Sweden, the energy use of the industrial sector was 140 TWh in 2015 which corresponds to 38% of the country's final energy use [1]. The dominating energy carriers within the industrial sector are biomass and electricity. The pulp and paper industry alone stands for 52% of the Swedish industrial final energy use. The average consumption of a modern Swedish pulp mill is about 25 GJ/ADt (air-dried tonne) of thermal energy and is mainly provided by biomass residues [3].

A fuel shift within the industry has the potential to both increase energy efficiency and reduce net greenhouse gas emissions [4]. By replacing fossil fuels with renewable electricity, positive climate effects will be achieved. When replacing bio-based fuels with electricity, large quantities of biomass can be saved. The saved biomass could be converted into bio-based fuels via, for example, gasification or decomposition, and used to replace fuels of fossil origin in other sectors, hence achieving a net reduction of greenhouse gas emissions.

The calcination process within the pulp and paper industry is essential since it enables the recovery and reuse of the pulp cooking chemicals. The calcination process requires 1.1 - 2.9 GJ of energy input per ADt of pulp produced [5] and is one of the main external fuel consumers in the pulp process powered by either biogenic or fossil fuel.

Alternative calcination processes using electric gas-plasma technology in the pulp and paper industry have been proposed in the literature and a process design for the electric plasma calcination has been developed [6]. The new concept was compared to the conventional calcination process and it is recommended to replace conventional lime kilns at the end of their lifetime with the new electric plasma technology. Several benefits such as faster reaction times, smaller equipment size and a resulting pure stream of carbon dioxide which could be further utilised were pointed out. The compatibility with steam slaking of lime was studied with promising results as the reaction heat of the steam slaker could be used to power the dryer needed for the electric plasma calcination [7]. The electric plasma technology has been investigated for similar applications such as the calcination of limestone in the cement industry as a measure for emission reduction [8]. Plasma arc furnaces opens for new possibilities of electrifying high-temperature and high-volume processes that are otherwise difficult to electrify [4]. However, the integration of electric plasma calcination with steam slaking to the rest of the pulp process has not been further investigated. Consequently, there is a lack of knowledge about how the integration of this concept affects the mass and energy balances of such plants, as well as the economic feasibility when integrating the concept.

The concentrated stream of carbon dioxide resulting from implementation of the electric plasma calcination could be used for further utilisation in a cost-effective way since no further concentration of the gas is needed. Previous studies suggest the integration of electro-fuel production to existing pulp/pulp and paper mills [9]. However, the production of electro-fuels is only one option for utilisation of the resulting stream of carbon dioxide. There are other possible options to use the carbon dioxide stream including extraction of lignin via the LignoBoost process, carbon capture and storage, *CCS*, or further usage on-site in pulp bleaching processes.

1.1 Aim

The aim of the project is stated as follows:

> *Get a better understanding of how an electric plasma calcination process with steam slaking can be integrated in different types of mills, and how that affects the on-site heat and power balance. Furthermore, investigate options for utilisation of the carbon dioxide resulting from the integration for possible further emission reductions.*

To investigate this, the project has four main objectives:

- Investigate the impact of integrating electric plasma calcination concept with steam slaking into existing pulp (and paper) mills. The mass and energy balances are studied and pinch analysis is used to compare the conventional process based on a lime kiln with the new process in terms of heat integration potential and demand for external utility. Steam cycle integration is used to investigate the impact of the on-site power production. This information, together with the electric power demand of the new process form the base for techno-economic and CO_2 emission assessment.

- Investigate the role of the surrounding energy system by using different energy market conditions. Assess the sensitivity of the results to changes in important energy market parameters, such as the electricity price and carbon intensity of grid electricity production.

- Inventory of different CO_2 utilisation options for the pure stream of carbon dioxide resulting from the implementation of the new calcination technology by a literature study.

- Detailed analysis of two CO_2 utilisation options; the production of electro-fuels, and lignin extraction via the LignoBoost concept.

The approach taken in this project is in line with PROCEL, a research project that aims to increase the knowledge base about opportunities for electrification of industrial processes [10].

1.2 Limitations

This project did not focus on optimisation or fine tuning of the concept by process modifications.

This project did not investigate all options of carbon dioxide handling on a deep technological level. The options considered to have most potential were further investigated, other possible options were discussed more briefly.

Process data from two specific mills were used for this study. Thus, this study was based on those specific plants. The results may differ between different plants due to different operating conditions and capacities of the plants.

This project did only compare the conventional process with a fired lime kiln and wet lime slaking with a complete replacement of the lime kiln for electric plasma technology with steam slaking.

As the heat capacities are temperature dependent, approximations are needed to use software for pinch analysis. Thus, linearisation of heat capacities or constant heat capacities was used for process streams.

2 Background

In this chapter, the fundamental theory of the pulp and paper industry is explained including the general background of the kraft pulping process along with the conventional calcination process used today. Furthermore, the new calcination concept using electric gas plasma technology along with steam slaking is introduced. In addition, possible carbon capture and utilisation options for the resulting pure stream of carbon dioxide are presented.

2.1 Pulp and Paper Industry

The pulp and paper industry is a large industry in Sweden. There are many different paper-based products that are produced from wood raw material, and several different processes to produce the paper. Different manufacturing methods are used to create products with different properties.

Pulp and paper are not necessarily produced on the same site but can be produced in stand-alone pulp or paper mills. However, it is normally more efficient to produce the pulp and the paper on the same site in an integrated pulp and paper mill [3]. Surplus heat from the pulp mill can thus be used in the paper drying process, which reduce the need for external utility. There is also a great advantage from an energy perspective in not needing to dry the pulp before transportation to the paper mill.

There are two commonly used pulping processes, namely chemical and mechanical pulping [3]. In the chemical pulping process, chemicals are used to release the cellulose fibres from the wood by dissolving the lignin. In mechanical pulping, the fibres are released from the wood by mechanical force by grinding of the wood or wood chips. The chemical process requires an efficient recovery system for reuse of the cooking chemicals. An overview of the chemical pulp process is found in Figure 1. The chemical pulp process will be in focus within this work and is further described in this section.

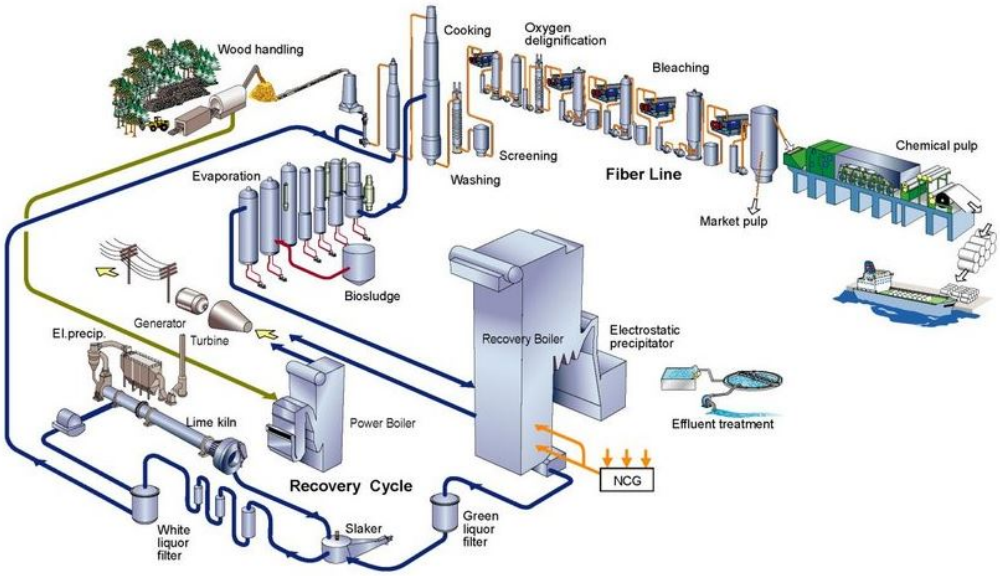

Figure 1: Process overview of a chemical pulp plant [11].

2.1.1 The Kraft Pulp Process

The dominating chemical pulp process, and the process investigated in this study, is the *kraft process* [12]. In the kraft process, a water-based solution with sodium hydroxide and sodium sulphide, *white liquor*, is used to dissolve and remove the lignin from the cellulose fibres in the wood [3]. After the cooking process, the cellulose fibres are removed from the lignin-containing solution, called *black liquor*, for further refining while the cooking chemicals in the black liquor are to be recovered in the recovery boiler and the causticising plant. In the causticising plant, calcium oxide (CaO) is needed which in turn is recovered in the calcination process by the use of heat. Figure 2 shows how these processes are connected.

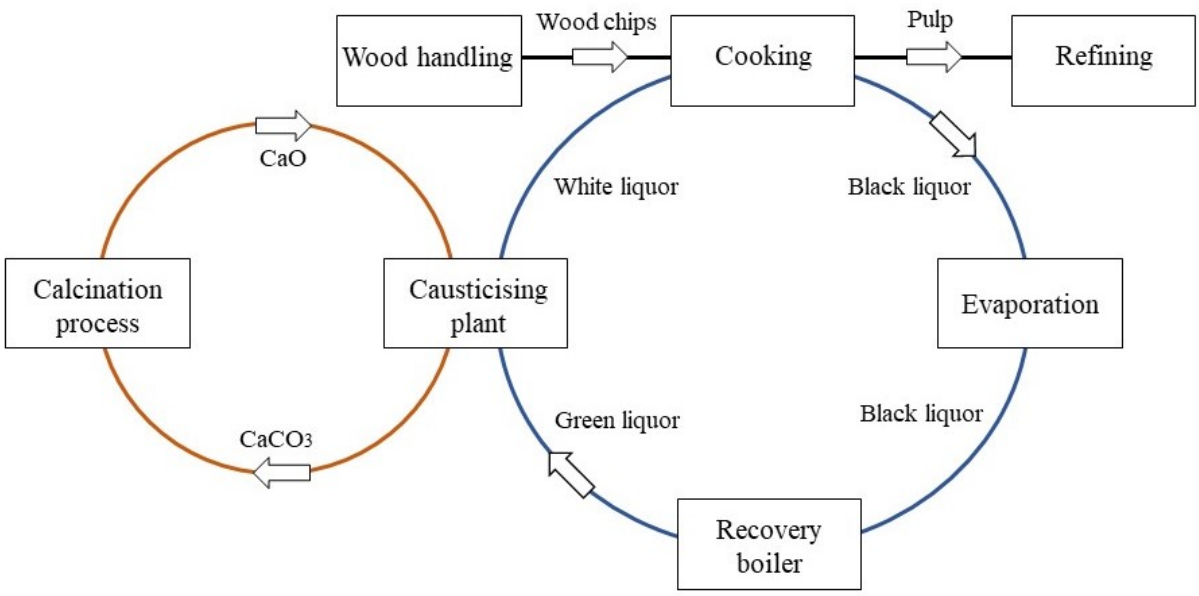

Figure 2: Visualisation of the cooking chemical recovery process.

The black liquor mainly consists of water, various organic compounds from the wood, and inorganic compounds originating from the cooking chemicals [3]. The black liquor passes several evaporators to reduce the water content before being burnt in the recovery boiler. The organic components are combusted to produce high pressure steam from water which is fed to a turbine to produce electricity and steam at lower pressures that is used to drive the processes in the plant. The inorganic components containing the cooking chemicals are found as a smelt in the bottom of the recovery boiler. This smelt is dissolved in weak white liquor to form so called *green liquor*. The weak white liquor is diluted white liquor that has been used for washing of the precipitated calcium carbonate ($CaCO_3$), *lime mud*. The green liquor is then transported to the causticising plant.

2.1.2 Existing Technology for Calcination and Cooking Chemical Recovery

The recovery of the cooking chemicals from the green liquor takes place in the causticising plant while calcium oxide is recovered in the calcination process. These processes are illustrated in further detail in Figure 3.

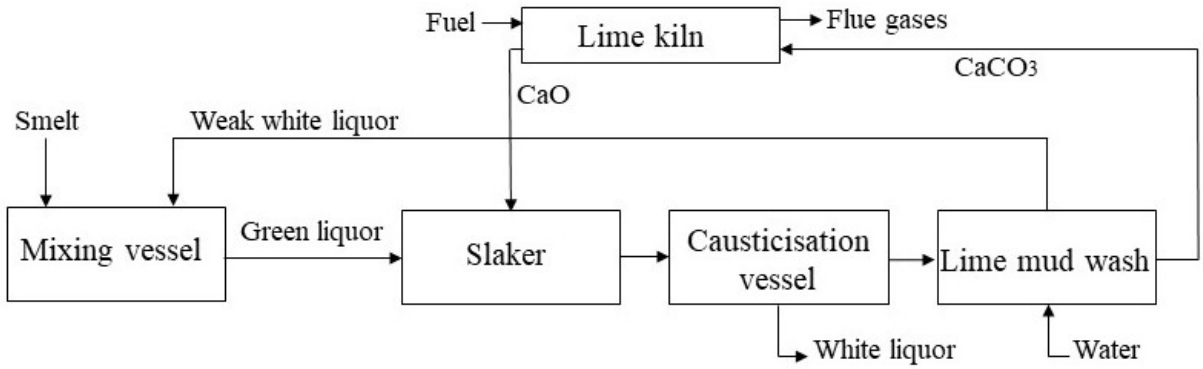

Figure 3: Visualisation of the existing calcination and causticising processes.

The green liquor is sometimes pre-cooled by heat exchanging it with water before entering the slaker unit to avoid boiling due to the exothermic hydration reaction. Calcium oxide, *burned lime*, is then added to the green liquor and the burned lime is slaked to calcium hydroxide, *slaked lime*, according to Reaction 1.

$$CaO(s) + H_2O(l) \rightarrow Ca(OH)_2(s), \quad \Delta H_r^{\circ} = -64.5 \ kJ/mol[13] \tag{1}$$

The slaked lime reacts with the carbonate ions from the green liquor forming calcium carbonate and sodium hydroxide ions, according to Reaction 2.

$$Na_2CO_3(aq) + Ca(OH)_2(s) \rightarrow CaCO_3(s) + NaOH(aq), \quad \Delta H_r^{\circ} = -5.3 \ kJ/mol[13] \tag{2}$$

The calcium carbonate precipitates, and the remaining solution is regenerated white liquor that can be reused in the cooking process. The reaction heat is recovered as hot white liquor, thus, reducing the energy demand in the cooking process. The precipitated calcium carbonate is washed from remaining white liquor and this washing solution, the weak white liquor, is recycled back for mixing with the smelt from the recovery boiler. The washed calcium carbonate is regenerated to calcium oxide by adding heat in the lime kiln according to Reaction 3, where 0.44 kilograms of CO_2 is released for every kilogram of $CaCO_3$. These emissions are biogenic as the carbon originates from the wood raw material.

$$CaCO_3(s) \rightarrow CaO(s) + CO_2(g), \quad \Delta H_r^{\circ} = +179.2 \ kJ/mol[13] \tag{3}$$

The rotary lime kiln is heated by combustion of oil, gas or biomass in air [6]. For the reaction to occur in a good way, temperatures of 1200°C is needed in the lime kiln. The hot flue gases are used to dry the calcium carbonate before reburning, which reduces the energy use of the lime kiln. A simplified illustration of a lime kiln can be seen in Figure 4. The flue gases are in some pulp mills further cooled after they leave the lime kiln in a heat recovery system where the flue gases heats low pressure steam or hot water [3]. The energy use for a lime kiln is typically between 5400 and 6800 kJ/kg CaO [14].

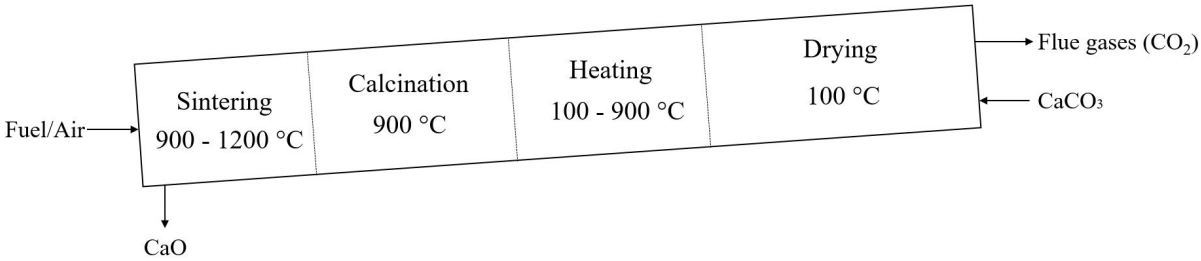

Figure 4: Illustration of a lime kiln with temperatures for the lime in each zone, data taken from [3].

2.2 Electric Plasma Calcination and Steam Slaking

An alternative calcination technology for reburning of lime has been proposed for use in the pulp and paper industry [6]. The idea is to replace the conventional, combustion driven lime kiln with an electric gas-plasma calcination reactor where carbon dioxide is ionised by an electrically generated arc. This is an innovative technology in the pulp and paper industry which has been tested in different scales during the last 15 years [7]. As the carbon dioxide is reformed, heat is released and drives the calcination process. The process occurs at 950 °C. The calcination reaction time is expected to be significantly reduced compared to the conventional calcination process.

The electric plasma calcination has several benefits compared to the conventional lime kiln. The equipment size is smaller, and the expected investment cost is lower compared to a rotary kiln [6]. The technology releases high quality heat and could improve the heat recovery of the plant. Furthermore, the electric reburning of lime results in a pure carbon dioxide exhaust stream. Thus, the carbon dioxide can be extracted in an energy and economically efficient way since no further concentration is needed. Some of the carbon dioxide is recycled back to the electric gas-plasma generator. The remaining carbon dioxide could be sold, emitted or used internally. The carbon dioxide resulting from the electric plasma calcination originates from the wood raw material and is of renewable origin. Thus, the emissions of the carbon dioxide resulting from electric plasma calcination is considered biogenic with net zero greenhouse gas emissions.

The electric plasma calcination can advantageously be combined with steam slaking of the lime [9]. A simplified process schematic of how the electric plasma calcination with steam slaking can be integrated in the causticising plant is presented in Figure 5. The slaking of lime is an exothermic reaction. In the traditional process, the burned lime is mixed directly with the water-containing green liquor [6]. Thus, the heat is recovered at low temperature and cannot be used further on site. With steam slaking, steam or wet air is used in the lime slaking process together with the hot lime from the calcination generator [7]. This means that the reaction heat can be recovered at higher temperatures and further used within the process.

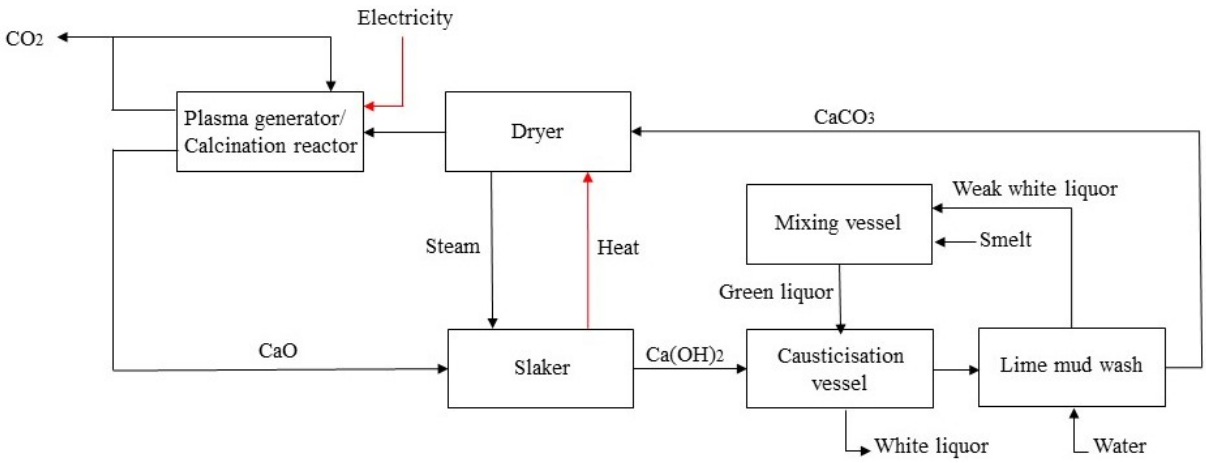

Figure 5: Simplified process schematic of causticising plant with electric plasma calcination and steam slaking.

The calcium carbonate needs to be dried before entering the electric gas-plasma calcination reactor to minimise the energy use of the reactor [6]. The water vapour from the calcium carbonate drier is used as slaking medium in the steam slaker. The reaction heat from the steam slaker is used to drive the calcium carbonate drier, excess heat is suggested to be used for production of process steam around 4 bar.

2.2.1 Electric Plasma Technology

Electric plasma technology has been used in the metallurgical industry for melting, remelting and refining of metals and alloys but it has not been applied within the pulp and paper industry [15]. The concept has potential to be part of a technological shift within the pulp and paper industry as a more efficient and climate neutral way of reburning limestone in the chemical recovery plant.

The electric plasma technology uses an electric arc furnace with a plasma generator and a calcination reactor where the reburning of lime takes place. The electric arc furnace has an electric current that flows between two electrodes, forming an electric arc. The carbon dioxide is heated to around 4000 °C when it gets in contact with the electric current, is partly ionised, and leaves the furnace as a gas-plasma mixture [6]. When leaving the arc, the ionised atoms starts to deionise and return to normal gas phase. The reforming of carbon dioxide releases large quantities of energy which is used to drive the calcium carbonate reaction in the calcination reactor.

A detailed process schematic of the electric plasma calcination with steam slaking is suggested by Bjoviet et al. and presented in Figure 6 [6].

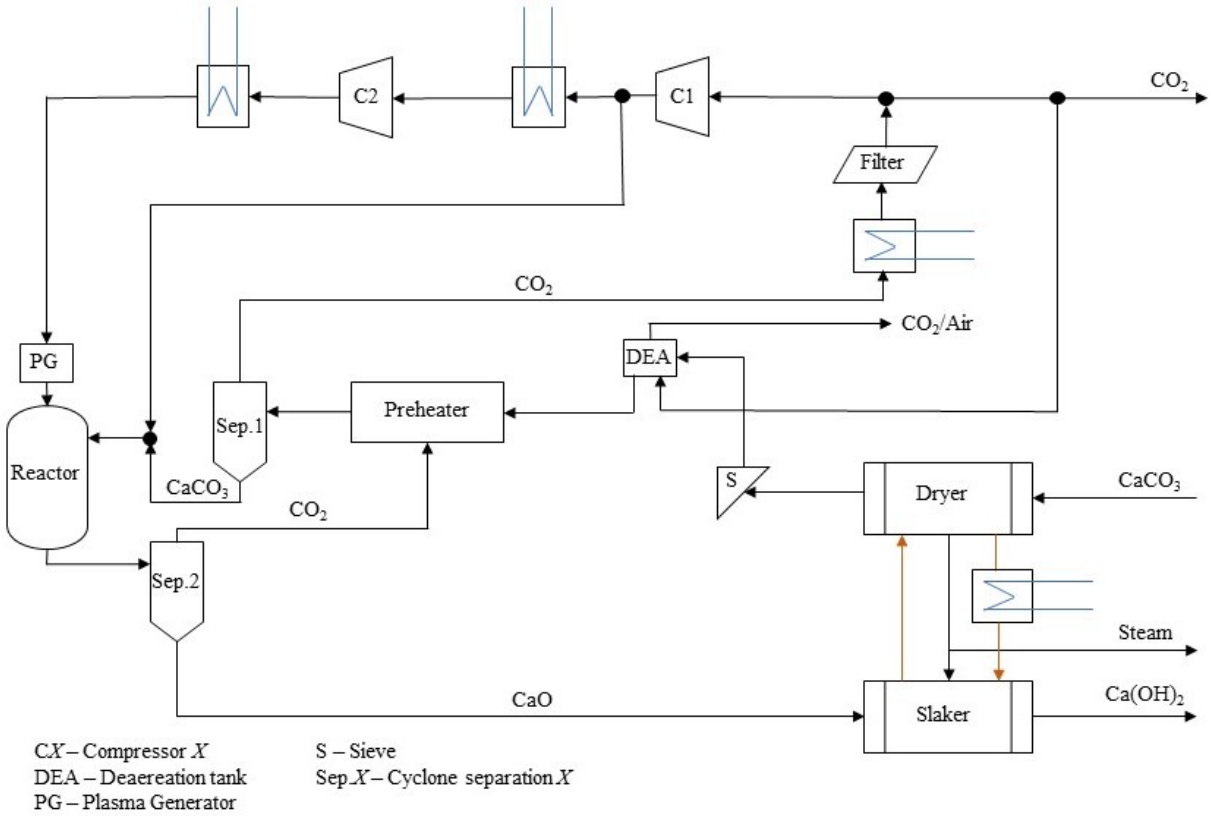

Figure 6: Detailed process schematic of electric plasma calcination with steam slaking [6]. Black lines represents mass flows and red lines represents heat flows.

