

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Biomass Gasification-Based Biorefineries in
Pulp and Paper Mills –
Greenhouse Gas Emission Implications and
Economic Performance

JOHAN ISAKSSON



Industrial Energy Systems and Technologies
Department of Energy and Environment
CHALMERS UNIVERSITY OF TECHNOLOGY
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ABSTRACT

The pulp and paper industry has a great opportunity to take advantage of the experience about forestry and wood handling for production of renewable fuels, chemicals and materials. Residues from forestry, i.e., tops, branches and stubs, have great potential to become a raw material that can replace crude oil or natural gas in many applications. By constructing a gasification-based production facility close to a pulp and paper mill, heat from the gasification process can be utilised in the mill, which replaces fuel for the boiler.

The overall aim of this thesis has been to demonstrate how the pulp and paper industry can help cut global greenhouse gas (GHG) emissions and increase revenues by efficient use of biomass via gasification. In this thesis, the impact of different end products and biomass pretreatment measures connected to integrated gasification processes were evaluated regarding efficiency, GHG emissions and economic performance.

The selection of end product from a gasification-based process (methanol, Fischer-Tropsch crude, synthetic natural gas or electricity, in this thesis) was demonstrated to significantly influence the required biomass input, if the sizing constraint was to replace heat production in the bark boiler. The size varied between approximately 200 MW_{biomass} and 600 MW_{biomass}. The selection of dryer type was shown to significantly impact the economic performance of a process despite its relatively low capital cost in relation to the total capital cost of 100 to 140 million Euros. With the preconditions in this study, thermal pretreatment (torrefaction or pyrolysis) of biomass may be beneficial with biomass-to-biofuel efficiencies reaching 68%.

Assumptions regarding the electricity production sector influence whether an integrated gasification process is better than a stand-alone unit and whether onsite co-generation of electricity should be prioritised in favour of increased yield of biofuels. Sequestration of separated CO₂ has been shown to improve the climatic performance of a system and increase its competitiveness against, e.g., co-firing biomass in a coal power plant, by increasing the potential from approximately 300 kgCO_{2,eq}/MWh_{biomass} to 700 kgCO_{2,eq}/MWh_{biomass}. Global GHG emission consequences are greatly affected by the assumed electricity production sector and selected end product.

Keywords: biomass gasification, heat integration, process integration, energy efficiency, biorefinery, CO₂ emissions, pulp and paper mill

Appended papers

This thesis is based on the work contained in the following papers:

- I. Integration of biomass gasification with a Scandinavian mechanical pulp and paper mill - consequences for mass and energy balances and global CO₂ emissions
Isaksson J, Pettersson K, Mahmoudkhani M, Åsblad A and Berntsson T (2012)
Energy, 44(1): 420-428
- II. Comparison between a detailed pinch analysis and the 'Heat Load Model for Pulp and Paper' – Case study for a Swedish thermo-mechanical pulp and paper mill
Isaksson J, Åsblad A and Berntsson T (2012)
Chemical Engineering Transactions, 29: 43-48.

Also presented at the 15th International Conference on Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction, PRES 2012, Prague, Czech Republic, 25-29 August, 2012.
- III. Influence of dryer type on the performance of a biomass gasification combined cycle co-located with an integrated pulp and paper mill
Isaksson J, Åsblad A and Berntsson T (2013)
Biomass & Bioenergy, 59: 336-347
- IV. Pretreatment methods for gasification of biomass and Fischer-Tropsch crude production integrated with a pulp and paper mill
Isaksson J, Åsblad A and Berntsson T (2014)
Clean Technologies and Environmental Policy, 16(7): 1393-1402
- V. Transportation fuel production from gasified biomass integrated with a pulp and paper mill - Part A: Heat integration and system performance
Isaksson J, Jansson M, Åsblad A and Berntsson T
Submitted to *Energy*
- VI. Transportation fuel production from gasified biomass integrated with a pulp and paper mill - Part B: Analysis of economic performance and CO₂ emissions
Isaksson J, Jansson M, Åsblad A and Berntsson T
Submitted to *Energy*

Co-authorship statement

Johan Isaksson is the main author of all appended papers. Professor Thore Berntsson was the main supervisor, and Anders Åsblad co-supervised the study. Dr Maryam Mahmoudkhani and Dr Karin Pettersson co-supervised the study in **Paper I**. Mikael Jansson contributed to the modelling work and co-supervised the study in **Paper V** and **VI**.

Related studies that are not included in this thesis

- Integration of biomass gasification with a Scandinavian mechanical pulp and paper mill
Isaksson J, Mahmoudkhani M, Åsblad A and Berntsson T (2011)
Proceedings of the 24th International Conference on Efficiency, Cost, Optimisation, Simulation and Environmental impact of Energy Systems, ECOS 2011, Novi Sad, Serbia: 3668-3679
- Influence of different pretreatment methods on biomass gasification and production of Fischer-Tropsch crude integrated with a pulp and paper mill
Isaksson J, Åsblad A and Berntsson T (2013)
Chemical Engineering Transactions, 35: 559-564. Also presented at the 16th International Conference on Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction, PRES 2013, Rhodes, Greece, 29 September - 2 October, 2013.
- Minimization of total drying costs for a continuous packed-bed biomass dryer operating at an integrated chemical pulp and paper mill
Holmberg H, Isaksson J and Lahdelma R (2014)
Biomass & Bioenergy, 71: 431-442

The first two papers comprise previous short versions of **Paper I** and **Paper IV**, respectively. The third paper is the result of a collaboration with Dr Henrik Holmberg and Professor Risto Lahdelma at Aalto University in Finland. The focus of that paper was on evaluating an optimisation procedure developed at Aalto University.

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1 Introduction

This thesis describes the technical, environmental and economic potential for the integration of a gasification-based biorefinery with an existing mill that produces pulp and paper.

1.1 Background

The pulp and paper industry has the potential to become an important player in biorefinery development. Research and development efforts focus on better utilisation of various by-product streams from pulp and paper mills and forestry, and on opportunities to integrate new biorefinery processes. One example of this development is the production of transportation fuels from forest residues. Tops and branches can be harvested in connection with the felling of wood for pulp and sawn goods. The majority of the approximately 14 TWh [1] which is currently harvested each year in Swedish forests is used for boiler-based combined heat and power (CHP) production. The potential for 2010-2019 has been estimated to range from 25-55 TWh/year, and combined with the harvest of stubs the potential increases to 54-140 TWh/year. The annual growth and thereby the potential harvest is expected to increase during the next 30 years [2]. In addition to residues from forestry, falling bark from pulp production and residues from sawmills, which is currently used to produce steam, can be added. These flows accounted for approximately 9 TWh in 2011 [3]. Fossil energy consumption for Swedish road transportation was 78 TWh in 2011 [4], which indicates that a minimum of 15-30% of the fossil energy input can be replaced by fuels that originate from domestic residues from forestry and pulp production (assuming 50% conversion efficiency).

The production of biofuels from forest residues has been suggested as a method of introducing renewable alternatives in the transportation system to hinder the build-up of greenhouse gases (GHGs) in the atmosphere. Biomass can be converted to fuels by gasification to a gas that is abundant in CO and H₂, which can be employed as building blocks for the synthesis of virtually any hydrocarbon. These processes, which have a conversion efficiency of approximately 50-70%, often have an excess of heat. To achieve improved overall efficiencies and economic competitiveness, this heat should be utilised.

By co-locating a gasification process with a heat demanding industry, the excess heat can be consumed throughout the year.

This study can be seen as part of the European efforts set up in one of the Europe 2020 targets, namely to decrease GHG emissions by 20%, have 20% renewables in the energy sector and use energy 20% more efficiently. In addition, an 80% reduction of emissions is targeted until 2050. The projected required decrease of emissions from different sectors in the European Union (EU) is shown in Figure 1. The modelling results show that the remaining GHG emissions in 2050, in addition to transportation, will derive from various non-CO₂ emissions and industrial processes. Biofuels in the transportation sector became particularly important for aviation and heavy vehicles; however, if the electrification of the transportation sector cannot be realised as predicted, biofuels will become increasingly important for cars to attain the 80% target. [5]

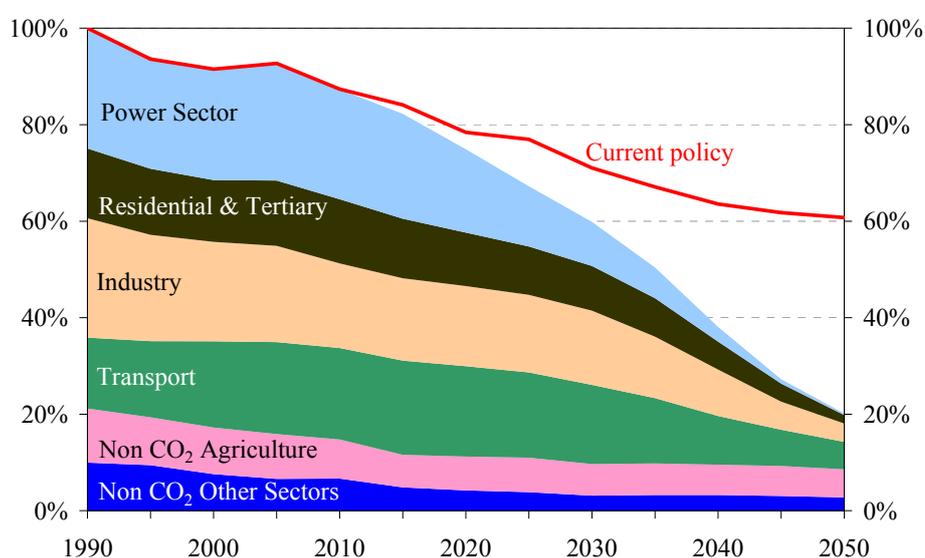


Figure 1. Projected GHG emissions in the EU with 80% reduction until 2050 based on levels in 1990 [5].

As shown in Figure 2, to create a large reduction of CO₂ emissions from the transportation sector in the EU, the main effort should be directed towards road transportation.

Currently, a few biofuels are available for Swedish consumers. Ethanol is used as a blend-in in petrol and as E85 - with 85% ethanol and 15% petrol - in modified spark ignition engines. Different blend-in solutions are also available for diesel engines, such as fatty acid methyl ester (FAME), and more recently, hydro-treated vegetable oils (HVOs). FAME and HVO are both referred to as biodiesel, which can cause some confusion in the debate. FAME is obtained by transesterification of vegetable oils or animal fats. HVO may have the same origin as FAME but is treated with hydrogen to create a fuel that is

identical to fossil diesel on a molecular level. HVO only slightly differs in density and heating value. More recently, cars that run on fossil or bio-based methane have gained increased attention, at least in the proximity of the national natural gas grid. Approximately 60% of the gas used for transportation originates from anaerobic digestion, whereas the remaining gas consists of fossil natural gas which has lower emissions of CO₂ and particles compared with petrol [6].

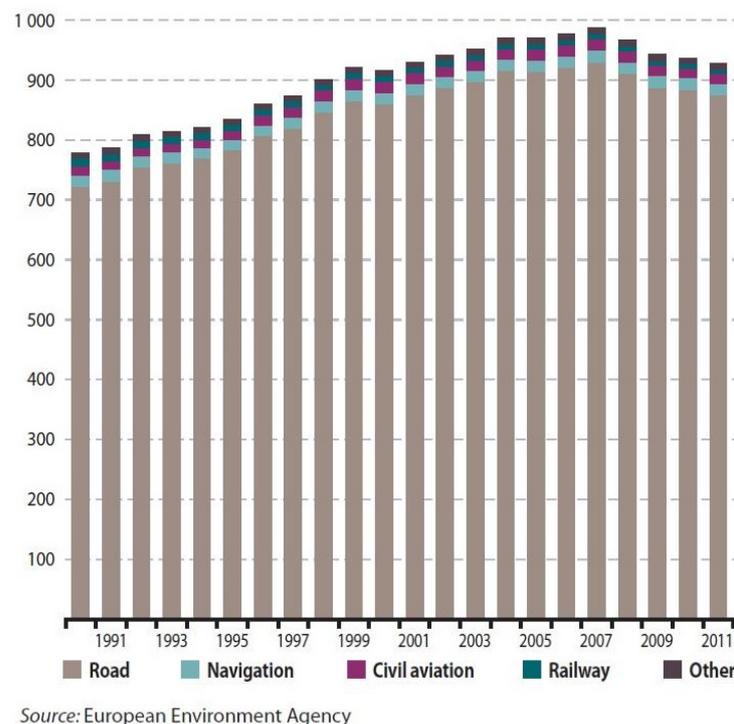


Figure 2. GHG emissions from transportation in the EU (Mt CO_{2,eq})

A study initiated by the Swedish government had the task of developing guidelines regarding how the Swedish transportation sector can become fossil-independent by 2030 [7]. In practice, at least one renewable alternative should exist for every vehicle. These alternatives can be fuels or electricity from a renewable source. The investigators emphasise that conditions should be set to ensure that the domestic process industry can be part of this transition and to promote solutions in which the Swedish industry is in the forefront. To reach a fossil-independent transportation fleet, the investigators believe that a CO₂ charge on its own would have to be set at a very (too) high level and that directed policy instruments would have to be complementary. To satisfy these goals, the production of biofuels has to be realised and is considered to be a potential future export products. The development of new technology that makes use of cellulosic biomass, especially such that can be classified as waste or residues, is crucial. Biomass gasification is emphasised as a promising emerging technology with a large number of different potential end products. Co-location with existing industry or a district heating network is mentioned as an option for using the generated excess heat. According to the report,

another factor to consider is that cars are produced in long series to pay for the high development costs. Selecting a non-standardised fuel (i.e., in addition to E85, petrol, diesel or methane) would require new fuel standards. Blend-in fuels or drop-in fuels can be used with conventional engines and existing distribution systems, whereas higher blends would require modified engines. The report concludes that expanded production of the current biofuels on the Swedish market based on local raw material, i.e., ethanol (based on sugar and starch), HVO, FAME and biogas (based on anaerobic digestion) is insufficient for reaching a fossil-independent transportation system. The addition of biofuels with a cellulosic origin will result in an increased demand for woody biomass from the forests, which is satisfied by extracting more biomass or requiring other users to use less. Residues from the production of pulp, paper and sawn goods are currently used to sustain the internal heat demand at the production sites but can instead be used for biofuel production via more efficient processes or other (non-fossil) sources of energy for these processes.

On a global level, the prognosis is less optimistic. A report from the IEA [8] states that biofuels can provide 27% of the total transportation fuel consumption in 2050. Conventional oil- and starch-based biofuels are assumed to be gradually phased out in favour of advanced biofuels.

The integration of gasification-based biorefineries in the pulp and paper industry can be beneficial on several levels. The declining availability of easily accessible fossil fuels and climate change caused by increased levels of GHG in the atmosphere are critical and well-known issues from a global perspective. National benefits from the implementation of renewable fuel production may include local jobs and less dependence on imported goods if biomass resources are nationally available. The local benefits can include a broader and more robust product portfolio for the pulp and paper mill, which is better adapted to withstand price fluctuations for individual products. The experiences of wood handling and processing, in addition to a commonly large demand for heat, make pulp and paper mills potentially well-suited hosts for the thermo-chemical conversion of biomass. A better utilisation of local resources, i.e., the use of forests for a more extensive range of products, improves resource efficiency.

Substantial efforts in studies about different biorefinery concepts in the pulp and paper industry have been made with the primary objective to diversify the product portfolio of the mill and to produce more value-added products, compared with combustion of the biomass. Different concepts include the extraction of hemicellulose and lignin, or gasification of black liquor. Hemicelluloses are macro-molecular sugars that can be used to produce an extensive variety of products, such as different alcohols, barriers and lactic acid. They can be extracted prior to pulping (mechanical or chemical) or from the black liquor. Lignin can be extracted from the black liquor and can subsequently be directly applied as a high heating value biofuel or for additional upgrading to chemicals and materials [9]. A recent area of interest is the cracking of lignin in near-critical conditions for the production of bio-oils [10]. Another example of an upcoming biorefinery product

with a high value is lignin-based carbon fibre. However, the current world market is limited to approximately 50000 t per year, i.e., it would be enough to convert one Swedish mill to cover the current world market. (assuming a mill with an annual wood input of 700000 t on a dry basis with 25 wt-% lignin content, of which 2/3 can be extracted [11], and 50% carbon fibre yield [12]). If costs associated with raw material and production can be reduced, the market has a very large potential to grow [13]. The gasification of black liquor has received substantial interest as a route to renewable fuels. Black liquor consists of spent cooking chemicals and organic material, i.e., lignin and hemicellulose, from the pulping process. Instead of combusting the black liquor in a conventional recovery boiler to recover heat and regenerate the chemicals, this stream can be gasified to convert organic content into a synthesis gas. The regenerated chemicals are sent back to the pulping process, and the gas can be synthesised into fuels or chemicals. The introduction of a new synthetic product will change the energy balance of the mill, which requires the introduction of another source of energy to maintain the heat balance of the mill [14]. Due to the higher pulp yield from mechanical pulping compared with chemical pulping and therefore small streams of by-products, biorefinery integration with mechanical mills is less explored. In a study by Jaaidi and Stuart [15], three potential biorefinery concepts for thermo-mechanical pulp (TMP) mills were explored: xylitol production from hemicellulose, bio-composites from part of the TMP wood fibres and fermentation of cellulose and hemicellulose to ethanol and phenols. They identify gasification-based processes as an opportunity for heat integration with the mill, with the possibility to share utilities and overhead costs.

1.2 Objectives

The objective of this study has been to assess the GHG reduction potential, efficiency and economic performance of co-locating a gasification-based biorefinery with a pulp and paper mill. The thesis aims to increase the knowledge about the possible synergy effects associated with this integration and to investigate how the key operating parameters and selection of equipment affect the performance indicators. The intended long-term effect of these efforts is to strengthen the role of the pulp and paper industry and to reduce global emissions of CO₂.

1.2.1 Scope

The scope of this thesis has been limited to integrated mechanical or chemical pulp and paper mills as the host process to which a gasification-based process can be linked. Only gasification of solid biomass, i.e., bark and forest residues, has been considered. Gasification of black liquor has not been included as an option in this thesis. Four different end products have been evaluated to limit the extent of this thesis. The biofuels include methanol, Fischer-Tropsch (FT) crude and synthetic natural gas (SNG), i.e., dimethyl ether (DME), gasoline, hydrogen and various alcohols, in addition to methanol, are excluded. Utilisation of the gas for electricity production in a gas turbine combined

cycle has been assessed previously in many studies as a promising technology and was included in this study as a fourth option. Considering feedstock for the gasifier, only residues from forestry and the production of pulp and saw goods have been considered. The reason for this selection is the focus of this thesis on the pulp and paper industry in northern Europe where this feedstock is readily available. Storage of CO₂ is considered to be a possibility in GHG evaluation studies; however, only the CO₂ that was separated as part of the gas condition process has been considered, i.e., CO₂ from the recovery boiler flue gases was excluded. The biorefineries are primarily evaluated for the timeframe near 2030, when gasification processes are assumed to be available on a commercial scale. Thermal integration of these new gasification-based systems alters the energy balance of the pulp and paper mill; these changes were evaluated from a European energy system perspective, assuming a future European market for electricity and biomass.

1.3 Appended papers

This thesis is based on six appended papers. A general overview of the papers is illustrated in Figure 3, which shows that the papers discuss different parts of a gasification-based system and integration with different types of pulp and paper mills. The papers are briefly introduced in this section.

In **Paper I**, a mechanical pulp and paper mill served as an integration host for different gasification-based biorefineries. The different end products (methanol, FT crude or increased power production) were assessed in terms of integration potential, efficiency and GHG emission consequences. The possible impact of co-locating a large sawmill with the biorefinery, which enables an exchange of heat for drying and woody biomass, was also assessed. The integrated biorefineries were compared with a corresponding stand-alone plant.

The predictability of the methodological tool named ‘Heat Load Model for Pulp and Paper’ (HLMPP) which was used to acquire mill stream data, necessary for the process integration study in **Paper I**, was assessed in **Paper II**. Automatically derived stream data results from the HLMPP tool were compared with manually extracted detailed stream data for a mechanical pulp and paper mill. Based on the comparison, suggestions regarding how to improve the possibility to accurately predict stream data with the tool were presented.

