

**LAPPEENRANTA UNIVERSITY OF TECHNOLOGY**  
Faculty of Technology  
Environmental Engineering

*Kaisa Manninen*

**EFFECT OF FOREST-BASED BIOFUELS PRODUCTION ON CARBON  
FOOTPRINT, CASE: INTEGRATED LWC PAPER MILL**

Examiners:           Professor Risto Soukka  
                          M.Sc. Helena Wessman

Advisor:             M.Sc. Katri Behm

## **ABSTRACT**

Lappeenranta University of Technology  
Faculty of Technology  
Degree Programme in Environmental Technology

Kaisa Manninen

### **Effect of forest-based biofuels production on carbon footprint, Case: Integrated LWC paper mill**

Master's Thesis

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86 pages, 29 figures, 11 tables and 9 appendices

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M.Sc. Helena Wessman

Keywords: carbon footprint, LCA, biofuel, biomass, LignoBoost, Fischer-Tropsch diesel

International energy and climate strategies also set Finland's commitments to increasing the use of renewable energy sources and reducing greenhouse gas emissions. The target can be achieved by, for example, increasing the use of energy wood. Finland's forest biomass potential is significant compared with current use. Increased use will change forest management and wood harvesting methods however. The thesis examined the potential for integrated pulp and paper mills to increase bioenergy production. The effects of two bioenergy production technologies on the carbon footprint of an integrated LWC mill were studied at mill level and from the cradle-to-customer approach.

The LignoBoost process and FT diesel production were chosen as bioenergy cases. The data for the LignoBoost process were obtained from Metso and for the FT diesel process from Neste Oil. The rest of the information is based on the literature and databases of the KCL-ECO life-cycle computer program and Ecoinvent.

In both case studies, the carbon footprint was reduced. From the results, it can be concluded that it is possible to achieve a fossil-fuel-free pulp mill with the LignoBoost process. By using steam from the FT diesel process, the amount of auxiliary fuel can be reduced considerably and the bark boiler can be replaced. With a choice of auxiliary fuels for use in heat production in the paper mill and the production methods for purchased electricity, it is possible to affect the carbon footprints even more in both cases.

# TIIVISTELMÄ

Lappeenrannan teknillinen yliopisto  
Teknillinen tiedekunta  
Ympäristötekniikan koulutusohjelma

Kaisa Manninen

## **Metsäperäisten biopolttoaineiden vaikutukset hiilijalanjälkeen, Case: Integroitu LWC-paperitehdas**

Diplomityö

2010

86 sivua, 29 kuvaa, 11 taulukkoa ja 9 liitettä

Tarkastajat: Professori Risto Soukka  
MMM Helena Wessman

Hakusanat: hiilijalanjälki, LCA, biopolttoaine, biomassa, LignoBoost, Fischer-Tropsch diesel  
Keywords: carbon footprint, LCA, biofuel, biomass, LignoBoost, Fischer-Tropsch diesel

Kansainväliset energia- ja ilmastostrategiat asettavat myös Suomelle velvoitteita lisätä uusiutuvien energialähteiden käyttöä ja vähentää kasvihuonekaasupäästöjä. Tavoitteisiin voidaan päästä esimerkiksi energiapuun käytön lisäämisellä. Suomen metsäbiomassapotentiaali on huomattava verrattuna nykyiseen käyttöön. Käytön lisääminen vaatii kuitenkin muutoksia metsänhoidossa ja puunkorjuutavoissa. Tässä työssä tarkasteltiin sellu- ja paperitehtaan mahdollisuuksia lisätä bioenergian tuotantoa sekä kahden bioenergiatuotantotavan vaikutusta integroidun sellu- ja LWC-paperitehtaan hiilijalanjälkeen tehdastasoisessa sekä kehdestä asiakkaalle -tarkastelussa.

Bioenergiateknologioiksi valittiin LignoBoost-prosessi ja Fischer-Tropsch dieselin valmistus. Ligno-Boost -prosessiin tiedot saatiin Metsolta ja FT-diesel-prosessiin Neste Oililta. Muut tiedot saatiin kirjallisuudesta sekä laskelmissa käytetyn KCL-ECO elinkaarimallinnusohjelman ja Ecoinventin tietokannoista.

Molemmissa tapauksissa hiilijalanjälki pieneni. Tuloksista voitiin päätellä, että LignoBoost-prosessin avulla voidaan saavuttaa fossiilisista polttoaineista vapaa sellutehdas. Hyödyntämällä FT-dieselin valmistuksesta syntyvää höyryä, lisäpolttoaineiden määrän tarve pienenee huomattavasti ja kuorikattila saadaan korvattua kokonaan. Paperitehtaalla lämmöntuotantoon käytettävien lisäpolttoaineiden sekä ostetun sähkön tuotantotavan valinnalla voidaan hiilijalanjälkeä pienentää entisestään molemmissa tapauksissa.

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Helsinki, April 20, 2010

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## ABBREVIATIONS

|                                 |  |
|---------------------------------|--|
| ADt                             | air dry ton  |
| BFB                             | bubbling fluidized bed boiler                      |
| BLG                             | black liquor gasification                          |
| CaCO <sub>3</sub>               | calcium carbonate                                  |
| CFB                             | circulating fluidized bed boiler                   |
| CH <sub>4</sub>                 | methane  |
| CHP                             | combined heat and power                            |
| CO                              | carbon monoxide                                    |
| CO <sub>2</sub>                 | carbon dioxide                                     |
| DME                             | dimethyl ether                                     |
| EFI                             | European Forest Institute                          |
| FRAM                            | Future Resource-Adapted Pulp Mill                  |
| FT                              | Fischer-Tropsch                                    |
| GHG                             | greenhouse gas                                     |
| GWP                             | global warming potential                           |
| H <sub>2</sub>                  | hydrogen   |
| H <sub>2</sub> S                | hydrogen sulphide                                  |
| HHV                             | higher heating value                               |
| IGCC                            | integrated gasification combined-cycle power plant |
| IPCC                            | Intergovernmental Panel on Climate Change          |
| LCA                             | life-cycle assessment                              |
| LHV                             | lower heating value                                |
| LWC                             | lightweight coated                                 |
| N <sub>2</sub>                  | nitrogen   |
| Na <sub>2</sub> CO <sub>3</sub> | sodium carbonate                                   |
| Na <sub>2</sub> S               | sodium sulphate                                    |
| NaOH                            | sodium hydroxide                                   |
| NCG                             | non-condensable gases                              |

|                 |   |
|-----------------|---|
| NO <sub>x</sub> | nitrogen oxide  |
| UNFCCC          | United Nations Framework Convention on Climate Change |

### **Most important terms used in the study**

|                           |  |
|---------------------------|--|
| First-generation biofuel  | biofuel made from the raw materials sugar, starch, vegetable oil or animal fats  |
| Second-generation biofuel | biofuel made from raw materials other than products used for food, e.g., wood, waste and field biomass   |
| Bioenergy                 | energy produced from biofuels  |
| Biofuel                   | fuel produced from biomass or biowaste   |
| Biogenic carbon dioxide   | carbon dioxide from biomass-based fuel   |
| Biomass                   | the biodegradable fraction of products, waste and residues from biological origin from agriculture (including vegetal and animal substances), forestry and related industries including fisheries and aquaculture, as well as the biodegradable fraction of industrial and municipal waste |
| Carbon footprint          | takes into account all greenhouse gas emissions produced throughout the product's value chain  |
| Fischer-Tropsch process   | biofuel production process via Fischer-Tropsch synthesis   |
| Forest chips              | biofuel produced with cleaving and shearing, commonly from harvesting residues   |
| Fossil carbon dioxide     | carbon dioxide from fossil fuels   |
| Heating value             | the heating value indicates how much heat is developed through the complete combustion of the fuel mass. The heating value is expressed in solid and liquid fuels, usually in megajoules per kilogram of fuel (MJ/kg).   |
| Kyoto Protocol            | an international agreement linked to the United Nations  |

## Framework Convention on Climate Change

|                       |   |
|-----------------------|---|
| Life-cycle assessment | compilation and evaluation of inputs, outputs and potential environmental impacts of a product system throughout its life cycle                         |
| LignoBoost            | a method in which lignin is removed from black liquor   |
| Pyrolysis             | thermal decomposition that occurs without oxygen  |
| Renewable energy      | energy from non-fossil energy sources: wind, solar, geothermal, wave, tidal, hydropower, biomass, landfill gas, sewage treatment plant gas and biogases |
| Syngas                | gas produced by the gasification process containing varying amounts of carbon monoxide and hydrogen   |

# 1 INTRODUCTION

## 1.1 Background

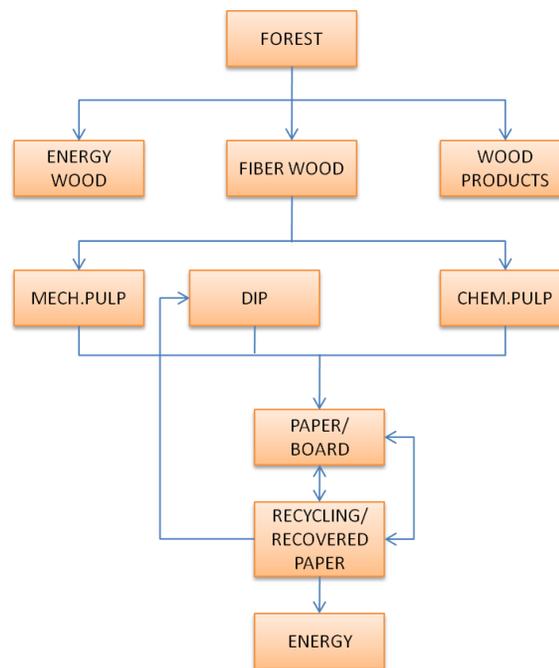
This master's thesis is part of KCL's project 1251 "Modeling fiber flows: Bioenergy potential and carbon footprint mitigation (FLOW)". The project was carried out together with the European Forest Institute (EFI). The aim of this project is to provide the industry with an approach with a scientific basis to communicate and calculate the specific biogenic carbon flows of products. The project will create new data on biocarbon by linking the competence of forestry experts and industry.

Climate change is the topic of the day. It has put growing pressure on all European industries. New solutions based on renewable raw materials are being developed. The carbon footprint is the key indicator that dominates the discussion, as carbon has become the most critical parameter of global well-being. So far, fossil carbon has been included in the carbon footprint calculation, but there is also bio-based carbon that is bound in forest and fibre products. Biocarbon flows need to be modelled in the product value chain in order to take a step forward in measuring product renewability.

The use of renewable energy is strongly linked to the carbon balance. The European pulp and paper industry is one of the drivers of bioenergy in Europe because it is one of the most important industrial producers and consumers of renewable energy. There is a need to clarify the impacts of alternative uses of fibre, different ways of producing pulp, side streams, recycling and end-use policies on the carbon balance. The objectives of the FLOW project are to model the wood-based carbon balance in different resource use intensities, to build a harmonized calculation procedure for the energy and carbon balance from the forest to end-use, to recognize the bioenergy potential through the value chain and to clarify the importance of forest- and fibre-based products in CO<sub>2</sub> mitigation.

## 1.2 Objectives and structure of the study

This master's thesis is one part of FLOW's subproject that examines alternative biomass and carbon flows and changes in CO<sub>2</sub> balances in the forest and along the product's value chain. The subproject represents both traditional and new or emerging products and technologies. The value chain takes into account the product's life cycle, from forestry to end-use. The product value chain is represented in Figure 1.



**Figure 1.** Product value chain

The objective of the master's thesis is to recognize the potential of bioenergy through the value chain and to clarify the importance of forest and fibre-based products in CO<sub>2</sub> mitigation. The aim is to clarify changes to the carbon footprint when the current pulp and paper production process produces more bioenergy and fossil fuels are replaced. The current usage potential of bioenergy and biomass in Finland, bioenergy policies and their requirements, and different technologies to produce bioenergy are explained in the master's thesis literature study. The main aim is to concentrate on wood-based bioenergy, which includes industrial by-products such as bark, sawdust, forest chips, forest logging residues and black

liquor. The thesis focuses on the potential of chemical pulp production to increase bioenergy usage. Some refined wood fuels such as wood pellets are therefore excluded. In contrast, the most important pulp manufacturing processes in which fossil fuels could be replaced by bioenergy are clarified. Existing and emerging bioenergy production technologies in Finland are also studied.

The carbon footprint and life-cycle assessment (LCA) are the tools used in the experimental part. Forestry changes and different bioenergy production methods are examined through five case studies. The first three cases are based on current forest management in Finland. The first case is a reference case that represents the typical integrated pulp and paper mill in Finland. The second case, the LignoBoost process, is added to the pulp mill. The third case is a reference case for the final two cases. It is the same as the first case but takes into account fossil fuel diesel production. The two final cases represent a situation in which forest management is energy-wood intensive, Fischer-Tropsch diesel (FT diesel) is produced with biomass and the available amount is increased during energy-wood intensive wood manufacturing. FT diesel is produced in order to replace fossil diesel production. The calculations will be performed with a KCL-ECO 4.1 life-cycle-assessment computing program.

## **2 BIOENERGY – POLICIES AND CURRENT USAGE**

Climate change is currently the topic of the day. The work to control climate change is ongoing, and new technologies are being developed to increase renewable energy production. An agreement system has been established to control climate change. The United Nations Framework Convention on Climate Change (UNFCCC) came into force in 1992 and an international agreement – the Kyoto Protocol, which is linked to UNFCCC – was adopted in 1997. The Kyoto Protocol commits industrialized countries to stabilizing their greenhouse gas (GHG) emissions. The Kyoto Protocol entered into force on 16 February 2005, and as of 3 December 2009, 189 countries and 1 regional economic integration organization (the EEC) have deposited instruments of ratification, accession, approval or acceptance. Mem-

ber states of the European Union, including Finland, ratified the Kyoto Protocol in 2002. (Berghäll et al. 2003, 3; Ministry of the Environment 2008; UNFCCC 2009.)

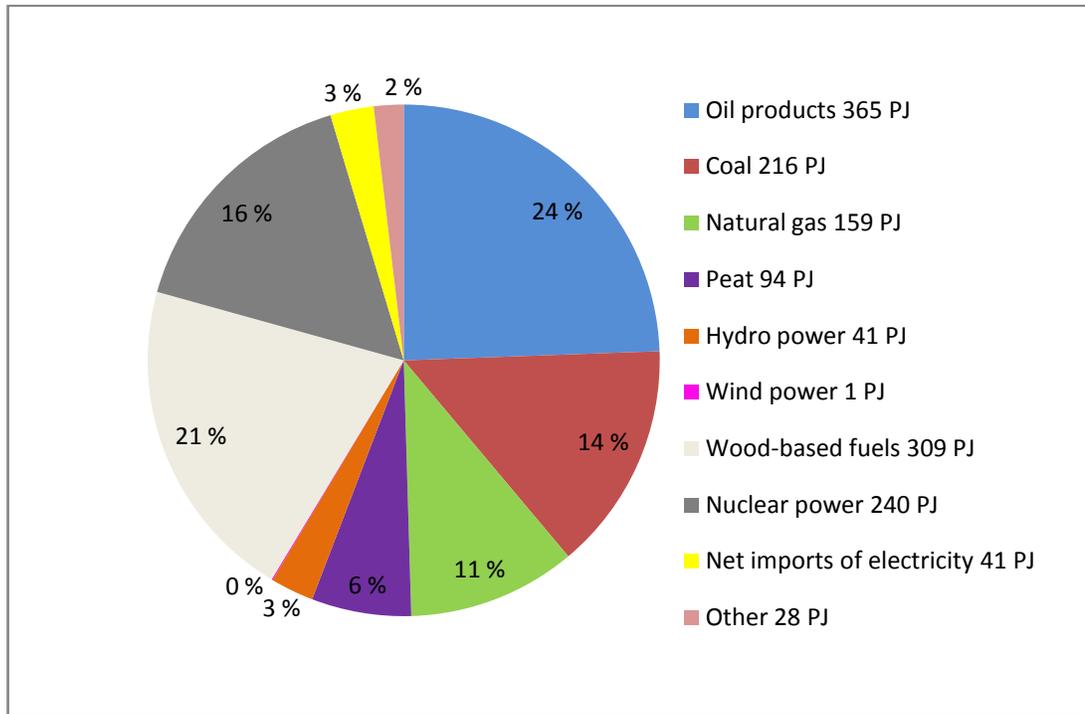
## 2.1 Policies

The bioenergy policy of the European Union is based on the Kyoto Protocol's target to reduce collective emissions of greenhouse gases in industrialized countries by 5.2% compared with the year 1990 in the first five-year period 2008-2012 (UNFCCC 2009). The protocol sets the EU countries a common target to reduce their emissions by 8% compared with the year 1990. The target has been shared out between the 15 member countries under a legally binding "burden-sharing agreement". Finland's target is to stabilize its emissions to the level of the year 1990 (Ministry of the Environment 2008.) In 2007, the European Union decided to reduce its greenhouse gas emissions by at least 20% by 2020 compared with the year 1990. In addition, the share of renewable energy sources will be increased by 20% by 2020 including 10% of the biofuel target for transportation (5.75% target in 2010). (Long-term Climate and Energy Strategy 2008, 13-14, 18; European Parliament and the Council 2009/28/EC.) The increase in the price of oil, coal and natural gas as well as the high price of emission allowances are affecting the price competitiveness of renewable energy sources, promoting emission reduction targets as well as increased their use (Long-term Climate and Energy Strategy 2008, 8).