The wet lime mud enters the dryer powered by the reaction heat from the steam slaker. The dry lime mud mainly consists of calcium carbonate and traces of inert material. The inert material originates from impurities from the wood raw material and mainly consist of compounds containing aluminium, silicone, sulfur and oxygen. The dried lime mud is sieved to remove large particles before entering a deaeration tank. Air is separated by a stream of carbon dioxide to avoid the formation of nitrogen oxides in the plasma generator. Nitrogen oxides are potent greenhouse gases and harmful to the ozone layer why an air-tight system is essential to reduce formation and emission of nitrogen oxides.

The lime mud is preheated with hot carbon dioxide from the calcination reactor. The carbon dioxide is separated from the lime mud in a cyclone and the lime mud enters the calcination reactor where the reburning of lime occurs. The burned lime enters another cyclone where the hot carbon dioxide is separated and sent to the preheating unit. The burned lime enters the slaker where steam produced in the lime mud dryer is used as slaking medium. The reaction heat is used to power the dryer. Excess heat could be used for low pressure steam production.

The hot carbon dioxide is cooled and filtered. Some of the carbon dioxide is compressed in two steps with inter-cooling for reuse in the plasma generator or as transport medium for the lime mud entering the calcination reactor. The remaining carbon dioxide is removed from the system as a pure carbon dioxide stream which could be emitted or further utilised.

The total power consumption of the electric plasma concept consists of the electric power demand of the plasma generator plus the power demand of the two compressors used to compress the carbon dioxide. The power consumption of the electric plasma concept is interpolated from reference [6] and found to be 5 300 kJ/kg CaO for the electric plasma generator and 140 kJ/kg CaO and 50 kJ/kg CaO for the two compressors respectively. The interpolation calculations can be found in Appendix A.

2.2.2 Steam Slaking

In the conventional slaking process, the burned lime is mixed directly with the green liquor and lime mud precipitates as the white liquor is recovered. The green liquor is sometimes pre-cooled to avoid boiling in the slaker due to the exothermic hydration reaction [7]. Thus, the heat is recovered as low temperature heat with little value.

Steam slaking is proposed as an alternative to the conventional slaking process. With steam slaking, the burned lime is slaked with steam before being mixed with the green liquor. The proposed slaking process has several advantages compared to the conventional slaking process from an energy perspective. By steam slaking, the reaction heat of the exothermal reaction can be recovered at higher temperatures, which enables production of high-quality steam. Furthermore, there is no need for cooling of the green liquor to prevent boiling in the causticising vessel as the lime will already be slaked when mixing with the green liquor. Excess heat from the slaker could instead be used for preheating of the green liquor to save energy in the cooking process [7].

Steam slaking works as a stand-alone process that is compatible with the conventional causticising apparatus but can advantageously be combined with the electric plasma calcination technology. By combining steam slaking with electric plasma calcination, the reaction heat from the slaker can be used to run the lime mud dryer and the evaporated water from the dryer can then be used as the slaking medium in the slaker [7].

The reaction for slaking of lime is an equilibrium reaction with an equilibrium temperature of 530 °C at 1 bar [16]. The reaction needs to be performed below the equilibrium temperature to achieve a good conversion of calcium hydroxide. However, it is desirable to have the reaction taking place at a as high temperature as possible, as valuable steam can be produced from the reaction heat. Experiments have found that the reaction rate decreases with decreased steam pressure and increased temperature. Thus, there is a trade-off between high reaction rates and possibility for high temperature heat recovery.

It is suggested to perform the hydration reaction at 250 °C, at most, to enable heat recovery of high temperatures while the reaction rate is still reasonable. Experimental work predicts reaction times of about 2 hours for these circumstances using reburned lime from the lime kiln [7].

2.3 Carbon Capture and Utilisation Options

The pure stream of carbon dioxide resulting from the electric plasma calcination can be captured for further utilisation. In this section, different utilisation possibilities for the carbon dioxide will be covered including production of electro-fuels, lignin extraction via the LignoBoost process, carbon capture and storage, *CCS* and further usage within the pulp process.

2.3.1 Electro-fuels

Electro-fuels are a way of storing electric energy in the form of chemical components. By hydrogenation of carbon monoxide or carbon dioxide from industrial flue gases, carbon-based electro-fuels such as methane or methanol can be produced. The electro-fuels can be used to replace fossil fuels within the industry or in other sectors (e.g. transport sectors) to reduce greenhouse gas emissions.

The carbon dioxide produced in the electric plasma calcination process could be used as raw material for electro-fuel production. A sustainable hydrogen source is essential for electro-fuel production to achieve the beneficial climate effects. In this work, hydrogen production by electrolysis of water was considered as the hydrogen source. Electrolysis of water is a very energy consuming process and thus, electricity may be the limiting factor for electro-fuel production. A simplified process schematic of electro-fuel production, with hydrogen produced by electrolysis of water, is visualised in Figure 7.

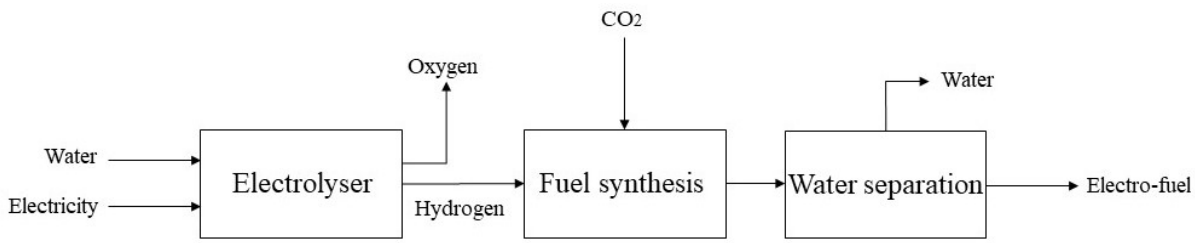

Figure 7: Simplified process schematic of electro-fuel process.

2.3.1.1 Electrolysis of Water

Electrolysis of water is an endothermic electro-chemical reaction that requires high input of electric energy. The water is split into hydrogen and oxygen according to Reaction 4. The hydrogen can either be stored and used directly as a fuel or further used for refined electro-fuels such as methane or methanol. The produced oxygen can be partially used on site or sold to the market. A typical mill uses 5 kg/ADt of oxygen in the production [9].

$$H_2O(l) \rightarrow H_2(g) + 1/2\, O_2(g), \qquad \Delta H_r^{\circ} = +286\ \ kJ/mol[9] \tag{4}$$

The electrolyser requires a total energy input of 286 kJ/mol, 237 kJ/mol of electricity and 49 kJ/mol of heat input [9]. There are different technologies available for electrolysis of water and the most established and technical mature technologies have an efficiency of

60-70 % and operate at a temperature of 60-90 °C. Less technically mature technologies could have both higher and slightly lower operating temperatures.

The equipment cost of an electrolyser is reported to be 250-600 euro/kW$_{el}$, the total investment cost is suggested 1 Meuro/MW$_{el}$ the year 2019 [9]. Electrolyser stack replacement is needed every 60 000-90 000 hour of operation at a cost of 50-60% of the equipment cost. Operation and maintenance are reported to 2-5 % of the annualised total investment cost.

2.3.1.2 Methane Synthesis

Methane is a gaseous fuel that is used both in private and industrial scale. Methane is the main component in natural gas which can be used for example as transportation fuel. It occurs as a fossil resource in the ground and underneath the seafloor but can also be produced from biogenic origin by decomposition of biomass.

Methane can be produced via the so-called Sabatier reaction, where carbon dioxide and hydrogen react to form methane according to Reaction 5.

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O, \qquad \Delta H_r^{\circ} = -165 \ kJ/mol[9] \tag{5}$$

The reaction takes place at 200-400 °C at pressures of 1-30 bar in presence of a nickel- or ruthenium-based catalyst [9]. The total energy efficiency of the process is 70-85%. The reaction is exothermic, thus heat is released at high temperatures which enables heat recovery of high quality steam.

The investment cost of catalytic methanisation plants has been reported to vary widely [9]. In general, small scale plants are more expensive than large scale plants per produced kW of methane. A 5 MW sized plant have been reported to cost 100-900 euro/kW methane with a median value of 600 euro/kW methane, a 50 MW plant 50-500 euro/kW methane with a median value of 300 euro/kW methane and a 200 MW plant 30-300 euro/kW methane with a median value of 200 euro/kW methane the year 2017. The operation and maintenance cost of the methane synthesis process is reported to be 4-10 % of the annualised investment cost.

2.3.1.3 Methanol Synthesis

Methanol is an alternative biofuel that can be used as a transportation fuel. Methanol is currently produced mainly from fossil sources but can also be produced from biogenic sources. The methanol production is similar to the Sabatier process and is summarised in Reaction 6.

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O, \qquad \Delta H_r^{\circ} = -49.5 \ kJ/mol[9] \tag{6}$$

The reaction occurs at a temperature of 200-300 °C at pressures of 30-50 bar in presence of a catalyst [9]. By recirculation of unreacted gases, the conversion of carbon dioxide

to methanol reaches up to 99%. The efficiency of the methanol synthesis is reported to 70-80 %. Water is co-formed with the methanol during the fuel synthesis which has to be separated from the methanol by distillation. The fuel synthesis process is exothermic and releases heat at high temperatures. The excess heat could thus be used in the distillation process for separation of water and light gases from the produced methanol.

The investment cost for catalytic methanol synthesis plants has been reported to vary widely and with the plant size [9]. The investment cost for a 5 MW methanol plant has been reported to be 600-1 200 euro/kW methanol with a median value of 1 000 euro/kW methanol and the investment cost for a 50 MW methanol plant has been reported to be 300-600 euro/kW methanol with a median value of 500 euro/kW methanol the year 2015. The operation and maintenance cost of the methanol synthesis process is reported to be 4-10 % of the annualised investment cost.

2.3.2 LignoBoost Process

LignoBoost is a process to extract lignin that is dissolved in the black liquor. Lignin is a macromolecule that composes around $33\%_{wt}$ of the dry wood raw material [17]. The extracted lignin has a lower heating value, LHV, of 24 MJ/kg and could be used as fuel [18]. The lignin could also be further refined for various products such as carbon fibres, fine chemicals such as vanillin or as an ingredient in bioplastics.

Lignin is the main energy carrier in the black liquor, thus, by extraction of lignin, the load of the recovery boiler will be reduced. A reduced load of the recovery boiler results in a decrease of steam production. However, if the recovery boiler capacity is constraining the load of the black liquor, extraction of lignin may enable that the pulp production can be increased if no other bottlenecks exist along the production line. LignoBoost has therefore been studied as a promising measure for mill capacity increase.

The LignoBoost process mainly consists of well-known unit operations including gas absorption, precipitation and solid liquid separation. A simplified process schematic is presented in Figure 8.

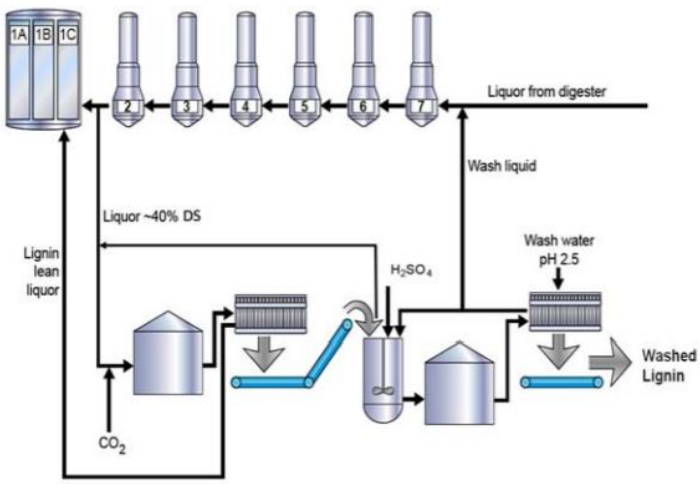

Figure 8: Simplified process schematic of LignoBoost process [19].

The solubility of lignin is highly dependent on the pH of the solution [20]. At high pHs, the lignin is soluble, but at a pH of 9 and below, the lignin will precipitate from the black liquor solution. By injection of carbon dioxide to the black liquor, carbonic acid is formed which lowers the pH and lignin precipitates [19]. The solid lignin is separated from the lignin-lean black liquor by filtration and the black liquor is sent back to the evaporation plant. The lignin filter cake is re-dispersed in washing water and sulphuric acid to further lower the pH. After a second filtration and washing step, the lignin is extracted and ready for any further refining.

The process is not particularly energy intensive but requires an input of 0.4 kWh of electric energy per kilogram lignin extracted [18]. No additional heat is required. However, the effluent streams from washing of the lignin filter cake is sent to the evaporation plant which increase the load of the evaporators and surface condensers. The increased steam demand of the evaporation plant is typically 0.9 GJ per tonne lignin extracted.

The maximum lignin extraction rate is around 135 kg per ADt pulp which corresponds to about 25% of the lignin present in the black liquor [19]. If more lignin is extracted, the heating value of the black liquor is decreased to the extent that the operation of the recovery boiler could be endangered.

The process requires an input of 0.5-0.6 kg of carbon dioxide and 0.3-0.4 kg of sulfuric acid per kilogram of lignin extracted [20]. Thus, parts of the pure carbon dioxide produced in the electric plasma calcination could be used for lignin extraction via the LignoBoost process.

Lignin from the kraft pulp process is currently a small-scale product with an uncertain market. Market price indications of lignin from kraft pulp processes range from 230-440 €/tonne [21].

The investment and operating cost of the LignoBoost process was estimated based on a commercial LignoBoost plant, producing 50 000 tonnes per year of dry lignin product [18]. The total investment cost, including equipment, construction and start-up costs, is estimated to 10 M€, the year 2008. The operating cost does mainly consist of the cost for carbon dioxide and other acids needed in the process. The total operating costs are estimated to range from 21-54 € per tonne of lignin extracted. The lower value represents a reasonable potential when spent acid from the pulp mill can be reused in the LignoBoost process to a large extent, and CO_2 is collected and concentrated from the flue gases in the mill.

2.3.3 Carbon Capture and Storage Processes

One way to handle the carbon dioxide is by carbon capture and storage, *CCS*, which is a process where carbon dioxide is captured and pumped deep into geological formations for storage [22].

Today, most of the costs for CCS are related to the collection and concentration of the carbon dioxide from flue gases [23]. With a pure stream of carbon dioxide resulting from the electric plasma calcination process, there is no need for further concentration of the CO_2 and the total cost for CCS could therefore be significantly reduced. However, the

infrastructure to handle and export the CO_2 to a storage site is generally not in place at a pulp/pulp and paper mill. Thus, there would be a need for the mill to invest in equipment for handling of the CO_2, including compressors, pipelines for transportation and possibly some intermediate storage capacity.

As CCS does not generate any products, the profitability must come from avoided fees by avoided greenhouse gas emissions, such as taxes, or revenues from green certificates or tariffs.

2.3.4 Carbon Dioxide Utilisation on Site

After the last bleaching steps in the pulping process, sulfur dioxide is added to lower the alkalinity and therefore prevent the pulp from losing its brightness that alkaline conditions can contribute to [24]. Sulfur dioxide must be used if chlorine dioxide is used as a bleaching agent. However, if other bleaching agents such as sodium hypochlorite or hydrogen peroxide are used, the sulfur dioxide aborts the bleaching process. Here, carbon dioxide could be used instead as it can lower the pH in the same way as sulfur dioxide, but it does not abort the bleaching process which leads to a brighter pulp.

Also in other processes where the alkalinity needs to be lowered many pulp mills use sulphuric acid. But some of that acid could also be replaced with carbonic acid which is formed from when carbon dioxide is dissolved in water [25].

Changing some of the sulfur-compounds to carbon dioxide will not significantly affect the process and can therefore be integrated without any major rebuildings or changes in energy consumption. CO_2 also have less contamination risks compared to sulfuric compounds as it is less toxic [24].

CO_2 can also be used when separating the cellulose fibres and the black liquor [26]. When adding CO_2 to the washing, the pH is lowered, and less water is needed in the washing as the fibres' swelling is reduced. The reduced pH also leads to less losses of sodium.

3 Methodology

The methodology of this work is described in further detail in this chapter, focusing on the work process as steps of methodology and pinch analysis, which is the main tool used within this work.

3.1 Steps of the Methodology

The work consisted of six main parts; *Data collection and GCC construction* for the new technology with electric plasma calcination and steam slaking, as well as for the conventional calcination process, *Integration studies of electric plasma calcination* with steam slaking, *Steam cycle integration*, *Evaluation of CO_2 emission reduction*, *Economic evaluation* and inventory of possible *CO_2 utilisation* options.

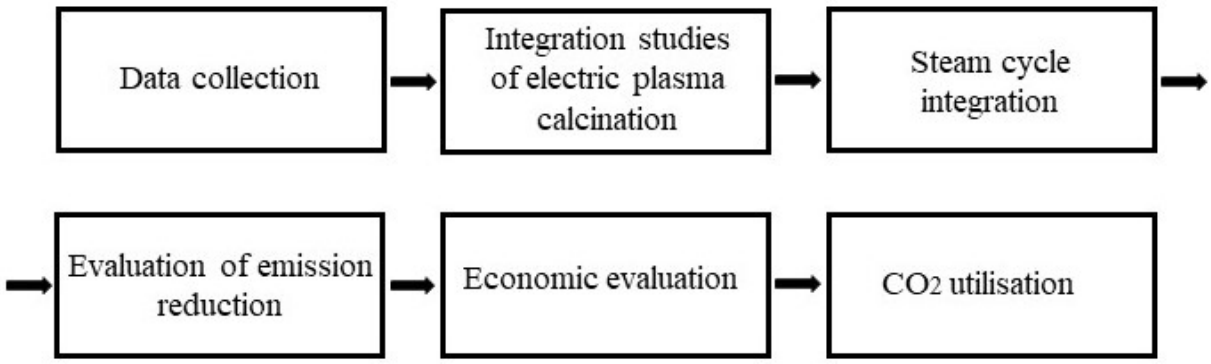

Figure 9: Flow sheet of work process.

The first step was to collect stream data for the electric plasma technology from literature and previous studies to be able to perform pinch analysis. From the pinch analysis data, the grand composite curve, *GCC*, of the process could be constructed using the Excel add-in application ProPi3. The stream data of the conventional process was collected from existing studies for an existing stand-alone pulp mill as well as an integrated pulp and paper mill. The streams connected to the calcination process were identified and removed from the mill process stream data to be able to generate the GCC of the remaining pulp/pulp and paper process without the conventional calcination process.

When the GCCs of the electric plasma calcination technology and the pulp/pulp and paper process without calcination were generated, it was possible to investigate integration possibilities of the new technology. To investigate how well the electric plasma technology can be integrated with the rest of the pulp/pulp and paper process, a foreground/background analysis with split-GCCs was performed with the electric plasma calcination process as foreground and the remaining pulp/pulp and paper process as background. With the results from the foreground/background analysis, the integration of electric plasma calcination with the pulp/pulp and paper process was evaluated from an energy perspective.

To investigate the possibilities for increased steam and power production, a steam cycle was integrated with the process using an in-house excel add-in software.

The carbon footprint of the conventional calcination process was compared with that

of the electric plasma calcination. In particular, the carbon dioxide emissions from the combustion in the lime kiln were assessed, as well as the off-site greenhouse gas emissions of the electricity production needed for the electric plasma calcination.

A simple economic evaluation, in terms of total annualised cost, was performed to investigate the economic profitability of investing in the electric plasma calcination technology compared to the investment of a new lime kiln for the conventional calcination concept.

A result of the implementation of electric plasma calcination is a pure carbon dioxide stream, which enables cost and energy-efficient capture and further handling of the carbon dioxide. There were a few possible options for the handling of the carbon dioxide including carbon capture and storage, *CCS*, conversion to electro-fuels or electro-chemicals, as input to a LignoBoost process for lignin extraction from the black liquor, or use as acid in the bleaching process of the pulp. A literature review of the possible carbon dioxide utilisation options was performed. The possibility to utilise the carbon dioxide for electro-fuel production was further investigated including process layouts, economic assessments and heat integration studies. The lignin extraction concept LignoBoost was also further investigated including sizing of the LignoBoost plant and a simple economic evaluation. These two options were chosen as they have the capacity to extend the product portfolio of the two mills and they are relatively new concepts. The other two options, CCS and on-site utilisation were not further investigated.

3.2 Pinch Analysis and Modelling

To perform heat integration studies with pinch analysis, a Foreground/Background analysis with splic-GCCs was performed using the excel add-in application ProPi3. Pinch analysis is a tool for targeting the energy requirement of a process and is described by Kemp [27]. By several graphical visualisations, the minimum energy requirements for a specific process can be found. Two of the most common tools within pinch analysis, which were used within this project, are the Grand Composite Curve and Foreground/Background analysis using split-GCCs.

To construct the GCC of a process, the stream data of the process had to be collected. Each stream with a heating or cooling demand was modelled as a hot or cold stream with a known start temperature and target temperature. A hot stream is defined as a stream with excess heat, thus requires cooling. Hence, a cold stream is defined as a stream with a heat deficit that requires heating.

Streams where the physical properties change drastically over the temperature range, such as evaporating or condensing streams were modelled in three steps, one step each for the different phases and one step for the transition between the phases where a constant temperature was assumed.

Reacting streams produce or consume heat in the form of reaction heat which also needed to be considered in the pinch analysis. An exothermic reaction releases heat to the surroundings and was thus considered as a hot stream with heat excess, hence, an endothermic reaction requires heat and was thus considered a cold stream with heat deficit.

Reacting streams were modelled as streams with constant temperature and with a heating/cooling capacity, Q, equal to the heat of reaction.

A minimum temperature difference for heat transfer, ΔT_{min}, was defined for the system to be able to construct the GCCs. All streams were then shifted in temperature according to Equation 7. Hot streams are shifted down, using the minus sign in equation 7, while cold streams are shifted up, using the plus sign in the same equation. In this way, hot and cold streams are easily matched for heat integration without interfering with the minimum temperature difference for heat transfer.

$$T_{shift} = T \pm \frac{\Delta T_{min}}{2} \tag{7}$$

Foreground/Background analysis using split-GCCs is a tool used for heat integration studies. The GCC of the process part of interest for integration studies is constructed along with the GCC of the remaining process, where the process part is to be integrated. The GCC of the remaining process becomes the background process and the GCC of the process part of interest is the foreground process. The GCC of the foreground process is mirrored and plotted in the same graph as the background process. The result is a graphical visualisation of how well the foreground process may be heat integrated to the background process.

4 Input Data and Assumptions

In this chapter, the input data and assumptions that form the base for calculations are presented.

4.1 Investigated Mills

Two mills were investigated and used as models to verify the proposed concepts. The two mills investigated are the integrated pulp and paper mill Holmen Iggesund, and the stand-alone pulp mill Södra Cell Värö, denoted *Iggesund* and *Värö* throughout this work.

There are significant differences between the two mills in terms of production capacity, energy need and electricity demand. The plant specifications are summarised in Table 1 and are taken from earlier works by Gustafsson [28] and Glader [29] for Iggesund and Pedersén et al. [30] for Värö. The amount of calcium oxide used per ADt pulp produced is assumed based on an average Swedish mill and assumed valid for both investigated mills. The assumed calcium oxide to ADt pulp factor is presented in the aforementioned table together with the mills specifications.

The main difference between the two mills is that Iggesund produces both pulp and paper while Värö produces only pulp. This leads to that Värö has a surplus of heat while Iggesund has a heat deficit due to the energy consumption of the paper production. Thus, Värö is self-sustaining with electricity and can even sell electricity to the grid while Iggesund consumes more electric energy than it can produce and therefore needs to purchase electric power from the grid to power the processes.