Paper III discusses the drying of biomass prior to gasification and utilisation in a gas turbine combined cycle that is integrated with a paperboard mill. Three different drying technologies (superheated steam dryer, flue gas dryer and low-temperature air dryer) were evaluated in terms of the integration potential with the subsequent gasification process and the mill. An economic evaluation of the three concepts were performed which also included a heat pump case and the impact of using air or oxygen as the oxidising agent in the gasifier.

In **Paper IV**, a gasification process was integrated with the same paperboard mill that was examined in **Paper III**; however, this study focused on torrefaction or pyrolysis as an alternative pretreatment measure to milling the biomass prior to gasification. The different technologies were evaluated in terms of conversion efficiency and GHG emissions reduction potential for FT crude as the end product.

Paper V and Paper VI comprise two parts of the same study. Different end products (methanol, FT crude, SNG or increased power production) from gasified biomass were assessed in terms of heat integration opportunities in **Paper V** and in terms of GHG emission consequences and economic performance in **Paper VI**. The evaluation was performed as a case study on a conceptual integrated kraftliner mill. The influence of energy efficiency measures, in connection with biorefinery integration, was also assessed.

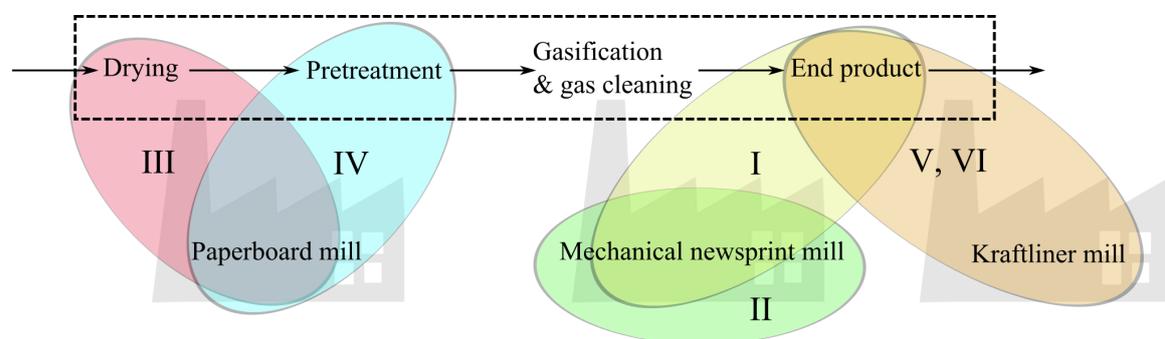


Figure 3. Visualisation of the relationship among the appended papers and the main focus regarding the case study mill and the process step in the gasification route.

1.4 Thesis outline

The thesis begins with a broad perspective of the future challenges that motivate the present work towards production of renewable transportation fuels in general, and particularly via biomass gasification. The background is followed by the objectives and scope of this study. An overview of the status of activities within the field of gasification of biomass in general and particularly connected to the forest industry is presented in Chapter 2. Previous studies about integration of biorefinery concepts with existing industries, such as pulp and paper mills, are also addressed, and the need for additional research is identified. Different pulp and paper mills and various gasification-based systems are addressed throughout the thesis. These are described in Chapter 3. The methodology employed for heat integration studies and assessments of GHG emissions and economic performance are described in Chapter 4. The most important results from the appended papers are summarised in Chapter 5. A general discussion in Chapter 6 is followed by general conclusions in Chapter 7. Chapter 8 includes a discussion about identified areas for future studies.

2 Literature review and previous studies

This chapter presents a review of previous studies in the field of biomass gasification and studies that are specifically directed towards the integration of these processes with the pulp and paper industry. Numerous studies have been carried out regarding biomass gasification systems. The review of studies about stand-alone gasification processes have been focused on papers and reports where detailed data has been provided, and therefore have been used to retrieve input data for the studies in this thesis. Based on the literature survey, the need for additional research is identified.

Gasification is the thermal breakdown of, in this thesis, biomass into volatiles and ash in the presence of less than the stoichiometric amount of an oxidising agent. It is a two-step endothermic process during which volatiles, i.e., different hydrocarbons, CO₂, CO, H₂ and water, are released by supplying heat, followed by the reaction of the remaining char with an oxidising compound or H₂. Some of the char reacts completely to CO₂ to supply heat for the endothermic reactions. Gasification of biomass is an interesting biorefinery concept due to its versatility of application area, which ranges from pre-combustion of waste to avoid toxins in the flue gases, to advanced synthesis of hydrocarbons. For example, the technology has been employed in Sweden for the propulsion of cars during the Second World War when access to oil was limited. Many research groups have investigated how to utilise biomass via gasification in an efficient and economical manner regarding process pathways and suitable end products. In a recent study by Hannula and Kurkela [16], four different transportation fuels were compared: methanol, DME, petrol and FT diesel. They concluded that methanol and DME are more competitive end products regarding production cost compared with FT diesel and petrol in a stand-alone plant due to the need for additional processing steps, which affect both efficiency and capital costs. They also note that other factors, in addition to price, may have an important role, such as compatibility with existing infrastructure. They also emphasise the importance of integrating this type of process with “existing processes” to lower capital costs and ensure efficient utilisation of heat and steam. Kreutz et al. [17] have investigated gasification of coal or biomass or a combination of both for the production of FT fuels. They conclude that the biomass route with CCS (carbon capture and storage, i.e., sequestration of CO₂) can be more profitable than a corresponding case using coal as

feedstock if the CO₂ charge is at least 65 \$/tCO_{2,eq} [17] (to be compared with 7.3 €/tCO_{2,eq} for the EU when this thesis was printed [18]). Issues for the biomass cases include the high feedstock cost and the steep economy of scale for both investments and the transportation of CO₂ to the storage site. For these reasons, the authors propose co-processing biomass with coal. Similar studies have been carried out by Hamelinck and Faaij [19] for the production of methanol and hydrogen, and by Hamelinck et al. [20] for production of FT fuels. Green FT diesel was found to be 40-50% more expensive to produce than methanol and hydrogen. Tock et al. [21] studied different alternatives for thermos-chemical production of liquid fuels. They employed process integration to estimate the potential for energy savings and the utilisation of excess heat. Simulation models were used to determine overall energy efficiencies and production costs for FT fuels, methanol and DME. A study of a flexible gasification-based system where electricity can be produced during peak hours while switching to chemicals during off-peak hours was presented by Meerman et al. [22, 23]. They also explored a mix of fossil and renewable feedstock, which can be adjusted depending on the current price levels of commodities and policy instruments.

2.1 Current status of biomass gasification projects

Currently, no commercial scale biomass gasification plants exist, with the exception of direct combustion applications. Biomass-fed gasifiers for heating or CHP operation connected to a boiler, i.e., basically a pre-combustion chamber, are being operated in a number of places on a commercial scale. When this thesis was printed, the largest biomass gasifier in the world was located at the Vaskiluoto CHP plant in Vaasa, Finland (refer to Section 2.3.1 for a description of different gasifier types). It has a thermal input of 140 MW, and the produced gas is combusted with pulverised coal in a steam boiler. It was taken into commercial operation in early 2013 [24]. The technology with air-blown gasification and direct combustion of the gas is relatively well proven. This type of gasifier was designed for firing of lime kilns especially in the 1980s; new units have been recently constructed [25]. Gasification systems for more advanced applications, such as biofuel production, are currently run on pilot or a demonstration scale. A step towards more advanced gasification systems was achieved by Sydkraft's demonstration plant in Värnamo, Sweden, where the gas was sufficiently purified to be combusted in a gas turbine. The technology was based on an 18 MW_{th} air-blown gasifier that was pressurised to 18 bar, followed by hot gas filters for particulate removal. The plant was operated from 1993 to 1999 [26]. More advanced processes have followed on a smaller scale, in which the gas is upgraded to biofuels. Synthesis of these products requires additional gas cleaning and a nitrogen-free gas that is achieved via either oxygen-blown or indirect gasification. Another notable facility is the gasification plant at the Gas Technology Institute (GTI) in Des Plaines, Illinois. The system is based on an oxygen-blown pressurised CFB gasifier with a dried biomass capacity of 4.3 MW_{th} followed by hot gas filters and a tar reformer, which was developed by Andritz-Carbona. The synthesis train

is Haldor-Topsø's TIGAS-system for production of petrol via methanol/DME [27]. Based on the same gasification technology, the energy company E.ON plans to construct a 200 MW SNG plant in the south of Sweden and has been granted NER300 support for the project [28]. The NER300 is a large funding program for low-carbon emitting demonstration projects in the fields of bioenergy and CCS. The fund is managed and jointly implemented by the European Commission, the European Investment Bank and the member states of the EU [29]. The GoBiGas plant in Gothenburg, Sweden is based on an indirect gasification technology developed by Repotec, followed by gas conditioning and methanation from Haldor-Topsø. The plant has a 20 MW SNG output from a thermal input of 32 MW when at full capacity. The plant has been operational since late 2014 [30]. The GoBiGas gasifier is similar to the 8 MW_{th} gasification plant in Güssing, Austria, where considerable research on the gasification technology and utilisation of the produced gas has been performed [31].

We are still waiting to see the first commercial-scale biomass gasification process with downstream synthesis. The technology should be proven in a stand-alone plant before integration with an existing industrial system can be realised due to technological uncertainties.

Regarding pulp and paper mill-based biofuel production from gasified biomass activities have been conducted on a demonstration scale. Black liquor gasification has been demonstrated at Smurfit-Kappa's pulp and paper mill in Piteå, Sweden where a 3 MW_{th} slip stream of black liquor from the recovery boiler is gasified in an entrained flow (EF) gasifier. The gas is synthesised into DME, at a capacity of approximately 4 t per day, which is used in converted diesel trucks [32]. The technology has been proven to work; however, as the recovery of chemicals from the black liquor is critical in a chemical pulp mill, no mill owner has yet been willing to relinquish the reliable recovery boiler. Regarding pulp and paper mill integrated gasification processes that use solid biomass as feedstock, Finland has achieved the largest number of advancements. NSE Biofuels Oy, which is a joint venture between Stora Enso and Neste Oil, has demonstrated FT fuel production using a 12 MW biomass gasifier. The plant is situated at Stora Enso's mill in Varkaus, Finland. The tests were successful but the large capital costs forced them to abandon their plans to construct a full-scale commercial plant [33, 34]. UPM has initiated a collaboration with GTI and Andritz-Carbona to produce FT fuels; they plan to construct a commercial plant at one of their mills in either Strasbourg, France or Rauma, Finland. The plans for Strasbourg have been granted NER300 support but the project remains on hold [35].

Methanol production may be particularly suitable for chemical pulp mills as methanol is also a by-product formed in the cooking procedure, ending up in the evaporation condensate. The total Swedish potential for methanol derived from black liquor has been estimated to be 90000-110000 t per year [36], which can be compared with the annual petrol consumption of approximately 2.8 Mt, including 5% ethanol blend-in [6]. The

heating value of petrol is approximately twice as high as it is for methanol on a mass basis.

2.2 Feedstock and pretreatment

As described in the Section 1.1 Background, the availability of forest residues has been estimated to be approximately 25-140 TWh per year, including the 14 TWh that is currently harvested. An older study by Lönner et al. [37] estimated that 43 TWh of tops and branches (i.e., excluding stubs) can be harvested each year when environmental and technical concerns have been considered. In this study, secondary biomass from forestry has been considered to be the preferred feedstock for the gasifier, in addition to bark from the pulp mill and shavings from sawmills. The technical concerns regarding the gasification of bark were assumed to have been overcome by the time the plant is about to be built. Conversely, wood that is too clean can pose difficulties in its use as gasification feedstock. Demolition wood or waste have not been considered as feedstock in this study but is not assumed to affect the results as the impact is more of an operational concern. The harvesting of forest residues may require that ash is returned to the forests to avoid depletion of nutrients [38]. The costs associated with ash-handling have not been considered.

2.2.1 Drying

Drying of biomass prior to gasification is performed for several reasons. First, to maintain a steady and reliable operation of the gasifier, the feed should be as homogenous as possible, including an even moisture content over time. Second, the evaporation of moisture using heat from the gasifier bed causes exergy losses. A high moisture content decreases the reaction temperature in the gasifier, assuming no additional external heat supply compared with drier fuel, which reduces the reaction rate. This reduction causes a higher tar content and a poorer gas quality with a higher water content, possibly requiring additional dewatering of the gas downstream. Many dryer types are available on the market for different types of applications and various heat sources. For woody biomass, three types can be considered: drying with a belt dryer, drying with flue gas in a rotating drum and drying with a superheated steam dryer. These types were compared in **Paper III** and are described in Section 3.2. Fagernäs et al. [39] reviewed the most promising technologies in biomass drying. The authors identified some benefits of steam dryers, especially regarding specific heat consumption; however, drying with flue gases or heated air on a moving bed offer simpler construction.

In a belt dryer, fans are used to force air through heat exchangers to increase the air temperature. The wet biomass is transported on a perforated belt while the heated air passes the material from either above or below [39]. This type of dryer consumes excess heat at low temperatures to heat the drying air; however, a low air temperature requires

large air flows and large equipment to achieve sufficient drying [40]. The overall efficiency can be improved by recovering heat from the moist exhaust air.

In flue gas dryer applications, a rotating drum is the most common technical solution. If a dedicated boiler is used to generate flue gases, the inlet temperature to the dryer is normally relatively high, approximately 250 °C to 400 °C. The high temperatures are often causing fires, which is the main operational concern with this dryer type [39].

Drying with steam occurs at an elevated pressure to enable utilisation of the latent heat of the evaporated moisture from the biomass. This process requires pressurisation of the biomass and feeding with a screw followed by a rotary valve [41]. Condensing steam at high pressure superheats the steam in the dryer loop, and the superheated steam causes the biomass moisture to evaporate. Steam serves to carry the biomass through the dryer and to transport heat to the biomass particle. Evaporated steam at dryer pressure is continuously extracted from the system for additional utilisation as process heat [42].

2.2.2 Torrefaction

Torrefaction, which is also referred to as mild pyrolysis, can be used to improve certain characteristics of woody biomass. The process is performed at approximately 200-350 °C at atmospheric pressure for a few minutes to a couple of hours [43]. The improved properties that result from torrefaction can facilitate both the logistics and end use for combustion or gasification. In addition, the torrefied biomass is hydrophobic and capable of withstanding fungus. These characteristics are beneficial for logistical purposes. The gases released during torrefaction consist of permanent (primarily CO, CO₂ and CH₄) and condensable gases, which can be upgraded but are usually combusted to sustain the endothermic torrefaction reactions. If the torrefaction process is co-located with a gasification process or a pulp and paper mill, the pre-drying of biomass can be achieved with excess heat from the mill, i.e., volatiles do not need to be combusted for drying. In certain studies, the entire flow of volatiles were mixed with raw gases from the gasifier and excess heat from syngas cooling via HP steam was used to supply heat for torrefaction [44].

Repellin et al. [45] investigated the brittleness of torrefied wood and demonstrated that the power demand for milling is reduced 4.7 times for an anhydrous weight loss of 8%, for both beech and spruce. Phanphanich et al. [46] note that torrefaction at 300 °C reduces the power consumption for milling six times for forest residues for a particle size of 1.5 mm. A study of torrefaction of spruce and birch was conducted by Tapasvi et al. [47]; the authors investigated the impact on particle size, hold-up time and temperature on the composition and energy content of the torrefied material. Temperature was proven to have the largest impact, whereas particle size only had an impact at the lowest temperature investigated (225 °C). Varying the hold-up time, however, only made a difference at higher temperatures (275 °C). The authors noted a decreased milling energy demand of 40-88% for the samples. Due to this technology's ability to decrease the

milling energy and create a more homogeneous material, torrefaction may become an interesting pretreatment step prior to EF gasification. EF gasifiers are employed when liquids or pulverised coal is gasified but attached to a torrefaction system it might become an interesting technology also for biomass (refer to Section 2.3.1 for more information regarding gasifier types).

Torrefaction as a means of increasing the volumetric energy density of biomass may help to release the full potential of biomass as an energy resource. Mobini et al. [48] discovered that overseas transportation from Western Canada to north-eastern Europe can be achieved at lower cost if the biomass is torrefied prior to pelletising. The CO₂ emissions associated with the transportation of pellets were also reduced compared with conventional pellets due to the improved bulk density.

A drawback of torrefaction is that the brittle characteristics require that torrefied biomass is pelletised prior to transportation, which requires energy. The “springback” of the pellets, i.e., the volume expansion relative to the maximum compression, was 40% for torrefied wood compared with less than 20% for untreated wood. The positive effect of increasing the volumetric energy density was partly eliminated by the lower pellet density. The strength of the pellets decreased by 40-55% after torrefaction, which causes logistical problems when the pellets break. Torrefaction has a negative impact on the pelleting process by increasing the power demand by 42% for forest residues due to higher friction in the press channel compared with untreated material [49].

2.2.3 Pyrolysis

Pyrolysis of biomass is a process that occurs between approximately 400 °C and 600 °C in an inert atmosphere. During pyrolysis the biomass particle decomposes into a number of different products in liquid, gaseous and solid form. Char and volatile compounds are typically used as fuel to supply the necessary heat. The resulting liquid consists of a large portion of water from the biomass moisture and pyrolysis reactions and a large fraction of oxygenated compounds. As a result, the heating value is approximately 50% of that of a pure hydrocarbon oil. Pyrolysis oil can also be unstable as compounds react into heavier molecules and water, which may eventually cause phase separation [50].

Pyrolysis may be a potential alternative to torrefaction as a pretreatment method prior to EF gasification. As the biomass is converted into a liquid it can be sprayed into the gasifier. Decentralised pyrolysis units have been suggested to densify residues from agriculture, for gasification in a central EF gasifier [51].

Recent developments of pyrolysis oil production have facilitated the co-processing of the liquid phase with fossil vacuum gasoil in existing fluidised catalytic cracking (FCC) units at a refinery for petrol and diesel production. Maximum proportions of 5% can be fed to produce on spec diesel and petrol. If this approach is proven to be successful using only

existing infrastructure, a considerable amount of resources can be saved by avoiding the procurement of dedicated processing equipment for upgrading pyrolysis oil [52].

2.3 Production of biofuels via gasification of biomass

As previously discussed in Section 1.1 Background, gasification of biomass is basically incomplete combustion and is a well-known pre-combustion technology. However, more advanced applications are less explored practically on a larger scale, which places tighter constraints on both the feed and the operation of the gasifier and downstream equipment, especially concerning tar cracking/removal. Once the syngas only consists of H₂ and CO, the remaining process steps have been demonstrated on a commercial scale for fossil feedstock.

2.3.1 Gasification, gas cleaning and conditioning

Gasification can occur in a fixed bed or fluidised bed or can be entrained in the flow of oxidant as a spray. Fixed beds are only suitable for small-scale application due to the inhomogeneity of the bed at larger scales, which rules out this technology in this context. Fluidised beds are currently used for large-scale biomass combustion and air-blown atmospheric gasification and should be well suited for more advanced applications. The intrusion of inert gases - primarily nitrogen - should be avoided to keep the necessary size of downstream equipment to a minimum. To prevent nitrogen in the stream of raw gas, either pure oxygen is employed as an oxidising agent or heat is indirectly supplied in a dual-bed (DB) system by burning char in a separate combustion chamber. The amount of char that is fed from the gasifier to the combustor can be adjusted to make the system self-sufficient in heat. Oxygen-blown fluidised beds are operated as either a circulating fluidised bed (CFB), i.e., sand is separated from the gas in a cyclone after the gasifier and is returned to the bed, or a bubbling fluidised bed (BFB). The oxygen-blown solutions are penalised by the necessity of an air-separation unit for the production of oxygen, whereas the more complex nature of the indirect system complicates pressurisation. A consequence of the different setups is that the formed CO₂ ends up in the syngas in a direct system, whereas some ends up in the flue gases in an indirect system. If CO₂ needs to be sequestered, this consequence can be either positive or negative. To capture the CO₂ in the flue gas stream, an additional CO₂ separation unit is required. The EF gasifier has some interesting benefits and distinct challenges. It is operated at a high temperature - approximately 1100 °C to 1350 °C - to allow the reactions to occur during a short residence time, which indicates that the syngas contains no tar and very little methane. Downstream tar reforming becomes unnecessary, which decreases the complexity and cost [53]. Another consequence of the short residence time is that the feed material has to be small enough for full conversion, which requires extensive milling or pretreatment of the material by torrefaction or pyrolysis.

A number of techniques exist to remove compounds that can compromise the function and life time of downstream equipment and to assure that the product satisfies specifications. These compounds primarily include alkali, dust and sulphur. Particulate matter can cause fouling, corrosion and erosion of equipment [54]. Commercial equipment exists for the various cleaning steps but development is ongoing to obtain more efficient solutions.