The share of renewable energy in Finland in 2006 was approximately 24% (Statistics Finland 2008). A renewable energy target will be shared between the member countries; Finland's target for 2020 is therefore 38% of the share (Long-Term Climate and Energy Strategy 2008, 8). A potential way to achieve the 38% target is to increase bioenergy use, especially the use of forest chips (Asplund, Flyktman & Uusi-Penttilä. 2009, 3). According to the Long-Term Climate and Energy Strategy (2008, 37) in Finland, the overall target set for forest chips is 12 million m<sup>3</sup> per year, which is approximately 24 TWh by 2020.

## 2.2 Usage in Finland

Energy produced from biofuels is called bioenergy. Biofuels are produced from biomass, which is the biodegradable fraction of products, waste and residues from biological origin from agriculture (including vegetal and animal substances), forestry and related industries including fisheries and aquaculture, as well as the biodegradable fraction of industrial and municipal waste (Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009). Peat is also a biomass, but because it is the slowest renewable energy source used in Finland, it is not included in bioenergy. (Helynen et al. 2002, 11.) The total energy consumption in Finland in 2006 was approximately 1494 PJ. Figure 2 shows the total energy consumption by energy source. The share of renewable energy from total energy consumption was 365 PJ (24%). Renewable energy includes hydro, solar, wind, geothermal, tidal and wave energy as well as bioenergy (Hakala & Välimäki 2003, 240). The bioenergy share of renewable energy sources was approximately 325 PJ (89%). Figure 2 shows that the share of wood-based fuels is significantly higher than that of other bioenergy sources. Wood-based fuels were the second most important energy source in the total energy consumption. The share of wood-based fuels of the total energy consumption was 21% (309 PJ). This figure includes fuel-wood consumption in small-sized dwellings, which have a share of approximately 49 PJ. This is not taken into account in this thesis when examining the use of wood-based fuels in energy production. (Finnish Forest Research Institute 2008, 290-292.) Wood-based fuels consist mainly of by-products from the forest industry and are discussed in more detail in Section 2.3.



**Figure 2.** Total energy consumption by energy source in Finland in 2006 (Finnish Forest Research Institute 2008, 289).

### 2.3 Bioenergy in the forest industry

The electricity consumption in Finland is approximately 90 TWh per year. The forest industry's share of electricity consumption was 31% (28 TWh) in 2006. The forest industry produces 40% of its electricity demand in its own power plants, and approximately 75% of the electricity and heat is produced from wood-based biofuels. (Finnish Forest Industries 2008, 4.) The modern chemical pulp mill can meet all of its heat demand in the recovery boiler, however, with heat left over for the condensing power plant. Electricity is produced at twice the demand by the modern chemical pulp mill. The extra electricity is sold. The chemical pulp mill generally produces heat and electricity in combined heat and power (CHP) plants. Heat is produced in the recovery and bark boilers by burning black liquor, bark and other wood residues. The steam generated is channelled into the steam turbine where it expands into back pressure. (Kivistö 2008, Chapter 3, p. 3-4, 10.) The forest industry is the biggest bioenergy producer in Finland with an 80% share of total bioenergy pro-

duction (Finnish Forest Industries 2008, 6). Bioenergy sources in the forest industry are discussed in more detail in the next chapter.

The forest industry is the main wood-based bioenergy producer in Finland, as the wood-based biofuels used in energy generation in Finland consist mainly of the forest industry's by-products and side streams. The total wood-based fuel consumption in energy production is 260 PJ. It can be divided into liquid, solid and other wood fuels. Most of the wood-based fuel is used directly as the forest industry's mill fuel. The amount of wood-based fuel used in the generation of electricity and heat and directly in industrial processes in forest industry was 207 PJ in 2006. This is almost 80% of the total wood-based fuel consumption in energy generation. All the generated black liquor, for example, is used in pulp mills, as its burning is part of the pulp mill's circulation of chemicals. (Helynen et al. 2002, 12.) The share of solid wood fuel consumption used as mill fuel in forest industry was also over half of the total solid wood fuel consumption of energy production in 2006. The solid wood fuel is mainly used in power boilers that produce heat and electricity. (Finnish Forest Research Institute 2008, 292, 296.) The forest industry also uses other mill fuels: peat, natural gas, heavy fuel oil and coal. Their share of total mill fuel consumption is 70 PJ, which is 25%. (Finnish Forest Research Institute 2008, 296.) These other mill fuels are used to start and stop boilers and as supporting fuels (Kivistö 2008, Chapter 3, p. 4-5). Although the forest industry is an important wood-based fuel producer, the amount of wood fuel can vary depending on the forest industry's output (Helynen et al. 2002, 12).

Table 1 shows a more detailed distribution of the wood-based fuels that are used for the generation of electricity and heat and directly in industrial processes. Consumption by small-sized dwellings is excluded.

**Table 1.** Wood-based fuels that are used for the generation of electricity and heat and directly in industrial processes in Finland in 2006. (Finnish Forest Research Institute 2008, 292).

| <b>WOOD-BASED FUEL CONSUMPTION<br/>IN ENERGY GENERATION</b> | <b>[PJ]</b> | <b>Share of the forest in-<br/>dustry's bioenergy<br/>source</b> |
|---|-------------|--|
| <b>Waste liquors</b>  | <b>156</b>  | <b>60%</b>   |
| <b>Other by-products and waste products</b>                 | <b>5</b>    | <b>2%</b>  |
| <b>Solid wood fuels</b>                                     | <b>99</b>   | <b>38%</b>   |
| Forest chips  | 22          |  |
| Industrial chips  | 7           |  |
| Sawdust   | 13          |  |
| Bark  | 54          |  |
| Other   | 3           |  |
| <b>Total</b>  | <b>260</b>  |  |

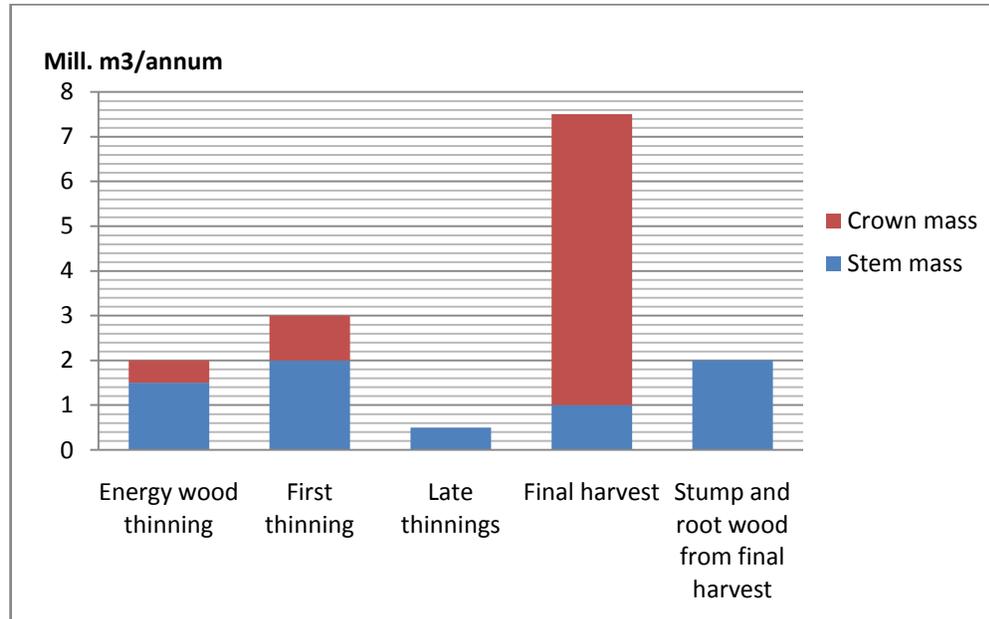
The chemical pulp industry is the most important producer of wood fuel. As Table 1 shows, the waste liquor share, which consists mainly of black liquor, is 156 PJ (60% of the forest industry's biofuels). It is approximately two thirds of the forest industry's by-products used in energy production. Other by-products and waste products consist of, for example, pine and birch oil, soft soap, methanol, biosuspensions and paper consumed in energy generation. Their share is only 5 PJ (2% of the forest industry's biofuels). Solid by-products are forest chips, industrial chips, sawdust, bark and other wood fuels, which include recycled wood, wood pellets, briquettes and other solid wood fuels. Their share is 99 PJ (38% of the forest industry's biofuels. (Finnish Forest Research Institute 2008, 291-292, 296.) Forest chips are mainly harvesting residues. Industrial chips, sawdust and bark are by-products of the forest industry. As Table 1 shows, bark makes up a significant share of the solid wood fuels. The biggest potential for increasing the use of solid wood fuel, however, is to intensify the production of forest chips. (Finnish Forest Industries 2008, 6.)

## **2.4 Biomass potential of Finnish forests and energy wood-based forestry**

Climate policy aims to prevent the harmful influences of climate change. International commitments control national operations to cut greenhouse gases. In Finland, wood is the fuel with the greatest potential to increase bioenergy. Wood energy only reduces greenhouse gases if fossil fuels can be replaced. In Finland, energy can be produced by burning wood without producing a significant amount of CO<sub>2</sub> emissions during the growing, harvesting and production chains. Changes are needed in forest management to increase the use of energy wood. Sustainable development is taken into account when using biomass. This is paradoxical when it is a case of intensifying energy wood harvesting. The increase in biomass recovery and usage prevents global climate change, but at the same time, there is an impact on the regional environment, caused by the significant reduction in biomass. (Kuusinen & Ilvesniemi 2008, 3.) This chapter discusses the biomass potential of Finnish forests and the changes and effects on forest management when the harvesting of energy wood intensifies.

As discussed in Section 2.3, the biggest potential for increasing forest-based energy production is to increase the use of forest chips. There has been research into the potential of biomass for the production of forest chips in Finnish forests. This theoretical potential includes all residue biomass generated when harvesting raw wood as well as the wood biomass that is generated during forestry operations like thinning. It is not possible to utilize all the theoretical potential due to technological, socio-economic and environmental factors, as well as the versatility of the forest and ownership-related subjects. The theoretical maximum annual production potential of forest chip biomass has been estimated at 45 million m<sup>3</sup>/annum, though the technically harvestable potential is only 15 million m<sup>3</sup>/annum. The energy content of this technically harvestable potential is approximately 108 PJ. (Hakkila 2004, 26-27.) In 2008, 4.03 million m<sup>3</sup> of forest chips were used in heat and energy power plants (Ylitalo 2009, 2). So, the technically harvestable potential is almost quadruple that

and the theoretical harvest about eleven-fold. Figure 3 shows the maximum technically harvestable potential separated for five different types of logging operations.



**Figure 3.** The maximum technically harvestable forest chip potential of Finnish forests separated for five different types of logging operations (Hakkila 2004, 27-28).

This estimation does not take the price assumption into account. In energy wood thinning, the trees are so small that the harvest of raw material for pulpwood is not profitable. The main product is therefore energy wood. The energy potential from energy wood thinning is 4 TWh or 14 PJ. In energy wood harvesting, the stands are typically 15-25 years old and most of the woods are pine. The cost of harvest is high and subsidies are necessary to make recovery possible. The first thinning is commercial harvesting from which the main product is pulpwood. A quarter of the stem wood does not meet the minimum dimension of pulpwood and it can be used as energy wood. The energy potential from the first thinning is 6 TWh or 22 PJ. In late thinning, the amount of residue wood is so small that it is not profitable to recover it. The recovery would also cause logging damage to standing trees and unnecessary nutrient loss at a critical development phase of the stand. In the final harvest, there is abundant crown mass, especially in spruce forest where the amount of crown is double that of pine or deciduous forest. The energy potential from the final harvest is 16

TWh or 58 PJ. The stump and root wood can be recovered from clear-cut areas of large spruce stands. The logging residues have usually already been recovered from these areas. The energy potential of stump and root wood is 4 TWh or 14 PJ. (Hakkila 2004, 27-28.)

When intensifying the production of forest chips, many forest management aspects have to be taken into account. The Forestry Development Centre Tapio has published the Forest Management Practice Recommendations, which present the principles and methods applicable in connection with the sustainable forestry practised in Finland's commercial forest. The recommendations are based on economically, ecologically and socially sustainable management and use of the forest. The aim is for the forest to be productive while at the same time ensuring biodiversity of the forest. Energy wood harvesting cannot cause significant damage to the environment and the development of the stand. The objective of energy wood harvesting is to support forest management and forestation of good quality commercial wood. When energy wood harvesting increases, the amount of rotten wood can decrease, and this has to be taken into account. (Tapio 2006, 5-6, 52.)

The Forestry Development Centre Tapio and the Finnish Forest Research Institute Metla have published the research report "The environmental effects of energy wood harvesting". The report brings out the latest available and updated research information on the economical, ecological and social impacts of energy wood harvesting. According to the report, the harmful effects of energy wood harvesting can be prevented beforehand if the recommendations of energy wood harvesting (Koistinen & Äijälä 2006) are followed. The recommendations, in general terms, are: 1) 30% of logging residues are not collected, 2) old stumps and parts of recently chopped stumps of different wood species are left  $\geq 25$  pieces per hectare, 3) existing rotten wood is left, 4) important living environments are left out of harvesting, 5) stumps are not collected from escarpments, cobble deposits and rocks, wetlands, border strips of water systems and the immediate surroundings of saved or rotten wood. (Siitonen 2008, 35.) The effects of energy wood harvesting are presented in Appendix I. In summary, it can be said that biomass harvesting has a positive effect on forest management. It has been estimated that the production of forest chips will reduce the costs

of forest management by over 2% and increase net income by 0.5% in 2010 if the targets of the national forest program on the use of forest chips are realized. (Saksa 2008, 40.)

### **3 MANUFACTURING PROCESSES OF WOOD-BASED BIO-ENERGY IN A KRAFT PULP MILL**

It is eco-efficient to generate heat and electricity from biomass in the pulp mill. Biomass can also be refined into solid, liquid or gaseous processed products. These products include traffic fuels, pellets and biogas. Processed products can be transported long distances and used to replace fossil fuels in energy production. CHP is now commonly used because of its high efficiency. New technologies are being developed to improve the competitiveness of biofuels. (Rintala et al. 2007, 11-12.) The manufacturing processes of wood-based bioenergy are discussed in more detail in the next chapters. First, the energy production processes in the pulp mill are discussed. Current technologies and potential bioenergy production technologies are represented. The objective is to examine the potential of wood-based bioenergy production technologies that could replace or be integrated into current pulp mill processes. These chapters concentrate on current combustion technologies as well as the potential bioenergy technologies that are being developed today.

#### **3.1 Bioenergy production from the kraft pulping processes**

The power plants in pulp mills are usually CHP plants. The main purpose of the power plant is to produce heat for pulp mill processes. Co-generation of heat and electricity is profitable in terms of energy economics because 80-90% of the fuel energy can be used. The recovery boiler is the main producer of heat in a pulp mill. The bark boiler is used when possible. These boilers operate as steam boilers in the pulp mill. Water is fed into one end of the tube in the boiler and exits from the other end as superheated steam. When the generated steam is channelled through the turbine, its pressure and temperature decrease and the energy that is released is converted into mechanical rotation energy for the turbine.