The stand-alone pulp mill Värö is equipped with both a back-pressure and a condensing turbine, while the integrated pulp and paper mill Iggesund is only equipped with a back pressure turbine. Thus, Iggesund cannot utilise any excess low-pressure steam for electricity production.

For the stand-alone pulp mill, the recovery boiler provides enough heat to cover the hot utility demand of the process. The surplus energy, plus the energy from the bark from the adjacent sawmill, is used for electricity production. In the integrated pulp and paper mill, the energy from the recovery boiler alone is not enough to cover the utility demand of the process. The recovery boiler has to be supported by an extra bark boiler to produce enough energy for the system. In this case, bark is not only a by-product from an adjacent sawmill but also a material that has to be purchased by the mill. The bark boilers are assumed to be used at maximum capacity.

Although the mills are self-sustaining, or partly self-sustaining with electric energy, the mills typically have a connection to the electricity grid that enables power supply equal to the mills power consumption and ensures power supply in case the internal power production is failing [9]. The equipment and contracts are often in place for the mill to buy the amount of electricity needed if their turbines need to be shut down. This power capacity is not used under normal operation and is thus considered available for other electric energy demanding processes, such as the electric plasma calcination technology or CO_2 utilisation processes, in this work. The total grid capacity of the investigated mills

based on this assumption is presented in Table 1.

The two mills use different fuels to power their lime kilns. Iggesund uses tall oil pitch (TOP) which is a by-product from refining of crude tall oil [14], and Värö uses saw dust and which comes from a nearby sawmill [31]. These fuels were compared to natural gas and fuel oil as these are common fuels to use in lime kilns internationally.

	Unit	Iggesund	Värö
Capacity	[ADt/yr]	405 000	694 000
Calcium oxide to ADt pulp factor, f_c[1]	[t/t]	0.3	0.3
Operating time	[h/yr]	8 000	8 000
Minimum hot utility demand	[MW]	204.7	231.1
Minimum cold utility demand	[MW]	97.1	80.3
Recovery boiler capacity	[MW]	190	341
Bark boiler capacity	[MW]	78	39
Electricity production	[MW]	43.4	122.9
Electricity demand	[MW]	59.6	77.4
Electricity sold	[MW]	0	45.5
Electricity bought	[MW]	16.2	0
Total grid capacity[1]	[MW]	59.6	77.4

[1] Qualified assumption

Table 1: Plant specifications for the two investigated mills.

The GCCs of the pulp/pulp and paper processes were constructed by Gustafsson [28] for Iggesund and Pedersén et al. [30] for Värö and are presented in Figure 10 and Figure 11. This was considered the starting point of this work. From the data collection and construction of these GCCs, the work followed the procedure described in Section 3.1. The stream data used for construction of the GCCs is presented in Appendix B. A global ΔT_{min} of 10 °C was used for Iggesund while individual ΔT_{min} specified for each stream were used for Värö. The individual ΔT_{min} used for GCC construction of Värö can also be found in Appendix B.

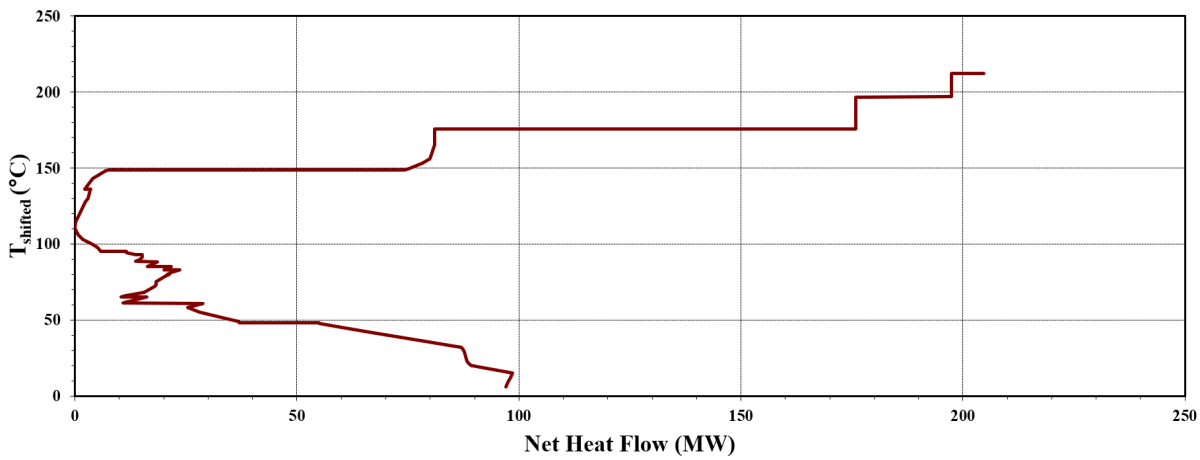

Figure 10: Grand composite curve for Iggesund pulp and paper mill [28].

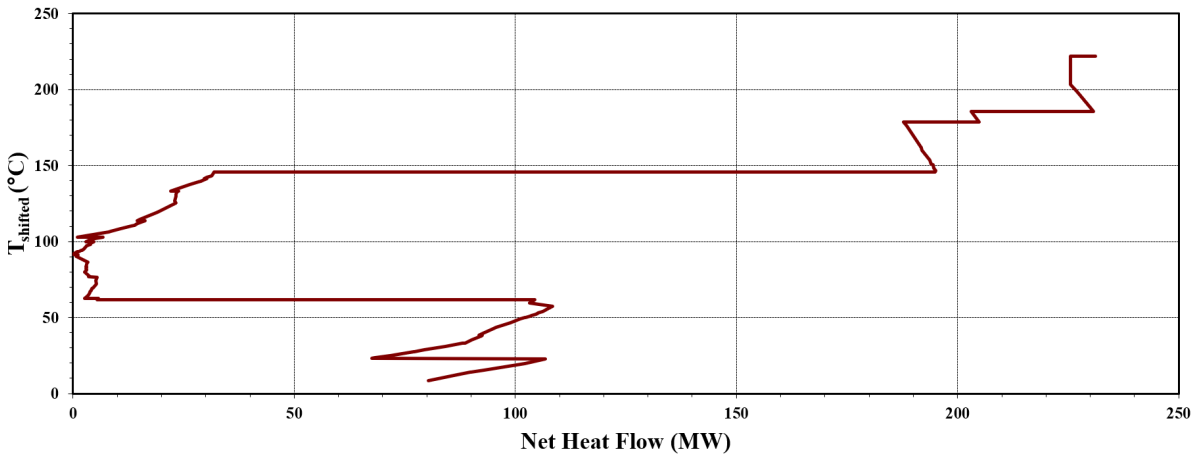

Figure 11: Grand composite curve for Värö pulp mill [30].

4.2 Electric Plasma Calcination

Assumptions and input data regarding the integration of electric plasma calcination, including GCC construction, steam cycle integration, emission and economic assessments are presented in this section.

4.2.1 Electric Plasma Technology

Assumptions and input data for establishing fundamental data regaring the electric plasma calcination technology, including process stream composition, process scaling and electricity demand is presented in the following section.

4.2.1.1 GCC Construction

The construction of the GCC of the electric plasma calcination with steam slaking was based on the process schematic presented in Figure 6, Section 2.2.1. The assumptions for calculations and construction of the GCC regarding material properties and equipment specifications are presented in Table 2. The specific heat capacity for each component was estimated for the mean temperature of each stream. The reaction temperature was assumed to be 200 °C, and the reaction heat was recalculated for the reaction temperature from the standard heat of reaction, taking the varying heat capacities into account. The calculation of the heat of reaction is found in Appendix C and physical properties of substances are found in Appendix D.

Composition lime mud	Unit	Value
Dry matter content	[%]	75
Calcium carbonate	[%]	95
Inert material (in dry lime mud)	[%]	5
Equipment specifications		
Slaker efficiency	[%]	95
Reactor efficiency	[%]	95
Reaction temperature	[°C]	200

Table 2: Assumptions related to the GCC construction of electric plasma calcination with steam slaking [6].

A ΔT_{min} of 10 K was assumed for the construction of the GCC of the electric plasma calcination.

4.2.1.2 Scaling of Electric Plasma GCC

The GCC of the electric plasma process with steam slaking was first calculated for 1 kg/s of wet lime mud to establish a general case. To be able to investigate integration with the rest of the pulp or pulp and paper process, the GCC had to be scaled to fit the production rates of the plants investigated. The scaling factors, f_s, were calculated according to Equation 8 for the two investigated mills based on the assumed ratio of calcium oxide per ADt pulp produced, f_c, and the mills production capacity, $\dot{m}_{ADt}$ [ADt/yr], found in section 4.1. The reference molar flow of calcium oxide, $\dot{n}_{CaO,ref}$ is based on the general case and was calculated to 7.1 mol/s. The scaling factors are presented in Table 3.

$$f_s = \frac{\dot{n}_{CaO}}{\dot{n}_{CaO,ref}} = \frac{\dot{m}_{ADt} \cdot f_c}{M_{CaO}} \cdot \frac{1}{3.6} \cdot \frac{1}{h} \cdot \frac{1}{\dot{n}_{CaO,ref}} \tag{8}$$

Plant	Scaling factor, f_s
Iggesund	10.57
Värö	18.12

Table 3: Scaling factors for investigated mills.

4.2.1.3 Electricity Demand

The electricity demand of the process consists of the electricity demand for the plasma generator plus the electricity demand for the two compressors used to compress the carbon dioxide used in the plasma generator. The specific electricity consumption for these units were interpolated from Bjotveit et al. [6] and are presented in Table 4. Interpolation calculations are presented in Appendix A.

Equipment	Unit	Specific power demand
Plasma generator	[kJ/kg CaO]	5 300
Compressor 1	[kJ/kg CaO]	140
Compressor 2	[kJ/kg CaO]	50
Total power demand	[kJ/kg CaO]	5 490

Table 4: Specific power demand of the electric plasma calcination concept.

The total electricity consumption was scaled to fit the production of the two investigated mills using the scaling factors presented in the previous section.

4.2.2 Steam Cycle Integration

Many of the processes in the mills are driven by steam that is generated by evaporating water in the recovery and bark boilers. The steam is expanded to different pressure levels in turbines that generate electricity. The heat is released when the steam condensates at a constant, known temperature given by the steam pressure.

The integration of a steam cycle was targeted by performing a foreground/background analysis with the steam cycle as foreground and the pulp mill and heat of combustion from the recovery and bark boilers as background. This gave an approximation on how much electricity that can be generated by the process and if some excess heat exists. By comparing the results for the original process and the process with the electric plasma calcination, changes in targeted electricity production and excess heat can be found.

An in-house add-in program to excel was used to approximate the integration of the steam cycle and how much electricity that could be generated. The stream data for the process together with the heat of combustion of the boilers was added as the background process. The temperature and pressure for the steam that leaves the boiler and up to three steam levels corresponding to turbine extractions was added. The steam cycle integration was done via an iterative procedure. The mass flows for the different steam levels were estimated and the steam cycle was calculated as the foreground process from tabulated steam data. The estimated mass flows of the steam cycle were then iterated until the foreground steam cycle was ideally fitted to be integrated with the background pulp mill processes.

The program allows only one inlet, which means that the recovery and bark boilers had to be modelled as one, even though they have slightly different pressures and temperatures for their outlet steam. Also, as the mills have more steam levels than three, some of the steam levels that are close to each other had to be modelled as one.

Two different approaches were used to integrate the heat from the electric plasma calcination with steam slaking; by integration with the existing utility system, or by a separate utility system for the electric plasma calcination process.

The first approach was to integrate the utility system of the electric plasma calcination process completely with the utility system of the remaining processes of the plant. This approach gives the maximum theoretical utilisation potential for the new electric furnace.

This was modelled by integrating the GCC for the electric plasma calcination process with the GCC of the remaining process. In practice, this could result in several streams flowing between the electric plasma calcination process and the recovery boiler, which could be impractical.

The other approach was to have a separate utility system for the electric plasma calcination apart from the rest of the processes, which is the approach that an earlier study had proposed [6]. A hot water stream at 90 °C enters the process and cools down the carbon dioxide stream after the two compressors. After that, the water stream is evaporated to low pressure steam by the hot carbon dioxide stream and the excess heat in the steam slaker. The modelling was divided into two parts. First, calculations on how much low-pressure steam that the electric plasma calcination process could produce were made. Secondly, the produced steam was integrated as a single stream in the GCC of the remaining process.

4.2.3 Greenhouse Gas Emissions

To be able to compare the greenhouse gas emissions from the conventional calcination process with the electric plasma calcination technology, the specific off-site emissions from electricity production as well as the emissions from the most common fuels used in a conventional lime kilns had to be specified. The fuels analysed were natural gas and fuel oil as these are common to use in lime kiln internationally, and wood and tall oil pitch as these are used by the studied mills.

The greenhouse gas emissions from the calcination process are the emissions released from the calcium carbonate as it is reburned to calcium oxide, and the emissions connected to the reburning unit. The carbon dioxide released from the calcium carbonate during the reburning process originates from the wood raw material and are thus of renewable origin. These emissions are independent of the reburning technology. The emissions from the conventional calcination process are either fossil or biogenic. The emissions from the electric plasma calcination are those associated with changes in electricity production due to changes in import or export of electric power between the mill and the grid.

The Swedish electricity production mainly consist of hydro power, nuclear power and wind power which are technologies with low greenhouse gas emissions [32]. However, the electricity system is interconnected with neighbouring countries and the Swedish electricity is traded on the Nordpool market, which includes the Nordic and Baltic countries. The average emissions from electricity production at Nordpool are significantly higher than the average of the Swedish electricity mix [33]. For the emissions from the electricity production, the two system boundaries, the Swedish electricity production mix, and Nordpool market place, are investigated and compared.

The specific emissions from the most common fuels used in the conventional calcination process and the specific average, fossil, emissions of the electricity mix in Sweden as well as in Nordpool are presented in Table 5. The presented numbers only account for the emissions from combustion for the fuels, upstream emissions are not considered.

Fuel	Specific emissions [kg CO2-eq/MWh]	Reference
Natural gas	202	[34]
Fuel oil	279	[34]
Tall oil pitch*	321	[35]
Wood*	395	[34]
Electricity: Sweden	20	[32]
Electricity: Nordpool	125	[33]

* Biogenic (net zero) emissions

Table 5: Specific emissions of power sources for the reburning unit.

The energy consumption of the calcination technologies was assumed, in line with the theory presented in section 2.1.2 and section 2.2.1, to be 6.0 MJ/kg CaO for the conventional lime kiln, and 5.5 MJ/kg CaO for the electric plasma calcination technology.

4.2.4 Economic Evaluation

The cost of the electric plasma calcination with steam slaking process in Section 2.2.1 were evaluated and compared to the cost for a new conventional lime kiln in order to identify the cheapest option if the investigated mills would replace their existing lime kilns. The total investment cost for both technologies, including equipment and installation costs, were based on values given in the report by Bjotveit et al. [6] and can be found in Appendix E. Note that the cost for the plasma generators were for generator modules with a capacity of 5 MW each that were placed in parallel until they could cover the required power demand and thus scaled linearly. The other values were exponentially rescaled to fit the production of the studied mills with Equation 9, where C is the total investment cost, CAPEX, C_{ref} is the total investment cost for the reference case, P the power consumption of the plasma generator, and P_{ref} is the power consumption for the reference case.

$$C = C_{ref} \cdot \left(\frac{P}{P_{ref}}\right)^{0.6} \tag{9}$$

All investment costs were adjusted with the Chemical Engineering Plant Cost Index (CEPCI) for the year 2019, see Appendix E. The expected lifetime for the equipment was set to 25 years, except for the lime kiln which was given an expected lifetime of 40 years. The currency conversion rates are presented in Appendix E.

The operating costs, OPEX, included operation and maintenance, O & M, fuel or electricity costs and a yearly investment cost for changing of electrodes in the plasma generators. The yearly investment cost is presented along with the total investment costs for the reference mill in Appendix E. The other assumptions are presented in Table 6.

Parameter	Unit	Value	Reference
O & M	[% of annualised investments]	4	[36]
Electricity price	[€/MWh]	30	[36]
Natural gas price	[€/MWh]	40	[37]
Fuel oil price	[€/MWh]	26	[38]
Tall oil pitch	[€/MWh]	33	[14]
Wood price	[€/MWh]	16	[39]
Interest rate	[%]	6	[9]

Table 6: Economic parameters for the calcination process.

To be able to compare the two alternatives with each other, the total annualised cost were calculated. This means that the investment costs were annualised with the interest rate over the lifetime of the equipment and added to the annual operation costs. The equation to calculate the annuity factor is presented in Appendix E.

4.3 Carbon Capture and Utilisation Processes

The possibility to utilise the pure, biogenic, carbon dioxide resulting from the integration of electric plasma calcination was investigated for the two studied mills. Two different utilisation options were investigated more in detail; the production of electro-fuels and usage of carbon dioxide for extraction of lignin via the LignoBoost concept. Assumptions made regarding production capacity and economy for the two technologies are presented in this section.

4.3.1 Electro-fuels

The input data and assumptions regarding electro-fuels were divided into production capacity assumptions and economic assumptions and are presented in this section.

4.3.1.1 Production Capacity

The possibility to produce electro-fuels from the pure carbon dioxide stream resulting from the electric plasma calcination process was investigated for the stand-alone pulp mill Värö and the integrated pulp and paper mill Iggesund. It was assumed that *either* electro-methane *or* electro-methanol is produced on-site by hydrogenation with hydrogen produced by electrolysis of water. The aim for the two mills was to produce as much electro-fuel as possible with the available carbon dioxide and electric power capacity.

The theoretical maximum production of electro-fuels, using all available carbon dioxide from the electric plasma calcination concept, and the corresponding electricity consumption of the electrolyser was calculated for the two investigated mills. The calculated electricity consumption of the electrolyser was compared to the available electric power capacity of the mills, to find the limiting factor for the electro-fuel production. The sizing of the electro-fuel plant was thereafter made based on the limiting factor to achieve maximum electro-fuel production.

For calculations of electro-fuel production capacity the following assumptions were made. The production was carried out according to the process schematic presented in Section

2.3.1. Biogenic carbon dioxide from the electric plasma calcination process was used as raw material along with hydrogen produced by electrolysis of water.

The theoretical maximum production of electro-fuel and the corresponding electricity consumption of the electrolyser, using all available carbon dioxide, was calculated based on the production rate of the mills and the ratio of calcium to produced pulp, ADt, see Section 4.1. The calculation procedure is presented below.

$$Electro-fuel\ production \begin{cases} \dot{m}_{CaO} = \dot{m}_{ADt} \cdot f_c \\ \dot{n}_{CaO} = \dot{m}_{CaO} \cdot \frac{1}{h} \cdot \frac{1}{3.6} \cdot \frac{1}{M_{CaO}} \\ \dot{n}_{CO_2} = \dot{n}_{CaO} \\ \dot{m}_{CH4/CH3OH} = \dot{n}_{CO_2} \cdot M_{CH4/CH3OH} \cdot X \end{cases}$$

$$Electricity\ requirement \begin{cases} Q_{CH4/CH3OH} = \dot{m}_{CH4/CH3OH} \cdot LHV_{CH4/CH3OH} \\ Q_{H2} = \frac{Q_{CH4/CH3OH}}{\eta_r} \cdot (1 + f_{H2}) \\ P_{el} = \frac{Q_{H2}}{\eta_{el}} \end{cases}$$

Where $\dot{m}_{CaO}$ is the mass flow of calcium oxide [t/yr], $\dot{m}_{ADt}$ is the mills production capacity of pulp [ADt/yr], f_c is the ratio of calcium oxide per tonne ADt and h is the operating time in [h/yr], see Section 4.1 for values. $\dot{n}_{CaO}$ denotes the molar flow of calcium oxide and $\dot{n}_{CO_2}$ is the molar flow of carbon dioxide [mol/s]. $\dot{m}_{CH4/CH3OH}$ denotes the mass flow of produced electro-fuel (methane/methanol) [kg/s] and $M_{CH4/CH3OH}$ is the molar mass of the electro-fuel [kg/mol]. $Q_{CH4/CH3OH}$ denotes the production of electro-fuels in [W], calculated via the lower heating value of the fuel, denoted $LHV_{CH4/CH3OH}$ [MJ/kg]. Q_{H2} denotes the hydrogen demand in [W] and P_{el} is the electric power requirement of the electrolyser. X is the conversion of carbon dioxide to methane/methanol, f_{H2} is the excess hydrogen factor, η_r is the fuel synthesis reactor efficiency and η_{el} is the electrolyser efficiency (electric energy to hydrogen).

Assumptions regarding process equipment specifications used for production capacity calculations are presented in Table 7. Physical properties of substances are presented in Appendix D.

Parameter	Value [%]
Electrolyser efficiency (energy-to-H_2), η_{el}	70
H2 excess, f_{H_2}	0.1
Hydrogenation reactor efficiency methane, η_r	80
Hydrogenation reactor efficiency methanol, η_r	75
Conversion of CO_2, X	99

Table 7: Electro-fuel synthesis process specifications.

The available electric power capacity for use in the electro-fuel production process was calculated by a power balance according to Equation 10.

$$P_A = P_P + P_{prod} + P_I - P_D \tag{10}$$

Where P_A is the available power capacity for use in electro-fuel production, calculated as the sum of the purchased power capacity (total grid capacity), P_P, the on-site power production, P_{prod} and the increased on-site power production by integration of electric plasma calcination, P_I, minus the total plant power demand after integration of electric plasma calcination, P_D.

4.3.1.2 Economic Evaluation

The economic potential of producing electro-fuels was investigated for the electricity price of today as well as for a future optimistic scenario. The fundamental economic assumptions are summarised in Table 8 and are taken from the report by Grahn et al. [36].

Parameter	Unit	base	future
Electricity price	[€/MWh]	30	20
Interest rate	[%]	6	
Operation time	[h/yr]	8 000	

Table 8: Fundamental assumptions for economic calculations.

For the economic assessment of electro-fuel production, the production cost of the respective electro-fuel was calculated considering the cost for hydrogen production, the fuel synthesis equipment and the value of the co-produced oxygen by electrolysis of water.

Regarding the hydrogen production it was assumed that hydrogen is produced by electrolysis of water and that two times the stoichiometric amount of water is needed for the electrolyser. The oxygen produced by electrolysis of water was sold for an income, however, it was assumed that 5 kg/ADt of oxygen are used on site and are thus not available for sale but still contribute to a cost reduction for the mill. The assumptions regarding process equipment is summarised in Table 9. CAPEX refer to the total investment cost and O & M refer to the operation and maintenance cost, fuel is not included.

Parameter	Unit	Value
CAPEX electrolyser	[k€/MW_{el}]	1 000
CAPEX stack	[k€/MW_{el}]	250
O & M (% of CAPEX electrolyser)	[%]	4
Lifetime electrolyser	[yr]	25
Lifetime stack	[h]	75 000
Stoichiometric water demand	[t/MW_{el}]	0.19
Oxygen production	[t/MW_{el}]	0.17
Water price [36]	[€/tonne]	1
Oxygen price [36]	[€/tonne]	50

Table 9: Equipment cost assumptions for electrolysis of water, year 2019.

The assumptions regarding the fuel synthesis equipment are in line with the theory presented in Section 2.3.1 and are summarised in Table 10. The costs are adjusted via CEPCI indexing, found in Appendix E, to apply for the year 2019.

Parameter	Unit	Value
CAPEX synthesis reactor methane	[k€/MW_{el}]	320
CAPEX synthesis reactor methanol	[k€/MW_{el}]	550
O & M (% of CAPEX synthesis reactor, methane)	[%]	4
O & M (% of CAPEX synthesis reactor, methanol)	[%]	4
Lifetime synthesis reactor	[yr]	25

Table 10: Equipment cost assumptions for electro-fuel synthesis reactor, year 2019.

4.3.2 LignoBoost

The extraction of lignin as a product to be sold for income via the LignoBoost concept was investigated for the two mills Iggesund and Värö. The assumptions for lignin production calculations are in line with the theory presented in Section 2.3.2 and summarised in table 11.