All types of gasifiers, with the exception of the EF gasifier, require some type of measure to eliminate tars. Current practice has involved washing out tars in a scrubber; for example, using rapeseed oil methyl ester (RME) as a solvent. The drawbacks are the reduction in overall conversion efficiency due to the loss of material and the additional cost for the purchase of RME; however, RME and tars can be combusted for heat generation. An alternative approach is to crack the tars into H₂ and CO in a reformer. Although no material is lost, energy is needed to increase the temperature or regenerate catalytic material. Particles that consist of inorganic compounds or residual carbon are removed in cyclones or filters, or a combination of both, before or after tar cracking. The approaches are frequently referred to as clean and dirty tar cracking, depending on the position of the filter in relation to the reformer. Filtering is preferably performed at the highest possible temperature to prevent losses associated with cooling and re-heating of the gas (if the reformer is positioned after the filter). The most promising technologies involve different types of ceramic or metallic candle filters, which refer to their geometrical shape [26]. The temperature for filtration prior to tar reforming is limited in the lower range by tar condensation at approximately 350 °C and filter blinding from sticky soot formation at temperatures above 600 °C. Research is now focusing on increasing the possible filtering temperature in order to enhance the overall efficiency [55]. If a scrubber is used for tar cleaning the gas is anyway cooled and a bag filter can be employed. Insufficient particle removal causes erosion or corrosion of downstream equipment.

Depending on the application of the gas, adjustments to the H₂/CO ratio may be necessary. A water-gas-shift (WGS) is used if a higher H₂ content is required, which is the case for all applications in this study. The syngas is passed through a WGS catalyst to convert CO and water into CO₂ and H₂ according to Equation 1. Steam is injected if the syngas moisture is too low.



Due to the reducing atmosphere during gasification, sulphur primarily exists as H₂S, with traces of COS and CS₂. Sulphur compounds are highly poisonous to catalysts and need to be reduced to low concentrations. Sulphur can also cause corrosion of metal surfaces. If sulphur is removed after the WGS, a sulphur tolerant shift catalyst has to be used [56]. If necessary, the remaining sulphur can be removed using activated carbon or a bed of ZnO. CO₂ is removed to increase the partial pressure of H₂ and CO in the synthesis steps and to

reduce the necessary size of the equipment. Removal of CO₂ is achieved through physical or chemical absorption, adsorption or membrane separation.

2.3.2 Syngas utilisation

Once the raw gas from the gasifier has been cleaned and conditioned, only H₂ and CO remains (if SNG is the desired end product methane has also been preserved). Virtually any hydrocarbon can be produced from H₂ and CO, and often with well-proven commercial technologies. The most commonly discussed end products are described in this section.

Methanol has a number of different applications, such as a bulk chemical, a low sulphur shipping fuel or a possible blend in petrol. The majority of the methanol that is currently produced in the world is synthesised into other chemicals: 40% is converted into formaldehyde, which is subsequently converted to plastics, paint etc. Global production is approximately 58 Mt [57]. The synthesis is performed at 250 °C to 280 °C and 60 to 80 bar [58]. In addition to the WGS reaction (Reaction 1), the methanol synthesis can be described by the following two reactions:



In addition to a possible end product from a gasification based biorefinery, methanol is a by-product from the sulphate pulping process. However, this methanol fraction is contaminated with sulphur, which causes odour problems in commercial applications [36].

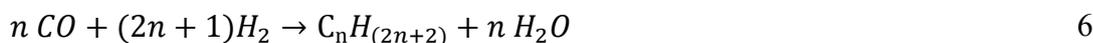
DME is commercially produced from methanol in a two-step process; however an emerging option is convert syngas to DME in one step. DME is in a gaseous phase at ambient pressure but changes to a liquid at 5 bar. The suggested application is a substitute for diesel in heavy trucks. DME is synthesised by dewatering methanol according to the following reaction:



The **Fischer-Tropsch** process, which was developed in the 1920s by the German scientists Fischer and Tropsch [59], is a path that is used to derive synthetic hydrocarbons - primarily paraffins - from a mixture of hydrogen and carbon monoxide. Historically and recently, this process has been used to produce liquid transportation fuels from gasified coal; however the interest in renewable feedstock has increased. In the FT process, hydrogen and carbon monoxide form carbon chains of various lengths approximated by the Anderson-Schultz-Flory distribution [60-62], which assumes a constant probability of chain growth that is independent of length and is expressed as

$$\frac{W_n}{n} = (1 - \alpha)^2 \alpha^{(n-1)} \quad 5$$

where W_n is the weight fraction of the molecules containing n carbons and α is the growth probability factor. The reaction can be expressed as



FT reactors are classified as either low temperature (200-240 °C) or high temperature (300-350 °C). For production of straight-chained hydrocarbons in the diesel range, the low temperature generates the higher yield. Iron or cobalt based catalysts are common in commercial applications. CO₂ primarily serves as a diluent in cobalt-based catalysts but acts differently in iron catalysts depending on their design [63]. Higher partial pressures of H₂ and CO enables higher C₅₊ selectivity [64]. An important benefit of FT diesel is the low soot formation and the very high cetane number, which exceeds general specifications for fossil diesel fuels and enables the usage of FT diesel where high grade diesel is necessary or for upgrading low quality diesel.

Biogenic methane is currently produced via anaerobic digestion of waste and manure; however, to achieve production on a larger scale, gasification of cellulosic biomass is a promising technology. This thesis distinguishes between biogas, which is methane from anaerobic digestion, and SNG, which is methane from gasification. A possible advantage of SNG compared with other biofuels is that the product can be injected to a natural gas grid for distribution to either heat and power production as industrial feedstock or as fuel for cars, trucks or ships. This process enables a flexible system in which policy instruments or commodity prices will direct the usage towards the most profitable solution. During the early phase of implementing SNG production, natural gas is available as an immediate backup. SNG is synthesised in either adiabatic or isothermal reactors according to Reaction 7 [65].



Electricity production in a gas turbine can be investigated as a potentially robust alternative to the previously described and more complex routes. The benefit of this kind of process is that the electricity-to-heat ratio can be increased from less than 0.5 to the range of 0.8-0.9 compared with a conventional CHP plant with a steam boiler [26]. The gas turbine exhaust is cooled in a heat recovery steam generator (HRSG) for additional electricity production in a back-pressure turbine.

2.3.2.1 Carbon capture and storage

CCS has been suggested as a method for drastically reducing emissions of CO₂ from large sources, such as coal power plants. This thesis was primarily concerned for European conditions and as the European pulp and paper industry is concentrated in the Nordic countries, the storage of CO₂ from these sites will have to be realised in this region. A

recent report from the Bastor project presents a study of opportunities for CO₂ sequestration in the Baltic Sea [66], around which the majority of the pulping capacity in Europe is located [67]. They conclude that transport by ship from coastal hubs to the storage site is a cost-effective solution as individual sources around the Baltic Sea are relatively small and more distributed than power plants in continental Europe and the United Kingdom, which are frequently referred in the context of CCS. The specific cost for the transportation of CO₂ via a coastal cluster hub to the storage site ranges from 12 to 20 €/tCO₂, depending on the cluster location and on the assumed capacity for individual storage wells. A similar study of the Skagerrak/Kattegat region in southern Scandinavia, where many oil refineries and fossil-based power plants are located, was performed. For a capacity of 14 MtCO₂/y, the resulting cost of transportation was 12 to 14 €/tCO₂.

The problem of reaching targets for CO₂ concentration in the atmosphere was recently addressed in a study by Azar et al. [68]. They claim that overshooting the temperature ceiling of 2 °C compared with pre-industrial levels, which was endorsed by the UNFCCC in 2010 [69], can be cost-effective using bioenergy with CCS (BECCS) if the target must be satisfied by the year 2150. Early implementation of zero emission technologies is, however, still crucial to reach the target in the long-term.

2.4 Integration of biomass gasification with pulp and/or paper mills

Gasification-based biorefineries in pulp and paper mills have been studied in a number of papers and reports that primarily focus on gasification of black liquor, but also on solid biomass gasification. Pettersson and Harvey [70], Larson et al. [71], and Joelsson and Gustavsson [72] focus on systems in which the recovery boiler has been replaced by a black liquor gasifier. These systems can offer higher efficiencies and better flexibility than conventional recovery boilers but are associated with a fair amount of risk. The regeneration of cooking chemicals is the core of the pulping process and the recovery boiler is currently a reliable piece of equipment. As black liquor gasification is not addressed in this thesis, the reader is referred to, e.g., Pettersson [14] for a review of studies on this topic. Pettersson and Harvey [70] evaluated the possibility of utilising excess heat from solid biomass gasification in integrated pulp and paper mills as part of a study on system aspects of black liquor gasification. They concluded that biomass gasification with DME production can be both profitable and able to reduce global GHG emissions under the assumption that CCS is applied (stored biogenic CO₂ was assumed to generate an income corresponding to the charge for emitting fossil CO₂). Other authors have studied gasification of wood, or hog fuel, integrated with pulp and paper mills, e.g. McKeough and Saviharju [73], Wetterlund et al. [74] and Consonni et al. [75]. McKeough and Saviharju emphasise the importance of producing pulp in a more efficient manner, i.e., decreasing the specific energy demand, and thereby increasing a mill's ability to export electricity or biofuels as the input of wood is essentially determined and

fixed by the pulp production. In addition, they stress the large potential of converting biomass residues into higher value products.

Tunå et al. [76] evaluated several cases of transportation fuel production from gasified biomass integrated with a pulp and paper mill. The effect of different setups of the biomass-to-fuel production chain was also evaluated in this paper, i.e., three different gasification technologies and three different end products. They concluded that integration of a gasification process with a pulp and paper mill only leads to more efficient use of resources for a few cases compared with corresponding stand-alone cases. In a study by Ljungstedt et al. [77] integration of FT crude with a chemical pulp mill that produces fine paper was evaluated. The integration was combined with energy-saving measures and lignin extraction in the host mill and was compared with stand-alone FT crude production. Both the environmental impacts (GHG emissions) and the economy were included in the evaluation. Their results show that energy-saving measures in the host mill can eliminate the possibility of heat integration of an FT crude plant with the evaluated mill type. Combined with lignin extraction, however, the potential for heat-integrated FT crude production at reasonable sizes ($>200 \text{ MW}_{\text{LHV}}$ biomass input) exists. Integrated FT crude production was shown to create a larger GHG emissions reduction given future electricity production in which the emissions are less than or equal to the emissions from a natural gas combined cycle (NGCC). In another study by the same authors [78], the results concerning a fine paper mill are compared with the corresponding results for a kraftliner mill. The results show that the kraftliner mill has a steam deficit even after extensive energy efficiency measures, which constitute a heat sink for a biomass-to-FT crude plant. Integrated production was shown to yield a higher biomass-to-FT crude efficiency, a lower production cost and a larger potential to contribute to GHG emission mitigation. Andersson et al. have examined pulp mill integrated biorefineries that are based on gasification in two recent studies. In [79], integrated ammonia production was compared with stand-alone production. In a similar study [80], integrated methanol production was compared with integrated production. A third case with black liquor gasification in parallel with biomass gasification was also considered as an option. Haikonen et al. [81] also discuss integration of FT crude production in a pulp and paper mill. The authors evaluated three integrated cases and compared them with a stand-alone FT unit with respect to emissions of CO_2 and mass and energy balances. Biomass was considered to be an unlimited resource and CO_2 emissions are consequently reduced as more biomass is introduced, as coal-based electricity can be replaced or off-gases can replace fuel oil in the lime kiln. Integration of biomass gasification and subsequent usage of the gas directly in the lime kiln or in a gas turbine combined cycle was investigated by Rofouieeraghi [82]. Both alternatives were found viable in terms of CO_2 emissions reduction. A study of gasification-based electricity production in a gas turbine integrated with a TMP mill was studied by He et al. [83] (published after **Paper I** which is dealing with a similar topic). They conclude that electricity production in a gas turbine combined cycle has improved economic performance because of lower specific investment cost compared with conventional boiler-based CHP under current Swedish

conditions. The studied integrated TMP mill was not able to be self-sufficient in electricity with a biomass gasification combined cycle (BGCC) even if the specific electricity consumption was reduced by 50%.

2.5 Research needs

Based on the literature review, the main objectives of this thesis, which are presented in Section 1.2, can be motivated. An economic and environmental comparison of different transportation fuels from pulp and paper mill-integrated thermochemical conversion of biomass has not been identified in the literature. There is a need to study the influence of biomass pretreatment prior to gasification-based forest biorefineries. These types of studies have previously focused on mills based on chemical pulping, while the implications for an integrated TMP mill concerning integration of biomass gasification have not been previously investigated.

3 Studied processes

In this chapter, the different systems described in the appended papers are briefly presented. The chapter begins with a description of the studied pulp and paper mills which is followed by descriptions of the various gasification-based systems that have been modelled. Input data for the evaluation of GHG emission consequences and economic performance, if these systems are realised, are presented in Chapter 4 Methodology.

3.1 Pulp and paper mills

In this thesis, two types of pulp and paper mills have been selected as the host industry for the integration of a gasification-based process, thermo-mechanical pulp production for newsprint and chemical Kraft pulp production with integrated production of liner or paperboard. Biorefinery integration with a market pulp mill without paper production would not provide the same integration opportunities as modern pulp mills are often self-sufficient in heat. A stand-alone paper mill offers a heat sink, but lacks the infrastructural benefits regarding biomass handling and does not offer as many heat integration opportunities as an integrated mill. Although the Kraft (sulphate) pulping process is not the only chemical pulping process, it is the only process considered here as it is the most important process in terms of production volume in Europe [67].

3.1.1 Thermo-mechanical newsprint mill

Thermo-mechanical pulp is produced by crushing and grinding wood chips in electrically driven refiners. Due to mechanical force, heat is generated and moisture evaporates which softens the lignin and facilitates fibre separation. As the lignin is not separated, the pulp yield is as high as 95 wt-%. Mechanical pulp is primarily used for newsprint, and several production lines have had to shut down production during past years due to the shift in how media is distributed. A possible response, which is associated with high risk, may be to diversify and expand the product portfolio. The studied mill in **Paper I** is a Scandinavian, integrated TMP mill with an annual paper production of 930000 t. The mill is co-located with a large sawmill. The placement of a sawmill and a pulp mill in close proximity to each other is relatively common and is therefore of interest also in a general context. The layout of the TMP mill and the sawmill and how they are integrated is shown in Figure 4. The TMP mill has a minimum hot utility demand of 131 MW, of

which 72 MW is provided via recovered steam from the refiners. Of the remaining 59 MW, a heat load of 40 MW has to be provided by low-pressure (LP) steam at 3.5 bar and 139 °C for technical reasons and the remaining 19 MW can be provided at lower temperature levels (refer to Figure 20). The sawmill requires another 16 MW for its dryer, which is constructed to utilise hot water from 120 °C to 95 °C. The total minimum primary heat demand for the site is 75 MW (59 MW for the TMP mill + 16 MW for the sawmill). In addition to steam, the mill consumes 194 MW of electricity, primarily for the refiners.

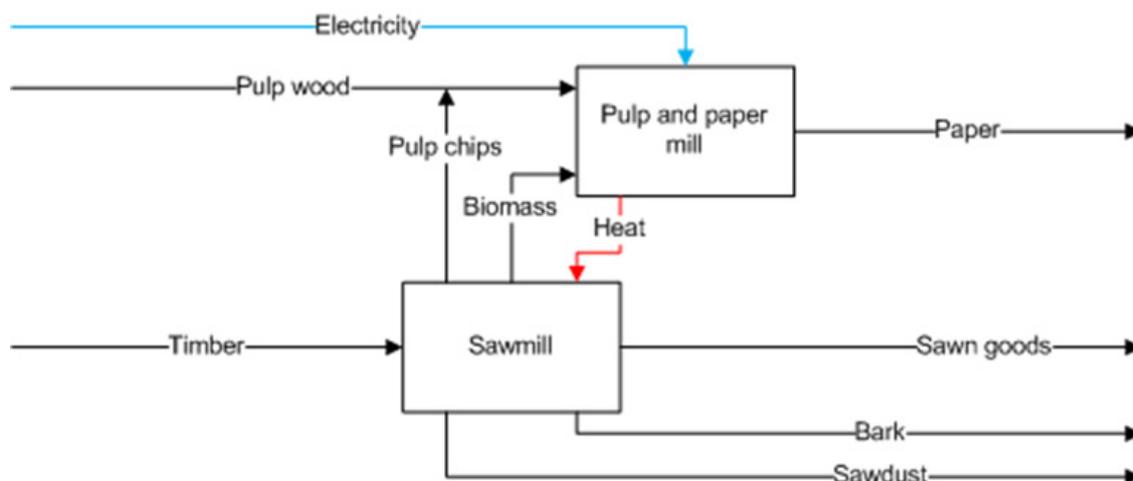


Figure 4. Flows of material and energy in a TMP mill co-located with a sawmill.

Jönsson et al. [84] used the HLMPP tool (presented in Section 4.1) to evaluate the potential for steam savings for four Scandinavian TMP mills. In **Paper II**, a detailed pinch analysis was performed for one of the mills (mill number 2 in the paper by Jönsson et al. [84]) to evaluate how well the model predicts the availability of excess heat from the mill and the potential for heat pumping. Mill number 2 is the mill with the lowest energy savings potential according to the HLMPP screening. The comparison showed that the stream data from the detailed pinch analysis did not entirely correspond with Jönsson's HLMPP results. The model predicted practically no steam savings potential for the mill compared with 17% steam savings by the detailed analysis, and the pinch point was estimated by HLMPP to be at 72 °C compared with 53 °C. The amount of excess heat below the pinch temperature to ambient conditions was estimated to be less. The study presented in **Paper II** was performed to examine the reasons for the discrepancies and to tune the model to better fit reality.

3.1.2 Chemical paperboard mill

Chemical pulp is produced by mixing chemicals with wood chips in a digester. The chemicals break down the lignin without significant degradation of the cellulose. The yield is only approximately 50% due to the removal of lignin but produces a stronger

product compared with mechanical pulp. The lignin/chemical slurry, which is referred to as black liquor, is sent to the recovery cycle where lignin is combusted to recover heat and the chemicals are regenerated. The host mill in **Paper III** and **Paper IV** is an integrated Kraft pulp and paper mill with an annual production of approximately 350000 t of paperboard, predominantly from pine and birch. The mill is equipped with a recovery boiler that produces steam at 110 bar and 515 °C and two supplementary boilers that produce steam at 61 bar and 460 °C from bark and oil. Approximately 14 MW is supplied by oil and 14.5 MW is supplied by tall oil pitch; a by-product from upgrading tall oil to bio-based chemicals. These three boilers are connected to a common steam turbine with two inlets and four outlets at different pressures. Methanol and waste gases from the process are incinerated in a separate boiler, producing another 2.5 MW of steam, at 13 bar. The average current electricity consumption is approximately 51 MW.

The level of excess heat is available at a low temperature - approximately 65 °C - which is within the lower range of what can realistically serve as a heat source for a belt dryer. The final drying of the wood particles when the surface is almost completely dry is difficult at low temperatures. According to an internal source at the studied mill, the bleaching plants can be improved to consume less hot water at temperatures near 80 °C by reusing water between the different bleaching units in a counter-current manner. As a result, hot water generated in the secondary heating system of the mill becomes available for the dryer instead of the bleaching plants. These two flows of warm water were used for the dryer comparison in **Paper III**.

3.1.3 Kraftliner mill

This mill is an integrated kraftliner model mill that was developed during the FRAM program [85] from 2003 to 2005 and was supposed to represent the best available commercially proven technology in the Nordic countries. The liner mill has two paper machines that produce unbleached liner and white top liner. Unbleached Kraft pulp is produced in the integrated Kraft mill, whereas the required amount of bleached pulp is purchased. In addition to the Kraft pulp, considerable amount of recycled fibres is used, especially in the unbleached liner. The Kraft pulp mill produces 2000 ADt/d and a total liner production of 3100 t/d including recycled fibres and purchased bleached pulp. The average electricity consumption is 110 MW. For detailed information about this mill, refer to **Paper V** or the complete report by Delin et al. [85].