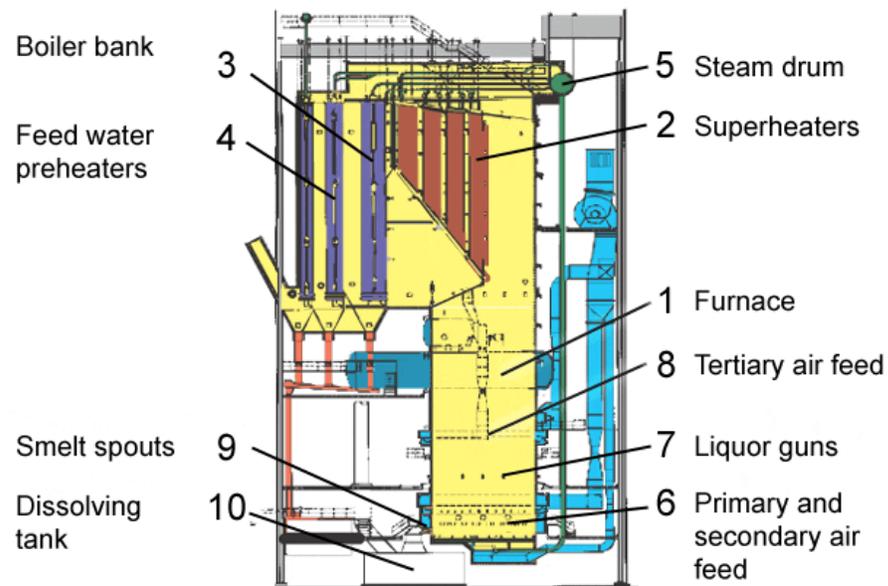
This mechanical rotation energy is converted into electricity in a generator. Steam is recovered for heating purposes to various heating points in the process. Steam releases its heat energy through condensation and after that, the steam is usually taken back to the power plant as condensate, and from there it is pumped back into the boiler as feedwater. (Huhtinen & Hotta 2008, 196-198; Kivistö 2008, Chapter 3, p. 4.)

### **3.1.1 Recovery boiler**

Concentrated black liquor is used as fuel in the recovery boiler. The purpose of the recovery boiler is to burn the organic part of the black liquor so that the generated heat can be used in steam production. It also recycles and regenerates chemicals in the black liquor and reduces emissions from several waste streams in an environmentally friendly way. (Vakkilainen 2008, 86.) Black liquor contains a large amount of water. The dry content of black liquor and the heating value affect steam production. The heating value of black liquor is the gross calorific value. The sources of the heating value of black liquor are organic matters: lignin, carbohydrates and extractives. (Kivistö 2008, Chapter 3, p. 5-6.) The dry content of black liquor is raised in the evaporating plant before being burnt in the recovery boiler. The gross calorific value ranges between 13 and 15 MJ/kg of dry solids when the dry content varies between 70% and 80%. (Energia Suomessa 2004, 381.) The generated heat energy produced in the combustion of black liquor is approximately 16 GJ/ADt. Of this, 3.2 GJ/ADt is used in the production of electricity and 13 GJ/ADt is used as process steam. (Know Pulp 2007.)

The quality of recovery boilers has improved due to the price, size of recovery boilers and demand for energy efficiency and environmental protection. The capacity of recovery boilers is 2500-5000 tons of dry solids per day. Some recovery boilers burn black liquor with a dry solid content of more than 80%. A traditional recovery boiler from 1985 is a double drum construction recovery boiler. Its main steam pressure is typically about 85 bars and the temperature 480°C. One of the structural changes was the development of the single drum constructive recovery boiler. This type of recovery boiler has more reliable control of the water system compared with the double drum construction. It also has better usability

and safety. The single drum construction withstands greater pressure and has a bigger capacity. The construction time is also shorter. Pressure and heat will continue to increase in future recovery boilers. The pressure will increase to 120 bars and heat to 520°C. This means that there will be an increase of approximately 7% in electricity generation. Figure 4 shows the single drum construction of a recovery boiler. (Know Pulp 2007; Vakkilainen 2008, 87-90.)



**Figure 4.** Single drum construction recovery boiler (Know Pulp 2007).

The evaporated black liquor is sprayed into the furnace in the form of droplets, usually by spoon-shaped guns. The black liquor is sprayed uniformly over the bottom of the furnace. The black liquor droplets are burnt in three phases. In the drying and pyrolysis phases, the droplets dry when all the water has evaporated and the volatile solids are burnt. The coal and chemicals drop to the bottom of the furnace where the coal burns itself out and the chemicals melt. The melted chemicals flow through the smelt spouts into the dissolving tank. Liquid chemicals consist mainly of sodium sulphate ( $\text{Na}_2\text{S}$ ) and sodium carbonate ( $\text{Na}_2\text{CO}_3$ ). The burning air is usually fed into the furnace in three stages. The primary air keeps the burning stable and at a high enough temperature in a hill. The purpose of the sec-

ondary air is to burn volatile gases. The burning of the remaining gas is controlled with tertiary air that is fed into the furnace above the liquor guns. Oil or natural gas is used as extra fuel in the recovery boiler at start-ups and shutdowns. (Kivistö 2008, Chapter 2, p. 22.)

### **3.1.2 Bark boiler technologies**

Bark and wood residues are used as the main fuels in the bark boiler. In addition, natural gas, peat, oil or coal is needed for boiler start-ups and as supporting fuels. The combustion technologies of the power boiler are fixed bed combustion and fluidized bed combustion. Fluidized bed combustion can be divided into a bubbling fluidized bed boiler (BFB) and a circulating fluidized bed boiler (CFB). Fluidized bed combustion has replaced other technologies due to its better fuel flexibility and improved emissions performance. (Huhtinen & Hotta 2008, 197-198.)

The forest biomass is burnt in controlled conditions in the combustion process of the boiler system. In the combustion process, the energy is released when the high-energy bonds between carbon and hydrogen are broken. Combustion produces CO<sub>2</sub> emissions. The CO<sub>2</sub> is the most important greenhouse gas causing climate change. The carbon content of the fuel affects the CO<sub>2</sub> released to the atmosphere in the combustion process. The amount of CO<sub>2</sub> emissions can therefore be affected by the choice of fuel. Woody biomass has a high ratio of specific emission, but if the biomass is a product of sustainable forestry, the actual emissions to the atmosphere are not permanent. This is because the growing of biomass binds the carbon so that the carbon produced in combustion circulates in a closed system. (Hakkila & Verkasalo 2009, 200-201.)

The generated energy can be used for the production of heat, steam or electricity (Hakkila & Verkasalo 2009, 200-201). CHP production has been an important option to increase the efficiency of power generation and the competitiveness of bioenergy. The typical annual efficiency of the CHP plant is 80-90%. In the next chapters, most of the combustion systems that are represented are the combustion technologies used in the CHP plant. CHP is typically the most profitable choice for power production from biomass. Technologies such

as fluidized bed boilers offer the benefits of using biomass. Biomass is used in big and small CHP plants, and new technologies have now been developed for even smaller plants. (Fagernäs et al. 2006, 81-82.) The other case in which combustion processes can be used is for refining biomass into liquid fuels such as biodiesel or bioethanol, which can be used in vehicles (Hakkila & Verkasalo 2009, 200).

### **Fixed bed combustion**

Fixed bed combustion technology is the oldest method of burning solid fuels. Until the 1970s, fixed bed combustion was the only combustion technology for burning bark and wood residues in the pulp and paper industry. Since then, fluidized bed combustion technology has replaced fixed bed combustion. There are many ways to classify grate types. They can be classified according to the method used to cool the grate or according to the method of feeding the fuel to the grate. The most common fixed bed combustion technologies that can be used to burn bark and wood residues can be divided into three groups as follows:

- stationary inclined or step grate
- travelling grate
- mechanical inclined grate

Moving grates are used in the pulp and paper industry where the power boilers are larger. These grates have an automatic fuel feed and ash removal. (Huhtinen & Hotta 2008, 214.)

The fixed bed combustion process can be divided into three systems:

- feeding-the-fuel system
- grate system
- air-of-combustion system

The function of the feeding-the-fuel system is to feed fuel onto the whole grate. It is important that the fuel is distributed evenly otherwise the escape of uncontrolled primary air can follow. Most of the grate's surface is used to evaporate moisture from the fuel. Fuel can include 10-60% moisture of the total weight. Decreasing the fragment size and using pre-heated air of combustion accelerate the evaporation of moisture. The combustion process is

based on pyrolysis. Combustion air is typically fed in two or three phases. Primary air is fed from below the grate. Secondary and possible tertiary air is fed into the afterburning space where volatile combustion gases are burnt. Heat power from the grate surface depends on the fuel and grate type and varies between 300 and 1000 kW/m<sup>2</sup>. (Helynen et al. 2002, 40.)

### **Fluidized bed combustion**

Fluidized bed combustion has become common. The advantages of fluidized bed combustion compared with fixed bed combustion are that it can be used for combustion of low-grade fuels with a high moisture or ash content, it allows different fuels to be burnt simultaneously, it is simple and cheap to remove sulphur by injecting limestone into the furnace and it has high combustion efficiency and low emissions of NO<sub>x</sub>. There are two types of fluidized bed combustion available for combustion of bark and wood residues. They are the bubbling fluidized bed boiler (BFB) and the circulating fluidized bed boiler (CFB). It is difficult to say which system is better for a given application. For most applications, BFB and CFB are both technically possible. The principle of fluidized bed combustion is based on a bed of sand particles. Fluidizing is a result of blowing air through the sand bed. (Huhtinen & Hotta 2008, 217-218.)

In BFB combustion the average particle size of the bed material is 1-3 mm and the depth of the bed is 0.4-0.8 m. With biomass, a maximum of 3 MW/m<sup>2</sup> of fuel power can be achieved per cross-sectional area of fluidized grate. The fluidizing velocity is typically 0.7-2 m/s. The bed is first heated to a temperature of 500-600°C with ignition burners to ensure safe burning. About half of the combustion air is fed evenly over the cross section of the bed by the air distributor or grid that forms the furnace floor. The rest of the air is fed into the afterburning space where NO<sub>x</sub> emissions can be reduced. There are usually evenly spaced drainpipes in the grid. The spent bed material and bottom ash are removed through these pipes. Refined sand is fed back into the boiler. The crushed fuel is fed into the top of the bed. When the fuel has dried, it burns in the sand bed and the furnace above the bed. The firing temperature is approximately 900°C so that it stays under the melting point of ash.

Due to the melted ash, the bed material can be sintered. (Helynen et al. 2002, 42; Know Pulp 2007; Huhtinen & Hotta 2008, 221-222.)

In some cases, CFB combustion differs from BFB combustion. A higher, fluidizing velocity and more powdery bed material are used in the CFB boiler. The fluidizing velocity is typically 3-10 m/s and the average particle size of the bed material is 0.1-0.5 mm. CFB combustion is typically the turbulence and confusion of the particles. There is no clear bed surface in the CFB boiler because the density of the bed decreases as it becomes higher in the boiler. There is also a solids separator in the CFB that captures entrained bed material and sorbent and unburnt fuel particles and returns them to the lower part of the furnace. The flue gases are fed through the convective heat exchanger or superheater, feedwater and air preheater. Coal, which contains only a little volatile matter, can also be burnt in a CFB boiler. (Know Pulp 2007; Huhtinen & Hotta 2008, 223.)

### **Gasification of solid woody biomass**

Gasification is a manufacturing process that occurs with partial oxidation. It is therefore not a combustion process. The gasification process produces syngas, which comprises mainly hydrogen (H<sub>2</sub>) and carbon monoxide (CO). Table 2 shows the differences between gasification and combustion of coal. (Gasification Technologies Council 2008, 3.)

**Table 2.** Gasification compared with combustion of coal. Gasification occurs with partial oxidation, therefore the produced syngas comprises mainly H<sub>2</sub> and CO (Gasification Technologies Council 2008, 3).

| <b>Constituents of coal</b> | <b>Gasification</b> | <b>Combustion</b> |
|-----------------------------|---------------------|-------------------|
| Carbon                      | CO                  | CO <sub>2</sub>   |
| Hydrogen                    | H <sub>2</sub>      | H <sub>2</sub> O  |
| Nitrogen                    | N <sub>2</sub>      | NO <sub>x</sub>   |
| Sulphur                     | H <sub>2</sub> S    | SO <sub>2</sub>   |
| Oxygen                      | -                   | O <sub>2</sub>    |

The gasification process consists of feedstock of raw material, an oxygen plant or air feeding, a gasifier, gas clean-up, syngas cleaning and the end use of syngas. Solid, liquid and gaseous raw materials can also be used as feedstocks in the gasifier. This chapter discusses gasification of solid woody biomass. (Gasification Technologies Council 2008, 4.) Gasification has proved an attractive option for producing power and heat. The gasification technology has been developed since the 1970's in Finland. (Kurkela 2002, 3.) Biomass gasification has many positive effects. The main reasons for using wood gasification are that fossil fuels can be replaced with CO<sub>2</sub>-neutral biofuel in energy production and that the power to heat ratio of combined heat and power production can be increased. Many types of gasifiers have been developed. There are three main types of biomass gasifiers: fixed bed, bubbling fluidized bed (BFB) and circulating fluidized bed (CFB) technologies. The fluidized bed and CFB have been used in the pulp and paper industry to produce biofuel for the lime kiln and the CFB has given the most practical experiences. (Vakkilainen & Kivistö 2008, 9.)

### **Fixed bed gasifiers**

In small-scale heat and power production (less than 15 MW), the most competitive gasifiers are based on fixed bed technology (Helynen et al 2002, 43). Updraft and downdraft gasifiers are the basic types of traditional fixed bed technology. Their technology is based on natural, slowly descending fuel caused by gravity. These gasifiers are only suitable for sized feedstocks that have a sufficiently high bulk density to guarantee a stable fuel flow. The fuel stays in the gasifier for a long time and the gas velocity is low. The function of the updraft gasifier is based on the lowering fuel. The fuel is fed into the top of the gasifier where it flows through drying, pyrolysis, gasification and combustion zones. The generated ash is removed from the bottom. The product gas of the updraft gasifier contains a large amount of oils and tars because the process does not include secondary decomposition reactions. The temperature of the product gas is also low (80-300°C for biomass fuels and 300-600°C for coal). Bottom ash is usually completely oxidized and does not contain significant amounts of unburnt carbon. Due to the low gas velocity and “filtering effect” of the drying

and pyrolysis zones, the product gas does not contain significant amounts of dust. (Kurkela 2002, 4-5.)

In contrast to the updraft gasifier, the downdraft gasifier's pyrolysis products have to flow co-currently through the hot combustion and gasification zones where tars are decomposed and oxidized. The downdraft gasifier therefore produces low-tar content syngas. Downdraft gasifiers had a long history as energy producers in cars, buses and boats during World War II. The downdraft gasifier can be used in internal combustion engines, but the ideal operation requires high-quality sized wood fuels. This has been a delay element of the development of downdraft gasifiers due to the high costs. (Kurkela 2002, 8-9.)

The Bioneer gasifier is the best-known fixed bed gasifier operating with a range of biofuels. It is an updraft gasifier that produces tarry and low-caloric value fuel gas. In the existing Bioneer plant, the generated gas is burnt to produce hot water for district heating. The Bioneer gasifier is suitable for applications like district heating 1-15 MW<sub>th</sub>, small-scale CHP 1-3 MW<sub>e</sub>, drying kilns, process ovens and diesel power plants after catalytic gas cleaning. The fuels, which are used in Bioneer gasification plants in Finland, have been sod peat and wood chips. Several potential fuels, such as crushed bark, sawdust and crushed demolition wood, have had some problems, for example, with flowing. The generated syngas contains tars that foul the pipelines and complicate its use. (Kurkela 2002, 5-7.)

The Novel gasifier combines the best features of the Bioneer updraft gasifier with the low-tar content typical of the downdraft gasifier. The function of the Novel gasifier is based on forced fuel flow that allows the use of low-bulk-density fibrous biomass residues. In contrast to the Bioneer gasifier, the Novel gasifier can be scaled up to more than 8 MW. The Novel has no problems with leaking feeding systems or blocking gas lines. The syngas has a high temperature and low tar content. It is suitable for various biomass residues and waste derived from forest wood residue chips, sawdust and wood shavings, crushed bark, demolition wood, residues from plywood and the furniture industry, recycled fuel manufactured from household waste and sewage sludge. (Kurkela 2002, 7.)

### **Fluidized bed gasifiers**

There are low-pressure and pressurized fluidized bed gasification technologies. The low-pressure technology is discussed first. Bubbling fluidized bed gasification (BFB) technology has been studied and appears to be more economically viable for medium-sized applications (15-40 MW). In contrast, the circulating fluidized bed (CFB) gasifier is most economic on a larger scale (40-100 MW). Bark and waste wood are used as fuel in CFB gasifiers in pulp mills to replace fuel oil in the lime kiln. Part of the generated gas is also utilized in drying plants. (Kurkela 2002, 10.) The CFB gasifier exploits the function of CFB combustion technology, but the burning uses less air or oxygen. An atmospheric gasifier is composed of a vertical refractory-lined steel cylinder in which the fuel is fed into the lower part of the gasifier. At this level, the upward-flowing gas stream does not contain free oxygen. In the reactor, the biofuel particles start to dry in the hot gas flows. The temperature is 850-950°C. After drying, pyrolysis occurs where the volatile compounds liberate from the fuel and form combustible gas. The gas is led through the cyclone, which separates the circulating material from the gas, directly to the burners via an air preheater and ducts. (Vakkilainen & Kivistö 2008, 9-10.)