Parameter	Unit	Value
Maximum lignin extraction rate	[kg/ADt]	135
Electricity demand	[MWh/t lignin]	0.4
CO_2 requirement	[kg/kg lignin]	0.55
Lifetime LignoBoost plant	[yr]	25
Increased heating demand evaporation plant	[MJ/kg lignin]	0.9
Lignin market price estimation	[€/tonne]	230

Table 11: Assumptions for lignin production calculations.

The limiting factor for LignoBoost concept could either be the maximum extraction rate of lignin that does not endanger the operation of the recovery boiler, or the maximum load reduction of the recovery boiler that does not endanger the steam supply for the hot utility system of the process, which is explained in Section 2.3.2. The aim for the two mills was to produce as much lignin as possible without changing the power balance or endanger the steam supply for the hot utility system of the process.

The lignin extraction by maximum extraction rate was calculated based on the mills production of pulp. The corresponding load reduction of the recovery boiler was then calculated according to Equation 11.

$$LR_{RB} = \dot{m}_{lignin} \cdot LHV_{lignin} \cdot \eta_{RB} \tag{11}$$

Where LR_{RB} denotes the load reduction of the recovery boiler in [MW] due to lignin extraction by maximum lignin extraction rate. $\dot{m}_{lignin}$ denotes the mass flow of extracted lignin in [kg/s], LHV_{lignin} is the lower heating value of lignin in [MJ/kg] and η_{RB} is the marginal efficiency of the recovery boiler.

The maximum allowed load reduction of the recovery boiler was calculated by a power balance of the two investigated mills. It was assumed that the power demand of the plant is primarily satisfied with the purchased electricity. The part of the electricity demand which cannot be covered by the purchased electricity, due to grid limitations, see Section 4.1, must be covered by electricity generation from steam production in the recovery boiler and/or by increased power production by integration of electric plasma calcination. The recovery boiler must also cover the increased steam demand of the evaporation plant and supply the electricity needed for the LignoBoost process itself.

Hence, the maximum allowed load reduction of the recovery boiler was calculated as the total available power minus the power demand for the LignoBoost process and the power demand of the pulp/pulp and paper plant after integration of electric plasma calcination, according to Equation 12.

$$LR_{RB,max} = (L_{RB} + P_P) - (P_L + L_{evap}) - (P_D - P_I) \tag{12}$$

Where $LR_{RB,max}$ denotes the maximum allowed load reduction of the recovery boiler with respect to fulfill the steam balance of the plant, L_{RB} the current excess load of the recovery boiler, P_D is the power demand of the plant after integration of electric plasma calcination, P_L is the power demand of the LignoBoost process, L_{evap} is the increased steam demand of the evaporation plant, P_P the purchased power capacity of the plant and P_I is the increased power production resulting from the integration of electric plasma calcination.

In Equation 12, the quantities denoted with L represent a steam demand in [MW_{th}] and the quantities denoted with P represent electric power demand in [MW_{el}]. By shifting the on-site heat and power balance so that steam that originally was used for power production instead is used as hot utility it was assumed that the heat to power ratio is 1, assuming no losses in the electricity production. The decreased electric power production by a shift of the on-site heat and power balance was covered by purchased power from the electricity grid. The equation was solved in an iterative way presented in Appendix F.

A simple economic analysis of the LignoBoost concept was performed. The fundamental economic assumptions presented in Table 8 in Section 4.3.1.2 were used for the LignoBoost calculations as well. The total investment cost and operation and maintenance cost was adjusted via CEPCI indexing, found in Appendix E, to apply for the year 2019. Furthermore, it was assumed that the investment cost scales exponentially with the lignin production with a factor of 0.6 according to Equation 13.

$$C = C_{ref} \cdot \left(\frac{\dot{m}}{\dot{m}_{ref}}\right)^{0.6} \tag{13}$$

Where C is the total investment cost, CAPEX, year 2019 calculated in [Meuro], C_{ref} is the CAPEX of the reference plant equal to 10.4 [Meuro] year 2019, $\dot{m}$ is the actual lignin mass flow in [t/yr] and $\dot{m}_{ref}$ is the lignin mass flow of the reference plant, equal to 50 000 [t/yr]. The operation and maintenance cost for the LignoBoost process was assumed, in accordance with the theory in Section 2.3.2 to 22.2 [€/t lignin], adjusted to apply for the year 2019 via CEPCI index found in Appendix E. The value was in the lower range since no further concentration of the CO_2 is assumed needed from the process configuration used in this work.

5 Results

The heat integration possibilities of the electric plasma technology with steam slaking to existing mills were investigated using pinch analysis. The greenhouse gas emissions of the conventional process were compared to those of the electric plasma technology and an economic evaluation, in terms of total annualised cost, of the two concepts was performed. Furthermore, the options to utilise the pure, biogenic carbon dioxide stream for electrofuel production, or for extraction of lignin via the LignoBoost process were investigated. The results are presented in the following chapter.

5.1 Integration of Electric Plasma Calcination

The integration of electric plasma calcination with steam slaking into an existing pulp/pulp and paper process was investigated for the two studied mills. The GCC of the electric plasma concept was constructed and heat integration studies using split-GCCs were performed. The integration of the electric plasma technology via the utility system of the plant was investigated by steam cycle integration. Furthermore, an emission and economic comparison between the new and the conventional calcination concept were performed. The results are presented in this section.

5.1.1 Electric Plasma Calcination with Steam Slaking

The GCC of the electric plasma calcination with steam slaking was constructed based on the process schematic presented in Section 2.2.1. The GCC was constructed for a reference case based on a flow rate of one kilogram of wet lime mud per second as described in section 4.2.1.1, and is presented in Figure 12. The stream data used to construct the GCC is presented in Table 12.

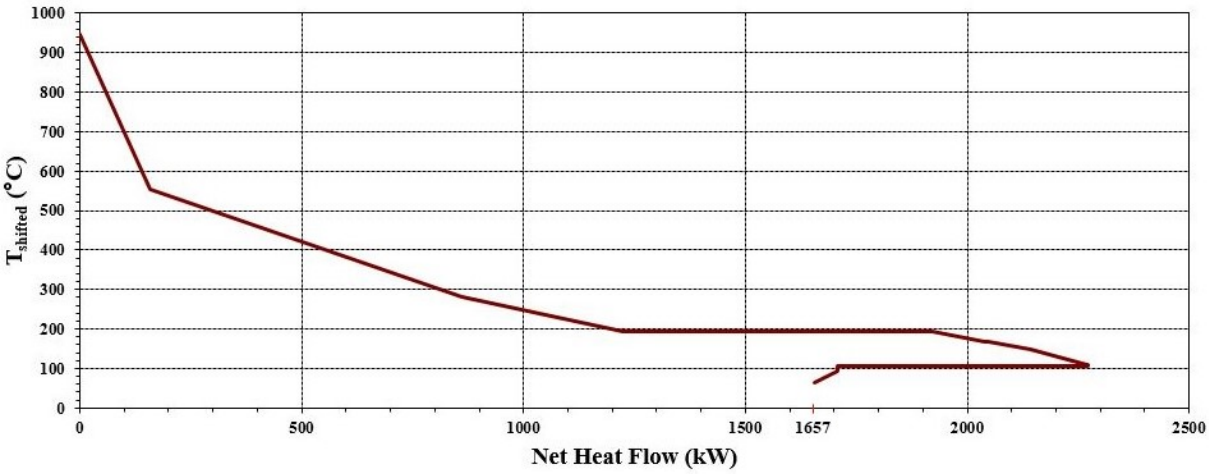

Figure 12: Grand composite curve of electric plasma calcination process based on a flow rate of one kilogram wet lime mud per second.

Type	T_{start} [°C]	T_{target} [°C]	F [kg/s]	Cp [kJ/kg °C]	Q [kW]	Comment
Cold	60	100	0.25	4.19	41.9	Dryer (heating)
Cold	100	100.1	0.25	-	564.4	Dryer (evaporation)
Cold	100	105	0.25	2.11	2.6	Dryer (superheating)
Cold	60	105	0.71	0.83	26.8	Dryer ($CaCO_3$)
Cold	60	105	0.04	0.80	1.4	Inert
Hot	950	200	0.40	0.94	281.6	Slaker pre-cooling
Hot	950	200	0.04	0.80	22.5	Inert
Hot	200	174	0.53	1.38	18.9	Slaker ($Ca(OH)_2$)
Hot	200	174	0.04	0.80	0.8	Inert
Hot	559	154	2.03	1.07	876.6	CO_2 cooling
Hot	286	114	1.67	0.99	283.2	Compressor 1 intercooling
Hot	172	100	1.67	0.95	113.7	Compressor 2 intercooling
Hot	200	199.9	-	-	696.8	Reaction heat

Table 12: Stream data for electric plasma calcination based on a flow rate of one kilogram wet lime mud per second.

From the GCC, it was found that the process generates 1.7 MW of high temperature excess heat per kg wet lime mud which is available for process heat integration.

5.1.2 Heat Integration of Electric Plasma Calcination

To investigate the heat integration possibilities of the electric plasma technology with the existing pulp/pulp and paper process, all process streams connected to the conventional calcination process were identified and removed from the original GCC, which was presented in Section 4.1. The identified process streams are visualised in Figure 13 and Figure 14 for Iggesund and Värö respectively. The corresponding stream data for the identified process streams are clearly marked in the stream data setup in Appendix B for the two mills.

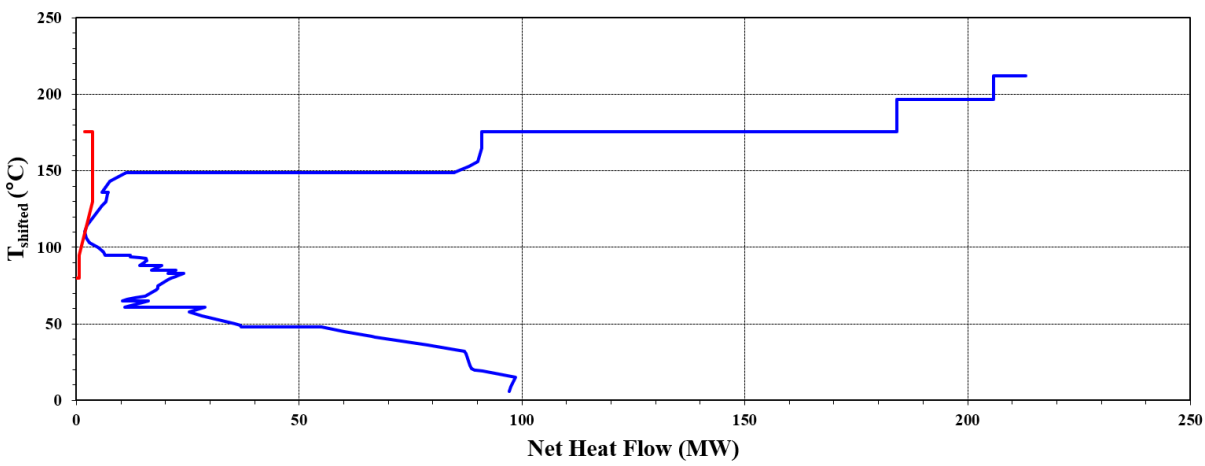

Figure 13: Visualisation of the identified process streams connected to the conventional calcination process at Iggesund by a split-GCC. Calcination process in red and background pulp and paper process in blue.

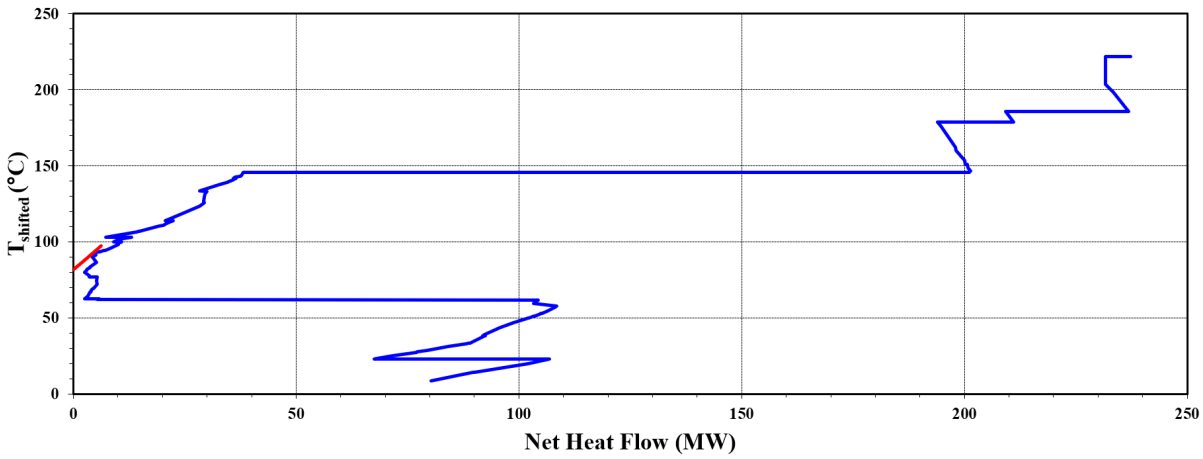

Figure 14: Visualisation of the identified process streams connected to the conventional calcination process at Värö by a split-GCC. Calcination process in red and background pulp process in blue.

The GCC of the electric plasma calcination, presented in Section 5.1.1, was scaled to fit the production of the two mills using the scaling factors presented in Section 4.2.1.2. The heat integration possibilities were then studied using foreground/background analysis with the electric plasma calcination as foreground and the remaining pulp/pulp and paper process, without calcination process, as background. The results are presented in Figure 15 for the integrated pulp and paper mill Iggesund, and in Figure 16 for the stand-alone pulp mill Värö.

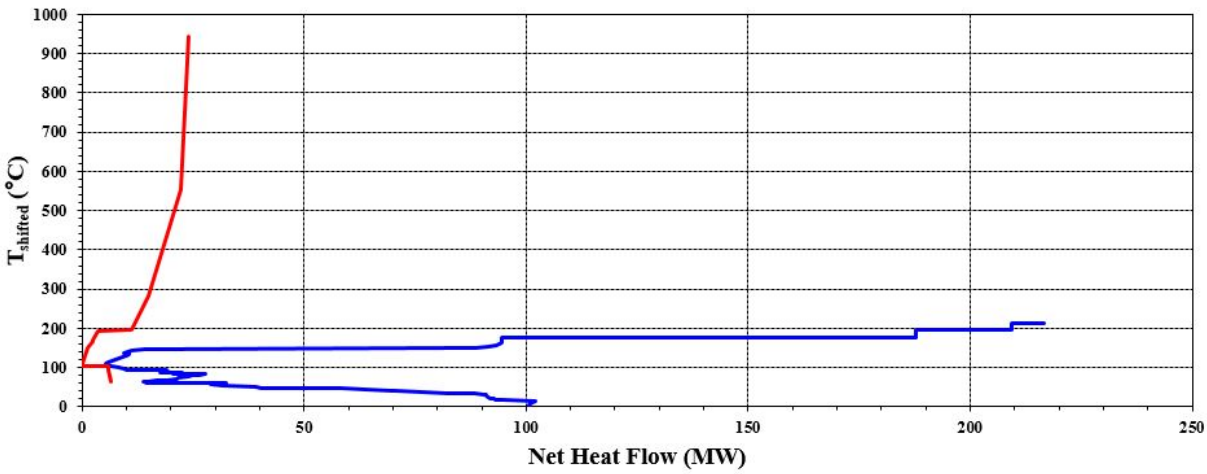

Figure 15: Visualisation of heat integration possibilities of electric plasma calcination at Iggesund mill by a split-GCC. Electric plasma calcination process as foreground (red line) and remaining pulp and paper process as background (blue line).

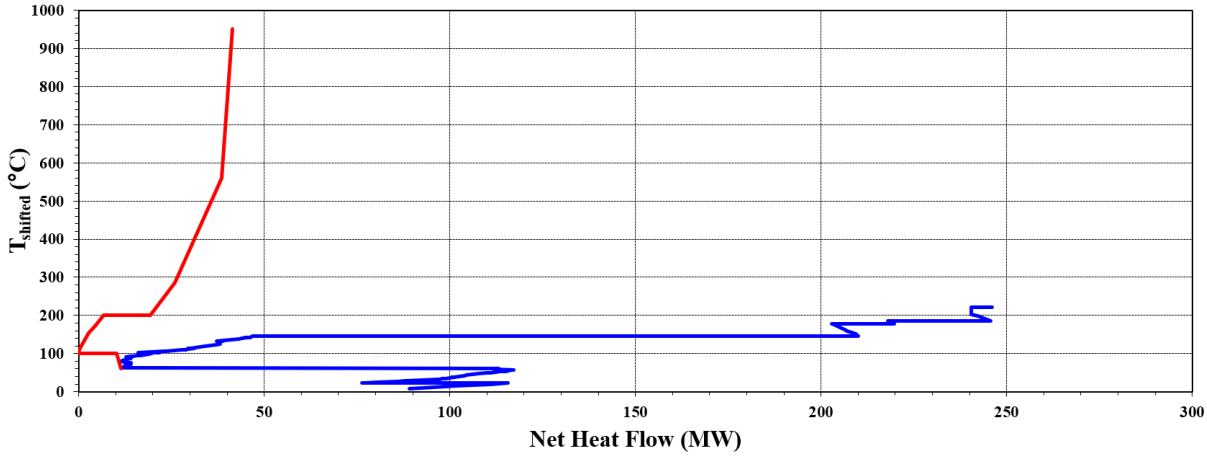

Figure 16: Visualisation of heat integration possibilities of electric plasma calcination at Värö mill by a split-GCC. Electric plasma calcination process as foreground (red line) and remaining pulp process as background (blue line).

The results from the foreground/background analysis show that it is theoretically possible to integrate the electric plasma calcination with steam slaking with the existing mills to achieve a reduction of utility demand for the integrated processes. The results are summarised in Table 13.

	Minimum hot utility [MW]	Minimum cold utility [MW]
Iggesund		
Original Plant design	211.1	97.1
After electric plasma integration	192.5	94.1
Total theoretical savings	18.6	3.0
Värö		
Original Plant design	231.2	80.3
After electric plasma integration	204.7	77.6
Total theoretical savings	26.5	2.7

Table 13: Theoretical utility savings by integration of electric plasma calcination.

5.1.3 Steam Cycle Integration

In this section, the possible integration of a steam turbine cycle is investigated. In the figures below, the solid red lines represent the pulp/pulp and paper process together with the heat generated in the recovery and bark boilers, and the dashed blue lines represents the steam cycle that can fit into the process.

In Figure 17 the steam cycles are presented for the original plants with lime kilns. For Värö, the excess heat is used to produce electricity in the condensing turbine. Since Iggesund uses purchased bark as a supplement to the recovery boiler to satisfy the process hot utility demand, see Section 4.1, the produced heat precisely covers the heat demand of the process.

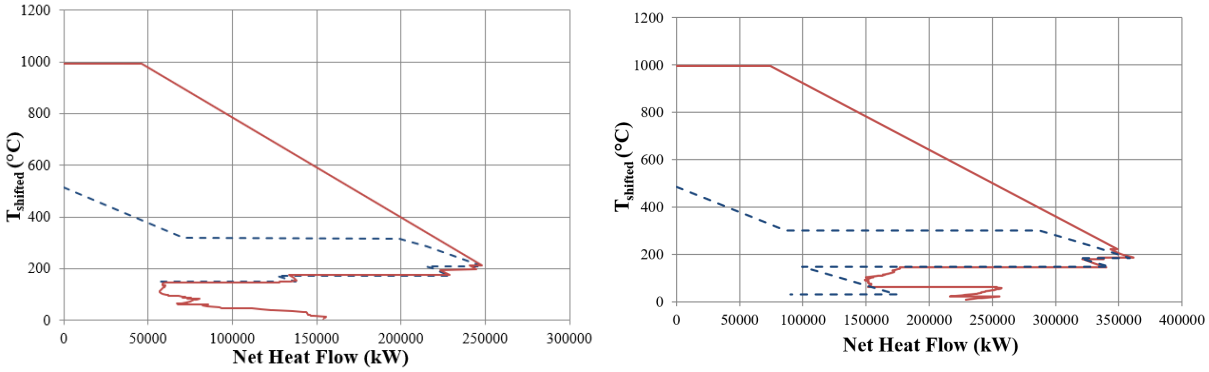

Figure 17: Integrated steam cycles (blue dashed lines) for Iggesund (left) and Värö (right) for the original mills (red lines).

The excess heat, resulting from the integration of the electric plasma calcination, can be integrated as a heat source to the utility system of the plant. This can be done in two different ways, as described in Section 4.2.2. The first approach is to integrate the utility system of the new process entirely with the utility system of the existing mill. This approach is presented in Figure 18 for the integrated pulp and paper mill Iggesund and the stand-alone pulp mill Värö respectively.

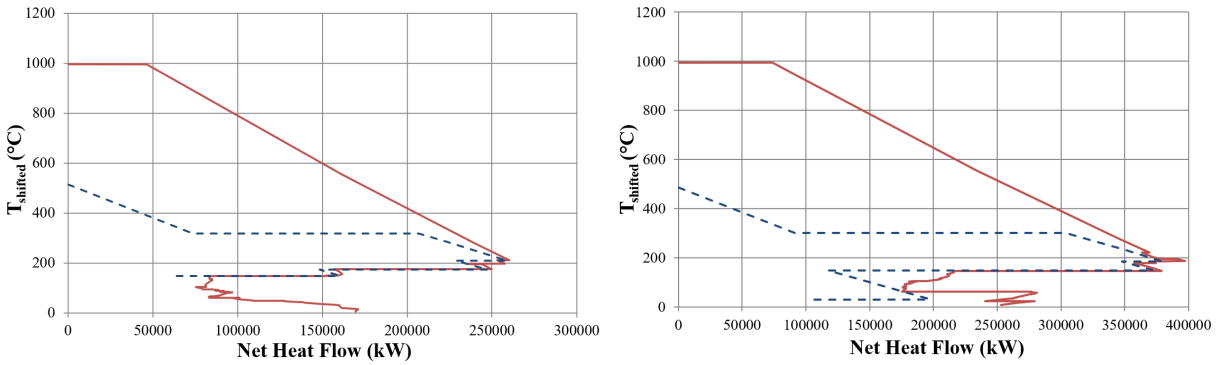

Figure 18: Integrated steam cycles (blue dashed lines) for Iggesund (left) and Värö (right) when the electric plasma calcination and steam slaking process is completely integrated with the rest of the mill (red lines).

This approach results in an increased electricity production of around 10 MW in Värö and around 4 MW in Iggesund, compared to the original plants. For Iggesund, this integration also results in an excess of low-pressure steam of around 15 MW, found by graphical approximation. The excess low-pressure steam cannot be used for electricity production with the available turbines of today, see section 4.1. To avoid the excess low-pressure steam, the load of the bark boiler could be reduced which results in saved biomass since less bark is burned. However, this would result in a decreased electricity production as less high pressure steam is fed through the turbine.

The second approach is to integrate the new process with the existing mill processes via the utility system, i.e. assuming no direct heat exchange takes place between streams in the new calcination process and the existing mill, but heat recovery is achieved via the steam network. This utility system integration approach is presented in Figure 19. In these figures, the electric plasma calcination and steam slaking process is represented in the background GCC by a hot stream representing the steam that can be generated by the electric plasma calcination concept. The steam cycle integration for the electric plasma calcination for both mills are presented in Appendix G.