3.2 Syngas production

With the exception of the study in **Paper IV**, a direct CFB was the selected gasifier type in all studies. Compared with the EF gasifier in **Paper IV**, the technology is fuel flexible and is relatively easy to pressurise in compared with a DB gasifier. It is also suitable for scaling up, which is particularly problematic with fixed-bed gasifiers. Data for the syngas composition for **Papers I** and **Paper III** were obtained from Hamelinck et al. [20]. For

Paper IV and **V** the syngas composition was based on Aspen Plus [86] models. The assumptions behind the CFB in **Paper IV** were obtained from Hannula and Kurkela [87, 88] and the EF gasifier in **Paper IV** was assumed to reach equilibrium due to the high operating temperature (1300 °C).

Before gasification can proceed, the biomass should be dried from its initial moisture content. This value varies depending on the weather and how the material is stored; however, a typical value of 50% for forest residues and bark has been used throughout this thesis. Although drying can be performed off-site, the purchased forest residues are assumed to have been dried onsite as falling bark from the pulp mill should be dried. Different dryer types were assessed in terms of efficiency, GHG emissions and economy in **Paper III**. The studied concepts included drying with warm air in a belt dryer, drying with flue gases in a rotating drum and steam drying. The warm air for the belt dryer was assumed to be heated with secondary heat from the pulp and paper mill. Certain improvements to the pulp and paper mill may increase the temperature of excess heat flows. This would result in hotter air and a lower air flow through the dryer (refer to Section 3.1.2). A lower air flow translates to lower electricity consumption for the fans. Using flue gases from the recovery boiler was not an option in the studied pulp and paper mill as it is already equipped with heat recovery from the flue gases, i.e., the temperature is too low to be used directly for drying. The exit temperature of flue gases from the dryer was set to 80 °C and the inlet temperature was a result of the heat demand of the dryer and the flow of flue gases.

The feeding issues of the EF gasifier for biomass applications are specifically addressed and discussed in **Paper IV**. In this paper torrefaction or pyrolysis are discussed as pretreatment methods to facilitate the feeding of biomass into the gasifier. The evaluated routes are shown in Figure 5. There are three paths included: extensive milling to small enough particle size to reach a sufficiently short reaction time for the fuel particle, torrefaction prior to milling to decrease the electricity demand for milling, or pyrolysis to produce a pumpable slurry to essentially avoid milling. The choice of pretreatment method affects the energy balance and the composition of the feed to the gasifier. The study did not assess the possibility of decentralised torrefaction or pyrolysis.

For the torrefaction process, information about the fraction of condensable species and the composition of permanent gases was obtained from an article by Tapasvi et al. [47]. The composition of the condensable phase was estimated for larch by Prins [89]; in **Paper IV**, it was assumed to represent an approximate composition for a coniferous wood species. The process was allowed to proceed until the energy requirement was equal to the energy contained in the released volatiles, which were used to provide heat for the endothermic reactions.

The composition of the pyrolysis products was predicted by applying equations suggested by Neves et al. [90], who performed a literature review of pyrolysis characteristics. From these data they developed empirical relationships, which were used in **Paper IV** to

calculate mass and energy balances for the pyrolysis step. The model calculates the heat requirement for the pyrolysis, the amount of ash, and the amount and composition of char and gases (condensable and permanent). Part of the char content was used to provide heat for the pyrolysis process, whereas the remaining part was milled and mixed with the liquid fraction into a pumpable slurry (Henrich and Weirich [91]).

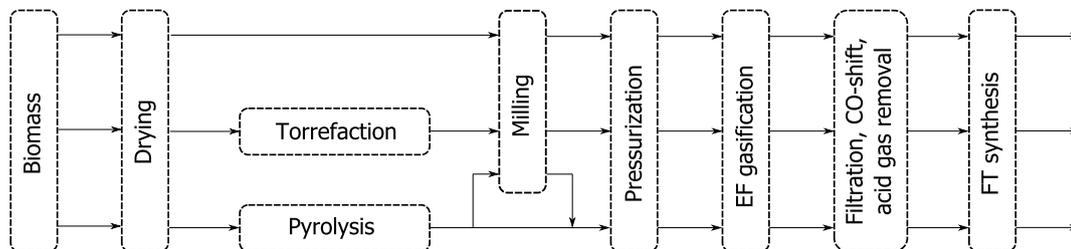


Figure 5. Production routes for the different pretreatment alternatives.

Untreated wood, milled to approximately 1 mm, was pressurised in lock-hoppers, using CO₂ from downstream acid gas removal and was fed to the gasifier with a screw feeder. Consumption of electricity for milling and feeding of the biomass was obtained from Svoboda et al. [92] and van der Drift et al. [93]. In the torrefaction route, the material was milled to a size of 400 µm compared with 1 mm for untreated biomass. The particle size may be smaller as the reactivity of the biomass decreases during the torrefaction process. The torrefied wood was pressurised in lock-hoppers and was pneumatically fed to the gasifier.

As previously described, the remaining char from the pyrolysis was milled (assuming the same specific electricity consumption as for torrefied wood) and mixed with the pyrolysis oil to create a slurry. The power consumption for pumping the slurry was neglected. The permanent gases were fed into the hot raw gas stream after the gasifier.

The WGS reactor was modelled in the same manner for all studies, namely addition of steam in excess assuming complete conversion of CO. The desired H₂:CO ratio was achieved by by-passing the shift reactor with part of the gas flow.

Removal of CO₂ and sulphur were performed by physical absorption in methanol, i.e., consistent with the operation of the Rectisol process [94]. Assumptions regarding steam and electricity demands were obtained from Liu et al. [95].

The process layout for **Papers V** and **VI** was inspired by the layout employed by Hannula and Kurkela [16], as shown in Figure 6, i.e., direct CFB followed by clean tar reforming, WGS, scrubbing of remaining impurities, acid gas removal and synthesis.

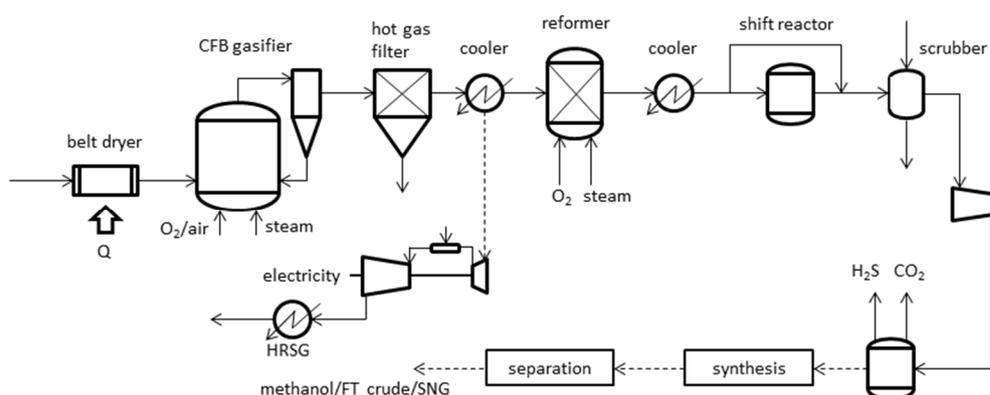


Figure 6. Flowchart showing the main equipment in the front end part of the process from biomass to cleaned syngas and the alternative route to the gas turbine. The dashed lines indicate different end-product alternatives. Steam is generated by cooling hot gases and the synthesis reactor or by cooling the exhaust from the gas turbine and is expanded in a back-pressure turbine for electricity generation.

3.3 Syngas utilisation

3.3.1 Enhanced power production

Electricity production in a gas turbine is a relatively simple way to utilise the produced gas compared with synthesis-based systems, alternatively it is considered to be a complicated method for producing combined heat and power. As mentioned in Section 2.1, commercial plants already exist where gasification-derived gas is used in boiler-based CHP applications, however, more stringent requirements are put on levels of impurities for use in a gas turbine. The benefit with a gas turbine cycle compared with conventional boiler-based CHP is the higher electrical efficiency and higher power-to-heat ratio. Because of that, more electricity can be produced against the same heat demand with tighter constraints on the gas quality. The cleaned product gas is fed to the gas turbine after particulate removal at high temperature to prevent tar condensation (above 350 °C [55]). Particles and some alkali are removed in candle filters or bag filters. Combustion of gas in a gas turbine was the final utilisation of the produced gas in **Paper III**, in which various drying technologies were evaluated. In **Paper I, V** and **VI** it was used as a comparative case in relation to the more complicated synthesis routes. The setups slightly differ between the papers, but look essentially as shown in Figure 7.

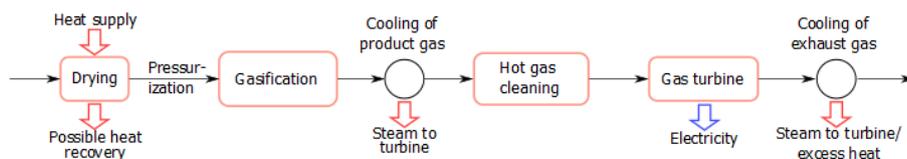


Figure 7. Process scheme of a gas turbine route based on gas production via biomass gasification.

3.3.2 Methanol

In **Paper I** a liquid phase reaction was assumed in order to omit the need for an acid gas removal unit. This assumption was employed to achieve three distinct cases in terms of process complexity. The synthesis was performed at 90 bar and 240 °C with a carbon monoxide conversion of 75%. Unreacted syngas was recycled with a recycle-to-feed ratio of two [19]. The synthesis in **Paper V** was performed in a gas phase reactor at 80 bar, and the purge flow was set to reach a recycle-to-feed ratio of four. The per-pass conversion of CO was limited to 30% [16]. Side reactions to other alcohols or formaldehyde were not modelled. Separation and purification of methanol was conducted using the same method for both systems, i.e., in two flash stages and two distillation columns.

3.3.3 Fischer-Tropsch crude

The Fischer-Tropsch synthesis steps in the appended papers are based on two different literature sources with slight differences. The first process is primarily based on a study by Ekbohm et al. [96] who evaluated the possibility of producing jet fuel from gasified biomass. In **Papers I** and **IV**, the front end of this step, i.e., syngas to FT crude, was utilised. In the two final papers, **Papers V** and **VI**, modelling data from Hannula and Kurkela [16] was instead used. In **Paper I**, the product stream was assumed to consist of solely paraffin, whereas some olefins were introduced in **Papers IV, V** and **VI** in accordance with findings by Fox and Tam [97].

The reaction suggested in the paper by Ekbohm et al. [96] occurs in an isothermal slurry phase reactor at 23 bar and 210 °C. Conversion of CO is assumed to be 90% per pass. Unreacted gas is recycled back to the ATR and the acid gas removal to increase the overall yield. Hannula and Kurkela used 80% per pass conversion at 30 bar and 200 °C. An alpha value of 0.9 was used in both studies.

3.3.4 Synthetic natural gas

In **Papers V** and **VI**, SNG was introduced as an additional alternative biofuel. The SNG is synthesised in three adiabatic reactors with intercooling and a recycle loop over the first reactor for temperature control [98]. Some final upgrading of the gas is necessary to satisfy the required specification for injection into the natural gas grid. The conditioning includes final drying of the gas and the removal of trace impurities.

4 Methodology

This chapter describes the methodology on which the work in the appended papers is based, and presents the background data in this study. The assumptions made to perform the economic and GHG emission evaluations are presented.

The objective of this thesis was to investigate integration opportunities between a gasification process and a pulp and paper mill and to analyse the performance of these processes. As the evaluated gasification systems are not commercially available, process modelling in Aspen Plus has been used to estimate the mass and energy balances of the processes. Using these balances, process integration opportunities have been investigated using a pinch analysis methodology. Certain performance indicators, such as energy efficiency, GHG emission implications and economic performance, have been used to rank the different options. In this chapter, the different elements of the methodology are described.

4.1 Process integration

In this study, two aspects of process integration have been analysed, namely material integration and heat integration. Material integration was primarily performed via the use of different woody residues from the debarking in the pulp and paper mill and from shavings and other residues from a sawmill. The potential for heat integration between the gasification process and the pulp and paper mill was assessed with heat recovery targeting tools using pinch analysis. Pinch analysis is a structured method that is employed to design energy efficient industrial processes with the goal of maximising internal heat recovery (or heat recovery between processes to be integrated) and minimising the need for heating and cooling by utilities. The concept is based on the first and second law of thermodynamics and was introduced in a paper by Linnhoff and Flower [99], which was extended into the first pinch analysis user guide [100]. Recent updates about new developments have been published by Smith [101] and Kemp [102]. The targets for heat recovery potential have been evaluated without constructing the heat exchanger network; instead, a temperature difference has been set for the minimum driving force for the heat transfer between two streams. The thermal streams that are included in the analysis are referred to as hot or cold. By definition, the hot stream is cooled (heat source) and the

cold stream is heated (heat sink). The thermal cascade for a process, or part of a process, i.e., how heat is transferred from higher to lower temperatures, can be visualised in a grand composite curve (GCC). The GCC provides information about minimum requirements for heating and cooling in the process, and at which temperature levels. In the GCC, the pinch temperature can be located at the point at which the net energy demand is zero, i.e., heat has to be supplied above and withdrawn below this level. An example of a GCC is shown in Figure 8.

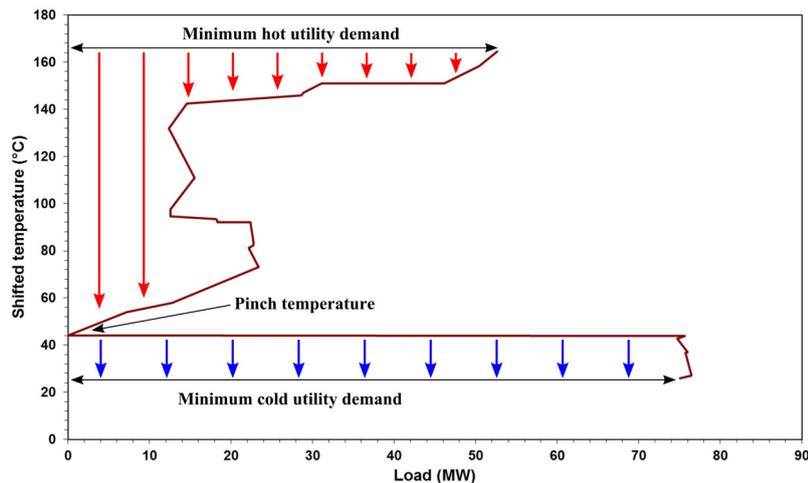


Figure 8. A grand composite curve with the indicated pinch temperature and minimum demand for hot and cold utilities.

Two GCCs can be combined into a split-GCC to determine the potential for heat integration between the processes or process parts. A split-GCC is achieved by mirroring one of the GCCs followed by shifting the two curves along the x-axis until they touch. The point where the curves touch is the pinch point of the system. The mirrored curve is referred to as the foreground curve and the other curve is referred to as the background. Split-GCCs are used to assess and visualise the heat integration opportunities between a gasification process and a pulp and paper mill and to assess the potential to co-generate electricity. An example of a split-GCC is shown in Figure 9; the two processes are depicted by the red-coloured and wine-coloured curves in the left figure. The opportunity to integrate a steam cycle is represented by the black line. This integration will affect the minimum hot utility demand due to the addition of electricity production.

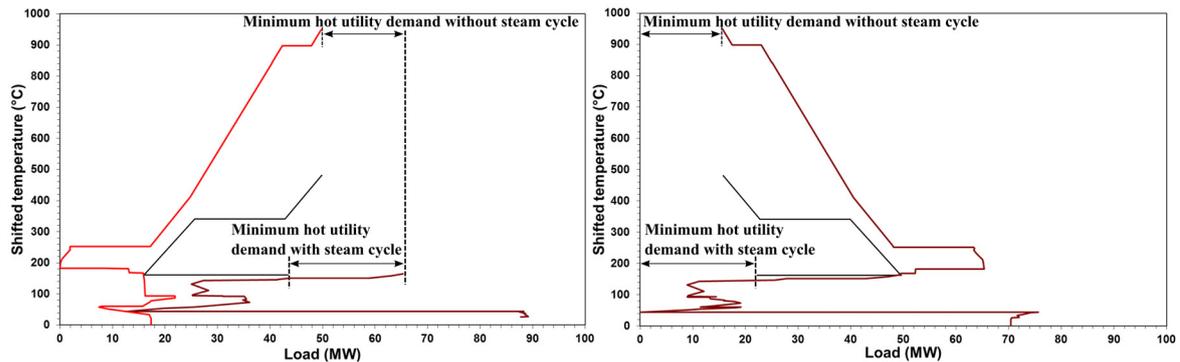


Figure 9. A split-GCC is shown in the left diagram; the background process is depicted in wine and the foreground process is depicted in red. An integrated steam cycle is represented by the black line. The two GCCs from the left diagram are merged into one GCC in the right diagram. The steam cycle is mirrored and represented as a foreground curve.

The split-GCC representation will provide the same results as if the processes were represented by the same curve (shown in the right diagram in Figure 9); however, separating the two curves enables the distinct parts or streams to be observed.

Integration of biomass gasification systems with a pulp and paper mill generally does not affect the pulping process but is integrated via a utility system. Excess heat at high temperatures can be distributed via the steam network, and excess heat at lower temperature from the mill can be used for preheating different streams through a water circuit. The definition of excess heat can be problematic as the amount can be increased by increasing the input of primary energy. This issue was addressed by Bendig et al. [103], who discussed the difference between avoidable and unavoidable excess heat.

The term integrated may cause some confusion due to its dual meanings in this study. The established term for a site with production of both pulp and paper without intermediate drying of the pulp is an integrated mill, compared with a (market) pulp mill or a paper mill. The alternate meaning refers to the situation in which a gasification process is integrated with a pulp and paper mill (i.e., an integrated pulp and paper mill) compared with a stand-alone gasification process. As the three case study mills addressed in this thesis are integrated pulp and paper mills, the word integration is only used to describe an exchange of heat and material between the gasification process and the pulp and paper mill.

The stream data for the pulp and paper mill studied in **Paper I** were retrieved using a software named ‘heat load model for pulp and paper’ (HLMPP). The tool was developed by Hakala et al. [104] to provide a method for a quick scan of heat integration opportunities in mechanical pulp and paper mills. The approach is based on energy balance modelling in BALAS [105], Microsoft Excel for data manipulation and Pro_PI [106] for construction of the heat load curves. The predictability of the tool has been

tested using a ground wood mill [104], a simulated TMP mill in [107] and a real TMP mill in **Paper II**.

4.2 Process performance

Process performances for different process alternatives have been evaluated based on their energy efficiency (**Papers IV and V**), GHG emissions balance (**Papers I, IV and VI**) and economic performance (**Papers III and VI**).

4.2.1 Thermodynamic performance

To compare different biorefinery process integration alternatives, the biomass resource efficiency of the processes is calculated as follows:

$$\eta_{biomass} = \frac{\text{biofuel}}{\Delta \text{biomass} + \frac{\Delta \text{electricity}}{\eta_{biomass-to-electricity}}} \quad 8$$

This equation compares the produced amount of biofuel with the increased use of biomass resources compared with the reference pulp and paper mill, i.e., compared with the biomass that would be required in the bark boiler. The change in electricity demand/production is recalculated to primary energy, i.e. a biomass equivalent, for a range of biomass-to-electricity efficiencies. This value indicates in what system the biomass is most effectively used. The conversion of electricity to biomass equivalent is not connected to the build margin technology for electricity production.

4.2.2 Economic evaluation

The biorefinery concepts were evaluated in economic terms using the net annual profit (NAP) defined according to Equation 9. The variables C_x and P_x are the price and energy flows for products, electricity and biomass feedstock; t is the operating time; I is the total capital investment (€); r is the annuity factor (1/y); m_{CO_2} is the flow of CO₂ (t/y) and PI_{CO_2} is the policy instrument to support CCS (€/t), i.e., the CO₂ charge. Cash flows are assumed to be the same each year, and the entire investment is completed within the first year, which means that this method gives the same result as the net present value (NPV) method. The annuity factor (or capital recovery factor) r is a function of project life time and interest rate (Equation 10); it was assumed to be 0.1 as a base case (for example, an economic life time of 20 years and a rate of return of 7.8%), which represents a strategic investment for the company.

$$NAP = t \cdot (P_{product} \cdot C_{product} + P_{el,net} \cdot C_{el} - t \cdot P_{biomass} \cdot C_{biomass}) - r \cdot I + \dot{m}_{CO_2} \cdot PI_{CO_2} \quad 9$$

$$r = \frac{i(1+i)^n}{(1+i)^n - 1} \quad 10$$

Capital costs for process equipment were obtained from the literature (refer to the appended papers for detailed descriptions). Cost scaling exponents (k) for different equipment were used to scale the reference capital cost (C_0) to the evaluated unit size (S), as shown in Equation 11.

$$C = C_0 \cdot (S/S_0)^k \quad 11$$

All equipment costs have been recalculated to the selected monetary value (2010 in **Paper III** and 2012 in **Paper VI**) for the study with the composite Chemical Engineering Plant Cost Index (CEPCI) [108]. For the economic evaluation in **Paper III**, a fixed economic support of 20 €/MWh for renewable electricity production was assumed to be established. In this case, the entire production of renewable electricity was assumed to be sold and the demand is re-purchased from the grid at a lower price. In **Paper VI**, no directed supports were assumed; only the CO₂ charge.