The flammable components of product gas on a dry basis for a CFB wood gasifier are N<sub>2</sub> (46-47 vol. %), CO (21-22 vol. %), CH<sub>4</sub> (5-6 vol. %) and tars. The product gas also includes CO<sub>2</sub>, steam, charcoal, ash and in air gasification nitrogen. The heating value in air gasification is 3-7 MJ/m<sup>3</sup> and in pure oxygen gasification 7-15 MJ/m<sup>3</sup>. The higher heating value in oxygen gasification is due to the dilution of product gas components, and air nitrogen is avoided. Oxygen plants are very expensive, so the oxygen technology has not been applied. If the content of chlorine and alkali metals or aluminium is high in the product gas (0.1-1.0 mass-%), the gas has to be cleaned before burning. If the product gas is used in a lime kiln, the bark or wood needs to be dry before gasification. The drying is required to increase the heating value so that the desired flame temperature can be achieved. Fuel drying decreases the amount of flue gas. (Vakkilainen & Kivistö 2008, 9-10.)

Interest in the integrated gasification combined-cycle power plant (IGCC) increased in Finland in the late 1980s. In a basic IGCC process, gasification is based on oxygen-blown and multistage wet gas cleaning. The focus is on the simplified IGCC process, the technology of which is based on pressurized air gasification and hot gas cleaning. The driving force for the development of the IGCC was the need for higher power-to-heat ratios in co-generation. The main market for biomass-based IGCC plants is in combined heat and electricity production in the medium-sized range (30-100 MW<sub>e</sub>). CFB and BFB can both be used as a gasification technology. The temperature of the reactor is 800-1000°C and the pressure 1.8-2.5 MPa. The produced fuel gas is first cooled to 350-550°C and then cleaned before leading into the combustion chamber of the gas turbine. (Kurkela 2002, 3, 14.) This technique is still under development. The first commercial plant is expected to be in the oil refinery processes. Traditional steam power plants and combined gas cycle power plants are still competitive compared with IGCC technology. (Helynen et al. 2002, 62.)

### **Co-firing**

This chapter discussed biomass and fossil fuel co-firing. The use of biofuel as a mixed fuel has become a research subject of interest in many countries. The costs of biofuels as mixed fuel in existing carbon boilers are lower than the building of new biofuel boilers. If biofuels are used on their own as primary fuels, the efficiency of energy production is lower than in co-firing boilers. The availability of biofuels fluctuates. Co-firing can therefore reduce the need for storage. If there are two or more possible fuel alternatives, the choice of fuel can be made flexibly on the grounds of fuel price rather than only using one fuel. (Helynen et al. 2002, 47.) In addition, by reducing CO<sub>2</sub> emissions, co-firing in CFP boilers allows low sulphur and nitrous oxide emissions to be achieved without flue gas cleaning (Fagernäs et al. 2006, 82).

In most CFB and BFB boilers, the possible use of coal was taken into account already at the design phase. Back-pressure power plants built in the 1990's were prepared for the use of coal. In these plants, the main fuel is peat, which has had a competitive price, so the use of coal has been insignificant. Environmental regulations have also restricted sulphur emis-

sions, and this has been a legal excuse for using coal. Some plants can limit their sulphur emissions with the addition of lime. There are also cases of biofuel use in pulverized coal-fired boilers in Finland, for example, the Kymijärvi power plant owned by Lahti Energy Inc. that uses a fluidized bed gasifier to produce biogas and, for example, wood and recovered fuel (REF) as fuel. The commissioning of the gasifier reduced coal use by approximately 60,000 tons in 2002. (Helynen et al. 2002, 47-48; Lahti Energy Inc.)

The following facts have to be taken into account for biomass and coal co-firing (Helynen et al. 2002, 48):

- The fragment size of biofuel has to be small enough to allow enough time for burning out.
- In many pulverized coal-fired burners, the burning temperature is 1000-1250°C. The biofuel therefore has to be chosen from those that have a high enough melting point for ash.
- The use of biofuel can affect the quality of ash when the use of ash is not necessarily possible.
- Biomass has a lower energy density than coal. The efficiency and power of the boiler can therefore decrease.
- Combustion stability, fuel feed technology and fuel delivery have also caused some problems (Rosillo-Calle 2006, 348).

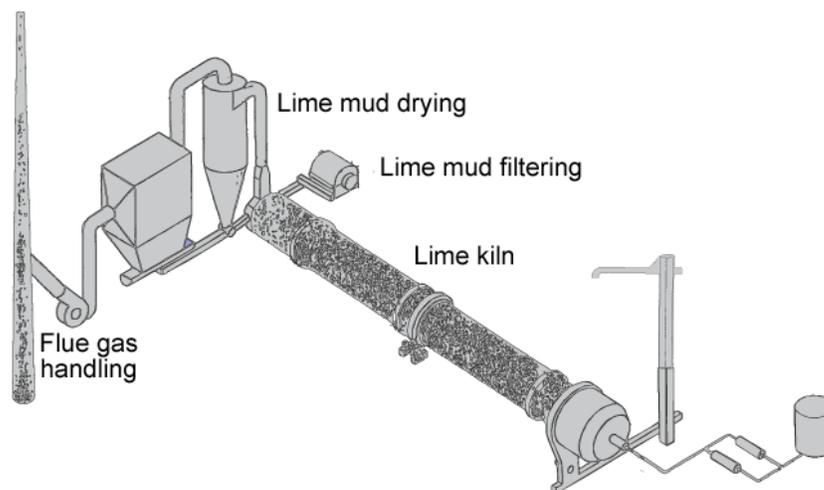
### 3.1.3 Lime kiln

Lime reburning is part of chemical circulation. Lime burning and causticizing constitute the lime cycle. In the causticizing process, sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) from the recovery boiler is converted into sodium hydroxide ( $\text{NaOH}$ ). The reaction occurs with calcium oxide ( $\text{CaO}$ ), which is a product from lime reburning. Calcium carbonate ( $\text{CaCO}_3$ ), which is called lime mud, is also generated in the causticizing process. Separated lime mud is burnt in a lime kiln to convert calcium carbonate back into calcium oxide. ( $\text{CaO}$ , Kivistö 2008, Chapter 2, p. 24-26.) Approximately 5.5- 7 GJ/t<sub>lime</sub> of heavy fuel oil or natural gas is used as a fuel in the lime kiln (Engdahl et al. 2008, 161; Fogelholm & Sutela 2008, 297). The

lime kiln is therefore the only part of the pulp mill that needs substantial purchases of fuel (Kivistö 2008, Chapter 2, p. 28). This is why the lime kiln is a potential process when examining energy production with bioenergy in the pulp mill. It is possible to burn bark and residue wood in the lime kiln if the fuel is gasified first. It is also possible to burn methanol and tall oil as well as non-condensable gases (NCG). (Engdahl et al. 2008, 175-176.)

A lime kiln is a cylinder-shaped steel construction lined with bricks. Its diameter is 2-4 m and its length 20-150 m. It is inclined horizontally by about 2.5%. The driving mechanism rotates the lime kiln at approximately 0.5-1.5 rpm. Figure 5 shows the lime burning process. The lime is fed into the lime kiln from the top. The lime mud leaks down to the bottom of the kiln. Flue gases flow in the opposite direction and heat the lime to the reaction temperature, which is approximately 1100°C. In this burning zone, chemical reactions take place that can be divided into four zones (Know Pulp 2007):

- drying, when water contained in lime mud evaporates
- heating, when lime mud heats up to the reaction temperature
- calcination, when calcium carbonate decomposes into calcium oxide and carbon dioxide
- final treatment, when lime cools prior to being removed from the kiln.



**Figure 5.** Lime kiln process (Know Pulp 2007).

## **3.2 Technologies to increase bioenergy production in conventional processes**

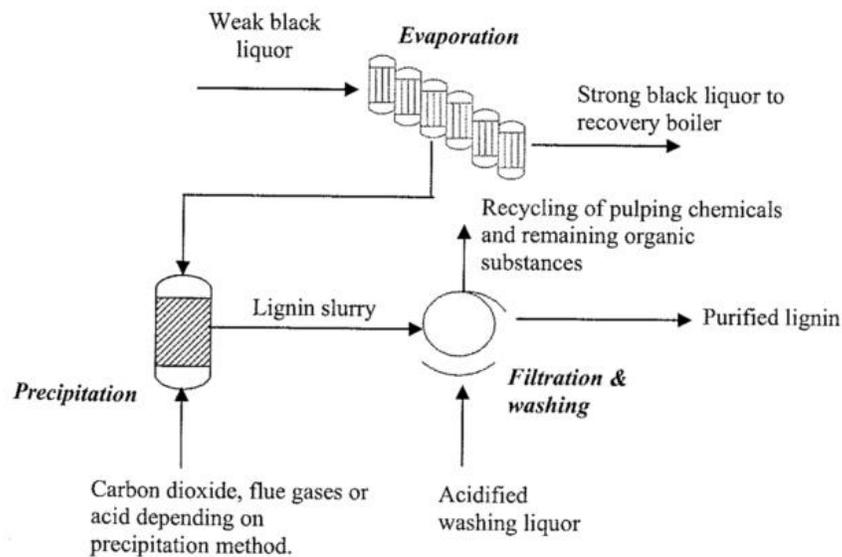
Potential bioenergy production technologies are discussed in the following chapters. The technologies represented are LignoBoost and black liquor gasification. The capacity of the recovery boiler can be increased with LignoBoost technology and the removed lignin can be burnt to produce energy (Öhman, Wallmo & Theliander 2007b, 188). In contrast, in black liquor gasification technology, the recovery boiler is completely replaced. The syngas that is produced can be used as fuel in a gas turbine or as a raw material in liquid fuel production. (Pettersson & Harvey 2009, 1.)

### **3.2.1 Lignin removal – LignoBoost**

Lignin removal from black liquor is a method to increase bioenergy use in pulp mills. The other reason for using lignin removal is that it decreases the heat load on the recovery boiler. The recovery boiler is often the bottleneck that limits production because of its heat transfer. The separated lignin could be used in a lime kiln to replace fossil fuels such as heavy fuel oil and natural gas or be burnt in a power boiler to produce energy. Modern pulp mills have an energy surplus. The extra energy generated can therefore be exported to other users in the form of solid biofuel. The separated lignin can also be used as a raw material in chemicals. (Öhman, Wallmo & Theliander 2007b, 188.) Wood includes approximately 15-30% lignin (Vakkilainen & Kivistö 2008, 5), and in the dry solid of black liquor, the proportion of lignin is 30-45% (Öhman 2006, 8). There are two ways to produce lignin: membrane filtration after digestion and precipitation, and water separation from black liquor at the evaporation stage. The latter is the so-called LignoBoost method, which is discussed in this chapter. (Vakkilainen & Kivistö 2008, 6.)

Figure 6 shows the traditional one-stage process of lignin removal. The black liquor is separated from the black liquor evaporation plant at approximately 30-40% dry solid content and led into the precipitation process to lower the pH. The pH is lowered with acidifi-

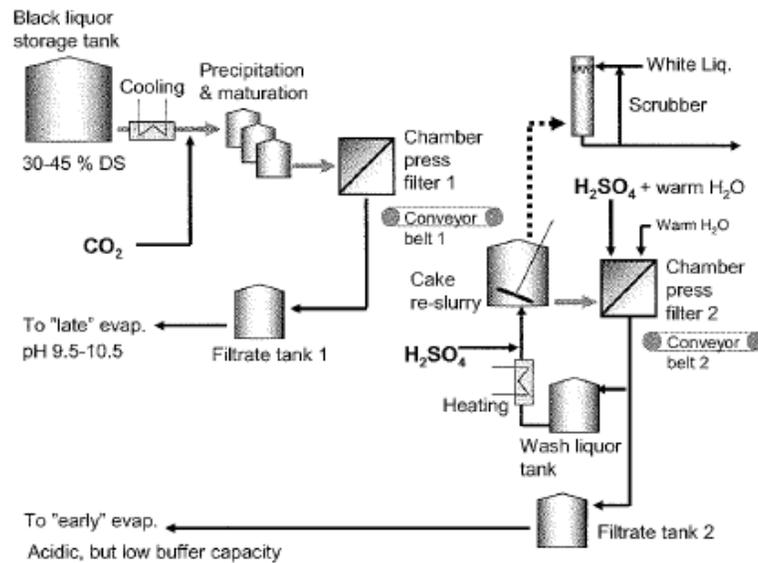
cation by injecting carbon dioxide, waste acid from older chlorine dioxide generators or sulphuric acid to the black liquor. The pH and temperature control is important to the success of precipitation. After precipitation, lignin precipitate is filtered and washed. Lignin slurry is washed with acidified water in one or several steps to remove adsorbed sodium and other contaminants from the black liquor. Wash water is returned to the black liquor recovery system to recycle the pulping chemicals and recover energy from the remaining organic compounds. (Öhman 2006, 15; Vakkilainen & Kivistö 2008, 6.)



**Figure 6.** Traditional one-stage process of lignin removal (Öhman 2006, 15).

This traditional method has some plugging problems, and a modified method for washing lignin precipitated from black liquor has therefore been developed. The filter cake plugging caused an extremely low flow of wash liquor through the filter cake as well as a very high concentration of impurities in the lignin. The plugging is assumed to be due to changes in lignin solubility. The changes in solubility are caused by excessive pH and the ionic strength gradient of the cake during the washing process. (Öhman, Wallmo & Theliander 2007a, 9-10.) The improved method of lignin removal is shown in Figure 7. The precipitation by acidification and filtering occurs as in the traditional method (“Precipitation & maturation” and “Chamber press filter 1” in Figure 7). Instead of washing the generated

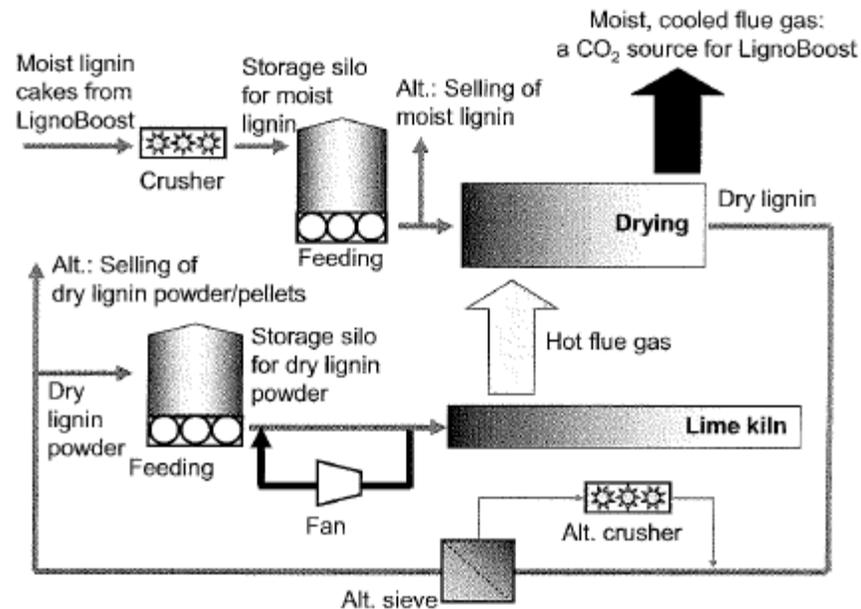
filter cake immediately, it is redispersed and acidified (“Cake re-slurry” Figure 7). The new slurry is filtered and washed using displacement washing (“Chamber press filter 2” in Figure 7). In this method, the plugging problem is avoided because the pH level and the temperature of the re-dispersed liquid are approximately those of the final wash liquor. The changes that occurred earlier in the filter cake or in the filter medium during washing have therefore already taken place in the slurry. (Tomani 2009, 460.)



**Figure 7.** The modified method for lignin removal from black liquor (Tomani 2009, 460).

The aim of the modified method is to decrease the black liquor content in the filter cake that leads to a lower consumption of acid for pH reduction. Using the spent wash water for re-slurry liquor reduces the usage of fresh liquid added to the process and keeps the energy and capacity demands in the evaporation realistic. The spent wash water includes ions that increase the ionic strength in the slurry tank however. Effective washing reduces the sodium content of lignin precipitate. The separated sodium is returned to the mill in order to avoid disturbing the sodium or sulphur balance. There is thus no excessive demand for make-up chemicals. The sodium content of lignin can also lead to corrosion problems and a low melting point of the ash if the lignin is burnt when using it as a biofuel. (Öhman, Wallmo & Theliander 2007a, 9-10; Vakkilainen & Kivistö 2008, 7.)