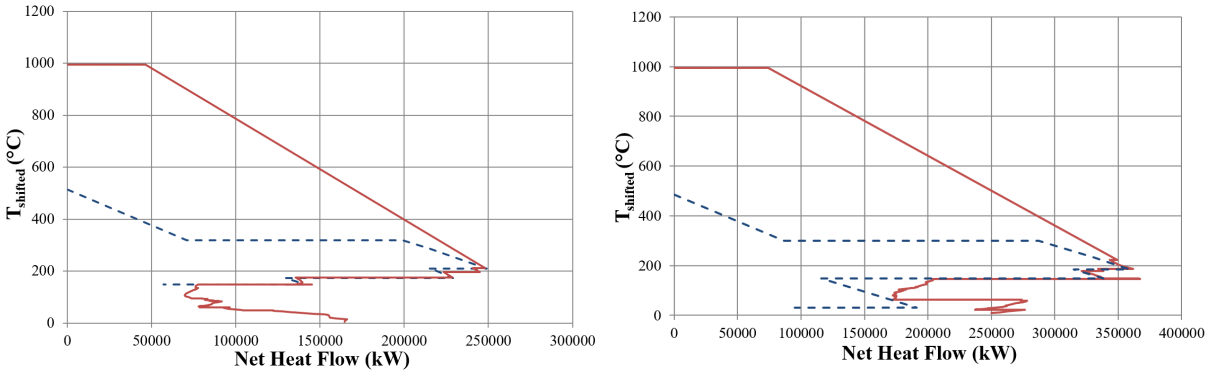

Figure 19: Integrated steam cycles (blue dashed lines) for Iggesund (left) and Värö (right) when the electric plasma calcination and steam slaking process is integrated with the rest of the mill via the steam system. The red line represents the pulp mill process with the new electric plasma calcination process represented by its potential steam generation.

This approach results in an increased electricity production of about 5 MW for Värö and no increase in electricity production for Iggesund. For Iggesund, the utility system integration also results in an excess of low-pressure steam of around 15 MW that cannot be used for electricity production due to turbine limitations. This means that Iggesund could reduce the load of the bark boiler to remove the extra steam, but that would have a negative effect on the electricity production.

The integration of the new process via the steam utility system is the approach that will be used for the rest of the report as it is considered more realistic.

5.1.4 Emission Comparison and Electricity Demand

The emissions from the calcination process consist of the carbon dioxide that is released from the calcium carbonate in the lime reburning process, and the emissions related to the reburning unit. The carbon dioxide released from the calcium carbonate is biogenic since it originates from the wood raw material. The carbon dioxide released from the calcium carbonate in the reburning process is estimated, based on the mills production capacity and the ratio of lime to ADt pulp produced, see Section 4.1, to be 95 000 tonnes/yr for Iggesund and 164 000 tonnes/yr for Värö and is independent of the reburning method. The emissions from the reburning unit on the other hand can be of either fossil or renewable origin.

The investigated fuels for the conventional reburning unit are natural gas, fuel oil, tall oil pitch and wood, where Iggesund uses tall oil pitch and Värö uses wood. A comparison of the annual greenhouse gas emissions from the reburning unit with these different fuels are presented in Table 14 for the two investigated mills.

Fuel	Iggesund [t CO_2/yr]	Värö [t CO_2/yr]
Natural gas	41 000	70 000
Fuel oil	56 000	97 000
Tall oil pitch*	65 000	-
Wood*	-	137 000

* Biogenic (net zero) emissions

Table 14: Emissions from the different fuels that a lime kiln can use. Iggesund currently uses tall oil pitch and Värö uses wood in the lime kiln.

With the electric plasma technology, the reburning process is powered by electricity and thus, the emissions from the reburning process are those associated with the electricity production. The electricity consumption was scaled to fit the production of the two investigated mills as presented in Section 4.2.1.3 and the results are presented in Table 15. The associated emissions for the two investigated mills are presented in Table 16. The emissions were calculated both for the Swedish average electricity mix and for the average Nordpool mix. The increased electricity production at Värö, resulting from the integration of electric plasma calcination, was used to cover part of the electric power demand for the electric plasma generator. The purchased power could thus be reduced which also reduced the emissions associated with the imported electricity. The net increase of on-site power production was therefore subtracted from the total electric power demand of the electric plasma calcination process presented in Table 15.

Actual electricity demands		Iggesund	Värö
Electricity demand Plasma generator	[MW]	22.4	38.3
Electricity demand compressor 1	[MW]	0.6	1.0
Electricity demand compressor 2	[MW]	0.2	0.4
Total electricity demand of the EPC[1] process	[MW]	23.2	39.7
Increased on-site electricity production	[MW]	0	5
Net increase in demand for purchased electricity	[MW]	23.2	34.7

[1] Electric plasma calcination

Table 15: Electricity demand of electric plasma calcination concept for the two investigated mills.

Electricity market		Iggesund	Värö
Sweden average electricity mix	[t CO_2/yr]	3 700	5 600
Nordpool average electricity mix	[t CO_2/yr]	23 000	35 000

Table 16: Emissions from electricity used in the electric plasma calcination with two different system boundaries.

By comparing the fossil emissions from the conventional reburning process with those from the electric plasma technology, it was found that a significant emission reduction can be achieved by integration of electric plasma technology. However, replacing conventional calcination processes driven with biomass results in increased greenhouse gas emissions since the biogenic emissions are considered net zero to the atmosphere. By replacing biomass driven calcination processes with electric plasma technology, significant amounts of biomass are released for use in other sectors which could lead to a net decrease of emissions in the larger system. The yearly amount of biomass that is released from the two investigated mills was calculated via the lower heating value of wood and tall oil pitch, found in Appendix D, and amounts to 81 000 tonnes/yr saved wood for Värö and 18 000 tonnes/yr saved tall oil pitch for Iggesund.

5.1.5 Economic Evaluation

An economic comparison of the new investment in a lime reburning unit was performed for the electric plasma calcination concept and the conventional calcination concept using a rotary lime kiln. The aim was to investigate the economic profitability of investing in the new technology compared to a re-investment in the conventional technology at the end of the lifetime of the existing reburning unit.

The cost related to the investment and installation, as well as operational cost for the electric gas plasma technology and the rotary lime kiln are presented in Table 17 and Table 18 respectively, and are based on the assumptions presented in Section 4.2.4. The increased electricity production at Värö, resulting from the integration of electric plasma calcination, was used to power the electric plasma generator thus reducing the net purchased power demand. The net purchased power demand was considered in the cost calculations.

		Iggesund	Värö
Investments			
Plasma generator	[M€]	9.5	15.0
Number of units á 5 MW	[-]	5	8
Cyclone CO_2 and $CaCO_3$	[M€]	0.8	1.2
Cyclone and reactor	[M€]	1.7	2.3
Heat exchanger CO_2 and steam	[M€]	0.3	0.4
Heat exchanger CO_2 and water	[M€]	0.1	0.2
Heat exchanger CO_2 and water	[M€]	0.1	0.2
heat exchanger slaker cycle and steam	[M€]	0.2	0.3
Filter CO_2	[M€]	1.5	2.1
Compressor 1 CO_2	[M€]	0.6	0.9
Compressor 2 CO_2	[M€]	0.6	0.9
Dryer and steam slaker	[M€]	7.7	10.6
Total investment cost (CAPEX)	[M€]	23.3	33.9
Annualised investment cost	[M€/yr]	1.8	2.7
Operating costs			
O & M	[M€/yr]	0.1	0.2
Electricity	[M€/yr]	5.6	8.3
Changing electrodes	[M€/yr]	0.5	0.9
Total operating costs (OPEX)	[M€/yr]	6.2	9.4
Total annualised cost	[M€/yr]	8.0	12.0

Table 17: Costs for investment, installation and operation of the electric plasma calcination process.

		Iggesund	Värö
Investments			
Rotary lime kiln	[M€]	55.8	77.0
Annualised investment cost	[M€/yr]	3.7	5.1
Operating costs			
O & M	[M€/yr]	0.2	0.3
Natural gas	[M€/yr]	8.1	13.9
Fuel oil	[M€/yr]	5.3	9.2
Tall oil pitch	[M€/yr]	6.7	-
Wood	[M€/yr]	-	5.6
Total annualised cost			
Natural gas	[M€/yr]	12.0	19.2
Fuel oil	[M€/yr]	9.2	14.5
Tall oil pitch	[M€/yr]	10.5	-
Wood	[M€/yr]	-	10.9

Table 18: Costs for investment, installation and operation of a lime kiln.

It was found that the total annualised cost for the two technologies were comparable and that the electric plasma calcination process was cheaper than the lime kiln fired with natural gas, fuel oil or tall oil pitch.

To investigate the impact of change in important economic parameters, sensitivity analysis was performed of the electricity price as well as the interest rate.

Figure 20 shows the total annualised cost of the electric plasma technology with steam slaking and the conventional lime kiln fired with different types of fuel for different electricity prices. The electricity price was varied between 20 to 60 €/MWh, while all other parameters were kept constant. The electric process is the cheapest option for electricity prices below 37 €/MWh for Iggesund and below 26 €/MWh for Värö. Thereafter, the lime kiln with fuel oil is the cheapest option for Iggesund, and the lime kiln with wood as fuel for Värö. The lime kiln fired with tall oil pitch at Iggesund becomes cheaper than the electric process when the electricity price exceeds 42 €/MWh.

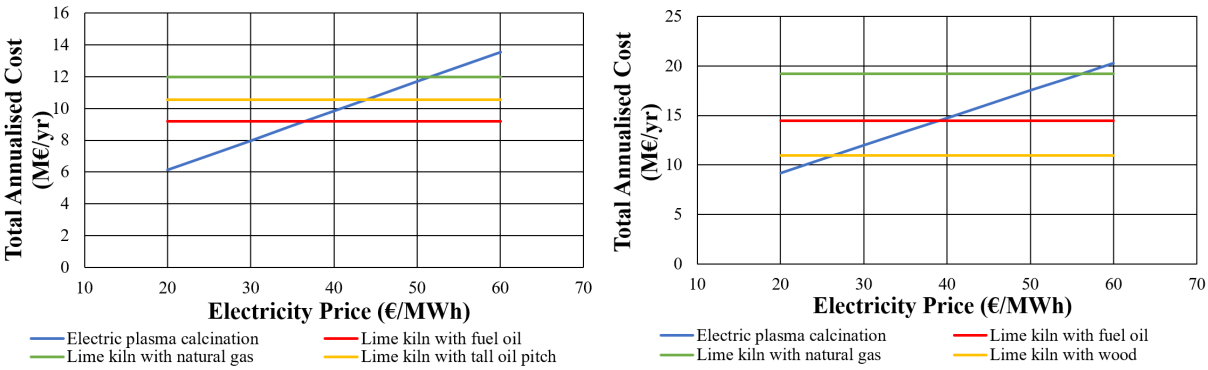

Figure 20: Sensitivity analysis of the total annualised cost with respect to electricity price for Iggesund (left) and Värö (right).

In Figure 21, the interest rate was varied to investigate how that would affect the total annualised cost for the electric plasma calcination process and for the lime kiln when fuelled with tall oil pitch or wood for Iggesund and Värö respectively. For interest rates below 9%, the lime kiln is the cheapest option for Värö. For Iggesund, the electric plasma calcination process is the cheapest option for all investigated interest rates.

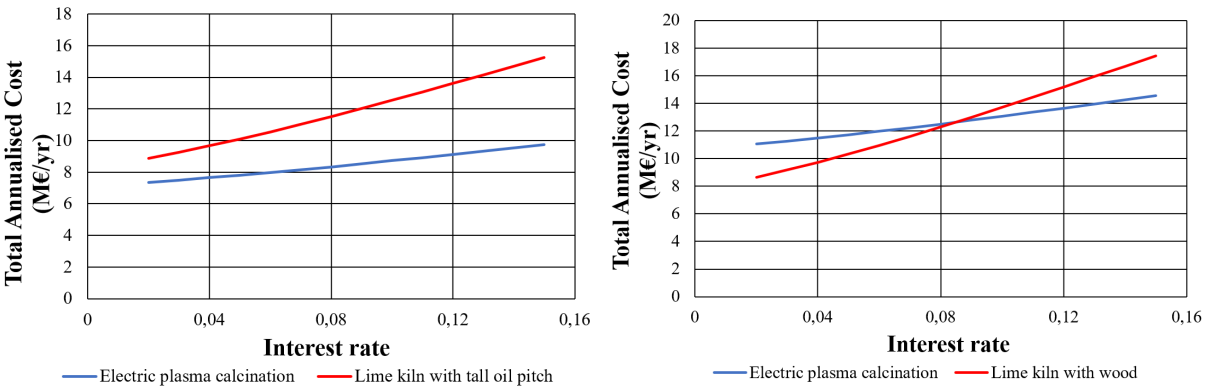

Figure 21: Sensitivity analysis of the total annualised cost with respect to the interest rate for Iggesund (left) and Värö (right).

5.1.6 Summary

A summary of the results of integration of electric plasma calcination with steam slaking is presented in Table 19. The theoretical, ideal change of utility demand resulting from the split-GCC analysis and steam cycle integration is presented along with the key results of the emission and economic assessments. Note that presented in the table is the difference between the electrified and the conventional calcination process. Hence, the net change of replacing a conventional lime kiln for electric plasma calcination with steam slaking is presented.

The emissions section in Table 19 shows only the change in fossil emissions. Since the biogenic fuels have net zero emissions, only the fossil emissions from the electricity production are considered in these cases. A comparison between the two different electricity markets is presented and compared to a lime kiln using natural gas, fuel oil, wood and tall oil pitch as fuel.

	Unit	Iggesund Δ	Värö Δ
Utility			
Minimum heating demand	[MW]	-18.6	-26.5
Minimum cooling demand	[MW]	-3.0	-2.7
Total plant power demand	[MW] *([GWh/yr])*	23.2 *(185.7)*	39.7 *(317.6)*
On-site power production	[MW] *([GWh/yr])*	0 *(0)*	5 *(40.0)*
Excess power production	[MW] *([GWh/yr])*	-	-34.7 *(-277.6)*
Imported power production	[MW] *([GWh/yr])*	23.2 *(185.7)*	-
Fossil Emissions			
CO_2 emissions (natural gas) el_{swe}[1]/el_{NP}[2]	[kt CO_2/yr]	-37/-18	-64/-35
CO_2 emissions (fuel oil) el_{swe}/el_{NP}	[kt CO_2/yr]	-52/-33	-91/-62
CO_2 emissions (wood) el_{swe}/el_{NP}	[kt CO_2/yr]	-	6/35
CO_2 emissions (TOP) el_{swe}/el_{NP}	[kt CO_2/yr]	4/23	-
Saved biomass	[kt/yr]	18	81
Economy			
Annualised investment cost	[M€/yr]	-1.9	-1.6
Operating costs (wood/electricity)	[M€/yr]	-	3.5
Operating costs (TOP/electricity)	[M€/yr]	-0.7	-
Total annualised cost	[M€/yr]	-2.5	1.1

[1] Swedish electricity mix

[2] Nordpool electricity market

Table 19: Summary of results in terms of utilities, emissions and economics of integration of electric plasma calcination with steam slaking.

It was found that the theoretical utility demand of the complete process could be reduced by integration of electric plasma calcination with steam slaking. The electric plasma technology is energy intensive which is why the total plant power demand is increased by integration of the new calcination concept which leads to a reduction of excess power production that could be sold to the grid for Värö and increased requirements for imported power at Iggesund.

It was shown that the emissions could be significantly decreased by replacing a lime kiln fired with fossil fuel for the electric plasma calcination technology provided sustainable electricity production. By replacing a lime kiln fired with biogenic fuel, a net increase of fossil emissions are achieved since biogenic emissions are considered net zero. However, large amounts of biomass could be saved (TOP at Iggesund and wood at Värö) that could be used to replace fuels of fossil origin in other sectors, hence, achieving a net reduction of greenhouse gas emissions.

Furthermore, it was found that the investment of the electric plasma calcination with steam slaking is in the same order of magnitude as the re-investment of a lime kiln.

5.2 Carbon Dioxide Utilisation

Further utilisation of the pure, biogenic stream of carbon dioxide resulting from the implementation of electric plasma calcination was investigated for the two studied mills. The two utilisation methods covered are the production of electro-fuels and the usage of carbon dioxide for lignin extraction via the LignoBoost process. This section covers the results in terms of electro-fuel production capacity, emission assessment, heat integration studies of electro-fuel synthesis process along with an economic analysis. Furthermore, the results from the LignoBoost concept are presented including the production capacity of lignin along with a simple economic analysis.

Results in this section presume investment and implementation of electric plasma calcination with steam slaking. Furthermore, results in this section assume that the ideal increase of power production at the respective mills calculated in Section 5.1.3 is available for use by CO_2 utilisation processes. Thus, the ideal increase of power production is added to the mills current power production in this section.

5.2.1 Production of Electro-fuels

The utilisation of the carbon dioxide resulting from the integration of electric plasma calcination for production of electro-fuels is further investigated and the results, including production capacity, emission assessment, heat integration possibilities and an economic evaluation, are presented in this section. The results are based on the assumptions stated in Section 4.3.1.

5.2.1.1 Electro-fuel Synthesis

The possibility to produce electro-fuels from the pure carbon dioxide stream resulting from the electric plasma calcination process was investigated for the stand-alone pulp mill Värö and the integrated pulp and paper mill Iggesund. It was assumed that *either* electro-methane *or* electro-methanol is produced on-site and a comparison between the production of these two fuels is presented in this section. If assuming that all carbon dioxide produced in the calcination process is used for electro-fuel production, the resulting theoretical production of methane and methanol and the corresponding electric power demand of the processes are presented in Table 20.

		Iggesund	Södra
CO_2 production	[kt/yr]	95.5	163.6
Theoretical methane production	[kt/yr]	34.4	58.9
Electric power demand	[MW]	106.7	182.6
Theoretical methanol production	[kt/yr]	68.7	117.8
Electric power demand	[MW]	91.5	156.6

Table 20: Theoretical maximum production of electro-fuels at the two investigated mills.

The available electric grid capacity for electro-fuel production after integration of electric plasma calcination was calculated as described in Equation 10 in Section 4.3.1.1. The results are presented in Table 21.

		Iggesund	Värö
Purchased power capacity (Total grid capacity), P_P	[MW]	59.6	77.4
Power production, P_{prod}	[MW]	43.4	122.9
Increased power production, P_I	[MW]	0	5
Total plant power demand, P_D	[MW]	82.8	117.1
Available electric capacity for electro-fuel production, P_A	[MW]	20.2	88.2

Table 21: Available electric power capacity for electro-fuel production.

It can be concluded that the electric power requirements for producing either methane or methanol from all carbon dioxide produced in the calcination process exceed the available electric power of the investigated mills. To be able to utilise all available carbon dioxide resulting from the integration of electric plasma calcination, the electric grid capacity would need to be extended by 86.5/71.3 MW (methane-/methanol production) for Iggesund and 94.4/68.4 MW (methane-/methanol production) for Värö. Thus, the electric power availability was identified as the limiting factor and only a share of the produced carbon dioxide can be used for electro-fuel production.

The resulting maximum production of methane and methanol and corresponding oxygen production and water demand with restricted electricity consumption are presented in Table 22.

		Iggesund	Värö
Electricity consumption electrolyser	[MW]	20.2	88.2
Electricity consumption electrolyser	[GWh/yr]	161.6	705.6
Methane production	[kt/yr]	6.5	28.4
Water demand	[kt/yr]	61	267
Hydrogen production	[kt/yr]	3.4	14.8
CO_2 demand	[kt/yr]	18.1	78.9
Percentage of available CO_2	[%]	19.0	48.3
O_2 production	[kt/yr]	27.1	118.5
Methanol production	[kt/yr]	15.2	66.3
Water demand	[kt/yr]	61	267
Hydrogen production	[kt/yr]	3.4	14.8
CO_2 demand	[kt/yr]	21.1	92.1
Percentage of available CO_2	[%]	22.1	56.3
O_2 production	[kt/yr]	27.1	118.5

Table 22: Maximum capacity for electro-fuel production at the two investigated mills.

5.2.1.2 Greenhouse Gas Emission Comparison

The greenhouse gas emissions from the production of electro-fuels were compared to the greenhouse gas emissions from combustion of corresponding fuels of fossil source, which the electro-fuels have potential to replace. Since the raw material for electro-fuel production is carbon dioxide of renewable origin, the emissions of the electro-fuels are solely those associated with the production, i.e., the emissions resulting from the electricity production.

The specific emissions from the electro-methane and electro-methanol are presented in Table 23 together with the carbon dioxide emissions of combustion of fossil methane and gasoline for comparison. The emissions from the electro-fuels were calculated based on the Swedish average electricity mix as well as the average emissions at Nordpool.

		Sweden	Nordpool
Electro-methane	[g CO_2-eq/MJ]	10.2	63.9
Electro-methanol	[g CO_2-eq/MJ]	10.7	66.9
Fossil methane	[g CO_2/MJ]	56.1	
Gasoline	[g CO_2/MJ]	69.3	

Table 23: Emission comparison of combustion of electro-fuels and equivalent fossil fuels.

It was found that by replacing fossil fuels with electro-fuels, there is great potential to achieve significant emission reductions. The emissions from electro-fuels are highly dependent on the emissions from the electricity production. Electro-fuels produced mainly by sustainable electricity sources have the potential to reduce emission to about one fifth, while electro-fuels produced from electricity with a larger share of fossil sources neither reduce, nor increase the emissions of carbon dioxide to any larger extent in the system.

5.2.1.3 Heat Integration of Electro-fuels

The heat integration options of electro-fuel synthesis with the remaining pulp/pulp and paper process were studied with foreground/background analysis using split-GCCs for the two investigated mills. The results for electro-methane synthesis are presented in this section. A similar analysis was performed for electro-methanol synthesis with similar results. The heat integration study of electro-methanol can be found in Appendix H.

The results of the heat integration studies are presented as split-GCCs with the electro-fuel synthesis process as foreground (red line) and the pulp/pulp and paper process with electric plasma calcination as background (blue line). The results for electro-methane synthesis are presented in Figure 22 and Figure 23 for the integrated pulp and paper mill Iggesund and the stand-alone pulp mill Värö respectively.

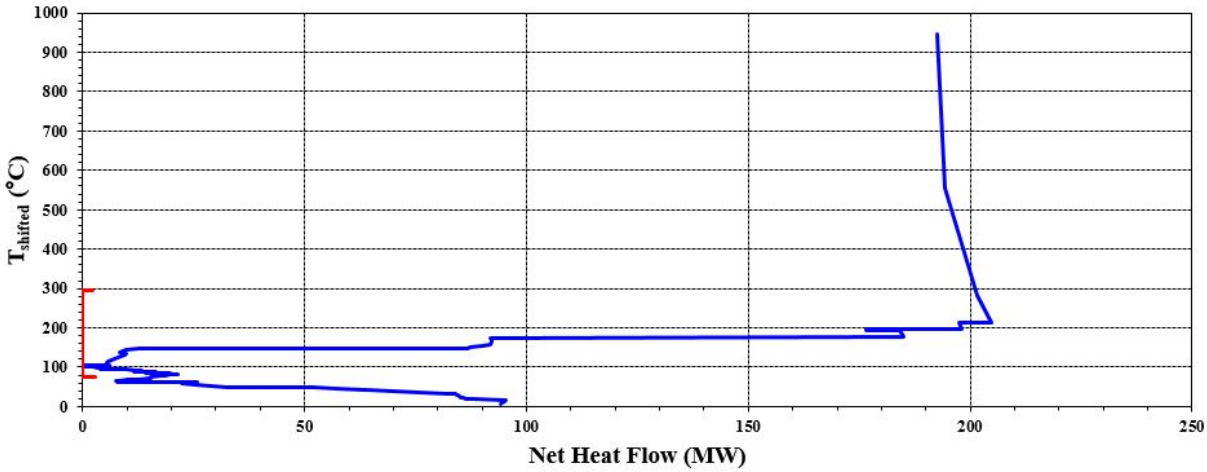

Figure 22: Visualisation of heat integration possibilities of electro-methane synthesis at Iggesund mill by a split-GCC.