The price of energy commodities, such as fossil fuels and biomass were retrieved from the Energy Price and Carbon Balance tool (the ENPAC tool) [109]. The tool uses built-in energy market models to determine prices for biomass, district heat and biomass-based fuels. The tool also determines the build margin technology for electricity production. The tool was used with a recent update in **Paper VI** [110], which includes new standard input data for fossil fuels. The projections for fossil fuel prices and level of CO₂ charge, which serves as input to the tool, was obtained from the World Energy Outlook [111]. The tool was also updated to include emissions from methane and nitrous oxide in addition to CO₂. Refer to Table 1 for some numerical values from the tool used in **Papers III** and **VI**. Given that no dedicated subsidies for electricity and biofuels are assumed to be established, the price for low-grade wood fuel (forest residues) is assumed to be set by the willingness to pay of a coal power plant co-firing biomass, as a result of positive CO₂ consequences when coal is replaced by biomass. As the potential demand for wood fuel from these plants is large in relation to the availability of biomass [112], they are possible candidates to become the price-setting users.

An important assumption in all studies in this thesis regarding the costs of CO₂ emissions is that the CO₂ charge is harmonised, i.e., it is assumed to be identical for all emitters across Europe.

Table 1. Economic figures concerning **Paper III** (in €2010) and **Paper VI** (in €2012) for 2030.

	Energy carrier				Policy instruments	
	Forest residues [€/MWh]	Fuel oil [€/MWh]	Electricity [€/MWh]	District heating [€/MWh]	Renew. el. Support [€/MWh]	CO ₂ charge [€/t]
low/low	25	44	53 ^a	42	20	27
high/high	48	82	76 ^a	75	20	85
new policies scenario	18.0	76.2	65.9	-	-	25.7
450 ppm scenario	32.7	80.8	91.8	-	-	73.9

^a Not including support for renewable electricity

4.3 System boundaries and climate performance

As discussed in Section 1.1, climate change is one of the major issues of our time; GHG emissions are an important process performance parameter in this study. The emissions of CO₂, CH₄ and N₂O were converted to CO₂-equivalents (CO₂eq) using global warming potential (GWP) factors over a 100-year period (GWP₁₀₀ conversion factors: CO₂ = 1, CH₄ = 25, N₂O = 298). The GHG reduction potential has been calculated using an expanded system that includes emissions reduction from replacing fossil energy with products from the biomass-based production route. Figure 10 illustrates this system.

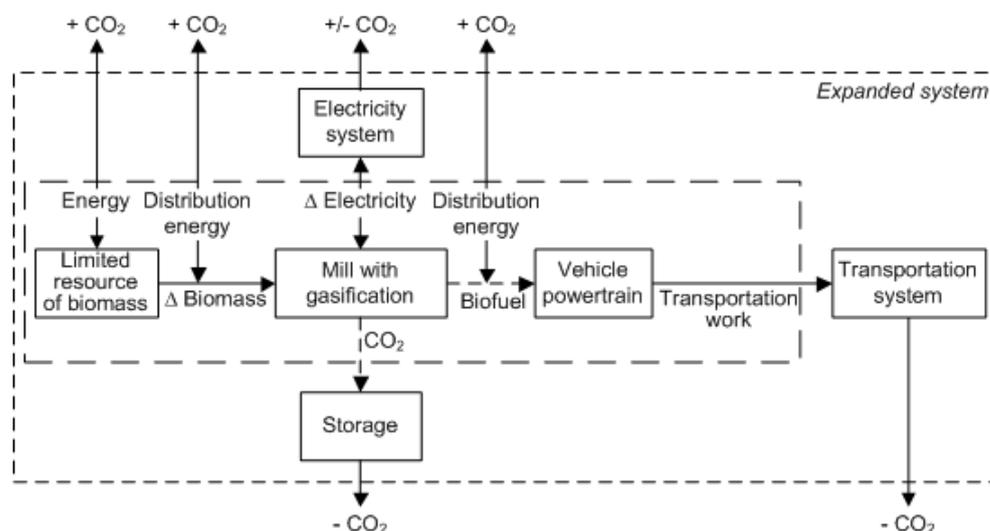


Figure 10. Schematic representation of net flows that enter or leave the mill and their interaction with the surrounding system. The CO₂ effect of each flow is indicated with +/-, where + denotes an increase and - denotes a decrease in CO₂ emissions.

Biomass

Emissions of CO₂ from biomass over a life cycle were assumed to be zero in this study, with the exception of fossil CO₂ emissions from biomass transportation. This assumption has been debated as it is dependent on how the forests are managed. In a recent report from the UK Department of Energy [113], the authors discuss GHG emissions from biomass-based electricity production from a lifecycle perspective. A large proportion of the biomass feedstock in the UK's electricity production sector is likely to be imported from North America; therefore, an evaluation of the GHG impact of this trade is critical. They claim that examining cultivation, harvesting, processing and transport is not sufficient for obtaining the true GHG intensities of different feedstocks and technologies. To obtain the complete picture, changes in carbon stock in a forest and indirect impacts should be considered. In this study, only the transportation of biomass was assumed to cause GHG emissions, which accounts for 7.9 kgCO₂/MWh [114]. Although biomass probably cannot be considered a limited resource today, it is likely to become limited in the near future due to an increased awareness of the problems connected to fossil fuel use. It is therefore important to compare GHG mitigation results from studies like the one presented in this thesis with emissions from alternative use. In this thesis, alternative use of biomass has been suggested to be a coal power plant co-firing biomass, thus reducing CO₂ emissions with approximately 401 kg/MWh_{fuel}. Biomass can however replace coal in other systems with the same reduction potential, assuming the same conversion efficiency for biomass and coal.

Biofuels

Biofuels are introduced on the market to replace fossil fuels and decrease overall GHG emissions. Fossil fuel consumption is assumed to decrease by the same amount (on an energy basis) as the biofuel supply increases. GHG emission consequences associated with biofuel are compared with the fossil equivalent which it can be assumed to replace to determine the net effect. Selecting the corresponding fossil alternative is sometimes straight forward, e.g. synthetic diesel is likely to replace fossil diesel, whereas SNG may replace natural gas, petrol or diesel.

As FT crude is the final product from the evaluated gasification systems and not a finished commodity, it cannot be considered to be a direct replacement of a fossil fuel. The emissions reduction is instead assumed to be the amount of "green" CO₂ that is released during complete combustion of the FT crude, which is approximately 255 kg CO_{2,eq}/MWh depending on the system. Additionally, it can be assumed that emissions associated with extraction of fossil crude oil can be avoided. Depending on the extraction method, this value can substantially vary but was assumed to be 23 CO_{2,eq}/MWh in this study [115]. Upgrading and distribution of the fuels in the refinery can be assumed to have the same GHG impact regardless of the source. Biomass-based FT crude could in fact have lower climatic impact at the refinery compared with the fossil equivalent as its lower sulphur content translates to a lower need for hydrogen processing [116].

In **Paper VI**, two possible fossil alternatives were employed for each renewable alternative, to capture some of the uncertainties connected to this selection. Regarding FT crude, diesel was also employed in addition to what was described in the previous paragraph. In **Paper I**, methanol was assumed to replace petrol and as a second alternative in **Paper VI**, it was assumed to replace fossil methanol derived from natural gas. GHG emissions for fossil methanol was calculated as the sum of emissions associated with processing from methane [57] in addition to complete combustion. SNG was assumed to replace either petrol or natural gas in **Paper VI**.

Electricity

A net surplus or deficit of electricity was assumed to affect the marginal electricity production. As the timeframe for the gasification projects is relatively distant, a base load build margin is considered instead of an operating margin. The base load build margin is defined as the type of electricity generation grid capacity addition that is affected by implementation of the gasification project in question. For GHG emissions associated with the different build margin technologies, refer to Table 2.

Table 2. Marginal electricity production technologies and associated CO₂ emissions [109]. Values from the updated ENPAC version [110] in **Paper VI** are shown in brackets.

	Coal power	Coal power w. CCS	NGCC
CO ₂ emissions [kgCO ₂ /MWh _{el}]	680 (805)	129 (240)	329 (376)

5 Results

In this chapter, the most important results from the appended papers are summarised and commented. The results are presented based on the focus of the process integration study, beginning with stand-alone versus integrated gasification routes followed by the influence of various pretreatment methods and end products. An evaluation of the impact from energy efficiency measures at the pulp and paper mill is included. The results from the study in which the pinch analysis tool is used to retrieve stream data in Paper I are presented.

5.1 Stand-alone versus integrated production of biofuels and electricity

The introduction to this thesis (Chapter 1) suggested that co-locating a gasification-based biorefinery with an integrated pulp and paper mill may be interesting due to the distinct opportunities to exchange heat between the two processes. Biomass residues from different parts of the pulp and paper mill and a possible adjacent sawmill can constitute part of the raw material demand for the new gasification-based process. Electricity can be co-produced to maximise overall efficiency, which was shown to impact the potential for CO₂ emissions reduction in **Paper I**. This paper suggested that the extra biomass needed for the new gasification process, compared with the base-case CHP operation in the mill may alternatively be used in a stand-alone plant for production of a biofuel and/or additional electricity. Excess heat from the stand-alone plant was used to generate electric power in a condensing turbine.

The two options, stand-alone and integrated biorefinery, were compared in terms of CO₂ reduction potential for three alternative build margin technologies for electricity production. As shown in Figure 11, an integrated process may not be better than a stand-alone plant from a climatic point of view. According to these results, the build margin electricity production technology determines whether integration is preferable, from a CO₂ emissions perspective. If the marginal electricity production is high-emitting (in this case a coal power plant), stand-alone operation has the best CO₂ reduction potential as the total green electricity production is higher in the stand-alone case due to the excess heat utilisation in the condensing turbine. In this case, the production of biomass-based

electricity on-site is preferred to purchasing from the grid. If build margin electricity is produced via low-emitting technologies, such as coal power with CCS or an NGCC, maximising biofuel production at the expense of producing less electricity seems to be better choice. For the gas turbine case, electricity is the only product and is better produced in an integrated unit than in a stand-alone plant, regardless of the build margin technology. The cleaner the build margin technology is, the better it is with integrated processes. Only the comparison between stand-alone processes versus integrated production is discussed in this section. The results for the different end products shown in Figure 11 are discussed in Section 5.3.2.

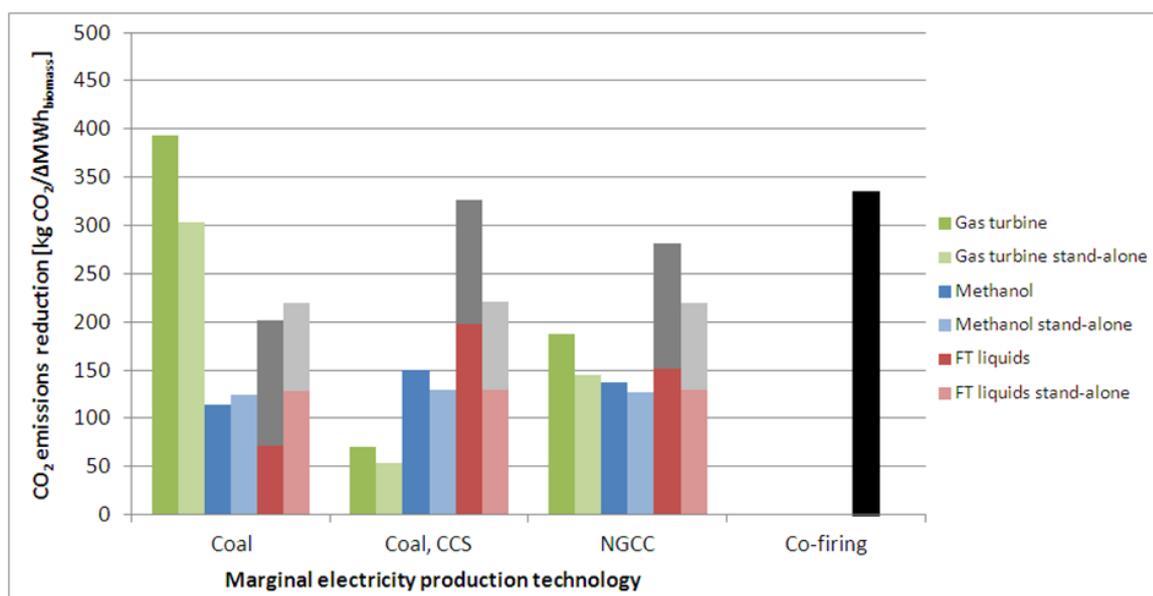


Figure 11. Specific CO₂ emissions reductions of integrated and stand-alone processes, considering the three marginal electricity production technologies presented in Section 4.3. The grey bar on top of a red bar indicates the reduction potential if the separated CO₂ from the FT process is captured and stored. The specific CO₂ emissions reduction of co-firing biomass in a coal power plant is also shown.

5.2 Influence of the selection of pretreatment method for gasification of biomass

Two studies about the choice and influence of biomass pretreatment method prior to gasification are discussed in the appended papers. **Paper III** focuses on various drying technologies and their impact on efficiency and economy, and **Paper IV** focuses on pretreatment methods that primarily facilitate the feeding of biomass into an EF gasifier. In **Paper III**, the end utilisation of the gas was increased electricity generation in a gas turbine combined cycle, whereas FT crude was the main product in **Paper IV**.

5.2.1 Impact of drying technology on efficiency and economic performance

In **Paper III** the impact in terms of economy and efficiencies of different drying technologies in connection with integration of biomass gasification-based electricity production were assessed. The evaluation in this paper also included a case in which heat pumping is used to increase the temperature of the drying air. Additionally, the choice of the oxidising agent was included as a parameter that affects the heating value of the gas and the steam and electricity balance of the biorefinery.

The three evaluated dryer types were belt dryer (at two temperature levels: 60 °C and 80 °C), drying with flue gases from the HRSG, and superheated steam dryer. If flue gases are used to dry biomass, less steam can be produced in the HRSG, i.e., less electricity will be produced in the back-pressure steam turbine (refer to Chapter 3 for assumptions). As the heat demand of the pulp and paper mill determined the size of the gasification system in **Paper III**, the solution with a flue gas dryer would have to be larger in terms of biomass input compared with the other two dryer options.

The steam dryer requires 18 bar steam and delivers steam back to the mill at 4 bar which causes an unavoidable penalty in terms of electricity production in the back-pressure turbine. The gasification system has to be slightly oversized in terms of steam generation, compared with the bark boiler which it replaces due to the efficiency of the steam dryer. A steam flow of 107 t per hour has to be extracted at 18 bar to enable the system to purge 103 t per hour at 4 bar, which is the requirement of the pulp and paper mill.

Using excess heat at 80 °C instead of 60 °C to heat the drying air for the belt dryer, the air flow passing through the dryer belt can be reduced by 20%, which reduces the power demand for the air fans and the size of the dryer. The electricity consumption of the fans is small in relation to other consumers, and the improvement of the electrical efficiency of the BGCC is therefore insignificant (0.1 %-points).

Table 3 summarises some key results for the dryer comparison. In terms of electrical efficiency, the penalty for the steam dryer and the heat pump can be seen. The flue gas dryer is less efficient than air dryers as less steam can be raised in the HRSG.

Under the preconditions for **Paper III** (presented in Section 3.2), air-blown gasifiers and subsequent equipment can be designed with a smaller throughput compared with oxygen-blown gasifiers for two reasons: the specific power output from the gas turbine is higher for oxygen-blown systems, i.e., first, less heat leaves with the exhaust; second, steam has to be extracted from the turbine to fluidise the gasifier bed material.

Table 3. Comparative figures for different pretreatment alternatives for BGCC integrated with a paperboard mill.

	Biomass input [MW]	Power production [MW]	Flow of drying media [m³/s]	Electrical efficiency [%]
Biomass boiler	93	19.3	-	20.7
OB air dryer 60 °C	158	64.3	673	40.7
OB air dryer 75 °C	158	64.6	521	40.9
OB flue gas dryer	176	70.5	237	40.1
OB steam dryer	163	60.0	-	36.8
OB heat pump	158	60.7	430	38.4
AB air dryer 60 °C	139	56.5	591	40.6
AB air dryer 75 °C	139	56.8	458	40.9
AB flue gas dryer	147	59.3	201	40.3
AB steam dryer	143	51.6	-	36.1
AB heat pump	139	53.3	378	38.3

The performance of the dryers can also be expressed based on the improved performance for each unit of biomass that is added to the system. Based on previous discussions, biomass is likely to become a limited resource in the future and should always be compared with alternative use. As shown in Figure 12, the air-blown systems utilise the incremental biomass input more efficiently than oxygen-blown systems, i.e., more power is produced in specific terms. Flue gas dryers and steam dryers lack power production. It should be noted that the higher power consumption for the heat pump cases compared with corresponding air dryer systems without a heat pump.

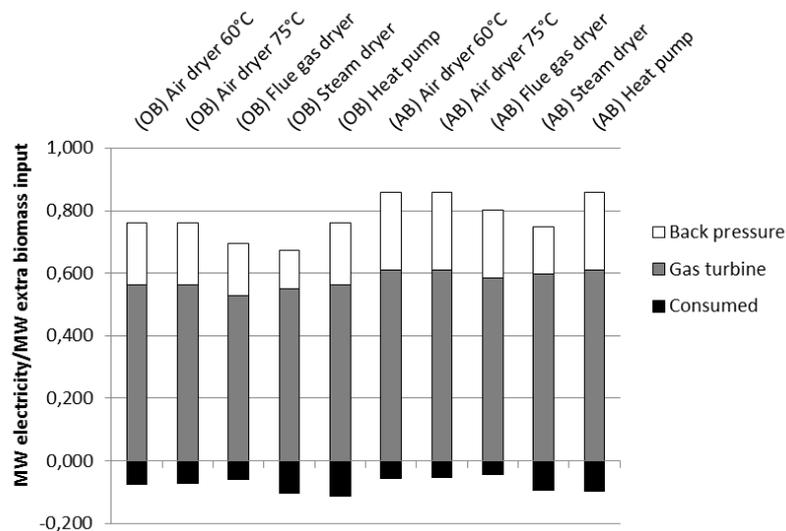


Figure 12. Electricity production/consumption per unit of biomass input.

The capital cost of equipment is detailed in the Appendix of **Paper III** and summarised in Figure 13, where the different fractional contributions are visualised. The overall investment cost ranges from 100 million Euros to approximately 140 million Euros. Although the choice of dryer technology affects dryer cost the dryer share is comparatively small based in the total investment cost. As the dryer is not the major contributor to expenses the savings in air flow at higher drying temperatures has a minor impact on the economy. The cost to upgrade or modernise the bleaching plant is difficult to estimate and is not included in the capital cost for the case with air drying at 75 °C. To break even with the more expensive dryer that operates at 60 °C, using the oxygen-blown case as an example, the allowed capital cost to upgrade the bleaching plant ranges between 650000 and 1010000 Euros depending on price levels (-15%/+30%). The ASU would be a major expense for the oxygen-blown case, whereas more expensive air compressor for the air-blown case compared with the oxygen compressor, compensates for this expenditure.