There have been demonstrations on firing separated lignin. Tomani (2009, 465) writes that three successful lignin firing tests were performed in different applications: co-firing with biomass, co-firing with coal and firing in a lime kiln. The tests results were good and there were no dramatic effects on the important combustion parameters. This master's thesis concentrates on the method of firing lignin in a lime kiln in order to replace fossil fuels and on the firing of lignin with biomass in a bark boiler. The test results showed that it is possible to burn lignin in a lime kiln and achieve the same burning qualities and parameters as with oil burning. There was no significant influence on the emissions of CO, H<sub>2</sub>S, NO<sub>x</sub> and SO<sub>2</sub> either. Figure 8 shows the LignoBoost method integrated with a pulp mill. Before lignin is fed into the lime kiln, it has to be dried. Hot flue gases from a lime kiln, for example can be used as drying gases. In a conventional application, the CO<sub>2</sub> used for lowering the pH to precipitate lignin in black liquor is purchased. It is also possible to produce in-house CO<sub>2</sub> from the lime kiln flue gases as shown in Figure 8. Lime kiln flue gases contain 15-30% of CO<sub>2</sub> related to moist flue gases. The use of CO<sub>2</sub> from lime kiln flue gases would lower the purchase cost of CO<sub>2</sub> by 40% to 50%. Separated lignin contains more carbon than other biofuels and has a higher heating value (HHV). The HHV was determined as 26.7 MJ/kg of dry lignin in the LignoBoost demo plant. In contrast, the HHV of wood or bark is 18 to 22 MJ/kg of dry solid. The lower heating value (LHV) of the dried lignin powder used in the demo plant was 24.4 MJ/kg of fuel at approximately 4% moisture content. (Tomani 2009, 461-465.)



**Figure 8.** The LignoBoost method integrated with a pulp mill. Flue gases from the lime kiln can be used as a CO<sub>2</sub> source. Separated lignin has to be dried before its usage as fuel in the lime kiln. (Tomani 2009, 461.)

Lignin can be burnt with biomass in a bark boiler. Burning tests in a fluidized bed boiler show that the combustion performance is normal and that lignin has not influenced it. The sulphur content of lignin has a reducing effect on the alkali chloride content in the deposit, leading to a reducing risk of sticky deposit and high temperature corrosion problems. When lignin is burnt with bark, the sulphur emissions increase compared with the situation when bark is burnt by itself. Most of the sulphur can be captured by calcium in the bark ash with the addition of limestone in the bed. The addition of lignin has no measurable effect on the sintering properties of the bed material. When limestone is added, however, the sintering temperature of the cyclone bed material decreases. The major challenges that arose in the burning tests when using lignin in a lime kiln or a bark boiler were the handling and feeding of the lignin fuel. The handling of the lignin is easier when the moisture content is low, i.e., below 10%. The dusting problem, however, increases with a rise in the dry content. This leads to a considerable risk of dust explosions, and precautions must be taken to minimize the risks of creating high dust concentrations. (Tomani 2009, 464-465.)

Although the LignoBoost method has many positive effects, it also affects the other processes in the pulp mill. The effects are caused by changes in the composition of the black liquor. The lignin removal dilutes the black liquor, resulting in a relatively higher content of inorganic material. The increase in inorganic material is also due to the inorganic compounds added during precipitation. As most of the organic part is removed, the heating value of the black liquor is lower. These changes have an effect on the evaporator line and recovery boiler operations. The important sodium and sulphur balance can also be changed. Due to the changes in the physical properties of the black liquor, there are several possible effects on the black liquor evaporators. When lignin is removed from the black liquor, the viscosity decreases. The other risk of lignin removal is scaling during evaporation. The re-alkalisation need of filtrates and spent wash water from the lignin separation process also needs to be taken into account. The increased amount of water from washing the precipitated lignin and the net removal of dry solid material lead to an increase in the demand on the evaporators. (Öhman 2006, 16-17.)

The important parameter influencing the recovery boiler is the heating value of the black liquor. There has been much research into the effects of lignin removal on the recovery boiler. In the FRAM-project (Future Resource-Adapted Pulp Mill), single droplet burning tests have been carried out, but it is difficult to translate the single droplet burning test results directly into the recovery boiler system. According to the test results, the low moderate lignin should not pose problems for recovery boiler systems. The combustion properties seemed to differ more with higher lignin removal. The gaseous emissions from the recovery boiler also differed. There were some changes in SO<sub>2</sub> formation in the single droplet test, but the NO formation was unaffected. (Öhman 2006, 16-19.) Vakkilainen and Kivistö (2008, 21) concluded that lignin removal does not have significant effects on the burning properties of black liquor. Naturally, more black liquor has to be burnt to meet the same heat requirement. The air and flue gas flows are instead close to the original values. Current recovery boilers are therefore suitable for the lignin-diluted black liquor combustion process without significant modifications. (Öhman 2006, 16-19.) According to Henrik Wallmo (11.12.2009) from Metso,  $0.3 \text{ ton}_{\text{lignin}}/\text{ADt}_{\text{pulp}}$ , which is roughly 50% of the total lignin in black liquor, can be removed without disturbing the operation of the recovery boiler.

The other alternative for separating lignin is to use it as a raw material for fuel applications, different material solutions and chemicals. As lignin is one of the most energy-rich compounds of wood, its use in fuel applications offers an interesting opportunity. The current pulp mill produces most of its energy in the recovery boiler. If lignin is extracted to reduce the load on the recovery boiler and there is no energy surplus in the mill, lignin can be burnt at the mill in, for example, a power boiler, to produce steam and electricity. Another internal utilization method is to replace fossil fuels in the lime kiln for firing lignin. The other fuel application is to export lignin from the mill if there is an energy surplus. Lignin can be co-fired with other fuels, either pellets, wood chips, coal and oil in CHP plants or in oil condensing plants. There is also an opportunity to process kraft lignin into more valuable fuel sources with fast pyrolysis, pyrolysis or gasification. Material applications that have been researched include the development of polymeric materials using lignin as a raw material, the production of a thermoplastic material consisting of 85% softwood kraft lignin and the production of carbon fibre. (Öhman 2006, 20-21.)

### **3.2.2 Black liquor gasification**

Black liquor is typically burnt in a recovery boiler, the operating principle of which is described in Section 3.1.1. Black liquor gasification (BLG) is an alternative process to recover chemicals and produce energy. The BLG can be carried out with almost unpressurized steam or with pressurized air or oxygen. The gasification processes can also be divided according to the operating temperature. The operating temperature determines the form in which most of inorganic compounds leave the reactor. In a high temperature gasifier, the temperature is 950°C or higher. The function is based on dust gasification. The inorganic compounds are in a melted form when leaving the reactor. The low-temperature gasifiers are instead based on fluidized bed gasification. The temperature is 700°C or lower and the inorganic compounds are in solid form when leaving the reactor. (Hepola & Kurkela 2002, 20.)

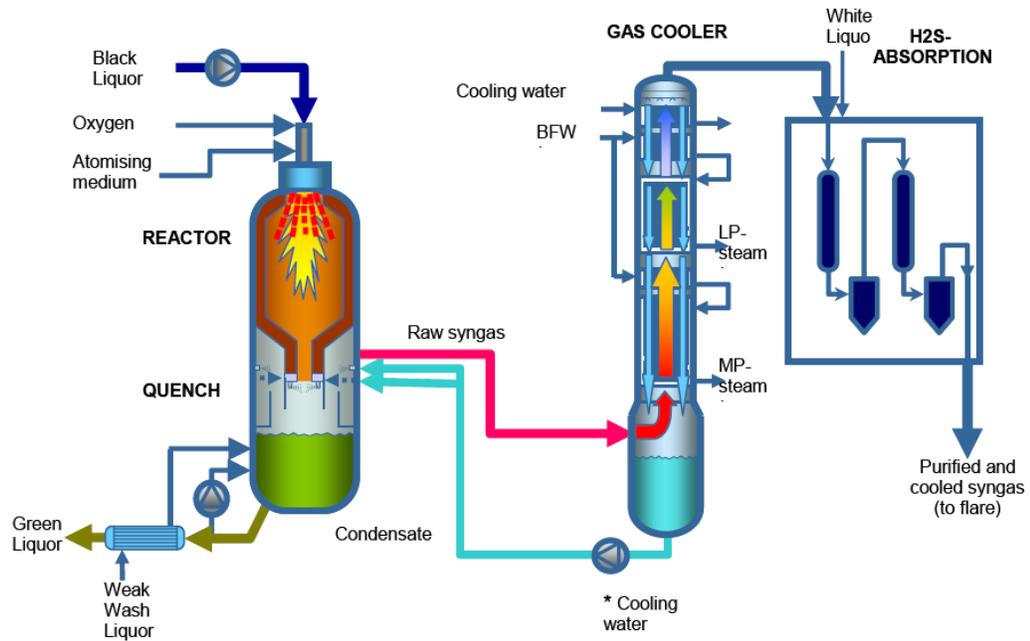
The BLG has many advantages compared with the conventional recovery boiler. It is possible to improve the power-to-heat ratio (electric power/heat power) of energy production. The BLG-IGCC process can reach a power-to-heat ratio of 0.70 and in later second-generation plants even a power-to-heat ratio of 0.83. The other advantages of BLG compared with a recovery boiler are (Hepola & Kurkela 2002, 21):

- 5-10% higher heat production efficiency
- lower NO<sub>x</sub>, sulphur, SO<sub>2</sub> and CO<sub>2</sub> emissions
- improved safety
- possibility to raise the fibre yield by approximately 2-4% because of better sulphur recovery.

Although the BLG power plant has been estimated to cost 30% more than the ordinary process, the cost of electricity production is lower than for a recovery boiler. The BLG process in the integrated pulp and paper mill may reduce heat production, and this has to be taken into account. The gasification enables intensification of chemical circulation without the investment in a new recovery boiler however. The profitability of gasification therefore has to dissect the view of the missing heat production costs. (Hepola & Kurkela 2002, 21.)

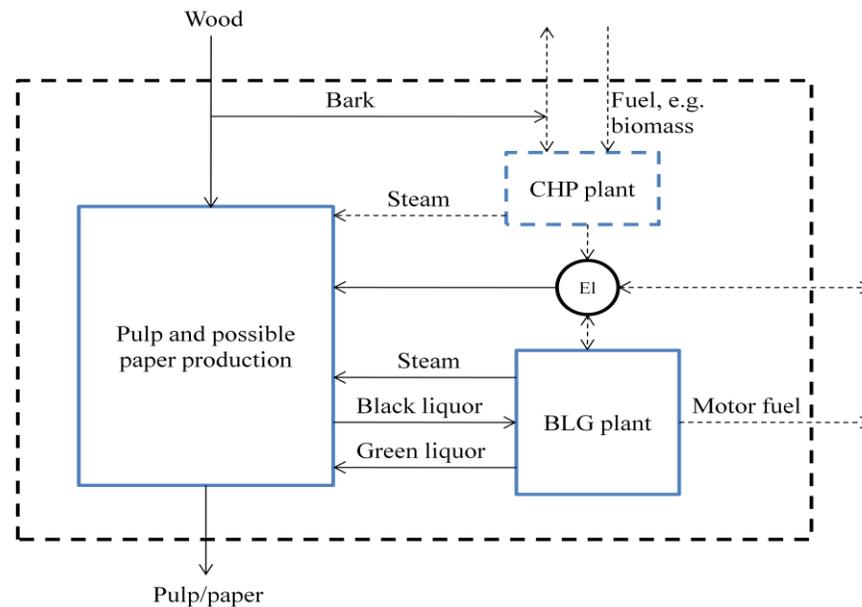
Research into BLG has been under way for decades in Finland, especially in 1989-1992 when BLG was studied at the pilot plant Äänekoski. Since then, the examination has continued on smaller scale pilot plants. (Hepola & Kurkela 2002, 22.) Nowadays, the most promising technology is the Swedish Chemrec's atmospheric and pressurized BLG technology. A new development plant was started up in 2005 in Piteå, Sweden. Figure 9 shows a drawing of the development plant. The plant consists of the reactor with the quench cooler, gas cooler and heat exchanger for cooling hot green liquor. The black liquor and oxygen are fed into the reactor from the top. The temperature of the reactor is just above 1000°C. The residence time in the reactor is about 5 seconds. The generated gas and melted inorganic salts are quenched by water spray. Salts separated by gravity are felt in the quench bottom and form green liquor. The generated raw gas is led into a countercurrent gas cooler. The cooled raw gas has to be refined in the H<sub>2</sub>S-absorption unit because it con-

tains 1.4-2.5% vol of H<sub>2</sub>S. In a commercial-scale BLG plant, some structural changes would be necessary. (Lindblom & Landälv 2007.)



**Figure 9.** The principal units of the oxygen-blown, pressurized black liquor gasification development plant 1 (DP1) in Piteå, Sweden. The plant consists of the reactor with the quench cooler, gas cooler and heat exchanger for cooling hot green liquor. (Lindblom & Landälv 2007.)

The BLG process produced syngas that can be used for electricity production in a cogeneration unit or in the production of motor fuels. Excess heat, which is produced in both processes, is used in steam production. Part of this steam is used internally in the BLG plant, but both processes have a large steam surplus that can be used at the pulp mill. It has to be taken into account, however, that less steam is produced in the BLG process than in a conventional recovery boiler. If the produced steam cannot cover the mill steam demand, the additional steam can be produced in a CHP plant by burning biomass. Figure 10 shows an overview of the main energy and material streams in a possible future pulp mill with BLG. (Pettersson & Harvey 2009, 2.)



**Figure 10.** Main energy and material streams in a market pulp mill/integrated pulp and paper mill. The produced steam from BLG plant can be utilized at the pulp mill. (Pettersson & Harvey 2009, 2.)

### 3.3 Additional bioenergy production processes in a pulp mill

Biofuels can be refined into a form that makes them easier to use or be used to replace liquid or gaseous fossil fuels. These kinds of solid processed products include wood dust, charcoal, wood pellets and briquettes. Liquid processed products are pyrolysis oil, ethanol, methanol, turnip rape diesel and the addition of gasoline. Solid processed products are not taken into account in this thesis when examining refined biofuel technologies. Natural gas can be replaced with biogas in different applications. Liquid fuels can be manufactured from solid raw materials with numerous methods. The thermochemical methods are pyrolysis, direct liquefaction and indirect liquefaction by gasification. Bioalcohols can also be manufactured for transportation use from sugars of biomass. These synthesis fuels are used in applications that can only use liquid fuels. (Helynen et al. 2002, 77.)

The motivation for increasing synthesis fuels has been the need and desire to reduce oil dependence. The applications of synthesis fuel include transportation and oil vessels. The advantages of synthesis fuels compared with solid raw materials are their higher energy den-

sity and easier processability and storability. Emission control in small applications is also easier when using synthesis fuels than solid fuels. The CO<sub>2</sub>-emission reduction can also be the reason for using renewable raw material to produce synthesis fuels or the overproduction of agriculture can be directed at energy production. The production costs of synthesis fuel are usually higher than for conventional fuel like oil or natural gas. The production of synthesis oil therefore presumes, for example, tax reliefs. Integrating the production into the pulp- or sawmill, the power plant or the food industry also improves the competitiveness of synthesis fuels. (Helynen et al. 2002, 77.)

### 3.3.1 Pyrolysis oil

Pyrolysis is a thermal decomposition that occurs without oxygen. Pyrolysis is the first step of combustion and gasification processes. It follows the total or partial oxidation of the primary product. (The IEA Bioenergy Biomass Pyrolysis Network.) In the combustion process, the pyrolysis of volatile matters begins at a temperature of 150-200°C. The most efficient pyrolysis occurs at a temperature of 250-500°C. At that temperature, plenty of tar and gaseous compounds are formed. Pyrolysis ends at a temperature of 800°C. In addition to inert gases, pyrolysis produces combustible and liquid phase tar compounds that burn if there is sufficient oxygen. (Helynen et al. 2002, 39.)

In the pyrolysis of biomass, the biomass is heated in an oxygen-free reactor, and gases, and liquid and solid char products are generated. The formation of liquid products can be affected by the temperature of the reactor and the residence time. Table 3 shows typical yields (dry wood basis) obtained by different modes of pyrolysis. As the table shows, fast pyrolysis is the most efficient process for producing pyrolysis oil. The temperature is approximately 500°C and the residence time in the reactor is short. The products of pyrolysis are cooled quickly. From dry biomass, 70-75 wg.-% liquid product can be produced. (Helynen et al. 2002, 78.) When the biomass decomposes in fast pyrolysis, vapours, aerosols and some charcoal are generated. When these products are cooled, a dark brown mobile liquid is formed. The generated liquid has approximately half the heating value (19 MJ/kg)

of conventional fuel oil (heating value 42-44 MJ/kg. (The IEA Bioenergy Biomass Pyrolysis Network.)