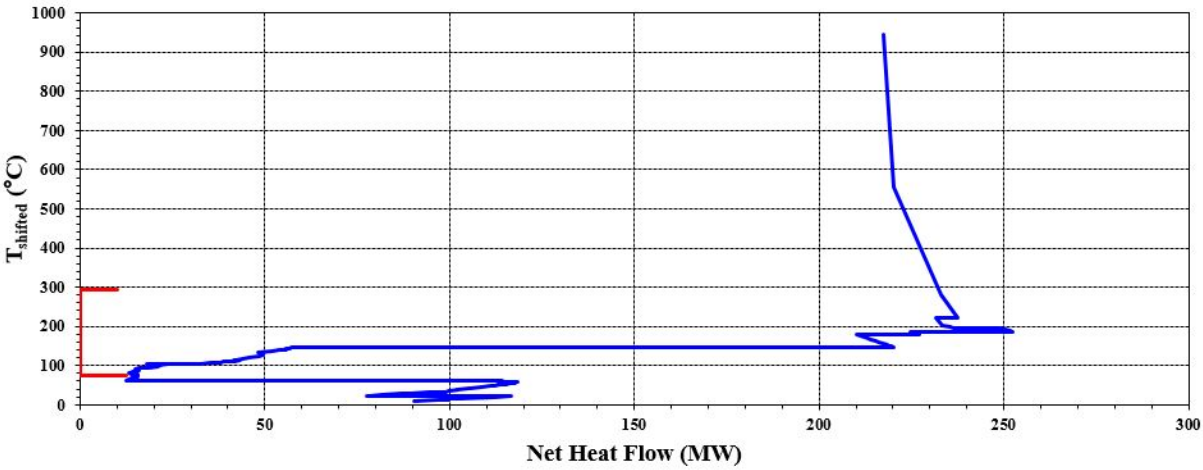

Figure 23: Visualisation of heat integration possibilities of electro-methane synthesis at Värö mill by a split-GCC.

The stream data used to construct the GCC of electro-methane synthesis consisted of a cold stream representing the heat demand of the electrolyser and a hot stream representing

the excess heat from the fuel synthesis reaction. The stream data is presented in Table 24.

T_{start}	T_{target}	Q Iggesund [MW]	Q Värö [MW]	Comment
69.9	70	2.9	12.6	Electrolyser
300	299.9	2.3	10.2	Fuel synthesis

Table 24: Stream data for electro-methane synthesis.

It was found that the theoretical minimum utility demands are further decreased by integration of electro-methane synthesis at the integrated pulp and paper mill Iggesund. However, integration of electro-methane synthesis at the stand-alone pulp mill Värö results in an increase of hot utility demand. The results are presented in Table 25.

	Minimum hot utility [MW]	Minimum cold utility [MW]
Iggesund		
Electric plasma integrated	192.5	94.1
After electro-methane integration	190.2	91.2
Total theoretical savings	2.3	2.9
Värö		
Electric plasma integrated	204.7	77.6
After electro-methane integration	207.2	77.7
Total theoretical savings	-2.5	-0.1

Table 25: Theoretical utility savings by heat integration of electro-methane synthesis.

The electrolyser temperature at 70 °C interfered with streams connected to the surface condenser at the evaporation part of the plant at Värö, making heat integration impossible between the two processes and resulting in increased utility demand. By lowering the electrolyser temperature to 50 °C, a better temperature match is achieved, thus enabling heat integration between the two processes. The results are presented in Figure 24 and in Table 26.

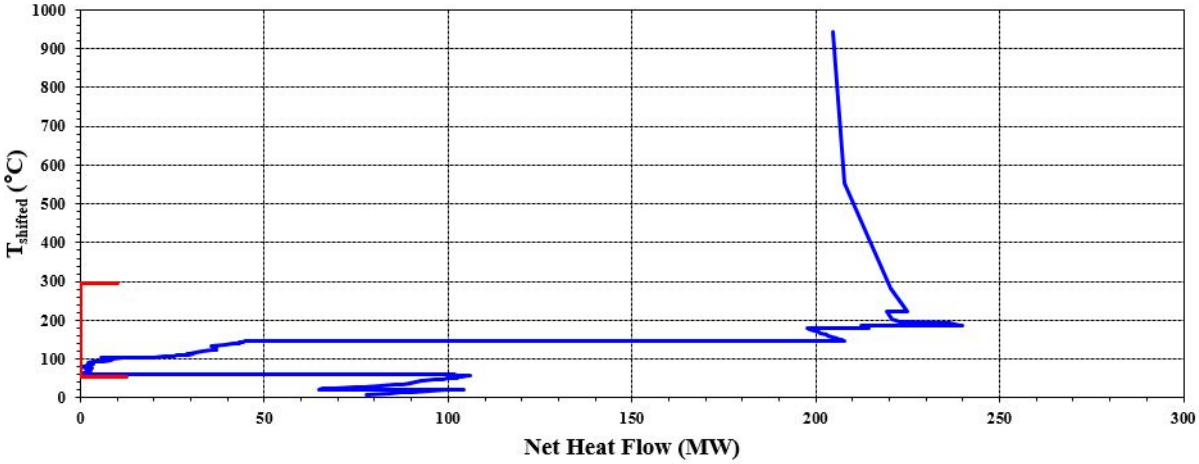

Figure 24: Visualisation of heat integration possibilities of electro-methane synthesis with an electrolyser temperature of 50 °C at Värö mill by a split-GCC.

	Minimum hot utility [MW]	Minimum cold utility [MW]
Stand alone pulp mill Värö		
Electric plasma integrated	204.7	77.6
After electro-methane integration (50°C)	194.6	65.1
Total theoretical savings	10.1	12.5

Table 26: Theoretical utility savings by heat integration of electro-methane synthesis with an electrolyser temperature of 50 °C at Värö mill.

5.2.1.4 Economic Evaluation

An economic analysis in terms of production cost calculation was performed based on the assumptions stated in Section 4.3.1.2 to investigate the economic feasibility of production of electro-fuels. The production cost of the electro-methane and electro-methanol was calculated and compared to the market price of the corresponding biofuels. The production cost was calculated for two scenarios, with the electricity price of today as well as for a future optimistic scenario with lower electricity prices.

Possible cost reductions from utility savings by heat integration of the electro-fuel process with remaining pulp/pulp and paper process, as described in Section 5.2.1.3, were not considered for this economic analysis.

The results are summarised in Table 27 for the base case, based on the electricity price of today, as well as for the future optimistic scenario with lower electricity price.

Expenses	Unit	Iggesund Base	Iggesund Future	Värö Base	Värö Future
Electrolyser					
Equipment cost	[M€]	20.2		88.2	
Stack replacement cost	[M€]	5.1		22.1	
Annualised equipment cost	[M€/yr]	1.6		6.9	
Annualised stack rep. cost	[M€/yr]	0.7		3.1	
O & M	[M€/yr]	0.06		0.28	
Water cost	[M€/yr]	0.06		0.27	
Electricity cost	[M€/yr]	4.8	3.2	21.2	14.1
H_2 production	[GWh/yr]	113		494	
H_2 production cost	[€/MWh$_{H_2}$]	64.3	50.0	64.3	50.0
Methane					
Hydrogen cost					
Hydrogen demand	[MWh$_{H_2}$/MWh$_{CH_4}$]	1.25		1.25	
H_2 production cost	[€/MWh$_{CH_4}$]	80.4	62.6	80.4	62.6
Methane synthesis					
Equipment cost	[M€]	3.6		15.8	
Annualised equipment cost	[M€/yr]	0.3		1.2	
O & M	[M€/yr]	0.01		0.05	
Methane production	[GWh/yr]	90		395	
Methane synthesis cost	[€/MWh$_{CH_4}$]	3.3		3.3	
Oxygen revenue					
Oxygen production	[kt/yr]	27.1		118.5	
Oxygen revenue	[€/MWh$_{CH_4}$]	15		15	
Production cost methane	[€/MWh]	68.7	50.8	68.7	50.8
Market price bio-methane [36]	[€/MWh]	56		56	
Methanol					
Hydrogen cost					
Hydrogen demand	[MWh$_{H_2}$/MWh$_{CH_3OH}$]	1.33		1.33	
H2 production cost	[€/MWh$_{CH_3OH}$]	85.8	66.7	85.8	66.7
Methanol synthesis					
Equipment cost	[M€]	5.8		25.2	
Annualised equipment cost	[M€/yr]	0.5		2.0	
O & M	[M€/yr]	0.02		0.08	
Methanol production	[GWh/yr]	85		570	
Methanol synthesis cost	[€/MWh$_{CH_3OH}$]	5.5		5.5	
Oxygen revenue					
Oxygen production	[kt/yr]	27.1		118.5	
Oxygen revenue	[€/MWh$_{CH_3OH}$]	16		16	
Production cost methanol	[€/MWh]	75.3	56.3	75.3	56.3
Market price bio-methanol [40]	[€/MWh]	53		53	

Table 27: Production cost calculations for electro-methane and electro-methanol production.

From the economic analysis it was found that the production cost of the electro-fuels are in the same order of magnitude as the current market price of corresponding fuels. The hydrogen production by electrolysis of water constitutes a large share of the total electro-fuel production cost where the electricity price is an essential parameter. Thus, the electricity price will has a large impact of the feasibility of electro-fuel production.

5.2.2 LignoBoost

Based on the maximum extraction rate of lignin from the black liquor and the production capacity of the two investigated mills, a theoretical maximum lignin production and corresponding load reduction of the recovery boiler was calculated according to Equation 11, described in Section 4.3.2. The results are presented in Table 28.

	Unit	Iggesund	Södra
Pulp production capacity	[kADt/yr]	405	694
Operating time	[h/yr]	8 000	8 000
Maximum lignin extraction rate	[kg/ADt]	135	135
Maximum lignin extraction, $\dot{m}_{lignin}$	[kt/yr]	54.7	93.7
Lower heating value lignin, LHV_{lignin}	[MJ/kg]	24	24
Recovery boiler marginal efficiency, η_{RB}	[%]	92	92
Reduced steam production, LR_{RB}	[MW]	41.9	71.8

Table 28: Theoretical maximum lignin production and corresponding load reduction of recovery boiler at maximum lignin extraction rate.

The maximum allowed load reduction of the recovery boiler with respect to ensure steam production for hot utility was calculated according to Equation 12, described in Section 4.3.2. The results are presented in Table 29. The current excess load of the recovery boiler after electric plasma integration, L_{RB}, was estimated using the GCCs for the two investigated mills, see Appendix F for excess load estimations.

		Iggesund	Värö
Current excess load of recovery boiler, L_{RB}	[MW]	70	170
Purchased power capacity, P_P	[MW]	59.6	77.4
LignoBoost power demand, P_L	[MW]	2.6	7.4
Increased load of evaporation plant, L_{evap}	[MW]	1.6	4.6
Plant power demand, P_D	[MW]	82.8	117.1
Increased on-site power production, P_I	[MW]	0	5
Maximum load reduction of recovery boiler, $LR_{RB,max}$	[MW]	42.6	123.3

Table 29: Maximum allowed load reduction of recovery boiler to ensure hot utility supply.

For both investigated mills it was found that the limiting factor for lignin extraction is the maximum lignin extraction rate, $LR_{RB,max} > LR_{RB}$. The LignoBoost plant was therefore sized for maximum lignin extraction. The excess heat from the recovery boiler can still be used for electricity production. The power demand that could not be covered by the recovery boiler was covered by purchased electricity.

The maximum lignin production for the two investigated mills was calculated and the results are summarised in Table 30. The production cost of lignin was calculated for two electricity market conditions with the electricity price of today and for a future optimistic scenario with lower electricity prices and compared to the market price indication of lignin. The calculated production cost of lignin includes the total investment cost, operation and maintenance, and the electricity costs and is presented in Table 30.

		Iggesund		**Värö**	
	Unit	Base	Future	Base	Future
Lignin production					
Lignin extraction	[kt/yr]	54.7		93.7	
CO_2 demand	[kt/yr]	30.1		51.5	
Percentage of total CO_2 available	[%]	31.5		31.5	
Power demand	[MW]	2.7		4.7	
Changes in utility demand					
Reduced steam production	[MW]	41.9		71.8	
Increased steam demand evaporation	[MW]	1.7		2.9	
Economy					
Total investment cost, CAPEX	[M€]	11.0		15.2	
Annualised CAPEX	[M€/yr]	0.9		1.2	
O & M	[M€/yr]	1.2		2.1	
Electricity demand lignoboost process	[GWh/yr]	21.9		37.5	
Electricity cost	[M€/yr]	0.7	0.4	1.1	0.7
Lignin production cost	[€/t]	49.9	45.9	46.8	42.9
Lignin market price indication	[€/t]	230		230	

Table 30: Summary of theoretical maximum lignin production and corresponding economic assessment, applies for year 2019.

6 Discussion

The obtained results are discussed in the following chapter, focusing on the validity of the assumptions and methodology. The discussion is divided into two parts. The first part is focusing on the integration of electric plasma calcination with steam slaking while the focus of the second part is on the CO_2 utilisation of the pure carbon dioxide stream resulting from the integration of the new calcination technology.

6.1 Electric Plasma Calcination

From the results presented in Section 5.1, replacing the lime kiln with the electric plasma calcination with steam slaking is beneficial for both investigated mills in terms of energy efficiency and total annualised costs.

For the chosen process design of the electric plasma calcination with steam slaking presented in Section 2.2, a temperature of 200 °C was chosen for the slaking reaction between steam and calcium oxide. This temperature was a compromise between reaction time and the potential to produce low pressure steam. A higher temperature increases the reaction time but also increases the possible steam pressure that can be utilised. This balance has not been studied in detail in this report so more studies should be performed to find an optimal reaction temperature.

Another key assumption is the factor of calcium oxide needed per ADt pulp produced. The assumption made is reasonable but there are indications of mills using less calcium oxide per ADt pulp than what has been assumed throughout this work. By reducing the amount of calcium oxide, the energy consumption of the reburning unit will be reduced as well as the carbon dioxide emissions. With a reduced mass flow, smaller equipment might be needed which reduces the cost. Furthermore, as the energy consumption of the reburning process decreases, more electric power would be available for use for a potential CO_2 utilisation technology. By changing the calcium oxide to ADt pulp factor, the scaling of the concept is affected, why this is an essential parameter to further investigate.

From the split-GCC analysis in Section 5.1.2 it was found that the electric plasma calcination with steam slaking has the potential to reduce the hot and cold utility demand for the two mills. These results are the ideal savings that can occur, and are not necessarily the real utility savings. This is due to the fact that it is not always feasible to get a perfect heat integration at an acceptable investment cost and heat losses often occur as total insulation is difficult to achieve.

A more realistic approach to integrate the new calcination technology is presented in Section 5.1.3 where the integration of a steam cycle was investigated. Värö can produce approximately 5 MW more electricity due to the energy from the electric plasma calcination with steam slaking. In this approach, the hot utility that results from the electric plasma calcination is used to replace hot utility from the boilers while that utility is used to generate electricity instead. These results are also ideal but can give a better indication of how Värö can utilise the energy from the electric plasma calcination. With the same approach, Iggesund has an excess of low-pressure steam that cannot be used to generate electricity as Iggesund only has a back-pressure turbine that cannot expand the steam

any further. It could be that if Iggesund invests in the electric plasma calcination with steam slaking, they might also consider other investments, such as a condensing turbine. If Iggesund does not invest in a condensing turbine, then the choices are either to use the excess steam to heat the district heating network or to reduce the load of the bark boiler and thus also reduce the electricity production. The most profitable option depends on the prices of bark, electricity and district heating.

When looking at the two different system boundaries for the electricity production, it can be seen that the Swedish electricity mix has a low level of greenhouse gas emissions while Nordpool has significantly higher levels. As the two investigated mills use biofuels that have been assumed to have net zero emissions, replacing the lime kilns with the electric plasma process will result in increased emission as the change in electricity production will generate emissions. However, if the saved biofuels in turn replace fuels of fossil origin, there will be a net reduction of emissions that would have a positive contribution to mitigate climate change.

Combusting biomass and biofuels has been assumed to have net zero emissions. This is because the forests that are cut down are replanted and the trees that grow up absorb the carbon dioxide that is released from the combustion. There are some emissions from transportation and possible refining that have not been taken into account in this work to simplify the calculations. These upstream emissions are usually small compared to the energy that the fuels contain and thus only makes a marginal difference.

It could be discussed how the emissions from electricity should be allocated for electricity consumed in Sweden. This is why both Nordpool and the Swedish electricity mix were compared. The results differ substantially and affect the advantages of the electric plasma calcination process. This report has only highlighted the differences between the energy market conditions and shown the importance of system boundaries. There is a trend that the average emissions for Nordpool will be reduced as more low emitting technologies are installed and high emitting technologies are decommissioned. This means that the Nordpool emissions in the future might be considerably lower than they are at present and the different system boundaries will give more similar results.

The two fuels natural gas and fuel oil were also studied as they are commonly used in lime kilns internationally. For both the Swedish electricity mix and for Nordpool, the electric plasma calcination process lead to decreased emissions. This shows that when replacing fossil fuels with electricity, the electricity production does not need to be totally emission free to have a positive impact.

The impact on the electricity system if a large-scale implementation of the electric plasma calcination technology and potentially also electro-fuel production should be realised has not been analysed. If theses implementations should occur, the electricity demand would increase and that could lead to higher emissions per produced kWh electricity as electricity production on the margin with higher emissions could be needed. These potentially higher emissions are likely short term as the base load of the electricity system will be adjusted to handle this new constant load.

Many of the costs presented are based on assumptions and estimations taken from different sources. Also, the prices on fuels and electricity have a tendency to fluctuate strongly depending on several aspects which can greatly affect the total costs, which is shown by the sensitivity analyses in Section 5.1.4. This means that it is difficult to estimate how the costs will be in the future. Also, as some of the equipment that are used are novel technologies, the prices of those might decline when they become more established and the production becomes more commercial. But as the total annualised costs for the two technologies are in the same order of magnitude, it indicates that electric plasma calcination will be a cost competitive alternative to the lime kiln.

For Värö it was found that around 81 000 tonnes of wood per year would be saved when replacing the lime kiln with the electric plasma calcination. As Värö uses wood residues from the nearby sawmill, these 81 000 tonnes that are not burnt must be handled on the site in a different way, which could be challenging. For Iggesund that saves 18 000 tonnes of tall oil pitch each year, it could be easier to handle the saved oil as the infrastructure for oil products is already well built due to the usage of fossil oil. The prices for the fuels are also an important factor. Sawdust is very cheap and could be difficult to handle and sell with a profit, while tall oil pitch has a higher market value and therefore it is a more interesting option to sell. One other option for Värö would be to invest in a biomass boiler and burn the sawdust to generate electricity and heat for district heating.

For all the calculations and assumptions made, it was assumed that no other rebuilding or investments are done except the ones linked to the lime kiln being replaced. It could be that the mills invest in other areas in the same time as the lime kiln is replaced that would remove some of the limitations that have occurred in this work. A possible investment is for instance a condensing turbine for Iggesund.

In this study, one proposed design of the electric plasma calcination process has been used. But there are other process designs that have been proposed in literature. Other designs will probably give different results and these designs should also be investigated so that the optimal process can be found.

6.2 Carbon Dioxide Utilisation

Provided that the proposed new calcination technology is implemented to the mill, a resulting, almost pure, biogenic stream of carbon dioxide is obtained. There are a few different options for utilisation of this carbon dioxide as presented in Section 2.3.1. Two options for CO_2 utilisation were further investigated, the production of electro-fuels and CO_2 utilisation for lignin extraction via the LignoBoost process. It is assumed that one of these technologies could be implemented along with the electric plasma calcination technology and therefore, the two options can be compared against each other.

6.2.1 Electro-fuels

The resulting pure stream of carbon dioxide can be used as raw material for production of either methane or methanol, fuels that could potentially replace corresponding fuels of fossil source in other sectors, such as the transport sector. The available electric power capacity for electro-fuels was shown to be the limiting factor for the electro-fuel production which results in that only a share of the available carbon dioxide could be used

for electro-fuel synthesis. These results are based on reasonable assumptions of available grid capacity of today. By grid capacity increase measure, larger shares of the available carbon dioxide may be utilised.

The integration of electro-fuel synthesis will initially result in increased greenhouse gas emissions resulting from the increased electric power demand. However, if the produced electro-fuels replace fuels of fossil origin, there is great potential to decrease the net emission of greenhouse gases to the atmosphere in the overall system. It is shown that the emissions of the electro-fuels are highly dependent on the electricity mix. The electro-fuel emissions are calculated for the average electricity mix of Sweden as well as for Nordpool, including the Nordic and Baltic countries. It is found that for the Swedish electricity mix, with low emissions, there is great potential to achieve net emission reductions by replacing fossil fuels with electro-fuels. For electro-fuels produced by electricity with Nordpool average emissions on the other hand, the emissions from the electro-fuels are similar to those of fossil fuels. The electro-fuel emissions are also highly dependent on the hydrogen source. In this work, solely hydrogen production by electrolysis of water is considered. There are other possible hydrogen sources which is less electric energy demanding but perhaps of fossil origin. In the case of electro-fuel synthesis is performed with other hydrogen sources, possible net emission reductions cannot be guaranteed.

The heat integration possibilities of the electro-fuel production process with the existing mills was investigated with varying results. It was found that the temperature of the electrolyser is an essential parameter for a beneficial heat integration as the process specifications at existing mills vary. However, the most developed and technical mature electrolysis process operate in a temperature range of 60-90 °C. Electrolysis at temperatures outside this range is less technical mature. To achieve successful heat integration of the electro-fuel synthesis at Värö mill, an electrolysis temperature of 50 °C or below had to be used, provided that no other process modification is done. The low-temperature electrolysis may cause difficulties to implement and could significantly increase the cost for electro-fuel synthesis.

The economic performance of producing electro-fuels was investigated by calculating the production cost of the electro-fuels and comparing that to the market price of corresponding biofuels. This was done for two scenarios, with the electricity price of today and for a future optimistic scenario with lower electricity prices. It is found that the production costs of electro-fuels were in the same order of magnitude as the market price of corresponding biofuels. For a future optimistic scenario with lower electricity prices there were even indications that the production cost could be lower than the current market price, which would result in a profitable process. However, there are several uncertainties in these estimations. The electro-fuel production process is not a commercial concept on a large scale. Cost estimations of equipment vary in large ranges. The estimated future electricity price is very low and is perhaps not realistic as an average price. It is however reasonable for a scenario with a domination of renewable energy sources and favourable weather conditions. It was assumed that the oxygen that is co-produced with the hydrogen in the electrolyser could be sold for an income which reduces the production cost of the electro-fuel, but the market for oxygen is uncertain. The quantities of oxygen produced if multiple mills implement this technology may exceed the market demand and cause the prices to fall, which would result in higher production costs than estimated in

this work. A successful heat integration of the electro-fuel synthesis with the existing mill could result in cost reductions by energy savings. Possible cost reductions by heat integration were not considered in the cost estimations in this work but could result in electro-fuel production cost reductions.

As more sectors and industries aim for decreased greenhouse gas emissions, it is possible that the demand for biofuels will increase in the future. A growing demand could eventually result in a increased market price for the biofuels, making the electro-fuel production more profitable. It is also possible that higher taxes of fossil fuels are introduced, making the biofuel option more profitable in the future.

6.2.2 LignoBoost

The other investigated option for CO_2 utilisation is extraction of lignin via the LignoBoost process. The LignoBoost process was sized to achieve maximum lignin extraction without changing the power balance or endanger the operation of the recovery boiler at the investigated mills. It was found that the limiting parameter for lignin extraction was the maximum lignin extraction rate that does not endanger the operation of the recovery boiler for both investigated mills. As for the electro-fuel synthesis option, it is not possible to utilise all of the available carbon dioxide for lignin extraction, the share of used carbon dioxide was found to be around 30% for both mills.

The economic potential of producing lignin via the LignoBoost process was investigated similarly to the economic performance of producing electro-fuels by calculating of the production cost of lignin which can be compared to market price estimations of lignin. Similarly, to the electro-fuel synthesis there are several uncertainties in the economic assumptions. The investment cost of the LignoBoost concept was based on a commercial LignoBoost plant. The estimated size of the LignoBoost plant for Iggesund is similar to the existing commercial LignoBoost plant, why these cost estimations could be more accurate. The size of the LignoBoost plant at Värö is almost twice the size of the existing commercial plant. Although the economy of scale has been accounted for, a LignoBoost plant of this size has never been built why unforeseen costs might occur. Furthermore, the market for lignin products is very limited since there are few commercial lignin extraction processes. The market price estimation is therefore more of an indication and could change if the market for lignin or lignin products grow.

6.2.3 Comparison

It was found that neither of the two investigated CO_2 utilisation options are able to utilise all of the carbon dioxide produced in the calcination process, but that different factors limit the utilisation processes. The electro-fuel synthesis is limited by the electric power capacity and could be extended by investments in capacity increase measures of the electricity grid. The LignoBoost process however is limited by the maximum lignin extraction rate to not endanger the operation of the recovery boiler. The share of the available CO_2 that can be used for lignin extraction can therefore not exceed about 30% for the two investigated mills. With respect to the aspect of potential to utilise the whole share of CO_2 available, the production of electro-fuels is the preferable option. The possibility to implement both electro-fuel synthesis and lignin extraction via the LignoBoost concept is theoretically possible, considering the amount of available carbon

dioxide, but would require grid capacity increase measures.