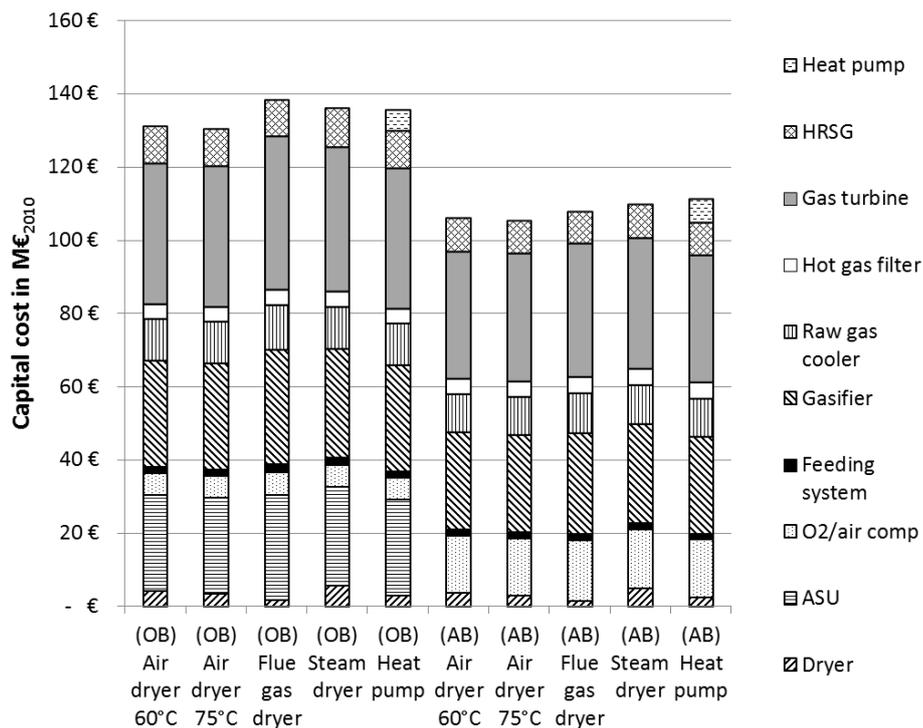


Figure 13. Distribution of capital investment cost according to the different process parts.

Figure 14 shows economic performance in terms of net annual profit with an annuity factor of 0.1. The oxygen-blown CFB encounters problems when competing with the air-

blown CFB despite the higher electricity production. The belt dryers performed well in the air-blown cases, whereas the flue gas dryer experienced difficulties. The lower specific investment does not compensate for the larger fuel input.

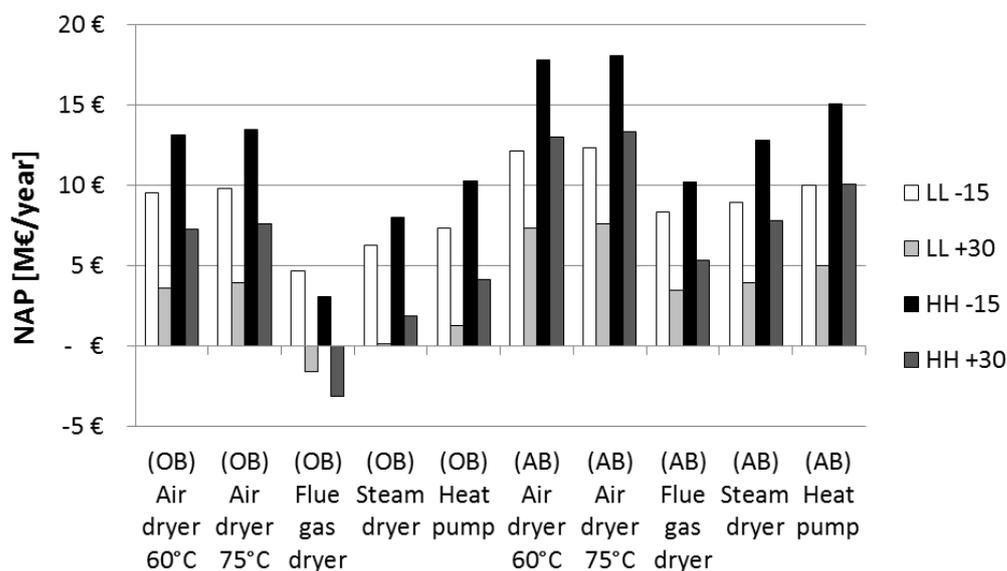


Figure 14. Net annual profit for different cases at different energy market levels and investment costs for an annuity factor of 0.1. (LL: low fossil fuel price level, low CO₂ emission cost; HH: high fossil fuel price level, high CO₂ emission cost; OB: oxygen-blown; AB: air-blown)

Although drying with steam offers interesting benefits in terms of pollution control and low risk of fires, it cannot economically compete due to the reduction in electricity production. A similar reasoning applies to the heat pump case, in which the cheaper dryer (due to the lower air flow) cannot compensate for the heat pump cost.

The assumption that excess heat in the exhaust gases from the HRSG can be used for district heating significantly impacts the results. The value of district heating for a maximum of CO₂ charge and fossil fuel price may be too optimistic (refer to Table 1). The way excess heat is valued affects the comparison, especially for the flue gas dryer as this dryer type does not enable any additional heat recovery.

5.2.2 Pretreatment to facilitate milling and feeding for EF gasification

In **Paper IV**, three different biomass pretreatment methods that were implemented prior to EF gasification and subsequent FT crude synthesis were studied (refer to Section 3.2 for pre-requisites). Additionally, the implementation of a recycle loop over the synthesis reactor to increase the CO conversion was examined. The purge gas was assumed to be combusted in a boiler to produce steam, i.e., the recycle loop increases the biomass-to-FT crude efficiency but decreases onsite electricity production. The process routes were

evaluated in terms of biomass resource efficiency and potential for global GHG mitigation. A biomass input of 300 MW_{HHV} was enough to completely satisfy the heat demand of the pulp and paper mill, which is currently provided by a bark boiler. The steam excess was expanded in a condensing turbine, as mentioned in Chapter 3. The contribution of the condensing turbine is too small to justify an investment, in some cases (refer to Table 4).

Table 4. Output of FT crude and off-gas and the electricity balance, are listed for the six cases. All cases have a biomass input of 300 MW_{HHV}.

	Milling		Torrefaction		Pyrolysis		
	Ref. case	Once-through	Recycl.	Once-through	Recycl.	Once-through	Recycl.
Biomass usage [MW _{HHV}]	85	300	300	300	300	300	300
CGE [%] ^a		77.9	77.9	78.2	78.2	77.2	77.2
FT crude [MW _{HHV}]	-	128.9	157.0	129.4	157.7	127.7	156.8
FT yield [%]	-	43.0	52.3	43.1	52.6	42.6	52.2
Off-gas [MW _{HHV}]	-	51.6	2.9	51.7	2.9	51.1	2.8
Electricity [MW]							
<u>Production</u>							
Back-pressure	15.7	20.1	12.7	20.1	13.0	21.0	13.7
Condensing	-	11.0	6.4	10.6	5.7	12.5	7.8
<u>Consumption</u>							
PP mill ^b	7.4	7.4	7.4	7.4	7.4	7.4	7.4
FT process (w/o or w/ CCS)	-	22.7/28.0	24.3/30.3	19.2/24.0	21.6/26.8	11.1/15.4	12.8/17.6
Excess/deficit	8.3	1.0/-4.3	-12.6/-18.6	4.1/-0.7	-9.5/-14.8	15.0/10.7	1.3/-3.5

$$^a \text{ Cold gas efficiency (CGE)} = \frac{\dot{m}_{\text{syngas}} \text{HHV}_{\text{syngas}}}{\dot{m}_{\text{biomass}} \text{HHV}_{\text{biomass}}}$$

^b Excluding the demand satisfied by electricity production from the recovery boiler (43.6 MW).

Consumption of electricity does not significantly vary depending on whether off-gases are recycled, with the exception of the electricity required for tar cracking and acid gas removal in the recycle loop. The production side, however, substantially differs depending on if off-gases are recycled to improve yield or if the gas is combusted for steam generation. The impact of this outcome on the biomass resource efficiency is shown in Figure 15. As the majority of cases have a deficit of electricity compared with the pulp and paper mill, they benefit from a high electrical efficiency. The exception is

the case with pyrolysis pretreatment and once-through synthesis of FT crude. Thus, if grid electricity is produced with low efficiency it is better to produce it on-site at the expense of a lower FT crude yield. The higher yield and lower electricity consumption of the torrefaction routes results in the higher efficiency compared with only milling the biomass. Future biomass-based electricity production is not expected to have an efficiency less than 35%, which suggest that high conversion into FT crude, i.e., recycling of off-gases, should be favourable. Implementation of CCS reduces the efficiency with four to five percentage points due to the added electricity demand for CO₂ compression.

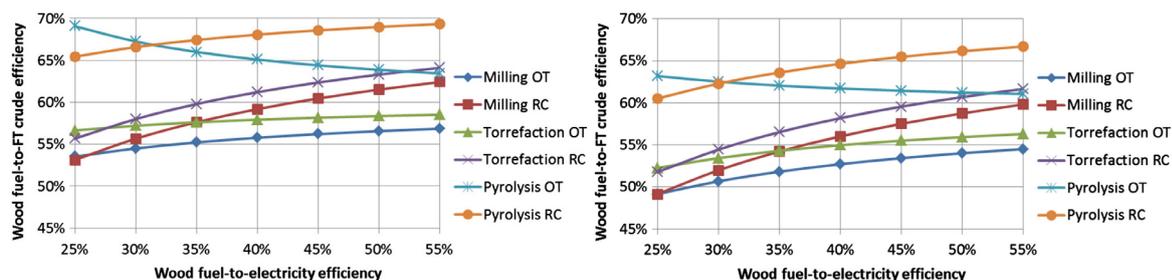


Figure 15. The left diagram shows the wood fuel-to-Fischer-Tropsch crude efficiency plotted against a range of efficiencies for wood fuel to electricity. The right diagram includes the influence of carbon capture and storage.

The various pretreatment options were compared regarding their climatic impact, which is presented as an absolute reduction potential of CO₂ emissions. In contrast to the CO₂ evaluation in **Paper I**, in which the cases were evaluated for three build margin technologies for electricity production (Figure 11 and Figure 22), the cases in **Paper IV** were evaluated for a range of CO₂ emissions associated with electricity from the grid. As shown in Figure 16, the difference between the cases increases when off-site GHG emissions increase. For comparison, a condensing coal power plant with CCS emits approximately 259 kgCO_{2eq}/MWh_{el} and an NGCC without CCS approximately 376 kgCO_{2eq}/MWh_{el} [109]. Given that the power production sector has the largest technical and economic potential for GHG mitigation in Europe [5], maximising yield by recycling off-gases seems to be the best option for the future, from a systems perspective. Only CO₂ from the syngas treatment was included in the CCS cases. The separation of CO₂ from flue gas streams was not included. The interrelationship between the cases regarding GHG emissions reduction potential shifts depending on the implementation of CCS. As a comparison, co-firing the same amount of biomass (215 MW in addition to original requirement in the pulp and paper mill) in a condensing coal power plant would decrease annual CO₂ emissions by 580000 t per year. As biomass can be assumed to be a limited resource in the future, this comparison is relevant. Resources should be utilised in cases in which their positive impact is maximised.

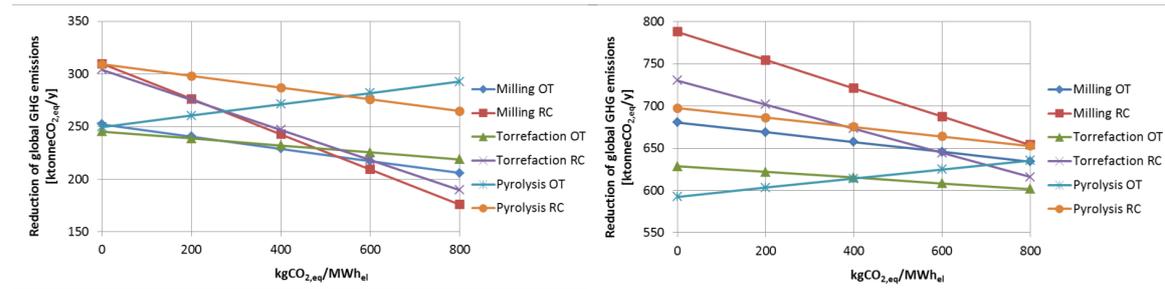


Figure 16. GHG emissions consequences of the various cases. Mitigation potential is shown as a function of GHG emissions associated with the production of grid electricity.

The results from the torrefaction study showed that the CO content in the gas increase from torrefied material, was evidenced by a recent study by Tapasvi et al. [117]. Consequently, a more extensive WGS is needed to satisfy the requirement of the FT synthesis. No economic investigation was performed for the mentioned pretreatment methods as both torrefaction and pyrolysis can be considered emerging technologies at the discussed scale; as a result, such a comparison would be uncertain.

5.3 Influence of different end-products

The influence of the chosen end-product on heat integration potential was evaluated in **Papers I** (TMP mill) and **V** (kraftliner mill). Different sizing constraints of the gasification trains were used in these papers. In **Paper I**, the processes were sized to ensure that excess heat matched the heat production in the bark boiler, i.e., the size will vary depending on which end-product that is produced. As excess heat levels differ between different routes, the processes will have different throughput. In addition to the variable size, a fixed and oversized system was used in **Paper V**. The required size (represented by the biomass input in MW_{LHV}) to exactly replace the bark boiler is dependent on both the conversion efficiency of the biorefinery and the shape of the GCC of the gasification process. For cases in which more energy ends up in the end product, less excess heat, which can produce steam for the pulp and paper mill, should be generated. This is valid for **Paper V** but is not entirely valid for **Paper I**. As was shown in the two papers, the end product from the gasification process has a significant effect on the necessary biomass input if the sizing requirement is to completely replace the steam produced in the bark boiler. Co-generation of electricity did not have a linear dependence on biomass input due to its influence by the shape of the GCC, as demonstrated by the comparison of methanol and SNG in Figure 17. Although the thermal input is identical, the lower cooling temperature of the isothermal methanol synthesis reactor compared with the cooling of the exhaust flow from the adiabatic SNG synthesis reactors at higher temperature results in more co-produced electricity.

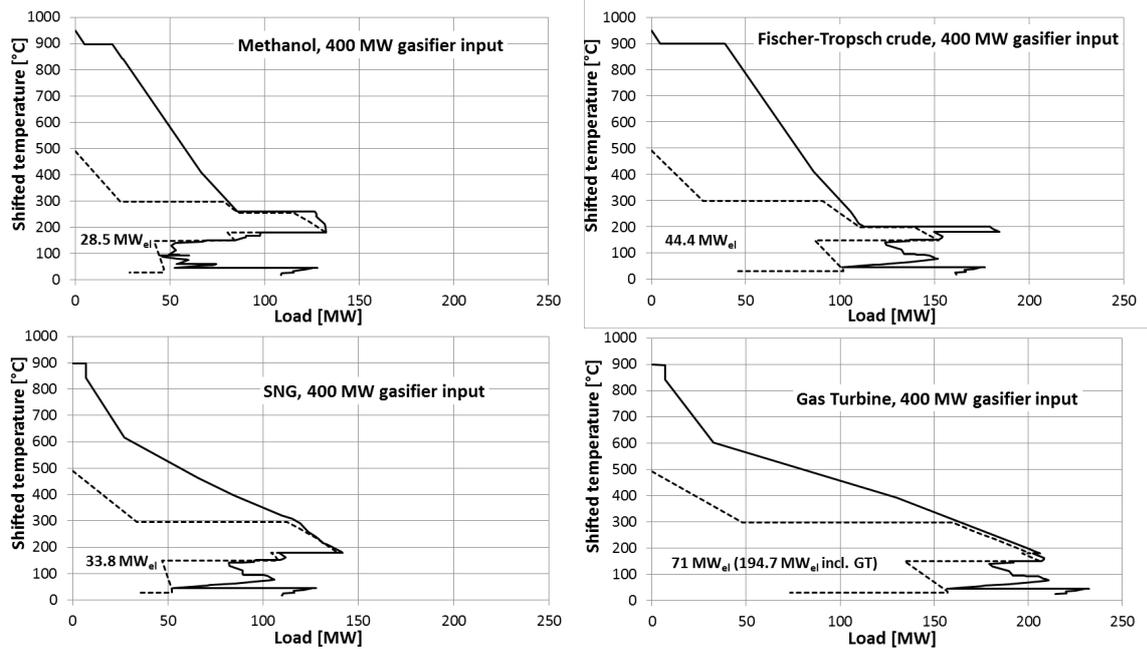


Figure 17. Evaluation of heat integration potential for the 400 MW_{th} cases using split GCC.

5.3.1 Kraftliner mill

The results from sizing constraint to exactly replace the bark boiler with excess heat from the gasification process are shown in Table 5. The end-product from the gasification process has a significant effect on the necessary biomass input if the sizing requirement is to completely replace the steam produced in the bark boiler. As previously mentioned, less excess heat should be generated, which can produce steam for the pulp and paper mill, in cases in which more energy is contained in the end product. This finding is confirmed by the fact that the FT process, which has a large off-gas flow, requires a biomass input of only 158 MW, whereas methanol and SNG production require a maximum biomass input of 280 MW.

Table 5. Summary of key results from **Paper V** (EM=efficiency measures). "Exact match" refers to the case in which the gasification process was sized to replace the heat production from the bark boiler.

		Biomass [MW]	Biofuel [MW]	Energy yield [%]	Co- generated electricity [MW]
Methanol	Ref - exact match	255	159	62.5	14.9
	400 MW biomass	400	250		28.5
Fischer-Tropsch	Ref - exact match	158	91	57.6	10.3
	400 MW biomass	400	230		44.4
SNG	Ref - exact match	280	198	70.8	19.8
	400 MW biomass	400	283		33.8
BGCC	Ref - exact match	120	-	-	49.8 (12.7)a
	400 MW biomass	400	-	-	194.7 (71.0)a

The results for GHG emissions balances and economy are presented in Figure 18. The upper diagram in Figure 18 shows the best use of each unit of biomass resource by presenting the GHG emissions per additional biomass usage, i.e., extra biomass required in addition the original requirement for the bark boiler. As described Section 4.3, the contributions originate from biomass (marginally depicted in the figure), the effect of the build margin for electricity generation, the fossil fuel substitute, and the sequestered CO₂ from the syngas conditioning. The results for the assumed conditions in the year 2030 and 2050 are presented in Figure 18 and in **Paper VI**, respectively. The results only marginally changed regarding GHG emissions between the two years as the only difference was seen in marginal electricity production technology in the *450 ppm scenario*. The marginal technology switched from NGCC to coal power with CCS and a comparison shows that the results are insensitive to these variations. Assuming zero emissions technology as the build margin would result in larger differences. CCS is assumed to be available for the power sector in 2050; thus, this option is more probable for the gasification processes for this year assuming that the necessary infrastructure will be constructed at the location of the pulp and paper mill. The high CO₂ charge in the *450 ppm scenario* in 2050 (144 €/tCO₂) results in increased competition for biomass from the power production sector, which increases the price for biomass. As the biomass price is determined by the willingness to pay of a coal power plant co-firing biomass, the biomass price is strongly connected to the CO₂ charge (refer to Chapter 6 for a discussion regarding this assumption). As a result, storage of CO₂ is a necessity to achieve a profitable case in this scenario.

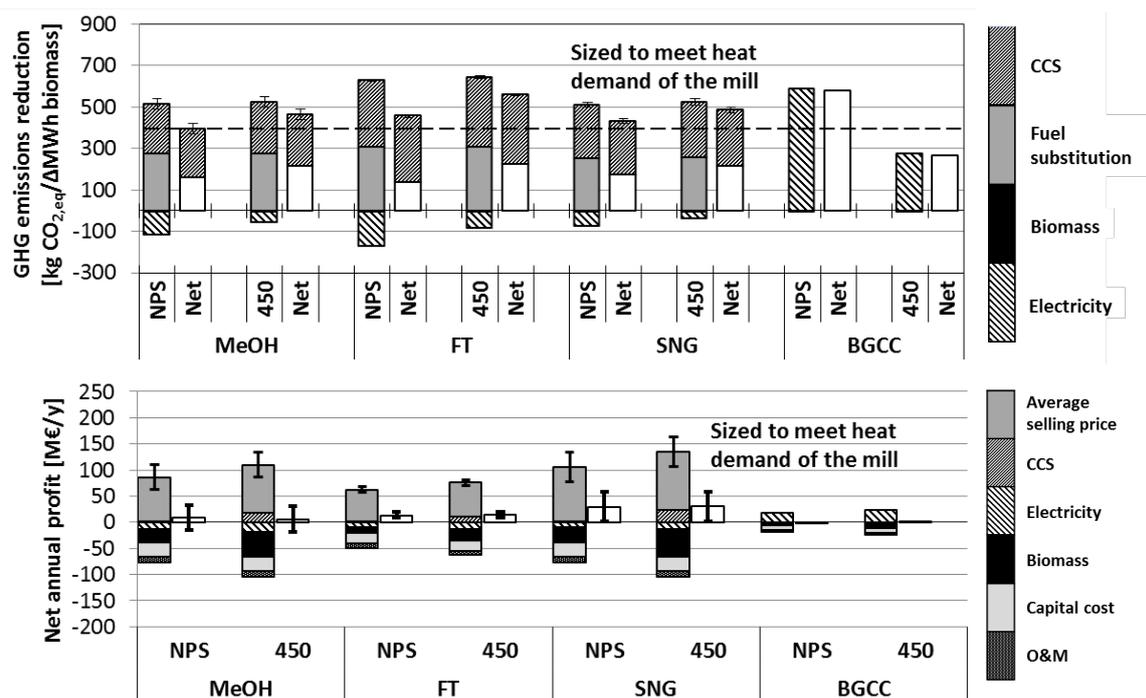


Figure 18. Specific greenhouse gas emissions reduction potential and net annual profit for the *new policies scenario* (NPS) and the *450 ppm scenario* (450) in 2030. The gasification routes are sized to satisfy the heat demand of the pulp and paper mill.