**Table 3.** Typical product yields (dry wood basis) obtained by different modes of pyrolysis of wood (The IEA Bioenergy Biomass Pyrolysis Network).

| <b>Mode</b>            | <b>Conditions</b>                                | <b>Liquid<br/>[Wg.%]</b> | <b>Char<br/>[Wg.%]</b> | <b>Gas<br/>[Wg.%]</b> |
|------------------------|--|--------------------------|------------------------|-----------------------|
| Fast pyrolysis         | ~500°C, short hot vapour residence time ~ 1 s    | 75                       | 12                     | 13                    |
| Intermediate pyrolysis | ~500°C, hot vapour residence time ~ 10-30 s      | 50                       | 25                     | 25                    |
| Slow torrefaction      | ~290°C, solids residence time ~ 30 min           | -                        | 82 solid               | 18                    |
| Slow carbonisation     | ~400°C, long vapour residence time ~ hours->days | 30                       | 35                     | 35                    |
| Gasification           | ~800°C   | 5                        | 10                     | 85                    |

The energy density of pyrolysis oil is four to five times higher than for original biomass, which has positive logistic advantages. The generated pyrolysis oil has many applications. It can be used for the production of heat and power, as transport fuel and for chemicals. The simplest and most straightforward application of pyrolysis oil is to combust it in a boiler or furnace to produce heat. Heavy and light oil can be replaced with pyrolysis oil in industrial boiler applications. The co-combustion process with natural gas has also been successfully demonstrated. Power production in gas turbines and diesel engines has been tested but needs further development. Different manufacturing processes have been developed to produce transportation fuels from pyrolysis oil. These are direct upgrading or gasification combined with gas-to-liquid synthesis to produce methanol, Fischer-Tropsch diesel or DME (dimethyl ether). Based on economic attractiveness, chemical manufacturing needs further development. (Biomass Technology Group.)

### 3.3.2 Liquid fuels from the forest industry's by-products

The most important by-products of the kraft pulp mill are black liquor, sulphate soap and tall oil. Tall oil, formed from extractives of wood, is isolated from sulphate soap by acidification (parcelling). The sulphate soap is resolved from black liquor in the evaporation plant of black liquor. Liquid bio-based fuels can be produced from these by-products. The alternative processes for producing liquid fuel or flue gas from black liquor have been examined. Gasification has proved the most competitive process. Black liquor gasification was discussed earlier in Section 3.2.2. The refining of sulphate soap into liquid fuel has turned out to be promising. Almost all sulphate soap is parcelled in Finland. After parcelling, the tall oil fatty and resin acid fractions can be refined from the sulphate soap. Tall light oil and tall-oil pitch can also be separated from it. The only alternative to parcelling is sulphate soap burning in a recovery boiler. (Helynen et al. 2002, 79-80.)

Tall oil, tall light oil and tall oil pitch can be used as liquid fuels in the raw as a substitute for heavy fuel oil. Their direct use as motor fuel for diesel power plants is a new alternative that has not been well examined. Tall oil fatty acid can be refined into ester, which can be used as good quality diesel fuel oil. Thermal treatment tests of sulphate soap have been executed in the laboratory and good results have been obtained. The end product of this process was hydrocarbonaceous light fuel oil. Its qualities as flue oil are better than those of raw tall oil. Thermal treatment can be applied to mixed sulphate soap (soap from pine and from birch) that is not suitable for tall oil production. Another interesting fuel oil production method is fast pyrolysis. (Helynen et al. 2002, 80.)

Tall oil has many applications as a raw material in the chemicals industry. The tall oil production capacity in 2004 was 250,000 tons (9.2 PJ). There are two existing tall oil refineries in Finland (in 2006). Of the upgrading products from tall oil, 90% are exported. The Finnish tall oil fatty and resin acid fraction share of the global market is 20%. The heating value of tall oil is 90% of the heating value of heavy fuel oil, which is 41.0-41.3 MJ/kg. (Heinimö & Alakangas 2006, 77; Energia Suomessa 2004, 381.) Due to its high heating value, an alternative use for tall oil is in the lime kiln, in energy production to replace fossil

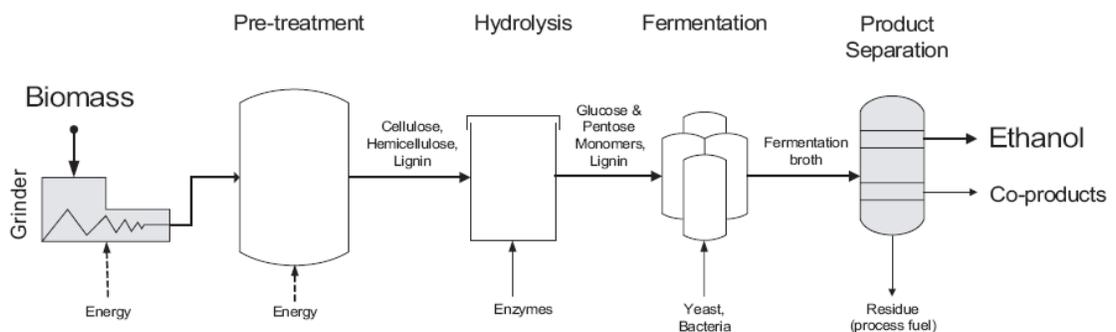
fuels. Due to emission trading, the competitiveness of tall oil as a raw material is weaker than its use in energy production. There has been discussion on tall oil usage in energy production because its use for that purpose reduces the raw material from the refineries. In order for tall oil refiners to obtain enough raw material and thereby continue their operation, they have to be able to pay a price that beats the cost of alternative fuels for pulp mills. (Heinimö & Alakangas 2006, 77.) These reasons have to be taken into account when examining fossil fuel replacement in pulp mills. In the case of tall oil, its use in energy production could end Finnish tall oil refinery operations completely.

### **3.3.3 Transport fuel**

Transportation produces approximately 18% of the greenhouse gases in Finland (Long-term Climate and Energy Strategy 2008, 67). The EU target to increase the share of renewable energy in transportation energy end-use consumption to 10% by 2020 is one option to reduce CO<sub>2</sub> emissions. The current share of biofuels of the total transport fuel use in Finland is 2-3% (Soimakallio, Antikainen & Thun 2009, 62). Current biofuels are still so-called first-generation biofuels for which raw material has been used: sugar, starch, vegetable oil and animal fats (Soimakallio, Antikainen & Thun 2009, 57). Finland cannot achieve the 10% target with only domestic raw materials and technology; imported raw materials and biofuels have to be taken into account. Second-generation biofuels are under development and offer a potential method to achieve the target. Second-generation biofuels differ from first-generation biofuels in the raw material. The raw materials used for second-generation biofuels are non-edible products such as wood, waste and residues from agriculture (Long-term Climate and Energy Strategy 2008, 39.) First- and second-generation biofuels also have other definitions. The Development Manager of Neste Oil, Seppo Mikkonen, says that the quality of the product defines the generation subject, not the production method (Lukkari 2007). The next chapters discuss the possibility of using wood-based biomass as a raw material for transportation fuel production and second-generation biofuel technologies. Most of the developed second-generation biofuel production technologies are still on a pilot or demo scale and have not yet been implemented on a full industrial scale. Second-

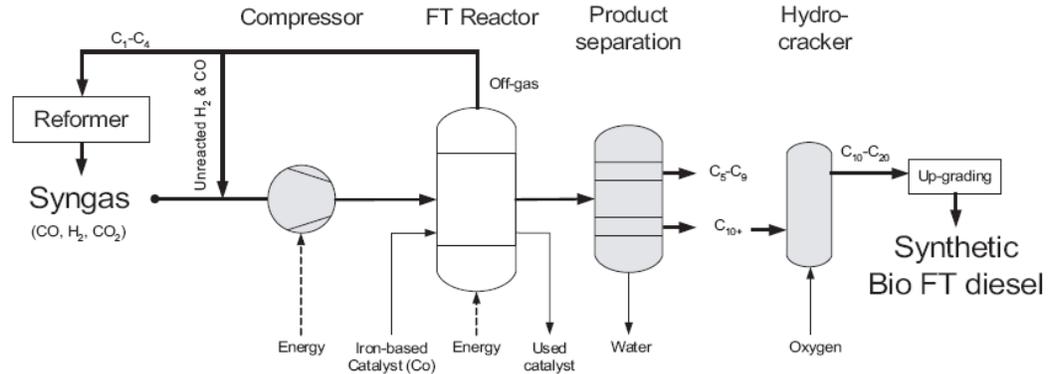
generation biofuels are also estimated to emit less greenhouse gases than typical first-generation biofuels. (Soimakallio, Antikainen & Thun 2009, 106.)

Forest-based raw materials suitable for the production of liquid biofuels are logging residues and stumps from clear-cut areas, and small trees from thinning. These raw materials are currently used for the forest industries' own needs, but their use for energy production and other purposes is increasing. (Soimakallio, Antikainen & Thun 2009, 69.) The potential of forest-based biomass is discussed in Section 2.4. There is one liquid biofuel that is suitable for transportation and that is bioethanol from lignocellulose. The woody biomass is used as a raw material in bioethanol production. The production of bioethanol requires many processing steps that the complex structure of hemicelluloses, cellulose and lignin have overcome to make them accessible to hydrolysis and fermentation. The first step is pre-treatment, when the cellulose structure is disrupted, the lignin seals are broken and the hemicellulose is partially removed. After pre-treatment, the forms of cellulose and hemicelluloses are more accessible to enzymes, which are needed in the hydrolysis process. In the hydrolysis process, the enzymes break the cellulose down into sugars. After that, the sugars are fermented into ethanol in the fermentation process. The fermentation takes place with wild type yeasts or genetically engineered bacteria or yeasts. The generated product after fermentation contains ethanol as well as water and residues that are separated in the final process step. (Schwietzke et al. 2008, 7-8.) Figure 11 shows the process flow diagram of bioethanol production from lignocelluloses.



**Figure 11.** Process flow diagram for ethanol production from lignocelluloses (Schwietzke et al. 2008, 8).

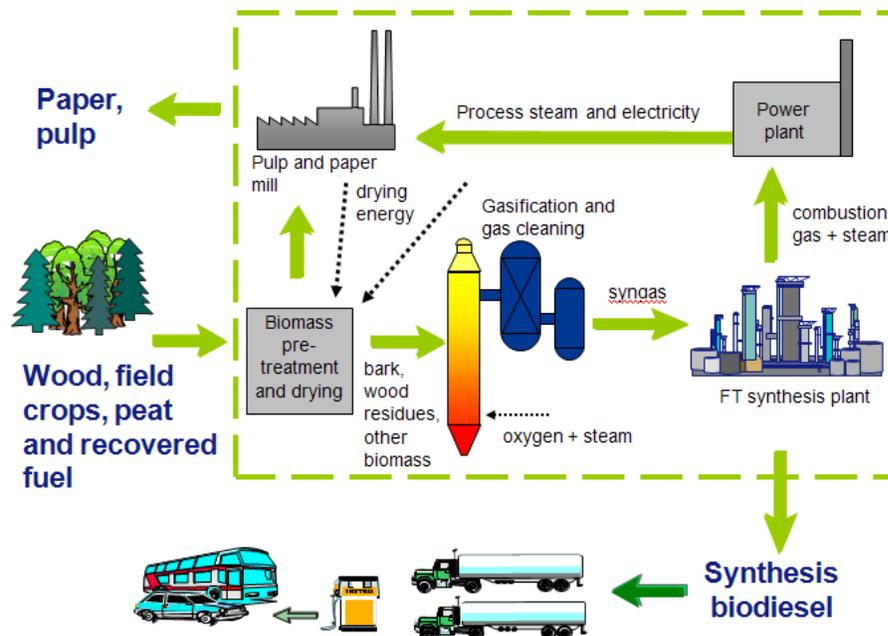
Coal and natural gas have been used as raw materials in commercial Fischer-Tropsch (FT) diesel and gasoline processes for decades in South Africa (Sasol) and Malaysia (Shell). FT technology is currently being developed for biodiesel production with biomass as the raw material. A demonstration plant has been built in Finland. The biomass-based FT process consists of pre-treatment, gasification, gas cleaning and conditioning, FT synthesis, and an upgrading and recycling section. The FT process diesel production flow diagram, in which syngas is used as a feedstock, is shown in Figure 12. (Soimakallio, Antikainen & Thun 2009, 106-107; Gust 2010.)



**Figure 12.** Process flow diagram for synthetic bio-FT diesel production using syngas as a feedstock (Schwietzke et al. 2008, 10).

First, the raw material is gasified and then the gas is conditioned and cleaned of catalyst impurities. Biomass gasification is discussed in more detail in Section 3.1.2. The generated product gas, which is a combination of hydrogen and carbon monoxide, also known as synthesis gas or syngas, is pressurized and then converted into long-chain hydrocarbons by the FT synthesis. The products of FT synthesis vary depending on the catalyst and reaction parameters used. In the upgrading process, the generated FT liquid is converted into FT fuels, one of which is FT diesel. FT diesel has similar qualities to conventional diesel except that it does not contain any sulphur or aromatics and thus will combust more cleanly than conventional diesel. It is therefore easy to use without engine modifications or the construction of a new fuel distribution system. FT diesel can also be used as a mixed fuel with conven-

tional diesel. The cost of FT diesel and other biofuels can be reduced by integrating their production into, for example, a pulp and paper mill. This enables efficient utilization of by-product energy from FT diesel production when the total efficiency of the process can be increased. In the integrated process, the heat produced in the FT process reduces the heat demand in the CHP plant. The electricity production is also lowered because of the reduced need for separately produced extra electricity. (Soimakallio, Antikainen & Thun 2009, 107-110; Gust 2010.) Figure 13 shows transport biofuel production integrated into the pulp and paper mill.



**Figure 13.** Transport biofuel production integrated into the pulp and paper mill. The excess heat from FT diesel process can be utilized in a pulp and paper mill. (Kurkela 2006.)

Other transportation fuels such as methanol, dimethyl ether (DME), methane and hydrogen can also be produced from biomass in similar kinds of processes, for example, FT synthesis (Soimakallio, Antikainen & Thun 2009, 107). Methanol is produced in a methanol synthesis reaction from syngas feedstock. The integration of the gasification process into the pulp and paper mill offers the advantage of providing low-cost black liquor as a feedstock. DME can be produced via methanol synthesis in a separate reactor or by using a single reactor. Methanol can be used directly as a transportation fuel in combustion engines or as a very

limited share of fuel shells of current vehicles. DME can also be used directly in diesel engines. Biosynthetic natural gas (Bio-SNG) is derived from biomass via thermochemical conversion. It contains mainly methane, but also some hydrogen, carbon dioxide and nitrogen. Bio-SNG production utilizes the off-gas from the FT synthesis process. In the future, it will be possible to use the Bio-SNG in transportation fuel in a similar way to compressed natural gas and liquefied natural gas. One alternative transportation fuel is green diesel, which is derived from pyrolysis. In the condenser, manufactured bio-oil is upgraded to convert the bio-oil into a fuel that can be used directly in diesel engines. Green diesel is not yet a commercial product, but interest in it is growing. (Schwietzke et al. 2008, 10-13.)

## **4 BACKGROUND INFORMATION FOR CALCULATIONS**

The life-cycle assessment (LCA) method is used in this master's thesis. The environmental impacts of different case studies have been examined during their life cycles. The carbon footprint is used as a tool to calculate systems' greenhouse gas emissions as carbon dioxide equivalents. The LCA and carbon footprint are presented in detail in the following chapters.

### **4.1 Life-cycle assessment (LCA)**

The awareness of the importance of environmental issues and the interest in environmental impacts of products and services has increased. Methods have been developed to understand these impacts better. Life-cycle assessment is a widely used tool, which was developed for this purpose. The methodology is standardized according to SFS-EN ISO standards 14040 and 14044. According to SFS-EN ISO 14040 (2006, 22), the aim of LCA is to address the environmental impact of a product during its life cycle, from cradle to grave, so that the LCA takes into account the impacts of manufacturing the raw material used, the disposal and recycling. The environmental aspects and impacts are taken into account, when economic and social aspects and impacts are typically excluded from the LCA. The LCA consists of four stages. These are:

- 1) Goal and scope definition, which defines the system under study, the functional unit, the product specifications of the systems and the system boundaries (SFS-EN ISO 14040, 31).
- 2) Life cycle inventory (LCI), which calculates all the inputs into the system and the outputs from the system. The LCI takes into account all the processes in the life cycle that produce inputs. The allocation of flows and releases are also included in the LCI. (SFS-EN ISO 14040, 35.)
- 3) Impact assessment (LCIA), in which the most significant environmental aspects are evaluated with the results of the LCI. The inventory results are associated with specific environmental impact categories and category indicators. The purpose is to understand these impacts. (SFS-EN ISO 14040, 35.)
- 4) Interpretation of the results. In this phase, the results of the LCI and LCIA are considered together. The results should be consistent with the defined goal and scope. Based on these results, conclusions can be reached, limitations explained and recommendations provided. (SFS-EN ISO 14040, 39.)