The economic potential of the CO_2 utilisation options shows promising results, especially for lignin extraction via the LignoBoost concept. However, the technical maturity of the concept is low and the market for lignin or lignin products is uncertain. The economic potential of electro-fuel production is highly dependent on the electricity price and could potentially be optimised by running the electro-fuel production only at times with low electricity prices.

The emissions of electro-fuel production are associated with the emissions from the electricity production and the hydrogen source while the emissions from the lignin extraction via LignoBoost process are solely dependent on the emissions associated with the electricity production. The environmental benefit of both CO_2 utilisation options however is that they avoid emissions from the fuels that they potentially will replace. The market for biofuels is growing and the demand for bio-methane and bio-methanol is expected to increase. The lignin market on the other hand is not as widespread and more uncertain. The investment in electro-fuel production as the CO_2 utilisation method could thus be a more certain option.

The resulting CO_2 could also be utilised by other possible CO_2 utilisation options presented in Section 2.3. By utilising the carbon dioxide on-site in the pulp bleaching process, the use of other acids such as sulphuric acid could potentially be reduced. The option of utilising the excess carbon dioxide for CCS could be an option in the future if certificates, tariffs or similar systems are in place to ensure a revenue for the avoided emissions of CO_2.

7 Conclusions and Future Work

The integration of electric plasma calcination with steam slaking into existing pulp and paper mills can result in reduced energy use in the overall process. The implementation of the new calcination concept would require the input 23.2 MW of electricity at Iggesund and 39.7 MW at Värö. However, it was found that the theoretical utility demand of the mills can be decreased with 18.6 MW of hot utility and 3.0 MW of cold utility at Iggesund and 26.5 MW of hot utility and 2.7 MW of cold utility at Värö by heat integration of the electric plasma calcination concept. The emission of greenhouse gases can be significantly reduced provided that the electricity production is fossil free and that the saved biomass is used to replace fuels of fossil origin. Furthermore, integration of electric plasma calcination with steam slaking results in a pure stream of biogenic carbon dioxide which enables cost-effective further utilisation. The production of electro-fuels and extraction of lignin via the LignoBoost process was further investigated with promising results. The production capacity of electro-fuels was found to be restricted by the available grid capacity and was calculated to 6.5 kt/yr of methane or 15.2 kt/yr of methanol at Iggesund and 28.4 kt/yr of methane or 66.3 kt/yr of methanol at Värö. The production costs were found to be in the same order of magnitude as the current market prices for equivalent biofuels. The LignoBoost production capacity was found to be limited by the maximum lignin extraction rate and was calculated to 54.7 kt/yr for Iggesund and 93.7 kt/yr for Värö. Both investigated utilisation options have the potential to contribute to further net emission reductions and possibly energy savings by replacing fuels of fossil origin in other sectors such as the transport sector.

The techno-economic performance of the new calcination concept was investigated and compared to a re-investment in a conventional lime kiln. In addition to the previously mentioned benefits of the electric plasma calcination concept, the economic assessment indicates investment costs in the same order of magnitude as the cost of re-investment in the conventional calcination concept. Although there are great uncertainties in the estimations, it is likely that the costs decrease as the technology becomes more mature and commercial.

It is concluded that the implementation of electric plasma calcination with steam slaking has a great potential to achieve energy savings and emission reductions when integrated into existing pulp and paper processes. However, the large quantities of saved biomass that is no longer combusted in the lime kiln need handling and further refinement before they can be utilised in other sectors. The logistics of handling the large quantities of biomass could be a challenge.

Further investigations should be done on the external utilities of the plant such as bark boiler load variation that could lead to saved biomass or increased electricity production, and extension of the electricity grid investments. Other interesting aspects for further research are other process configurations and possibly higher reaction temperatures for the slaking reaction which could enable high-temperature heat recovery. Different ratios of calcium oxide to ADt pulp could also be further investigated as it would have an impact of the scaling of the concept. The implementation of electric plasma calcination along with the conventional lime kiln as a capacity increase measure could be further investigated as a transition from the conventional to the new technology.

For the CO_2 utilisation options, it could be interesting to further investigate future energy market scenarios to investigate the profitability of electro-fuel or lignin production. The possibility to run the CO_2 utilisation process only at times when the electricity price is low could be investigated both as a profitability measure for the mill but also as a demand side management for the electricity system.

References

[1] Energiläget 2017. Eskilstuna: Swedish Energy Agency; 2017. Available from: www.energimyndigheten.se/statistik/energilaget/.

[2] The Swedish climate policy framework [Internet]. Bromma: Ministry of the Environment and Energy; 2018. Available from: www.government.se/information-material/2018/03/the-swedish-climate-policy-framework/.

[3] Ek M, Gellerstedt G, Henriksson G. Pulp and paper chemistry and technology. Volume 2, Pulping chemistry and technology. Berlin: Walter de Gruyter; 2009.

[4] Zero Carbon Industry Plan: Electrifying Industry. Beyond Zero Emissions; 2018. [cited 2020-05-05]. Available from: www.bze.org.au/electrifying-industry-2018/.

[5] Bajpai P. In: Pulp and Paper Industry: Chemical Recovery. Elsevier; 2017. p. 1–23. Doi: 10.1016/b978-0-12-811103-1.00001-2.

[6] Bjotveit S, Gordon A, Ohlsson K, Rutqvist E, Williamsson E, Westermark M, et al. Förstudie : Eldriven kalcinering. Stockholm: Royal Institute of Technology; 2003. Available from: www.carnot.se/pdf/KTH-rapporten,Eldrivenkalcinering.pdf.

[7] Lundqvist R. Ångsläckning av kalk - kinetik och teknik Ny energieffektiv teknik för släckning av mesakalk i sulfatmassaindustrin. Stockholm: Värmeforsk Service AB; 2008.

[8] CemZero - För en klimatneutral cementtillverkning [Internet]. Cementa AB;. [cited 2020-01-30]. Available from: www.cementa.se/sv/cemzero.

[9] Jannasch AK. Integration of the electro-fuel concept in pulp and paper industry for a future electricity system in balance and a sustainable energy system with minimal carbon foot print. Lund: RISE Energy and Circular Economy; 2019. Available from: www.energimyndigheten.se/forskning-och-innovation/projektdatabas/sokresultat/?projectid=26421.

[10] PROCEL Opportunities for decarbonisation of industrial processes through increased electrification; 2018. [cited 2020-02-21]. Available from: http://research.chalmers.se/en/project/7264.

[11] Pettersson K, Mahmoudkhani M, von Schenck A. Opportunities for biorefineries in the pulping industry. Systems Perspectives on Biorefineries. 2012;Björn A. Sandén (ed.):48–58.

[12] Britt KW. Papermaking - Processes for preparing pulp. Encyclopeadia Britannica [Internet];. [cited 2020-01-22]. Available from: www.britannica.com/technology/papermaking/Processes-for-preparing-pulp.

[13] Zeman F. Energy and material balance of CO_2 capture from ambient air. Environ Sci Technol. 2007;41(21):7558–7563. Doi: 10.1021/es070874m.

[14] Olsson D. Optimering av kalkcykeln på Iggesunds bruk med avseende på specifik energianvändning avseende på specifik energianvändning [master's thesis on the Internet]. Swedish University of Agricultural Sciences. Uppsala; 2020. Available from: `http://stud.epsilon.slu.se/15335/`.

[15] Nytt sätt att bränna kalk minskar klimatpåverkan och spar pengar. Movexum; 2019. [cited 2020-01-29]. Available from: `www.movexum.se/nytt-satt-att-branna-kalk-minskar-klimatpaverkan-och-spar-pengar/`.

[16] Criado YA, Alonso M, Abanades JC. Kinetics of the CaO/Ca(OH)2 hydration/dehydration reaction for thermochemical energy storage applications. Ind Eng Chem Res. 2014;53(32):12594–12601. Doi: 10.1021/ie404246p.

[17] Lignin. Encyclopeadia Britannica [Internet];. [cited 2020-03-16]. Available from: `www.britannica.com/science/lignin`.

[18] Tomani P. The lignoboost process. Cellul Chem Technol. 2010;44(1):53–58.

[19] The next generation LignoBoost – tailor-maid lignin production for different lignin bioproduct markets. Valmet. 2017;May 26:1–8.

[20] Nilsson K. Raising the efficiency of black liquor lignin extraction [master's thesis on the Internet]. Lund University; 2017.

[21] Ludmila H, Michal J, Andrea Š, Aleš H. Lignin, potential products and their market value. Wood Res. 2015;60(6):973–986.

[22] What is CCS? [Internet]. The Carbon Capture & Storage Association (CCSA);. [cited 2020-02-21]. Available from: `www.ccsassociation.org/what-is-ccs/`.

[23] Dadhich P, Dooley J, Fujii Y, Hohmeyer O, Riahi K. Cost and economic potential. IPCC Spec Rep Carbon Dioxide Capture Storage. 2005;p. 341–362.

[24] Lachapelle RC, inventor; CANADAIN LIQUID AIR Ltd, assignee. Process for preparing a paper pulp using carbon dioxide as an acidifying agent for a bleached pulp; United States patent US5139613A 1992-08-18. Available from: `http://patents.google.com/patent/US5139613A/en`.

[25] Speed up your success – with gas applications. Innovative solutions in papermaking [pamphlet on the Internet]. Stadl-Paura,: Linde Gas GmbH;. Available from: `www.linde-gas.de/de/images/Speed_up_your_success_with_gas_applications_Innovative_solutions_in_papermaking_en_tcm565-110522.pdf`.

[26] Pulp & Paper Brownstock Washing [Internet]. Praxair;. [cited 2020-03-04]. Available from: `www.praxair.com/industries/pulp-and-paper/washing`.

[27] Kemp IC. Pinch analysis and process integration : a user guide on process integration for the efficient use of energy. 2nd ed. Oxford: Butterworth-Heinemann; 2007.

[28] Gustafsson MS. Industrial excess heat for district heating - Comparison of potentials from top-down and bottom-up studies for energy-intensive process industries [Master's thesis on the Internet]. Gothenburg: Chalmers University of Technology; 2013. Available from: http://hdl.handle.net/20.500.12380/182322.

[29] Glader K. Measures for increased energy efficiency at Iggesund mill: Pinch analysis of the pulp production lines at a paperboard mill [Master's thesis on the Internet]. Gothenburg: Chalmers University of Technology; 2011. Available from: https://hdl.handle.net/20.500.12380/144146.

[30] Pedersén A, Larsson A. Energy Efficiency Study at a Softwood Kraft Pulp Mill [Master's thesis on the Internet]. Gothenburg: Chalmers University of Technology; 2017. Available from: http://hdl.handle.net/20.500.12380/250873.

[31] Södra har beslutat om energiinvesteringar i Värö [Internet]. Södra;. Cited 2020-05-18. Available from: www.sodra.com/sv/se/om-sodra/pressrum/pressmeddelanden/sodra-har-beslutat-om-energiinvesteringar-i-varo/.

[32] Energiföretagen. Energiåret 2017 - Energibranschens klimat– och miljöpåverkan [Internet]. Stockholm: Energiföretagen; 2018. Available from: www.energiforetagen.se/globalassets/energiforetagen/statistik/energiaret/energiaret2017_miljo_vers180904.pdf.

[33] Vägledning i Klimatklivet-Beräkna utsläppsminskning [Internet]. Stockholm: Swedish Environmental Protection Agency; 2018.

[34] Quaschning V. Specific carbon dioxide emissions of various fuels [Internet]. Berlin;. [cited 2020-02-25]. Available from: www.volker-quaschning.de/datserv/CO2-spez/index_e.php.

[35] Weidner E, Rajendran VK, Breitkreuz K, Kraft A, Maga D, Brucart MF. Analysis of The European Crude Tall Oil Industry – Environmental Impact, Socio- Economic Value & Downstream Potential [Internet]. Oberhausen: Fraunhofer Institute for Environmental, Safety and Energy Technology UMSICHT; 2016. May.

[36] Grahn M, Jannasch AK. Electro-methane: integration aspects and cost estimates. In: 5th Int. Conf. Renew. Energy Gas Technol; 3-4 May 2018. Toulouse; 2018. p. 4.

[37] Natural gas price statistics - Statistics Explained [Internet]. Eurostat; 2019. [updated 2020-01-28; cited 2020-03-11]. Available from: www.ec.europa.eu/eurostat/statistics-explained/index.php?title=Natural_gas_price_statistics.

[38] Pris på eldningsolja (heating oil) - Aktuellt terminspris med diagram [Internet];. [cited 2020-03-11]. Available from: www.ravarumarknaden.se/pris-eldningsolja-heating-oil-diagram-ho/.

[39] Trädbränsle och torvpriser, per kvartal exklusive skatt, från och med 1993, kronor/MWh fritt förbrukare, löpande priser. Swedish Energy Agency;. [cited 2020-03-20]. Available from: http://pxexternal.energimyndigheten.se/pxweb/sv/Trädbränsle-ochtorvpriser/Trädbränsle-ochtorvpriser/EN0307_1.px/.

[40] Hannula I, Arpiainen V. Light olefins and transport fuels from biomass residues via synthetic methanol: performance and cost analysis. Biomass Convers Biorefinery. 2015;5(1):63–74. Doi: 10.1007/s13399-014-0123-9.

[41] Fuels - Higher and Lower Calorific Values;. [cited 2020-02-25]. Available from: www.engineeringtoolbox.com/fuels-higher-calorific-values-d_169.html.

[42] Lozowski D. Economic Indicators. Chem Eng. 2009;116(13):63–64.

[43] Economic indicators. Chem Eng. 2020;127(4):68.

[44] Valutor - aktuella valutakurser och valutaomvandlare [Internet]. Stockholm: Dagens Industri;. [cited 2020-03-10]. Available from: www.di.se/valutor.

A Electricity Demand of Electric Plasma Concept

The electricity demand of the electric plasma concept was interpolated from values calculated by reference [6] and were used for calculations of the electricity demand is presented in Table 31.

Parameter	Unit	Value
Electric plasma generator power demand	[kW]	14 049
Compressor 1 power demand	[kW]	368.9
Compressor 2 power demand	[kW]	126.9
Mass flow of calcium oxide (CaO)	[kg/s]	2.657

Table 31: Electric plasma calcination specifications taken from Bjotveit et al. [6].

The power demand of the electric plasma calcination concept consist of the power demand of the electric plasma generator plus the power demand of the two compressors used to compress the carbon dioxide needed in the plasma generator. A specific power demand in terms of kJ per kg of calcium oxide was calculated by dividing the power demand with the mass flow of calcium oxide. The results are presented in Table 32.

Equipment	Power demand [kJ/kg CaO]
Electric plasma generator	5 287.5
Compressor 1	138.8
Compressor 2	47.8

Table 32: Interpolated specific electricity demand of the electric plasma calcination concept.

B Stream Data

Below is the stream data for the original process for the two investigated mills. The streams that are related to the lime kilns, and thus removed when using the electric plasma calcination process, are marked in grey. The stream data originates from the work by Pedersén et al. [30] for Värö and Gustafsson [28] for Iggesund. In these tables, a comma (,) is used as decimal separator.

B.1 Värö

Type	T_{start} [C]	T_{target}[C]	Q[kW]	ΔT/2[K]	Process part	Decription/Comment
Cold	95	120	10000	3,5	Cooking	White liquor to digester and impbin
Cold	145,1	145,2	89600,0	0,5	Evaporation	Evaporator plant LP process steam demand
Cold	185,0	185,1	19200,0	0,5	Evaporation	Super concentrator MP process steam demand
Cold	32,9	93,3	34700,0	0,2	Condensate	Feed water from BB filter 1-3 to HX
Cold	25,1	77,5	12400,0	0,0	Condensate	Feed water from VKT-tank to mixing point (mix)
Cold	77,5	123,0	34600,0	2,5	Condensate	Feed water to feed water tank
Cold	145,1	145,2	11300,0	0,5	Condensate	Feed water tank LP process steam demand
Cold	47,4	64,5	0,0	2,5	Local heating	FoU facility heating
Cold	64,1	80,7	400,0	2,5	Local heating	Office and workshop heating
Cold	139,8	144,1	3000,0	2,5	Recovery boiler	Feed water from feed water tank to economizer 1
Cold	221,3	221,4	5700,0	0,5	Recovery boiler	Feed water heating between economizer 1 and 2
Cold	120	140	4000	3,5	Cooking	White liquor to digester
Cold	49,9	132,2	500,0	7,0	Recovery boiler	Weak gas to GA-cyclone
Cold	6,2	43,1	2400,0	8,0	Recovery boiler	Outdoor air to recovery boiler
Cold	44,7	190,0	5500,0	8,0	Recovery boiler	Primary air to recovery boiler
Cold	44,7	168,1	3800,0	8,0	Recovery boiler	Secondary air to recovery boiler
Cold	31,0	179,9	4400,0	8,0	Recovery boiler	Quaternary air to recovery boiler
Cold	128,7	136,2	1500,0	3,5	Recovery boiler	Strong black liquor to nozzles
Cold	5,9	51,6	10900,0	2,5	Recovery boiler	Production of warm water, recovery boiler house
Cold	6,2	111,3	10500,0	8,0	Wood handling	Air to bark dryer
Cold	44,2	50,4	200,0	2,5	Wood handling	Deicing/wood handling water
Cold	94,2	110,7	18200,0	0,0	Saw mill	Heat demand timber kilns
Cold	145	150	1000	3,5	Cooking	Liquor mix to digester
Cold	77,4	95,0	8700,0	0,0	Saw mill	Heat demand sawdust dryer
Cold	46,5	65,0	700,0	2,5	Tomato farm	Heat demand tomato farm
Cold	185,0	185,1	2600,0	0,5	Bleaching	Bleaching step 2 MP process steam demand
Cold	185,0	185,1	5700,0	0,5	Bleaching	Bleaching step 4 MP process steam demand
Cold	5,9	67,1	17200,0	2,5	Bleaching	Warm water production for filter 4
Cold	87,0	90,9	2100,0	3,5	Bleaching	BB2 filtrate from filtrate tank 2 to screw feeder
Cold	86,8	90,7	2100,0	3,5	Bleaching	BB4 filtrate from filtrate tank 4 to screw feeder
Cold	5,9	25,0	100,0	2,5	Causticizing	Lukewarm water production for polymer preparation
Cold	5,9	69,5	22000,0	2,5	Causticizing	Hot water production for lime filter
Cold	185,0	185,1	200,0	0,5	Oxygen bleaching	O2 reactor MP process steam demand
Cold	130	140	10000	3	Cooking	Weak black liquor 2 from digester to impbin
Cold	6,2	84,7	800,0	8,0	Bark boiler	Air to bark boiler
Cold	6,2	121,5	700,0	8,0	Bark boiler	Air to kablitz in bark boiler
Cold	145,1	145,2	1000,0	0,5	Chemical plant	Chemical plant LP process steam demand
Cold	5,9	24,9	6100,0	2,5	Chemical plant	VKT production
Cold	5,9	55,0	400,0	2,5	Chemical plant	NaOH dilution water
Cold	6,1	30,0	1200,0	8,0	Paper room	Air to paper room 1
Cold	6,1	23,0	1600,0	8,0	Paper room	Air to paper room 2
Cold	6,1	25,0	2400,0	8,0	Paper room	Air to pulp cooling
Cold	23,0	98,9	5100,0	8,0	Paper room	Air feed to drying machine
Cold	145,1	145,2	48500,0	0,5	Paper room	Drying machine LP process steam demand
Cold	178,1	178,2	17000	0,5	Cooking	Digester MP process steam demand
Cold	5,9	49,9	19900,0	2,5	Paper room	Water 1 for paper room
Cold	49,9	61,7	3600,0	2,5	Paper room	Water 2 for paper room
Cold	145,1	145,2	3000,0	0,5	Paper room	Paper room LP (MP) process steam demand
Cold	6,1	155,6	14900,0	6,3	Paper room	Air feed to cyclone dryer
Cold	40,9	95,8	16200,0	2,5	District heating	District heating Varberg
Cold	145,1	145,2	9000	0,5	Cooking	Impbin LP process steam demand
Cold	67,2	108,3	14000,0	1,9	Evaporation	KLB from tank to stripper column
Cold	70,8	103,3	4700,0	3,5	Evaporation	KLS from tank to stripper column
Cold	145,1	145,2	600,0	0,5	Evaporation	MeOH column LP process steam demand

Type	T_{start} [C]	T_{target}[C]	Q[kW]	ΔT/2[K]	Process part	Decription/Comment
Hot	150	130	4000	3,5	Cooking	Weak black liquor 1 from digester to flash
Hot	63,8	63,7	99000,0	2,0	Evaporation	Surface condenser evaporation plant
Hot	77,2	77,1	1900,0	0,5	Evaporation	Evaporation vapor from line 5
Hot	92,9	92,8	1200,0	1,5	Evaporation	Evaporation vapor from line 3
Hot	115,9	115,8	1800,0	2,0	Evaporation	Evaporation vapor from line 1C
Hot	102,0	101,9	1900,0	2,0	Evaporation	Evaporation vapor from line 2, 2B
Hot	112,3	33,2	34700,0	0,2	Condensate	Fresh condensate to BB filter 1-3
Hot	93,3	77,5	12400,0	0,0	Condensate	Feed water from HX to mixing point (mix)
Hot	85,4	36,8	20900,0	3,5	Recovery boiler	Cooling to dissolving tank scrubber
Hot	210,3	140,0	23000,0	7,0	Recovery boiler	Flue gases from recovery boiler
Hot	143,6	143,5	600,0	2,0	Recovery boiler	Blowdown steam from steam drum
Hot	130	110	10000	3,5	Cooking	Weak black liquor 2 from digester to flash
Hot	89,9	37,0	21800,0	3,5	Bleaching	Alkaline effluent from bleaching to biocleaning
Hot	87,0	37,0	22900,0	3,5	Bleaching	Acid effluent from bleaching to biocleaning
Hot	75,0	49,9	1900,0	7,0	Bleaching	Weak gas from bleaching
Hot	100,6	85,0	6200,0	3,5	Causticizing	Green Liquor
Hot	34,8	15,5	2000,0	0,0	Turbine hall	Turbine cooling TB31
Hot	151,3	151,2	400,0	0,5	Turbine hall	Leakage steam from TB31
Hot	25,5	25,4	39400,0	2,5	Turbine hall	Condenser TB31
Hot	23,5	19,9	2000,0	0,0	Turbine hall	Turbine cooling TB21
Hot	99,0	98,9	400,0	0,5	Turbine hall	Leakage steam from TB21
Hot	50,0	25,1	300,0	0,0	Bark boiler	Bark boiler roster cooling
Hot	105	90	18000	3,5	Cooking	Weak black liquor from flash to evaporation tank
Hot	38,7	32,1	100,0	3,5	Chemical plant	Sulfuric acid
Hot	69,3	54,8	0,0	3,5	Chemical plant	Tall oil
Hot	97,3	66,4	4400,0	7,0	Paper room	Moist air from drying machine step 1
Hot	66,4	64,5	6400,0	7,0	Paper room	Moist air from drying machine step 2
Hot	97,3	66,4	100,0	7,0	Paper room	Moist air from drying machine split to cyclone dryer
Hot	133,7	133,6	1800,0	0,5	Paper room	Flash steam from drying machine and cyclone dryer
Hot	133,6	117,1	1300,0	2,5	Paper room	Flash steam condensate from drying machine and cyclone dryer
Hot	139,8	113,9	1500,0	2,5	Bark boiler	Bark boiler feed water
Hot	105	104,9	6000	2	Cooking	Turpentine gas from flash to primary condenser
Hot	105	60	1000	3,5	Cooking	Turpentine liquid to decanter
Hot	160	140	3000	0	Cooking	Weak black liquor 1 from digester to impbin (mix)
Hot	112,1	84,8	14000,0	1,9	Evaporation	KLR from stripper to tank (stream split)
Hot	112,1	84,8	4700,0	3,5	Evaporation	KLR from stripper to tank (stream split)
Hot	64,6	64,5	3200,0	2,0	Evaporation	MeOH gas from MeOH column
Hot	40,3	40,2	800,0	2,0	Evaporation	MeOH from primary condenser