The NAP for the various production routes for an annuity factor of 0.1 are shown in the lower diagram in Figure 18; it represents a strategic investment for a pulp and paper mill owner. The error bars represent the span between the high level and the low level for the fuel selling price, as discussed in Section 4.3. All biofuel routes yield positive NAPs for an average fuel selling price, whereas the BGCC route is reduced by high specific capital costs and the purchase of wood fuel. The FT crude process is capable of providing good return on invested capital in absolute numbers in relation to its relatively small throughput.

Increasing the biomass input to 400 MW_{th} results in an excess of steam for all four cases (previously visualised in Figure 17), which is expanded in a condensing turbine to increase onsite electricity production. For production of methanol, the added electricity production from the condensing turbine approximately satisfies the increased demand caused by the larger size of the gasification process, whereas the FT crude production route achieves significantly improved electricity balance. The specific GHG emissions reductions for all cases are lower for the option with the condensing turbine as the GHG emissions are based on the increased use of biomass compared with a conventional bark boiler. However, the differences are marginal for methanol and SNG. Regardless of the sizing constraint, BGCC achieves a GHG performance that is similar to the biofuel cases

with CCS in the *new policies scenario*, whereas the performances of all processes are similar in the *450 ppm scenario* without CCS.

Regarding economic performance, all options benefit from the economy of scale, as shown in the lower diagram in Figure 19. The income from CCS should be utilised to the same level of NAP in both the *450 ppm scenario* and the *new policies scenario*.

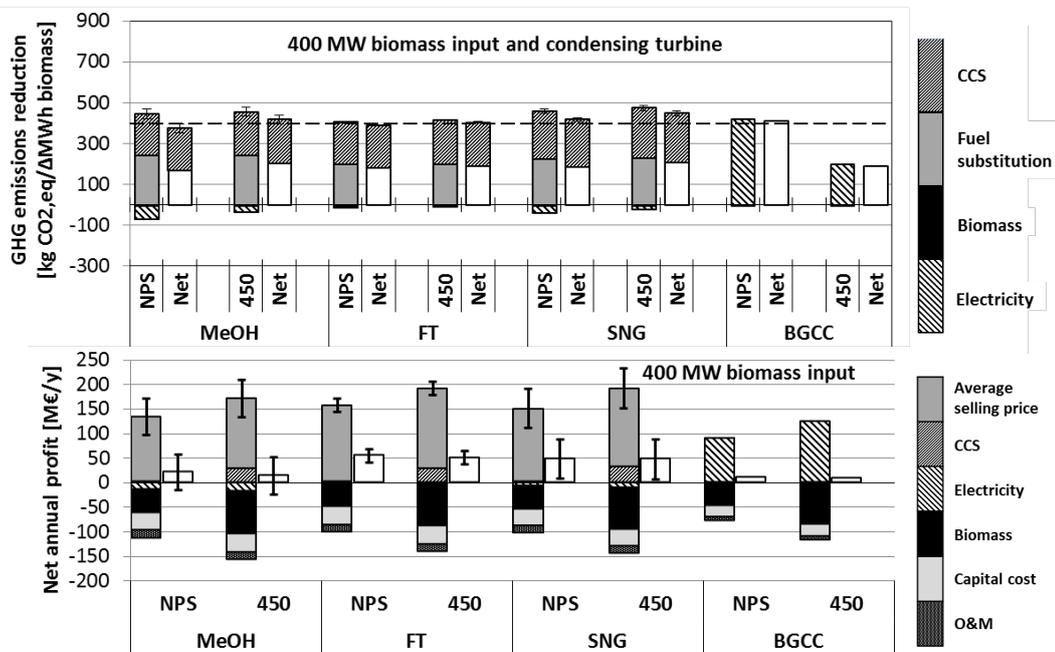


Figure 19. Specific greenhouse gas emissions reduction potential and net annual profit for the 400 MW_{th} gasification routes for the *new policies scenario* (NPS) and the *450 ppm scenario* (450).

5.3.2 Mechanical pulp and paper mill

The host pulp and paper mill in **Paper I** was based on mechanical pulping, as described in Section 3.1.1. In this paper, no thorough pinch analysis was conducted for the case study mill. The HLMPP methodology was employed to approximate the mill's stream data, which was used to construct the GCC. These two methods are described in Section 4.1.

The case study mill in **Paper I** had a hot utility demand of 131 MW of which 72 MW can be provided with steam from mechanical refiners. A 40 MW share of the remaining 59 MW has to be provided by LP steam at 3.5 bar for technical reasons, whereas the level of heat supply for the remaining part is determined by the cold streams (shown in Figure 20). The remaining heat was supplied by throttling HP steam from a bark boiler to the LP level. A simple measure for increased exergetic efficiency of the combusted biomass is to expand the steam in a turbine. CHP operation was selected as the base case scenario for the study presented in **Paper I**. In this study, the gasification processes were sized to

exactly be able to match the heat load currently supplied by the bark boiler. The corresponding stand-alone plants were evaluated for comparison.

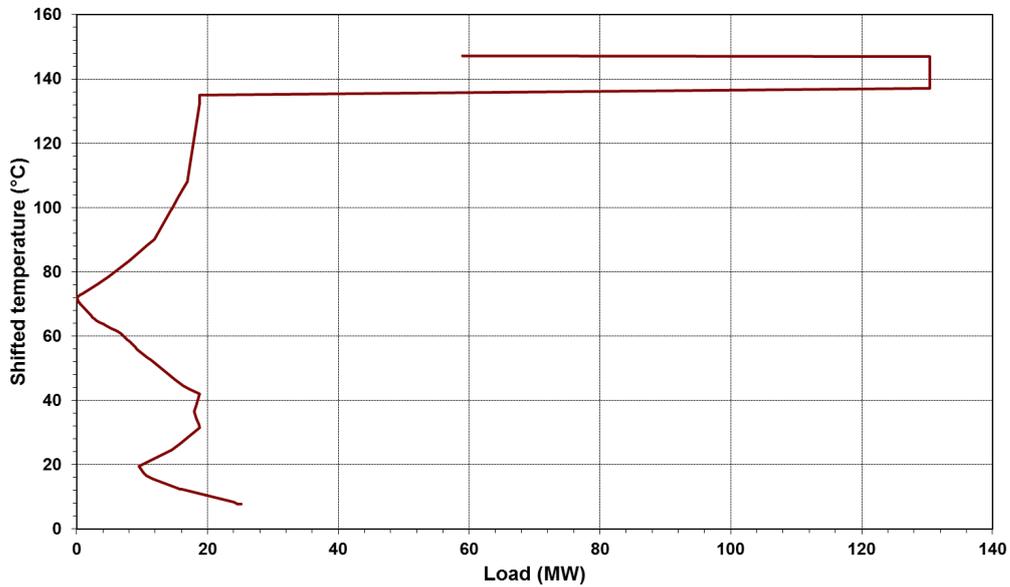


Figure 20. GCC of the integrated TMP mill without the sawmill using the HLMPP tool.

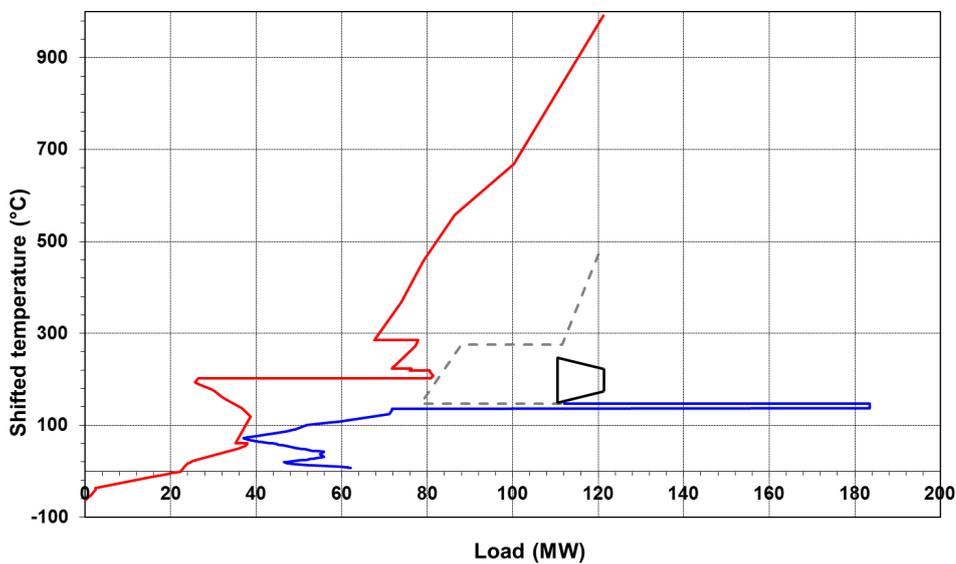


Figure 21. GCC for the FT process (upper red line), the TMP mill and the sawmill (lower blue line) and the steam cycle (dashed line). Reformed TMP steam is represented by the additional line at the LP level.

It was found that the solution with a gas turbine required a fuel input of 220 MW, the solution with methanol production required 635 MW and the solution with FT crude production 285 MW. This particular mill, which is co-located with a large sawmill, has

98.6 MW of biomass available for combustion or gasification on-site; the remaining part was assumed to consist of forest residues. Figure 21 provides an example of how the heat integration study was performed with the FT crude case using a split GGC with an integrated steam cycle. In **Paper I** the influence of the sawmill was also evaluated as it affects both the necessary heat generation for drying of the sawn goods and the availability of on-site biomass. If the pulp and paper mill is assumed to not be co-located with the sawmill, the heat demand would decrease (in this case by 16 MW). However, the amount of biomass available on-site decreases (here, from 98.6 MW to 35.3 MW).

Table 6. Summary of results for integrated cases including the sawmill.

	CHP	Gas turbine	Methanol	FT liquids
Biomass [MW_{LHV}]				
Demand	112.2	220	635	285
Available on site	98.6	98.6	98.6	98.6
Surplus/deficit ^a	-13.6	-121.4	-536.4	-186.4
Electricity [MW]				
TMP/sawmill	-194	-194	-194	-194
Energy mill	20.3	83	-14.4	-18.6
Surplus/deficit ^b	-173.7	-111	-208.4	-212.6
Compared with base case ^c	-	-62.7	34.7	38.9
Biofuel [MW_{LHV}]	-	-	322	148

^a [available on site - demand]

^b [TMP/sawmill + energy mill]

^c [surplus/deficit (base case) - surplus/deficit (energy mill)]

Figure 22 shows the absolute annual potential for CO₂ emissions reduction for the three evaluated integrated gasification routes. A methanol plant with the proposed configuration can be constructed on a larger scale compared with the other routes if the size in terms of biomass input is determined by the steam consumption of the mill and the CO₂ emissions reduction for methanol is therefore larger in this comparison. The comparison of FT crude production and increased power production indicates that the choice is dependent on the assumptions made about the electricity market. The assumption of a future low-emitting build margin technology suggests the selection of biofuels production over electricity production, especially if storage of the separated CO₂ is economically feasible. The absolute GHG results shown in Figure 22 can be compared with the results presented on a specific basis in Figure 11.

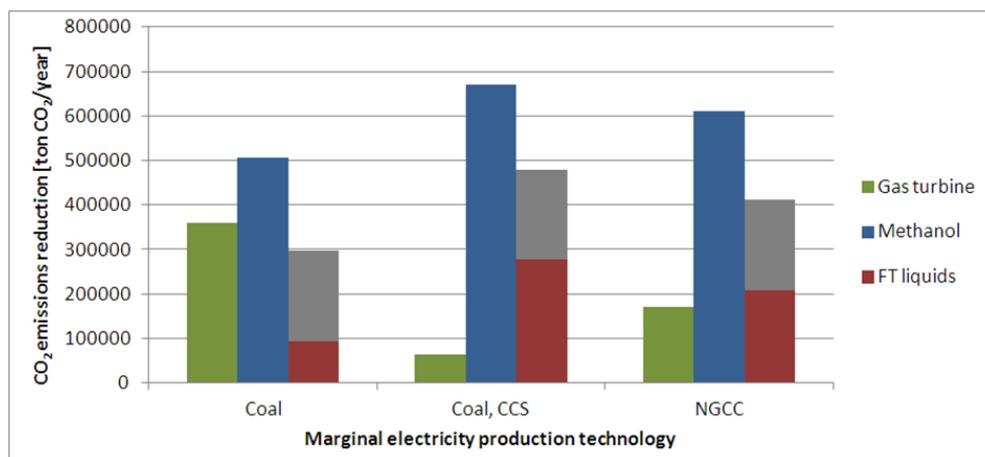


Figure 22. Absolute CO₂ emissions reduction for the three evaluated integrated energy mills. The grey bar on top of a red bar indicates the reduction potential if the separated CO₂ from the FT process is captured and stored.

5.4 Energy efficiency measures at the mill

Measures can be implemented to decrease the need for external supply of fuel to sustain the steam demand of the pulp and paper mill while maintaining the same output of paper products. How these measures may affect the biomass resource efficiency if a gasification process is integrated with the pulp and paper mill was quantified and compared with the case in which measures were not implemented. Note that the gasification process becomes smaller in terms of throughput in the case with the more efficient pulp and paper mill as the required heat demand is smaller. An additional case may be created in which the mill was more effective and the gasification process remained sized as if it was not. Excess heat can then be utilised for electricity production in a condensing turbine. Electricity is, however, more effectively produced in a dedicated power plant; this case would have lower overall efficiency (74.4% for the FT case - compared with 75.8% in Figure 23). The left part of Figure 23 represents the base case pulp and paper mill with a bark boiler. Alternative 1 and 2 represent integration of a gasification process with and without energy efficiency measures at the mill, respectively.

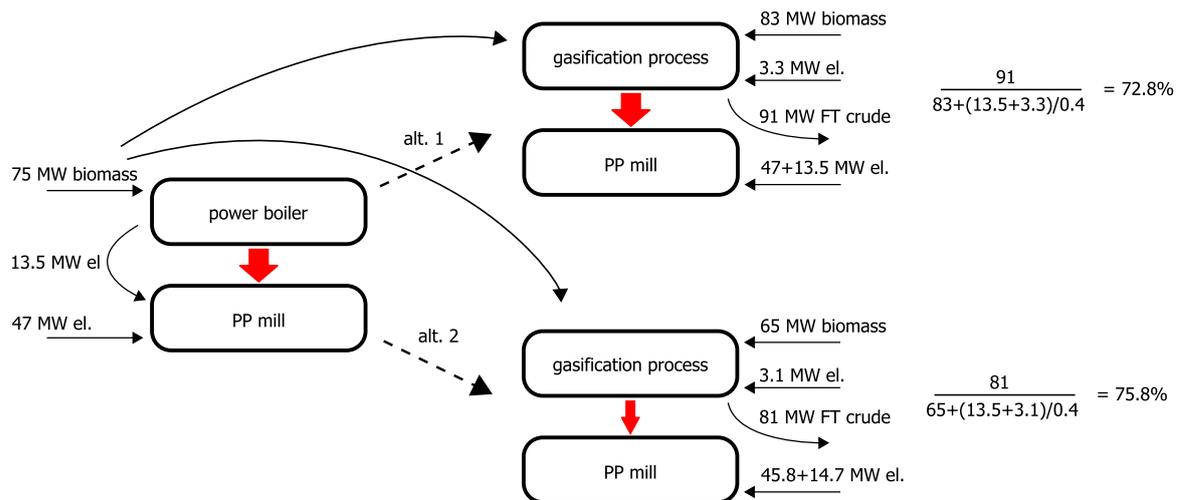


Figure 23. Energy balances for a pulp and paper mill with and without efficiency measures and subsequent integration of a gasification process which replaces the power boiler in terms of steam production. The red bold arrows represent the steam flow. Energy efficiency measures are not assumed to affect the electricity consumption, i.e., the total demand from the pulp and paper mill is identical in alternatives 1 and 2.

Resulting transportation fuel and/or electricity production are shown in Table 7. The required biomass input was decreased by 6% to 13% compared with the reference case.

Table 7. Heat integration study results (EM=efficiency measure).

		Biomass [MW]	Biofuel [MW]	Energy yield [%]	Co-generated electricity [MW]	Electrical efficiency ^b [%]
Methanol	Ref - exact match	255	159	62.5	14.9	5.8
	EM 70%	241	151		14.1	5.9
Fischer-Tropsch	Ref - exact match	158	91	57.6	10.3	6.5
	EM 70%	140	81		9.0	6.4
SNG	Ref - exact match	280	198	70.8	19.8	7.1
	EM 70%	244	173		17.3	7.1
BGCC	Ref - exact match	120	-	-	49.8 (12.7) ^a	41.5
	EM 70%	106	-	-	44.1 (11.3) ^a	41.6

^a power output from back-pressure turbine in brackets

^b does not include parasitic power demand

The resulting biomass resource efficiencies are presented in Figure 24 for a range of electrical efficiencies to demonstrate its influence. For routes with lower throughput, i.e., FT crude, the denominator is dominated by the constant electricity demand of the pulp and paper mill, which causes these smaller cases to be more influenced by variations in the assumed electrical efficiency. Integration of a smaller gasification process will

generate less biofuel as output but the incremental use of biomass compared with the 75 MW used for the bark boiler will be smaller, which has a positive influence on the efficiency.

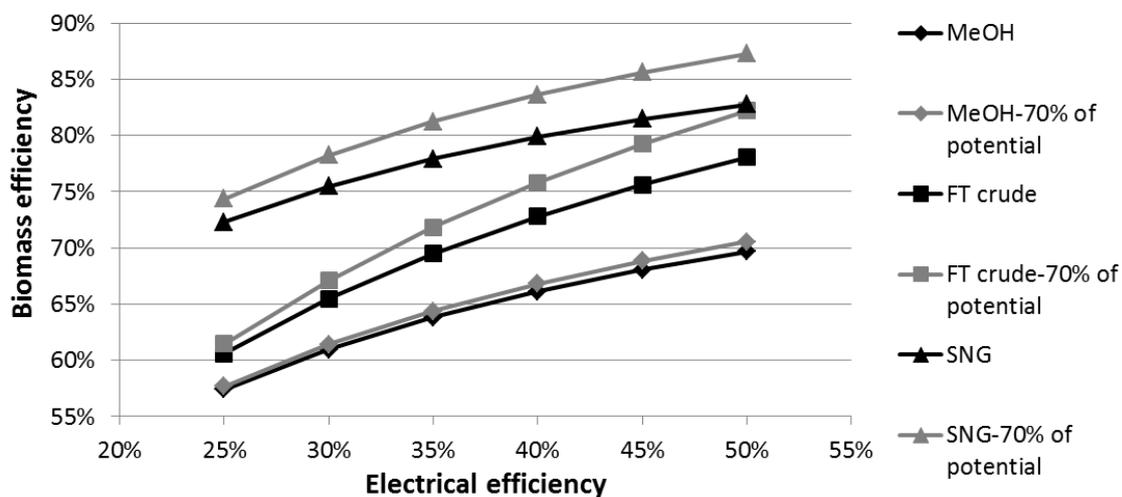


Figure 24. Biomass resource efficiency for a range of electrical efficiencies for external electricity. The black lines represent the reference cases with the current heat demand of the pulp and paper mill, whereas the grey lines represent the cases in which 70% of the theoretical heat savings potential is achieved.