## **4.2 Carbon footprint**

The carbon footprint is one part of the LCIA. In this thesis, the environmental effects are examined using the carbon footprint; other methods are excluded. The carbon footprint calculation procedure is based on life-cycle thinking and LCA methodology. The carbon footprint takes into account all greenhouse gas (GHG) emissions (usually the most important gases CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O) produced throughout the product's value chain. GHG emissions are reported as CO<sub>2</sub>-equivalent emissions, which are calculated by multiplying the mass of a given GHG by its global warming potential (GWP). The GWP describes the radioactive forcing impact of one mass-based unit of a given greenhouse gas relative to an equivalent unit of CO<sub>2</sub> over a given time period. Table 4 presents the GWPs for a 100-year period produced by the IPCC. (PAS 2050:2008, 2-3, 26.)

**Table 4.** GWPs for CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O

| <b>GHG</b>       | <b>GWP</b> |
|------------------|------------|
| CO <sub>2</sub>  | 1          |
| CH <sub>4</sub>  | 25         |
| N <sub>2</sub> O | 289        |

The current carbon footprint calculation only includes greenhouse gases from fossil sources. The calculations in this thesis were also supposed to include biogenic CO<sub>2</sub>. The European Forest Institute (EFI) calculated the effects of wood harvesting on the forest carbon balance but noted that the calculation procedure is not, as yet, exploitable in a product-specific calculation. It was therefore decided to exclude biogenic CO<sub>2</sub> from this master's thesis. In general, the inclusion of biogenic CO<sub>2</sub> in the carbon footprint poses many challenges because there are several issues of concern. The forest carbon balance and time scale are a consideration when including biogenic carbon in the carbon footprint. The issue of carbon storage in the product is also challenging when dealing with its role in the carbon footprint. (Kujanpää, Pajula & Hohenthal 2009, 31.)

PAS 2050:2008 (Publicly Available Specification), which was prepared to specify requirements for assessing the life-cycle greenhouse gas emissions of goods and services, discusses biogenic carbon. It lays down that biogenic carbon should be excluded from carbon footprints and that only carbon storage in products and the impact of land use change can be calculated in carbon footprints. (PAS 2050:2008, 7.) Methods have been developed to include forest carbon sequestration in the carbon footprint (Kujanpää, Pajula & Hohenthal 2009, 33). With regard to the carbon storage of the products, PAS 2050:2008 states that the impact of carbon storage shall be taken into account “if more than 50% of the mass of carbon of biogenic origin in the product remains removed from the atmosphere for one year or more following production of the product”. (PAS 2050:2008, 8.)

## 5 THE CASES

Case studies of different bioenergy production technologies are examined in the following chapters. The bioenergy production technologies that are studied are LignoBoost and Fischer-Tropsch diesel. In the LignoBoost case, the process is integrated into the connection of the pulp mill. The process is compared with the conventional pulp mill situation in the integrated pulp and paper mill. The effect of the LignoBoost process on the carbon footprint of the integrated pulp and paper mill is examined. The functional unit of the calculations is 1000 kg LWC paper.

In the FT diesel case, two co-products, LWC paper and FT diesel, are produced. The replacement of fossil diesel with FT diesel is included in the examination. The ISO 14044 Standard suggests avoiding allocation whenever possible. System expansion is therefore used to allow comparisons of FT diesel production and fossil diesel production. Although in cases 2b and 2c the FT diesel production is integrated with pulp and paper mill, it is not possible to say, who can receive the advantage from CO<sub>2</sub>-emission reduction, the pulp and paper producer or FT diesel producer. Therefore although the emissions related to pulp and paper production and the emissions related to fossil diesel or FT diesel production are presented in the same columns (production sites -level), it has to be taken into account that the CO<sub>2</sub>-equivalent emission reduction is not only the carbon footprint reduction of pulp and paper mill. The calculations are based on the typical Finnish pulp and paper mill which annual paper production capacity is 700,000 tons. The functional unit of the calculations is 1000 kg LWC paper (lightweight coated paper) and 214 kg diesel (i.e. fossil diesel in case 2a and FT diesel in cases 2b and 2c). The integrated pulp and paper mill was chosen by a reference case to avoid the additional final product, which would be energy if only the pulp mill was examined.

The environmental impacts from cradle-to-grave are usually taken into account in an LCA examination. As the purpose of this thesis is to examine the effects of different biofuel production technologies, the LCA will be carried out in two examination phases in which sys-

tem boundaries are limited to the transportation of the product to the customer and the usage of the products is excluded. In the LignoBoost process the mill-level examination is taken into account first and after that, the whole life cycle is researched with a cradle-to-customer approach. Thus, only the effects of the current technology on the pulp and paper mill carbon balance can be found. In the fossil diesel case 2a, the production sites of paper and fossil diesel are examined together even though the production sites would be unconnected in reality. In the FT diesel cases 2b and 2c the integrated production sites of FT diesel and paper are taken into account. In cradle-to-customer approach the CO<sub>2</sub>-equivalent emissions of fossil diesel production can be separated from the carbon footprint, but in the FT diesel cases it is not possible to allocate the total CO<sub>2</sub>-equivalent emissions to FT diesel and paper separately. That is why the results are presented in the same columns but it has to be taken into account that the results are not only the carbon footprint of the integrated pulp and paper mill. The whole life cycle considers, for example, the effects of chemical production, wood logging, electricity purchasing, transportations and other impacts. The greenhouse gas emissions are presented as CO<sub>2</sub> equivalents containing CO<sub>2</sub> fossil, CH<sub>4</sub> and N<sub>2</sub>O emissions to the air. The GHG emissions are calculated by KCL-ECO. Table 5 shows brief case descriptions.

**Table 5.** Case descriptions.

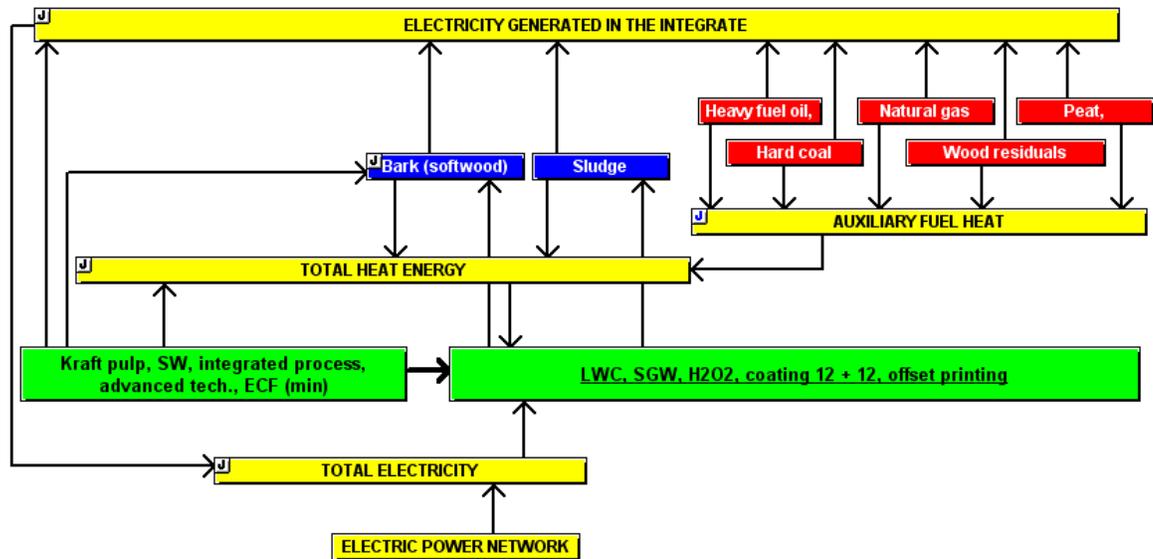
| <b>Name</b>    | <b>Forest management</b>  | <b>Mill process</b>                | <b>Fuel production</b> | <b>Fuel production information</b> |
|----------------|---------------------------|------------------------------------|------------------------|------------------------------------|
| <b>Case 1a</b> | Current                   | Integrated LWC mill                |                        |                                    |
| <b>Case 1b</b> | Current                   | Integrated LWC mill and LignoBoost |                        |                                    |
| <b>Case 2a</b> | Current                   | Integrated LWC mill                | Fossil diesel          |                                    |
| <b>Case 2b</b> | Forest residues collected | Integrated LWC mill                | FT diesel              | Condensing                         |
| <b>Case 2c</b> | Forest residues collected | Integrated LWC mill                | FT diesel              | Back pressure, no bark boiler      |

Some assumptions and simplifications are made in the calculations. The modules used in the calculations are from the KCL-ECO databases. Chemicals with amounts below 1 kg per ton of paper in the pulp and paper manufacturing process are excluded. The production of wood residuals used in auxiliary fuel heat production is also excluded from the cradle-to-customer approach because of the small amounts. Only the reference product (1 ton of LWC paper) is taken into account. The other by-products like tall oil, turpentine and solid wastes are excluded. The transport distance of chemicals and final products is chosen as 200 km and of wood as 100 km. The modules of heavy fuel oil and natural gas production already include transportation, though they miss out the peat and hard coal modules, so a 200 km transport profile is added to these modules. Auxiliary fuel transportation is not separated in this examination, but it is included in the code of auxiliary fuel manufacturing. A 25-ton truck is used in all transportation modules. This thesis uses the auxiliary heat module from the EcoData database in which the distribution of auxiliary fuels was defined by the Finnish Forest Industry Federation in 1997. This is the average share of auxiliary fuels used in paper mill heat production in 1997. In this approach, the module of used electricity represents the average Finnish five-year electricity production. The proportion of pine and spruce chips (67.7%/32.3%) to the pulp mill is chosen based on the earlier calculation made in KCL.

## **5.1 Case 1a: Integrated LWC mill**

The reference pulp and paper mill is a typical Finnish integrated SW kraft pulp and paper mill taken from the KCL-ECO EcoData database. The annual paper production capacity is 700,000 tons. The pulp mill produces 240 kg of pulp per ton of paper. The reference pulp mill includes the pulping process, bleaching, activated sludge plant and sludge combustion. In this thesis, the pulp mill used includes the elemental chlorine-free (ECF) bleaching process. The properties of pulp are kappa number 8 and brightness 89 ISO. The dry substance of black liquor is at 80% when feeding it into the recovery boiler. The efficiency of the recovery boiler is 69% and the flue gas cleaning system is a high-efficiency electrostatic precipitator (ESP). The back-pressure construction factor is 0.22. The paper mill includes wood

handling, mechanical pulping, a paper machine and an activated sludge plant. The most important chemicals used in the processes are  $H_2O_2$ ,  $NaOH$  and  $O_2$  and they amount to more than 20 kg per ton of pulp. Figure 14 shows the flowsheet of reference pulp and paper mill integration. Appendix II shows the flowsheet with flow values.



**Figure 14.** Case 1a: Mill-level examination of reference pulp and paper integration mill. The functional unit is 1000 kg LWC paper. The annual production capacity is 700,000 tons of paper

The pulp mill is self-sufficient in energy production and produces extra energy for use by the paper mill. Although auxiliary fuels are needed to produce heat and electricity, extra electricity is purchased from the grid to cover the energy needs of the paper mill. When only the mill-level examination (Figure 14) is taken into account, the carbon footprint in the reference case is 355 kg. These emissions are generated during heavy fuel oil burning in a lime kiln and during auxiliary fuel burning to produce the necessary heat.

Next, the modules, presented in Figure 14, are agglomerated into one module, and chemical production, wood logging, electricity purchasing, transportation and other impacts are taken into account in order to find all  $CO_2$ -equivalent emissions during the life cycle of paper production. In this thesis, the approach is called the cradle-to-customer approach. The new flowsheet is presented in Figure 15.

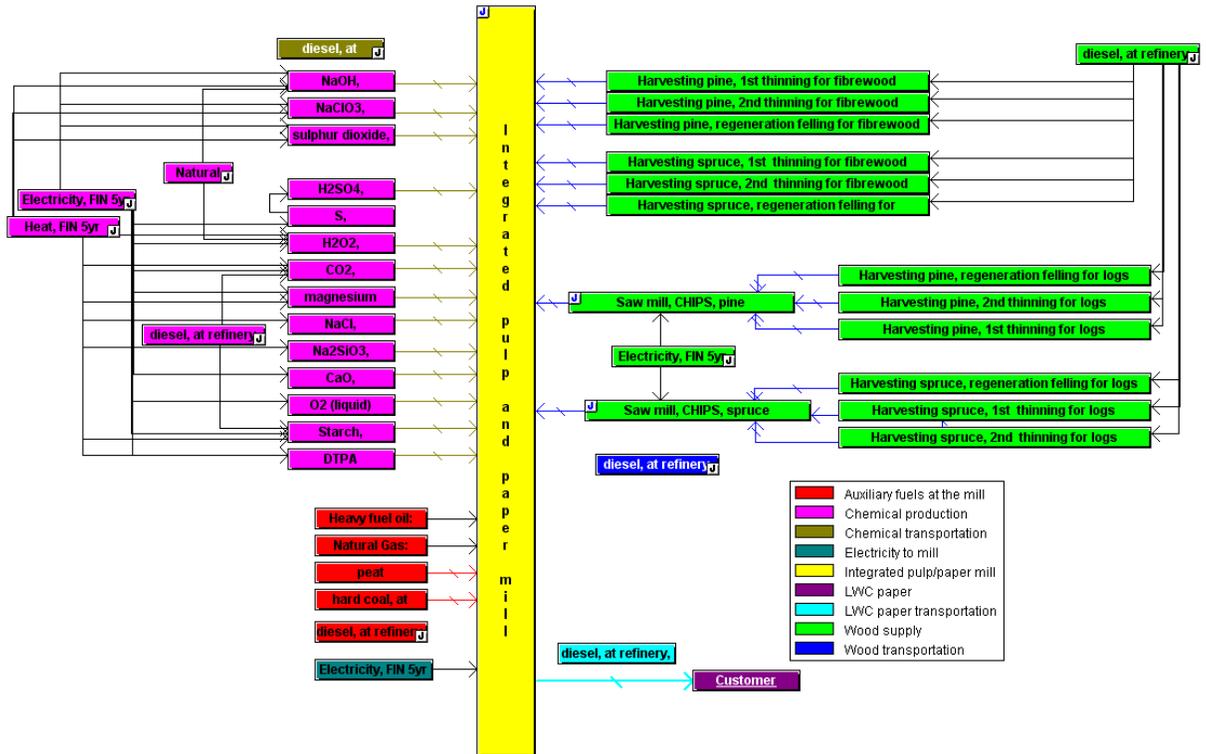
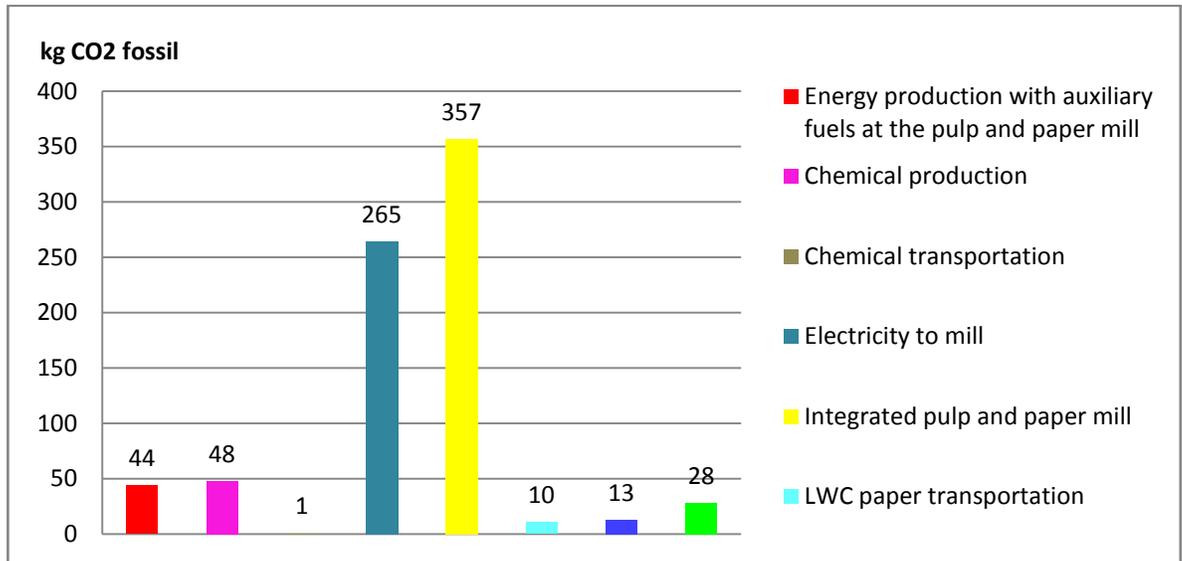


Figure 15. Case 1a: Reference mill in the cradle-to-customer approach.