B.2 Iggesund

Type	T_{start} [C]	T_{target}[C]	Q[kW]	ΔT/2[K]	Process part	Decription/Comment
Cold	61	85	2 200		Digester 3	Warm water production
Cold	138	144	2 016		Digester 3	Cooking circulation
Cold	148	151	1 535		Digester 3	Cooking circulation
Cold	109,2	144,5	4 822		Digester 3	Cooking circulation
Cold	61	92,5	3 900		Digester 3	Warm water production
Cold	122,5	160	4 194		Digester 4	Cooking circulation
Cold	143	148	3 622		Digester 4	Pre impregnation
Cold	61	75	1 200		Digester 4	Warm water production
Cold	61	92	3 500		Digester 4	Warm water production
Cold	45	76	1 698		Digester 4	Black liquor
Cold	10	31	455		Bleaching 4	Chlorine dioxide, pre heating
Cold	75	86,5	2 683		Bleaching 4	Pulp water
Cold	14,7	44	18 800		Evaporation 3	Warm water production
Cold	18	25,5	854		Evaporation 3	warm water production
Cold	40,1	90	5 267		Gas&cond	Cooling after stripper
Cold	14,7	44	18 800		Evaporation 4	Warm water production
Cold	4	76,4	600		Kaust	Warm water production
Cold	42,8	63	36 369		Water	Warm water production
Cold	14,7	45	6 700		Gas&cond	Warm water production
Cold	37	80	5 700		Gas&cond	Cooling of imcondensor
Cold	1	56	3600		KM1	Chemical cleaned water
Cold	1	60	5500		KM2	Chemical cleaned water
Cold	207,1	207,2	7239		MT17	Soot blowing, recovery boiler
Cold	191,6	191,7	822		MT12	Soot blowing, economizer
Cold	191,6	191,7	561		MT12	Bleaching 3
Cold	191,6	191,7	6862		MT12	Digester 4
Cold	191,6	191,7	1542		MT12	Bleaching 4
Cold	191,6	191,7	2212		MT12	KM1
Cold	191,6	191,7	804		MT12	KM2 coating
Cold	191,6	191,7	6166		MT12	TM4
Cold	191,6	191,7	2614		MT12	Unaccounted and losses
Cold	170,4	170,5	1690		MT7	Lime kiln
Cold	170,4	170,5	34874		MT7	KM1
Cold	170,4	170,5	51389		MT7	KM2
Cold	170,4	170,5	1630		MT7	TM4
Cold	170,4	170,5	5291		MT7	Misc, and losses
Cold	143,6	143,7	1095		LT3	Digester 3
Cold	143,6	143,7	1582		LT3	Bleaching 3
Cold	143,6	143,7	1978		LT3	Digester 4
Cold	143,6	143,7	44942		LT3	Evaporation
Cold	143,6	143,7	1030		LT3	Chemical preparation
Cold	143,6	143,7	11840		LT3	The saw mill
Cold	143,6	143,7	4056		LT3	Unaccounted and losses
Cold	15	125	11601		Kraft	Dilution water
Cold	105,37	125	3545		Kraft	Condensate
Cold	144	144	6385		Kraft	Steam feed water tank

Type	T_{start} [C]	T_{target}[C]	Q[kW]	ΔT/2[K]	Process part	Decription/Comment
Hot	99	98	2 200		Digester 3	Turpentine condenser
Hot	108	96	3 900		Digester 3	Weak liquor
Hot	141	140,9	1 398		Digester 4	Vapor from cyclone
Hot	98,1	98	1 200		Digester 4	Turpentine condenser
Hot	111	94	3 500		Digester 4	black liquor
Hot	84	77	1 698		Digester 4	Warm water production
Hot	70	38	455		Bleaching 4	Pulp water
Hot	66	65,9	18 126		Evaporation 3	Evaporation vapor
Hot	65,9	46,6	674		Evaporation 3	Sub cooling of evaporation vapor
Hot	26	24	854		Evaporation 3	Cooling of secondary condensate
Hot	80	78	788		Evaporation 3	Evaporation vapor
Hot	93,4	93	5 267		Gas&cond	Cooling after stripper
Hot	53,1	53	18 000		Evaporation 4	Condensation
Hot	53	33,9	800		Evaporation 4	Sub cooling of evaporation vapor
Hot	85	84,9	600		Kaust	Green liquor cooling
Hot	60	37	4 830		Bleaching 3	Effluent
Hot	60	37	4 562		Bleaching 3	Effluent
Hot	65	37	6 533		Bleaching 4	Effluent
Hot	60	37	4 562		Bleaching 4	Effluent
Hot	72	37	25 521		Bleaching 3	Effluent
Hot	63	20	86275		Water	From scrubber
Hot	80	37	9 486		Water	
Hot	70,1	70	5 978		Gas&cond	Methanol condenser
Hot	70	17	722		Gas&cond	Methanol sub cooling
Hot	100,1	100	5 700		Gas&cond	Imcondenser
Hot	140	105	6 000		Kraft	Flue gas cooling, recovery boiler
Hot	135	100	3 000		Kaust	Flue gas cooling, lime kiln
Hot	88,1	88	3600		KM1	Flash
Hot	90,1	90	5500		KM2	Flash

C Heat of Reaction

The heat of reaction for the lime slaking reaction was recalculated for the temperature at which the reaction occurs within the process (T_2). To account for the changes of heat capacity with temperature, the heat of reaction was calculated according to equation 14.

$$\Delta H_{reac}(T_2) = \Delta H_{reac}(T_{ref}) + \int_{T_{ref}}^{T_2} c_p dT \tag{14}$$

Using the presented equation for calculation of heat of reaction, it is assumed that all reactants enter at the temperature of reaction. The reacting steam does not enter the slaking vessels, in which the reaction occurs, at the reaction temperature which has to be accounted for in the calculations. Thus, the enthalpy of the fictive super heated steam is withdrawn from the heat of reaction. The heat of reaction was calculated using the computational software MATLAB. The MATLAB code for calculation of reaction heat is presented below.

```
%%———CALCULATIONS OF REACTION HEAT FOR STEAM SLAKING OF LIME———%%
% Reaction heat calculation of slaking of burned lime (CaO) with steam
% at other temperatures than the refecrence temperature.
% Temperature differences of CaO and Steam is accounted for.

clc, clear
% Parameter for heat capacity function on the form [CaO H2O CaOH2]
a=[49.95403 30.092 130.8253];
b=[4.887916 6.832514 -82.69216];
c=[-0.352056 6.793435 122.769];
d=[0.046187 -2.53448 -50.3921];
e=[-0.825097 0.082139 -2.513146];

M_CaO=0.056;                % [kg/mol]

T1=25;                      % Reference temperature [C]
T2=200;                     % Reaction temperature [C]
k=T1:T2;
Hr0=-109.18;                % Standard heat of reaction [kJ/mol]
DH=0
DHsteam1=0
DHsteam=0
diff=abs(T1-T2)+1;

for j=1:diff
    t=k(j)+273.15;          % Celcius to Kelvin
    T=t/1000;
```

```
    Cp_CaO=(a(1)+b(1)*T+c(1)*T.^2+d(1)*T.^3+e(1)./T.^2)/1000;
        % Heat capacity [kJ/mol]
    Cp_H2O=(a(2)+b(2)*T+c(2)*T.^2+d(2)*T.^3+e(2)./T.^2)/1000;
        % Heat capacity [kJ/mol]
    Cp_CaOH=(a(3)+b(3)*T+c(3)*T.^2+d(3)*T.^3+e(3)./T.^2)/1000;
        % Heat capacity [kJ/mol]

    dcp=Cp_CaOH-Cp_H2O-Cp_CaO;

    DH=dcp*1+DH;          % Heat of reaction at temperature T2
end

Tw=105;                   % Temperature of reacting steam [C]
kw=Tw:T2;
diffw=abs(Tw-T2)+1;
for n=1:diffw
    tw=kw(n)+273.15;      % Celcius to Kelvin
    Tw=tw/1000;
    Cp_H2O=(a(2)+b(2)*Tw+c(2)*Tw.^2+d(2)*Tw.^3+e(2)./Tw.^2)
        /1000;

    DHsteam=Cp_H2O*1+DHsteam;        % Fictive enthalpy of steam
end
Hreac=Hr0+DH+DHsteam                 % Heat of reaction,
    temperature difference of reactants accounted for [kJ/mol]
Hreac_kg=Hreac/M_CaO                 % Heat of reaction,
    temperature difference of reactants accounted for [kJ/kg CaO]
```

D Physical Properties

Physical properties of important substances are presented in Table 33.

Physical property	Unit	Value	Reference
Molar mass			
Water	[g/mol]	18	
Carbon dioxide	[g/mol]	44	
Calcium oxide	[g/mol]	56	
Calcium carbonate	[g/mol]	100	
Calcium dihydroxide	[g/mol]	58	
Lower heating value			
Lignin	[MJ/kg]	24	[18]
Wood	[MJ/kg]	15.4	[41]
Tall oil pitch	[MJ/kg]	40.6	[35]
Fuel oil	[MJ/kg]	40.6	[41]
Natural gas	[MJ/kg]	48.6	[41]
Methane	[MJ/kg]	50	[41]
Methanol	[MJ/kg]	20	[41]

Table 33: Physical properties.

E Economic Data

The Chemical Engineering Process Cost Index (CEPCI) is an index to show how the costs for process equipment increase over time. The index values that have been used in this report are presented below in Table 34.

Year	2003	2007	2008	2012	2015	2017	2019
Index	402.0	525.4	575.4	584.6	556.8	567.5	607.5

Table 34: The Chemical Engineering Production Cost Index for the years that occur in this report. The values are taken from references [42] and [43].

The currency conversion values used in this report are presented in Table 35.

Conversion	Unit	Value
$ to SEK	[SEK/$]	9.44
€ to SEK	[SEK/€]	10.74

Table 35: Currency conversion [44].

The costs to invest in electric plasma calcination with steam slaking or a lime kiln presented in Bjotveit et al. [6] are shown in Table 36 below. These values was used as a base for calculations, but was rescaled and adjusted.

Equipment electric plasma calcination		Cost [MSEK]
3 Plasma generators á 5 MW		53.1
Cyclone		4.54
Cyclone and reactor		9.08
Heat exchanger CO_2 and steam		1.57
Heat exchanger CO_2 and water		0.71
Heat exchanger CO_2 and water		0.71
Heat exchanger slaker cycle and steam		1.19
Filter CO_2		8.09
Compressor 1 CO_2		3.35
Compressor 2 CO_2		3.35
Dryer and steam slaker		41.3
Invest in electrodes	[MSEK/yr]	2.3
Equiment lime kiln		Cost [MSEK]
Lime kiln		300

Table 36: Total investment costs presented in reference [6] for the electric plasma process and the lime kiln.

In Equation 15 below is the annuity factor described. It is in the form that a investment cost is divided by the annuity factor to get the annualised investment cost. A is the annuity factor, r is the interest rate, and t is the expected lifetime in years.

$$A = \frac{1 - \frac{1}{(1+r)^t}}{r} \tag{15}$$

F LignoBoost

The maximum allowed load reduction of the recovery boiler, with respect to satisfy the mills hot utility demand, was calculated according to equation 12, repeated below.

$$LR_{RB,max} = (L_{RB} + P_P) - (P_L + L_{evap}) - (P_D - P_I) \tag{12}$$

Where $LR_{RB,max}$ denotes the maximum allowed load reduction of the recovery boiler, L_{RB} the current excess load of the recovery boiler, P_D is the power demand of the plant after integration of electric plasma calcination, P_L is the power demand of the LignoBoost process, L_{evap} is the increased steam demand of the evaporation plant, P_P the purchased power capacity of the plant and P_I is the increased power production resulting from the integration of electric plasma calcination.

The current excess load of the recovery boiler, L_{RB}, was approximated as the total hot utility production minus the hot utility demand of the pulp/pulp and paper process and is found graphically using the grand composite curve of the process after electric plasma integration. The utility integration of the electric plasma calcination is done according to the integrated approach described in section 5.1.3, the grand composite curves used for the graphical approach is thus the same grand composit curves as presented in Figure 19 previosly mentioned section. The graphical approach is visualised in Figure 25. The current excess load of the recovery boiler is approximated to 70 MW and 170 MW for Iggesund and Värö respectively.

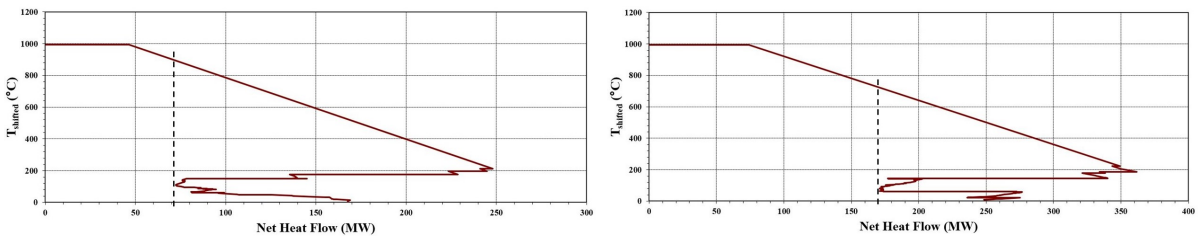

Figure 25: Visualisation of graphical approximation of current excess load of recovery boiler. Iggesund to the left and Värö to the right.

The equation is then solved in an iterative way by guessing the maximum load reduction, $LR_{RB,max}$, available for LignoBoost concept. The corresponding maximum lignin extraction was calculated based on the lower heating value of lignin. From the mass flow of extracted lignin, the power demand of the LignoBoost concept could be calculated as well as the increased load of the evaporation plant. The iterative process is described below.

$$\begin{aligned}
&Guess \quad LR_{RB,max} \\
&m_{lignin} = \frac{LR_{RB,max}}{LHV_{lignin}} \\
&P_L = m_{lignin} \cdot 3.6 \cdot f_{PL} \\
&L_{evap} = m_{lignin} \cdot l_{evap} \\
&newLR_{RB,max} = (L_{RB} + P_P) - (P_L + L_{evap}) - (P_D - P_I)
\end{aligned}$$

Constants	Unit	Value
Lower heating value lignin, LHV_{lignin}	[MJ/kg]	24
Electricity demand LignoBoost process factor, f_{PL}	[MWh/t]	0.4
Load increase evaporation plant factor, l_{evap}	[MJ/kg]	0.9

Table 37: Parameters for recovery boiler load reduction calculations.

G Steam Cycle Integration

For the second approach to steam cycle integration presented in Section 4.2.2, steam is heated in the electric plasma calcination process before being combined with the utility system of the mills. In Figure 26 presented below are the steam production visualised for both mills and the black dashed lines represent the amount of steam that is produced.

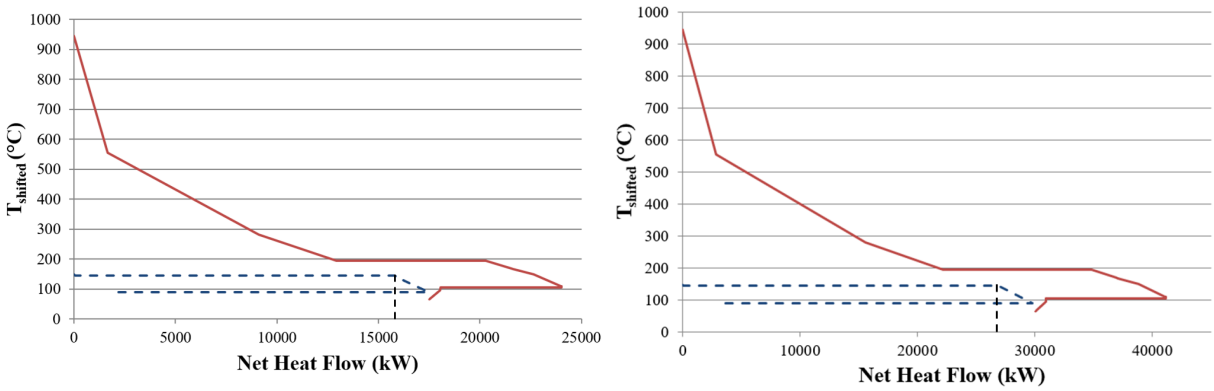

Figure 26: Steam production from electric plasma process for Iggesund (left) and Värö (right). The black dashed line represents the amount of steam delivered to the utility system.

H Heat Integration of Electro-methanol

A similar analysis for methanol syntheses was performed and the results are presented in Figure 27 for Iggesund, and Figure 28 for Värö.

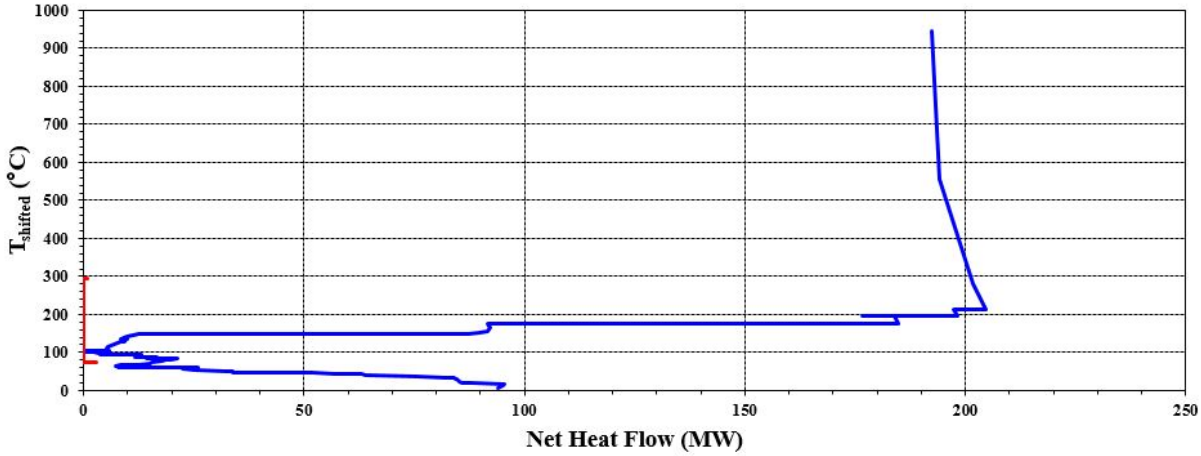

Figure 27: Visualisation of heat integration possibilities of electro-methanol synthesis at Iggesund mill by a split-GCC.

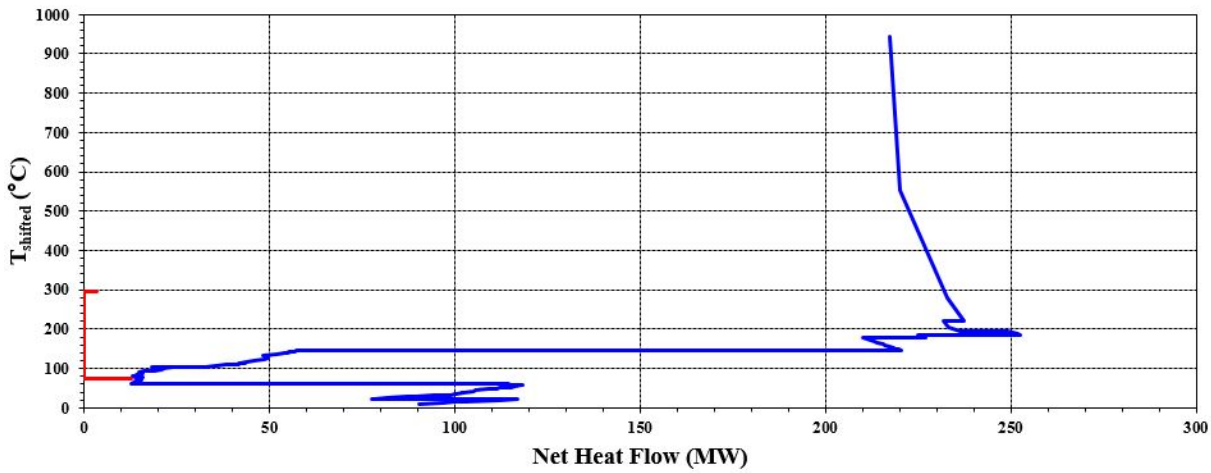

Figure 28: Visualisation of heat integration possibilities of electro-methanol synthesis at Värö mill by a split-GCC.

The stream data used to construct the GCC of electro-methanol synthesis consist of a cold stream representing the heat demand of the electrolyser and a hot stream representing the excess heat from the fuel synthesis reaction. The stream data is presented in Table 38.

T_{start}	T_{target}	Q Iggesund [MW]	Q Värö [MW]	Comment
69.9	70	2.9	12.6	Electrolyser
300	299.9	0.8	3.6	Fuel synthesis

Table 38: Stream data for electro-methanol synthesis.

It is found that the theoretical minimum utility demands are further decreased by integration of electro-methanol synthesis at the integrated pulp and paper mill Iggesund. However, integration of electro-methanol synthesis at the stand alone pulp mill Värö results in an increase of hot utility demand. The results are presented in Table 39.

	Minimum hot utility [MW]	Minimum cold utility [MW]
Iggesund		
Electric plasma integrated	192.5	94.1
After electro-methanol integration	191.7	91.2
Total theoretical savings	0.8	2.9
Värö		
Electric plasma integrated	204.7	77.6
After electro-methanol integration	213.8	77.7
Total theoretical savings	-9.1	-0.1

Table 39: Theoretical utility savings by integration of electro-methanol synthesis.

Once again it is found that the electrolyser temperature at 70 °C interfere with streams connected to the surface condenser at the evaporation part of the plant at Värö. By lowering the electrolyser temperature to 50 °C, a better temperature match is achieved, thus enabling heat integration between the two processes. The results are presented in Figure 29 and Table 40.

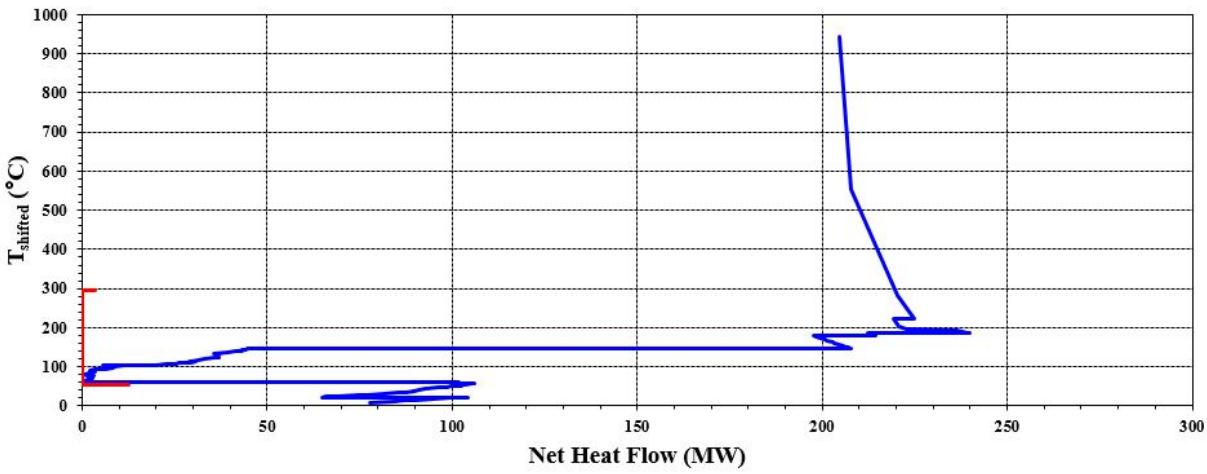

Figure 29: Visualisation of heat integration possibilities of electro-methanol synthesis with an electrolyser temperature of 50 °C at Värö mill by a split-GCC.

	Minimum hot utility [MW]	Minimum cold utility [MW]
Värö		
Electric plasma integrated	204.7	77.6
After electro-methanol integration (50°C)	201.2	65.1
Total theoretical savings	3.5	12.5

Table 40: Theoretical utility savings by heat integration of electro-methanol synthesis with an electrolyser temperature of 50 °C at Värö mill.