The cases in which the energy efficiency measures were implemented at the pulp and paper mill have a higher biomass efficiency as long as electricity from the grid is produced with higher efficiency than the efficiency originally achieved with the bark boiler at the mill. The results shown in Figure 24, which indicate that cases with implemented efficiency measures perform more efficiently after biorefinery integration, are supported by the GHG results shown in Figure 25. **Paper V** and the GHG emissions balances in **Paper VI** reveal that the integrated biorefineries performed more efficiently when the efficiency measures were applied to the mill, for the condition in which excess heat from the gasification route exactly satisfied the heat demand of the pulp and paper mill. The economic evaluation, however, does not support this path (refer to Figure 25). By applying energy efficiency measures at the mill, while maintaining the size of the gasification process as if measures were not implemented the NAP is improved.

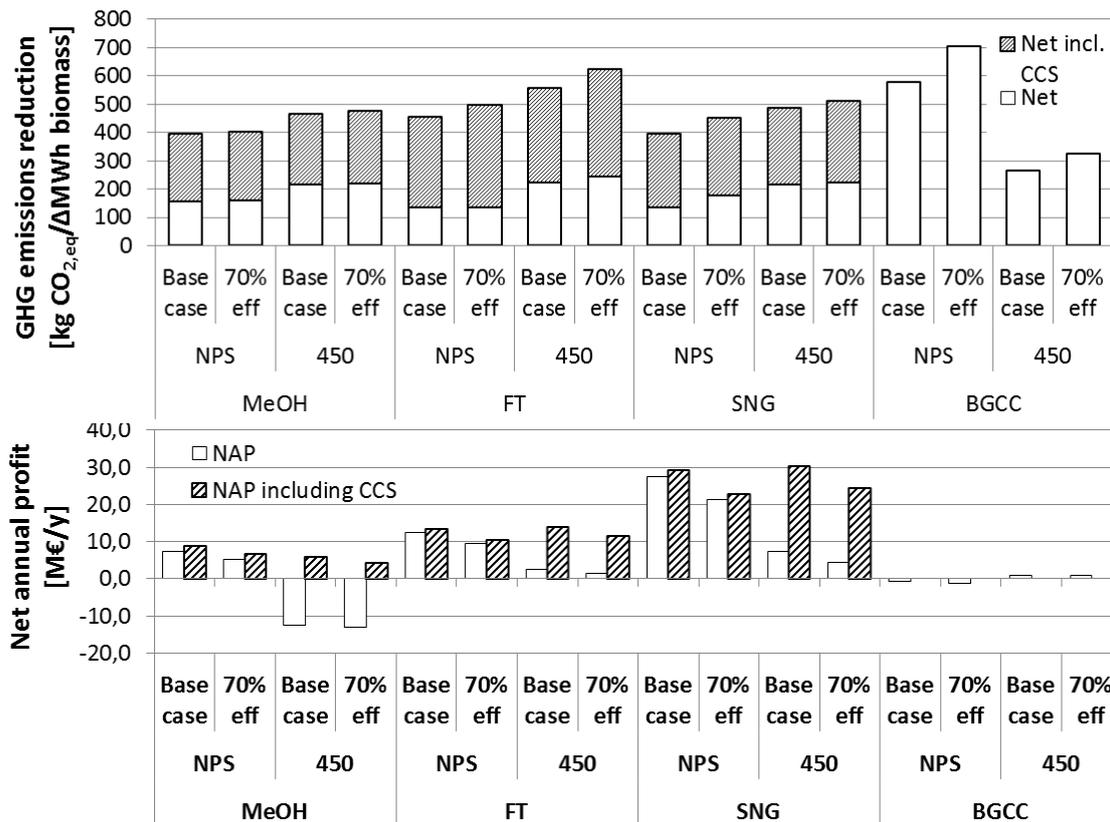


Figure 25. Comparison between GHG emission consequences for the reference case pulp and paper mill and the implemented efficiency measures that correspond to 70% of the theoretical potential, which were evaluated for two scenarios (*new policies scenario* – NPS, *450 ppm scenario* – 450). Net annual profits are shown in the lower diagram.

5.5 Comparison between a detailed pinch analysis and HLMPP

As discussed in Section 3.1.1, an evaluation of the HLMPP method was performed to validate the stream data for the heat integration analysis in **Paper I**. Table 8 lists important parameters from the two different methods - HLMPP and detailed pinch analysis – which were both evaluated for the same paper production. As this study was only conducted for one specific mill, general conclusions should not be formed based on these results. However, differences in the results may indicate the parts of the model that need additional attention and development. Estimation of this system by the model is difficult as it has three pinch points, as shown in Figure 26, and is sensitive to variations in the streams near the pinch points.

Table 8. Comparative figures for the HLMPP and the detailed pinch analysis

	HLMPP	Detailed pinch analysis
Minimum hot utility demand	86.6 MW	78.2 MW
Heat savings potential	7.7 MW (8.2%)	15.9 MW (17%)
Pinch temperature	72 °C	53 °C
Excess heat above 60 °C ^a	6 MW	0 MW

^a Heat that is possible to use in a new, integrated process, or a district heating network. Heat below 60 °C is assumed to be too low for utilisation. It has here been accounted for a contribution of 3 °C to the minimum temperature difference.

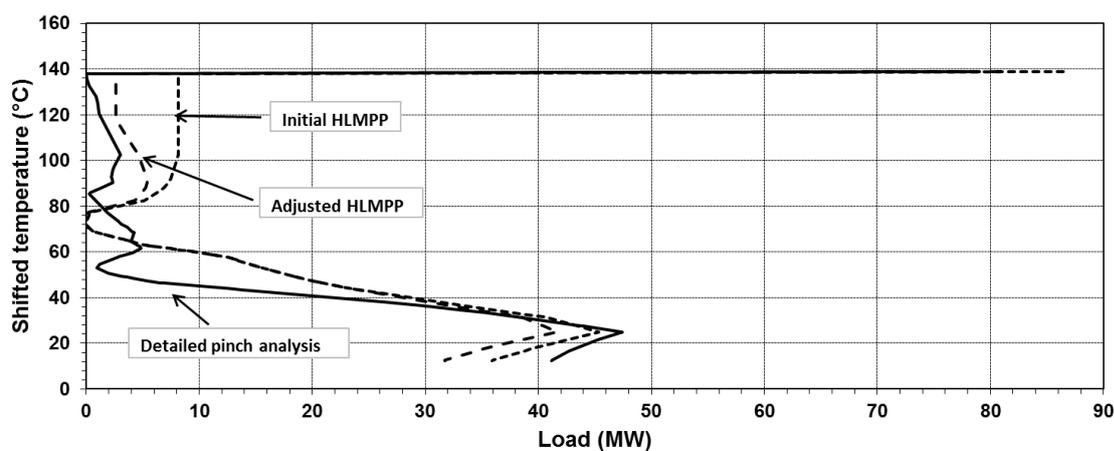


Figure 26. HLMPP GCC (dashed line) and detailed pinch GCC (continuous line).

An analysis was performed to explain the discrepancies between the two methods, such as the difference between the predicted minimum hot utility demand and the deviating pinch temperature. The streams at different locations in the mill were individually compared, and the most important streams are presented in the following list:

Pulp mill

- Chips warming and chips preheating comprise cold streams in the HLMPP tool, which range from 9 °C to approximately 75 °C. In the mill, part of this heat duty is satisfied with refiner steam with the combined purpose of heating and steaming (drive off air that is trapped in the wood).
- Dirty condensate at approximately 130 °C, from the reformation of TMP steam into clean LP steam, is available according to the detailed stream data extraction. These streams are not included by default in the HLMPP, which explains the difference between the HLMPP GCC and the detailed pinch GCC above the pinch in Figure 26. The stream is generated by HLMPP to a minimum of 78 °C and can be manually added to the analysis. It is not obvious to the HLMPP user if the colder part of the dirty

condensate (from 78 °C) from the reformers should be included in the TMP effluent or if it is automatically accounted in a similar manner as for the warmer part. By checking the stream data sheet, which is generated by the HLMPP, no such stream is available; therefore it should be added to the effluent in the HLMPP input form or separately added to the stream data sheet.

- Atmospheric steam is generated in the steam reforming process and can be utilised in the process. No such stream is generated by the HLMPP.

De-inked pulp plant

- 1.6 MW of heat for dispersing is estimated by the HLMPP to increase the temperature of the dispersed de-inked recycled pulp mixture from 55 °C to 75 °C. However, given the real flow of white water that collects the de-inked pulp, three times this amount is needed. The difference reduces the pinch point by 0.7 °C.

Paper mill

- HLMPP calculates the heat content in the moist exhaust based on start and target temperature and moisture content supplied by the user. The model underestimates the heat content in the moist exhaust by approximately 3 MW.

According to the bulleted list, dirty condensate from TMP steam has a large energy content at high temperatures after the regeneration of clean steam. Figure 26 shows that a pocket, similar to the pocket presented in the detailed pinch GCC, is partly created between 120 °C and 75 °C when this stream is incorporated, which makes the curves more similar.

A sensitivity test was performed for the detailed pinch analysis to verify what influences the pinch point. The streams with the highest energy content near the lower pinch (at 53 °C), which is the most influential, are the inlet air and outlet air from the paper machines and the heated fresh water. As the flow of moist exhaust air is underestimated by the HLMPP, increasing this flow in the detailed analysis makes no sense. The flow of drying air cannot explain the differences as the streams demand minimal energy. A decreased fresh water supply to the paper machines will decrease the heat demand in the region between 10 °C and 60 °C and will affect the shape of the GCC, which causes the results to resemble the HLMPP results. Whether the error is attributed to the HLMPP or the detailed stream data set cannot be explicitly determined.

6 Discussion

This chapter provides some general reflections on the research results obtained in this thesis.

A modern pulp and paper mill steadily operates throughout the year with minimal downtime and planned stops for maintenance at certain intervals and it is of great importance that implementation of a biorefinery process do not interfere with the operation of pulp and paper production. Biomass gasification has not been demonstrated on a commercial scale, at least not for advanced applications, but at least two things make the opportunities for this technology to be implemented more likely than gasification of black liquor, which has not yet been realised: First, unlike the black liquor gasifier, which is part of the chemical recovery cycle, the integration of biomass gasification can be achieved via the utility system. For example, the bark boiler can be retained as backup capacity in the event of failure in the gasification system or an off-gas/char boiler can be sized to handle these loads. Second, the technology can be proven as a stand-alone process prior to integration with existing industry, which significantly reduces the risk for the mill owner.

The implementation of biomass gasification with an existing pulp and paper mill will affect the demand for purchased electricity and biomass. Depending on the capacity of the biofuel production, the demand for electricity may either decrease or increase, which will affect the power production sector. The demand for biomass will always increase in the cases studied due to the production of biofuel or an increased production of power, thus competing for a presumably limited resource.

The studies in this thesis have been performed as case studies at two real mills and one model mill. Although these mills have certain individual characteristics, they represent typical mills of their type.

Constraints such as space and controllability have not been included in the analysis as the heat exchanger networks have not been designed. These constraints can potentially reduce the heat integration opportunities. A likely consequence is that the processes have to be constructed with larger throughput to satisfy the heat demand if the sizing constraint is to match the heat demand of the mill.

Storage of CO₂ has been assumed to be a viable option for the mitigation of GHG emissions throughout this thesis. Several of the appended papers indicate that the storage of CO₂ may become an important measure for reducing emissions and to compete with an alternative use of biomass. Potential problems with CCS are the lack of public acceptance towards onshore storage or uncertainties regarding the legal framework concerning offshore wells. The future political means to mitigate CO₂ emissions regardless of origin is another concern. Studies have shown that BECCS (bio-energy with CCS) plants are important and may be necessary in the future; therefore, the implementation of a CO₂ charge, which is technology neutral, is crucial.

This study demonstrates that the best climatic impact of biomass is frequently achieved by co-firing biomass in a coal power plant. On the other hand, there are more potential alternatives for renewable electricity than for renewable fuels, at least with the current fleet of cars. Carbon-based fuels are likely to be the predominant solution for the foreseeable future unless a rapid expansion of the electrified fleet occurs. A comparison of the results with the production of biomass-based materials, chemicals or other types of biofuels may be more interesting than the results for co-firing.

Regarding cost estimations, many inherent uncertainties exist as cost data typically has to be collected from multiple sources. Therefore, comparison of both input and output data with alternative sources is desirable. An Nth plant has been assumed for the studied cases.

Throughout the GHG emission studies, ENPAC has been employed to retrieve consistent prices for biomass and electricity based on fossil fuel prices and the CO₂ charge. It was found that production of biofuels were less profitable in a scenario with a high CO₂ charge, despite intuitive assumptions. The reason was that a high CO₂ charge causes an increased willingness to pay for biomass for coal power plants, co-firing biomass, which was the marginal user and price setting technology of biomass in these studies. This finding also necessitates capture and storage of CO₂ from the biofuel plant to compensate for the higher costs for biomass and electricity. Additional analysis of the factors that impact the biomass price is necessary to understand the interrelationships between these commodities. The ENPAC tool does not consider that coal power plants are generally located where biomass is dispersed and not always near harbours. The willingness to pay can be considered to be optimistic.

The HLMPP method can provide a quick and efficient scan of the potential for energy savings and/or the availability of excess heat in a TMP mill; however, the results should be analysed with some caution. The results from the comparison made after the data input had been synchronised demonstrate reasons to perform a complete pinch analysis; however, the HLMPP only indicates marginal opportunities for improvements. Another source of uncertainty, which requires additional attention, is the estimation of how much live steam is necessary for technical reasons, such as for the drying cylinders in the paper machines. The tool is capable of reasonable estimations of the mill profile, and the results can be rapidly achieved. The level of accuracy can only be established once a detailed

pinch analysis is established. Although it lacks precision in estimating flows of a particular mill, the HLMPP tool is convenient if the objective is to evaluate the trends and impacts of different process modifications on a more general basis.

7 Conclusions

In this Chapter, general conclusions based on the results of the appended papers are presented.

- Heat integration of biofuel production via biomass gasification with a pulp and paper mill is beneficial because of the good match between the heat source (gasification process) and the heat sink (pulp and paper mill), visualised by split-GCCs in this thesis. This can be seen for the TMP mill in Figure 21 or in Figure 2 in **Paper IV** for a chemical pulp and paper mill.
- Assumptions regarding the build margin technologies on the electricity market were shown to influence the preference of the integrated gasification processes to a stand-alone operation from a GHG perspective. If the build margin technology has emissions that correspond to a coal power plant, stand-alone biofuel production exhibits better potential for GHG emissions reduction as more renewable electricity is produced. If the marginal technology is relatively low-emitting, it is better to maximise the biofuel production and co-produce less electricity.
- Although it is a comparatively small investment, in relation to the total capital cost of 100 to 140 million Euros, the selection of dryer type influences the economic performance of an integrated BGCC unit. Using oxygen as an oxidising agent to increase the heating value of the gas exhibited poor economic performance due to increased costs for capital and for the operation of the ASU.
- The end product of a gasification-based biorefinery has a significant impact on the heat integration potential with a pulp and paper mill if the sizing constraint is to replace the current bark boiler, even though the front-ends are similar. The biomass input varied between approximately 200 MW and 600 MW for biofuel production. Generally, end products with high conversion efficiency will require a larger process throughput if the heat demand from the pulp and paper mill is to be satisfied, as less excess heat is generated per unit of product. In addition, downstream separation equipment for product purification, which also requires heat for their operation, may necessitate a larger process to be self-sufficient in heat. A large process, with a condensing turbine, has better economic performance

compared with a smaller process due to the economy of scale, but has less potential for GHG emission reduction if the size of the heat sink is constant.

- The energy efficiency measures that were implemented in the pulp and paper mill in connection with the integration of a gasification-based biorefinery will result in a more efficient use of biomass for the biorefinery, with a negative impact on the economy if the sizing constraint is to match the heat load for the assumed conditions.
- Thermal pretreatment of biomass may be beneficial in terms of wood-to-FT crude efficiency, with maximum efficiencies of 68%, assuming 40% electrical efficiency to convert electricity to biomass equivalents. Pretreatment using pyrolysis achieved the best performance with regards to GHG emissions, due to its low electricity consumption, if CO₂ from acid gas removal was vented, whereas milling the biomass prior to gasification yielded best results if the CO₂ was captured and sequestered. The reason was that all CO₂ was retained in the syngas stream for that case and was therefore available for separation.
- In a typical integrated TMP mill no excess heat exists at the required level to be utilised for example for biomass drying. The heat integration typically consists of one-way integration, i.e., from the gasification process to the pulp and paper mill (and the sawmill). Two-way integration can be realised in chemical pulp and paper mills, in which excess heat may be available from bleach filtrate or cooling water effluents. These sources have traditionally been used for district heating but may provide extended use for biomass drying prior to gasification during the entire year.
- Assumptions regarding the electricity market are highly influential in GHG emission consequences regarding the implementation of gasification-based biorefineries, which is particularly pronounced if various cases affect the electricity balance differently, as the impact of the assumption is amplified. Similarly, the assumption regarding the electrical efficiency of grid electricity is highly influential in the biomass resource efficiency.
- GHG emissions were considered on a specific basis, i.e., based on the extra biomass required compared to the original amount needed to sustain the heat balance. As discussed in the Results section, this amount favours smaller a gasification unit as the external share of the biomass input is smaller. This result can be explained by comparing the CHP operation with a heat-only boiler. The efficiency of the electricity production based on the added biomass is equivalent to the boiler efficiency.
- Capture and storage of CO₂ from acid gas removal is necessary for gasification-based biofuel production to compete with biomass co-firing in coal power plants

regarding potential for the reduction of GHG emissions. CCS has an important role in terms of emissions and economy (if biogenic CO₂ is included in the trading system) and was able to increase the reduction potential in the studied cases from approximately 300 kgCO_{2,eq}/MWh_{biomass} to 700 kgCO_{2,eq}/MWh_{biomass}.

8 Further work

In the GHG investigations in this thesis, the transportation and storage of CO₂ has been evaluated where applicable. In addition to the stream from the acid gas removal, which was accounted for in this thesis, other sources of CO₂ exist. Although the largest source of CO₂ is contained in the recovery boiler flue gases, the lime kiln and other boilers can be included. The effect of these measures can be further elaborated to better understand the complete potential for GHG emissions reduction for the pulp and paper industry.

The economic evaluations in this study did not specify the investor in these projects or describe the ownership. The study has implied that the mill owner would have the full interest in the facility; however, recent projects connected to gasification-based biorefineries in pulp and paper mills have been formed as joint ventures between the mill owner and a specialist in fuel production (see Section 2.1). New business models are necessary to determine future directions.

The studies in this thesis have not included flows of waste water and low temperature cooling water, i.e., the current systems at the pulp and paper mill are expected to be capable of handling an increased amount. Increased knowledge about the accuracy of this assumption is necessary.

In a pulp and paper mill, the gasification of other by-product streams in addition to bark may be interesting. Currently, sludge from the processing of recycling fibres is combusted with fuels with a higher heating value as it is no longer allowed to deposit. A gasification unit may leave sufficient excess heat to dry the sludge and to gasify it with bark and forest residues for advanced applications.

Based on the results of this thesis, more detailed integration could validate the accuracy of the estimated potentials. A comparison of the results with other opportunities for GHG emission mitigation and an evaluation of whether a combination of different measures can yield a corresponding incremental increase in GHG reduction potential may also be interesting.

As mentioned in Chapter 6 Discussion, the factors that influence the price of forest residues should be examined to accurately estimate the economic performance of these systems.

Abbreviations

ADt	air-dried tonne
ASU	air separation unit
ATR	auto-thermal reformer
BECCS	bio-energy with carbon capture and storage
BGCC	biomass gasification combined cycle
BLG	black liquor gasification
CC	composite curve
CCS	carbon (carbon dioxide) capture and storage
CEPCI	chemical engineering plant cost index
CFB	circulating fluidized bed
CHP	combined heat and power
DB	dual bed
DME	dimethyl ether
ETS	emission trading system
EU	European Union
FAME	fatty acid methyl ester
FT	Fischer-Tropsch
GCC	grand composite curve
GHG	greenhouse gas
GWP	global warming potential
HHV	higher heating value
HLMPP	heat load model for pulp and paper
HP	high pressure (steam)
HVO	hydro-treated vegetable oil
HW	Hot Water
IEA	international energy agency
LHV	lower heating value
NAP	net annual profit
NGCC	natural gas combined cycle
RME	rapeseed oil methyl ester
SEK	Swedish kronor
SNG	synthetic natural gas
TMP	thermo-mechanical pulp
WGS	water-gas-shift

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