CO<sub>2</sub>-equivalent emissions in the cradle-to-customer approach are presented in Figure 16. The total CO<sub>2</sub>-equivalent emissions are approximately 760 kg. When examining the CO<sub>2</sub> equivalent emissions in the cradle-to-customer approach it can be noted that the share of the mill-scale operations causes the biggest emissions with a share of 47% of the total emissions (about 355 kg CO<sub>2</sub>eq). The biggest individual emission source outside the integrated pulp and paper mill is electricity production to the mill. It is responsible for approximately 35% (about 265 kg) of the total emissions.

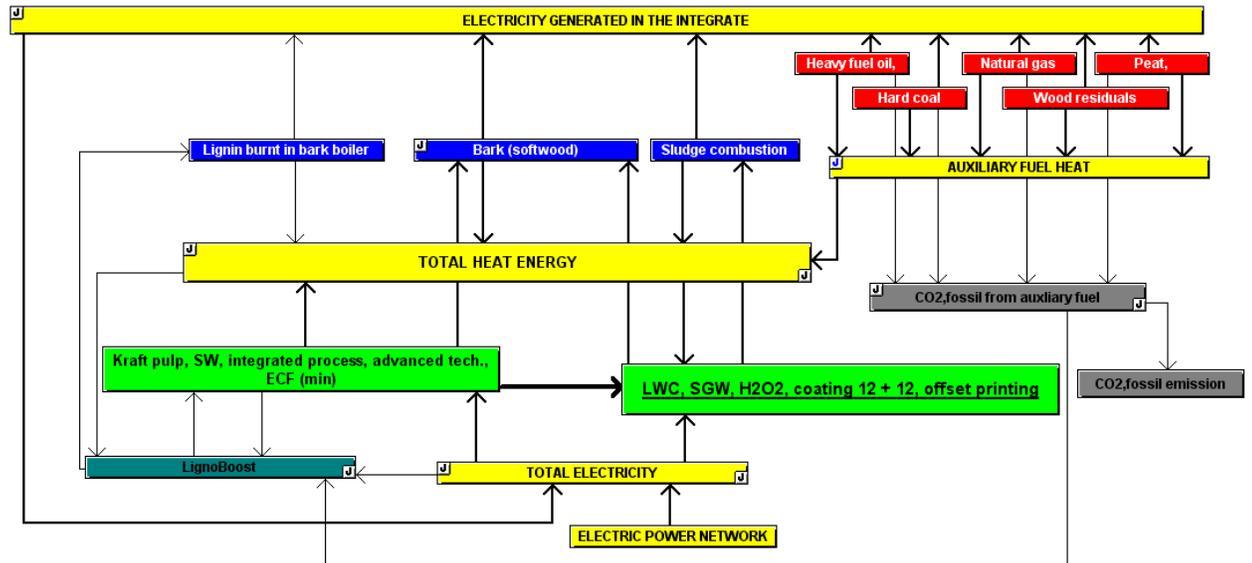


**Figure 16.** CO<sub>2</sub> equivalent emissions in cradle-to-customer approach by emission sources in case 1a. Case 1a is a typical Finnish pulp and paper mill which annual paper production capacity is 700,000 tons. The CO<sub>2</sub>-equivalent emissions are calculated per ton of paper.

All the chemicals are necessary for the processes. It is therefore difficult to reduce the emissions from chemical production. The transportation share is insignificant when compared with the total emissions. The biggest potential from alternatives to reduce CO<sub>2</sub>-equivalent emissions is therefore electricity production and integrated pulp and paper mill processes. CO<sub>2</sub>-equivalent emissions can also be affected by the choice of auxiliary fuels. Their usage, however, often depends on the location of the mill, fuel prices and fuel availability. The emissions from electricity production can be reduced by favouring electricity produced from renewable energy sources. The next case focuses on the emission reduction alternatives that are internal in the integrated pulp and paper mill.

## 5.2 Case 1b: Integrated LWC mill and LignoBoost

The LignoBoost case represents the situation in which the lignin removal unit is integrated into a pulp mill. The process data are based on information from Metso. Data are collected via e-mail (11 December 2009 and 18 January 2010) with Henrik Wallmo. Some assumptions and simplifications have been made in the calculations. Figure 17 shows the integrated pulp and paper mill with LignoBoost. Appendix III presents the flowsheet of the cradle-to-customer approach. Appendix VIII shows the calculation procedure used in these cases.



**Figure 17.** Case 1b: The mill-level approach of integrated LWC mill and LignoBoost. In case 1b the LignoBoost process is integrated with pulp and paper mill.

Black liquor from the evaporation process is led with 40% dry solid content to the LignoBoost process. The chemicals and the amounts used in the process are presented in Table 6.  $\text{CO}_2$  is assumed to be produced in-house from the lime kiln flue gases. It is assumed that if  $\text{CO}_2$  were purchased, it would be produced from fossil fuels. The amount of  $\text{CO}_2$  needed in the process can therefore be reduced from the fossil  $\text{CO}_2$  emissions to air, although the emissions from the lime kiln are biogenic  $\text{CO}_2$ . Approximately 0.3 ton of lignin per ton of produced bleached pulp can be withdrawn without disturbing the operation of the recovery

boiler. This amount is used in the calculation. As shown in Figure 7, the black liquor is taken from the evaporators and led to the LignoBoost unit. Lignin is separated and the filtrates are sent back to the evaporators. First filtration and, in particular, second filtration contain large amounts of water and influence the evaporation capacity. Roughly  $2 \text{ m}^3/\text{t}_{\text{lignin}}$  of “additional water” is generated during the LignoBoost process. A general 6-effect evaporator consumes approximately  $450 \text{ MJ}/\text{t}_{\text{H}_2\text{O}}$ . In a new modern green field mill, the energy consumption in the evaporation is lower (about  $390 \text{ MJ}/\text{t}_{\text{H}_2\text{O}}$ ). The extra energy needed to evaporate the “additional water” produced is thus  $900 \text{ MJ}/\text{t}_{\text{lignin}}$ . (Wallmo 2009.)

**Table 6.** Consumable used in the LignoBoost process (Wallmo 2009).

| <b>Consumable</b>              | <b>Consumption per ton of lignin</b> |
|--------------------------------|--------------------------------------|
| CO <sub>2</sub>                | 220 kg                               |
| H <sub>2</sub> SO <sub>4</sub> | 190 kg                               |
| NaOH                           | 65-130 kg                            |
| Electricity                    | 50 kWh                               |
| EVAP, steam                    | 900 MJ                               |

Separated lignin is used as a fuel to replace fossil fuels in a lime kiln. The results are presented per ton of produced pulp, corresponding to 300 kg of lignin. Before lignin is fed into the lime kiln, it has to be dried. It is assumed that the separated lignin has a 70% dry solid content after the LignoBoost process. According to Tomani (2009, 462), the theoretical demand for energy to obtain completely dry lignin (4% dry solid content) is  $0.3 \text{ kWh}/\text{kg}$  of dry solid lignin when the dry solid content is 70%. Thus,  $1080 \text{ MJ}/\text{t}_{\text{lignin}}$  of heat is needed to dry lignin, i.e.,  $324 \text{ MJ}/\text{t}_{\text{pulp}}$ . Dried lignin is burnt in a lime kiln as a substitute for heavy fuel oil. The reference mill needs 54 kg of heavy fuel oil per ton of pulp. The energy content of the replaced heavy fuel oil is 2214 MJ. The energy content of separated lignin is 7260 MJ. When 2214 MJ lignin is burnt in a lime kiln, 5046 MJ lignin can thus be burnt in a bark boiler to produce energy. Energy from the bark boiler is calculated with a total efficiency of 0.86. Thus, the energy produced from lignin in the bark boiler is  $4340 \text{ MJ}/\text{t}_{\text{pulp}}$ . This energy is separated into electric and heat power with a power-to-heat ratio of 0.22. The increasing

electric production in the bark boiler is  $0.217 \text{ MWh/t}_{\text{pulp}}$  and the heat production  $3.557 \text{ GJ/t}_{\text{pulp}}$ . Table 7 collects the energy flows in the LignoBoost case. The values are presented per ton of pulp.

**Table 7.** Case 1b: Energy flows consideration per ton of pulp. In case 1b the LignoBoost process is integrated with pulp and paper mill.

| Energy flow  | Values per ton of pulp |
|--|------------------------|
| Heat for lignin drying [GJ]  | 0.324                  |
| Energy content of lignin burnt in a lime kiln [GJ]                   | 2.214                  |
| Lignin burnt in a bark boiler [GJ]                                   | 5.046                  |
| Electricity production in a recovery boiler in case 1a/case 1b [MWh] | 0.17/ - (needs 0.08)   |
| Heat production in a recovery boiler in case 1a/case 1b [GJ]         | 4.8/0.69               |
| Electricity produced in a bark boiler by burning lignin [MWh]        | 0.217                  |
| Heat produced in a bark boiler by burning lignin[GJ]                 | 3.557                  |

When lignin is separated from black liquor, the heating value of the black liquor decreases. It is assumed that the lignin separated from the black liquor has the same energy content as the decreased heating value of the black liquor. This means that the total energy from the pulp mill is reduced in the module of kraft pulp presented in Figure 17. The kraft pulp module includes both a recovery boiler and a lime kiln. When the part of lignin is burnt in the lime kiln after the LignoBoost process, the total energy reduction in the recovery boiler (and in this case also the kraft pulp module) is the energy content of the lignin that is burnt in the bark boiler. The lignin separation thus reduces the electricity production in the recovery boiler by  $0.251 \text{ MWh/t}_{\text{pulp}}$  and heat production by  $4.11 \text{ GJ/t}_{\text{pulp}}$ . The new heat production in the kraft pulp process is therefore  $0.69 \text{ GJ/t}_{\text{pulp}}$  (without the LignoBoost process it is  $4.8 \text{ GJ/t}_{\text{pulp}}$ ). The new electricity production is  $-0.08 \text{ MWh/t}_{\text{pulp}}$  (without the LignoBoost process it is  $0.17 \text{ MWh/t}_{\text{pulp}}$ ), which means that in addition to the paper mill, the pulp mill also needs external electricity.

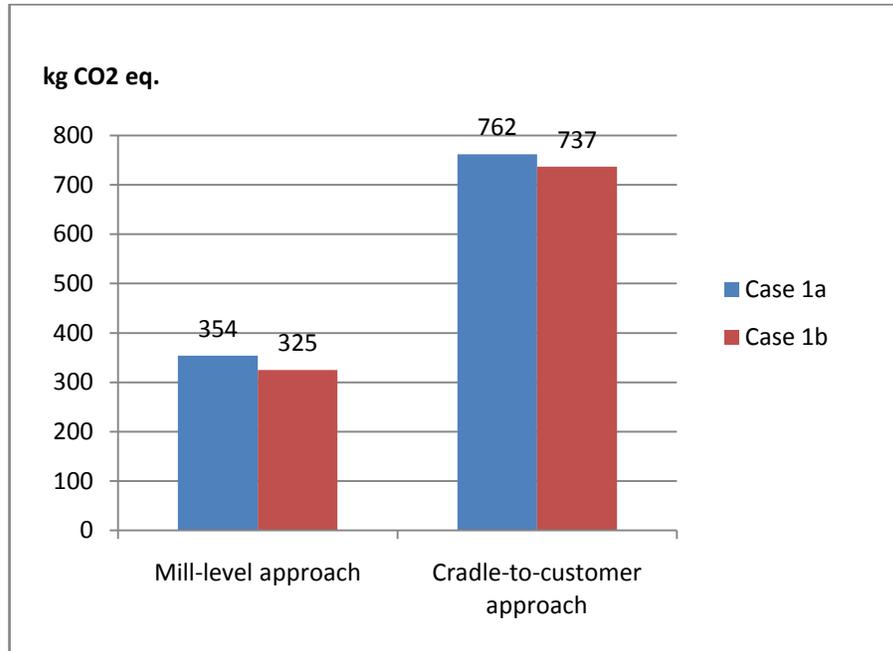
As the bark boiler is more efficient than the recovery boiler, more energy can be produced with lignin in case 1b commensurate with case 1a, but because the LignoBoost process also consumes energy, and the total energy production in the kraft pulp process is reduced due to the replacement of heavy fuel oil, more energy is needed outside the kraft pulp process in case 1b than in case 1a. The paper mill needs  $6 \text{ GJ}/t_{\text{paper}}$  of heat when heat production in the kraft pulp mill is reduced and heat is used in the LignoBoost process. The missing heat is therefore covered by auxiliary fuels. More electricity is therefore produced from auxiliary fuels. This means that the electricity demand from the electric power network decreases even though the total electricity demand in the integrated pulp and paper mill increases. The energy flows from energy sources per ton of paper in case 1a and case 1b are compared in Table 8. The plus sign before a value means that energy is produced and the minus sign means that energy is consumed. Appendix IV presents the flow values per ton of paper with a mill-level approach.

**Table 8.** Energy flows per ton of paper in case 1a and case 1b. The plus sign before a value means that energy is produced and the minus sign means that energy is consumed.

| Energy source            | Case 1a   |                   | Case 1b   |                   |
|--------------------------|-----------|-------------------|-----------|-------------------|
|                          | Heat [GJ] | Electricity [MWh] | Heat [GJ] | Electricity [MWh] |
| <b>Pulp mill</b>         | +1.152    | +0.041            | +0.167    | -                 |
| <b>Paper mill</b>        | -6.000    | -1.500            | -6.000    | -1.500            |
| <b>Auxiliary fuels</b>   | +3.109    | +0.316            | +3.384    | +0.342            |
| <b>Bark boiler</b>       | +1.683    | +0.089            | +2.537    | +0.141            |
| <b>Sludge combustion</b> | +0.056    | +0.003            | +0.056    | +0.003            |
| <b>LignoBoost</b>        | -         | -                 | -0.143    | -0.004            |
| <b>Electric network</b>  | -         | +1.052            | -         | +1.036            |

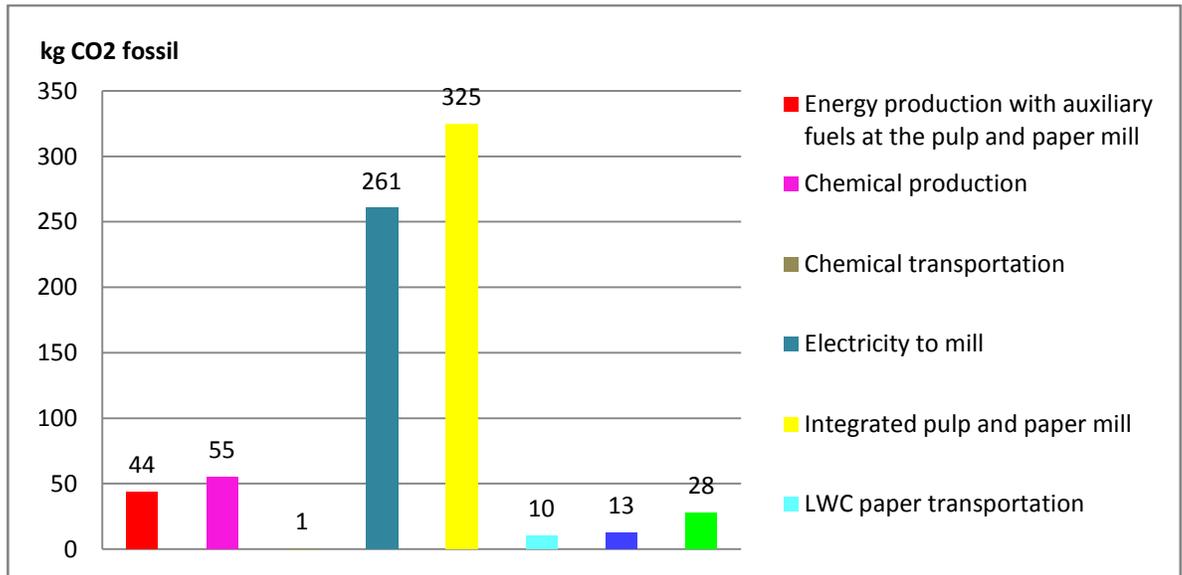
The CO<sub>2</sub>-equivalent emissions in cases 1a and 1b are very similar. When only mill-level emissions are taken into account, the carbon footprint in case 1b is approximately 325 kg. The greenhouse gases are thus reduced by 8.5% (30 kg) compared with the mill-level approach in case 1a. In the cradle-to-customer approach in case 1b, the carbon footprint only

decreases by 3.3% (25 kg) compared with case 1a, which is approximately 735 kg. Figure 18 shows the carbon footprints in cases 1a and 1b.

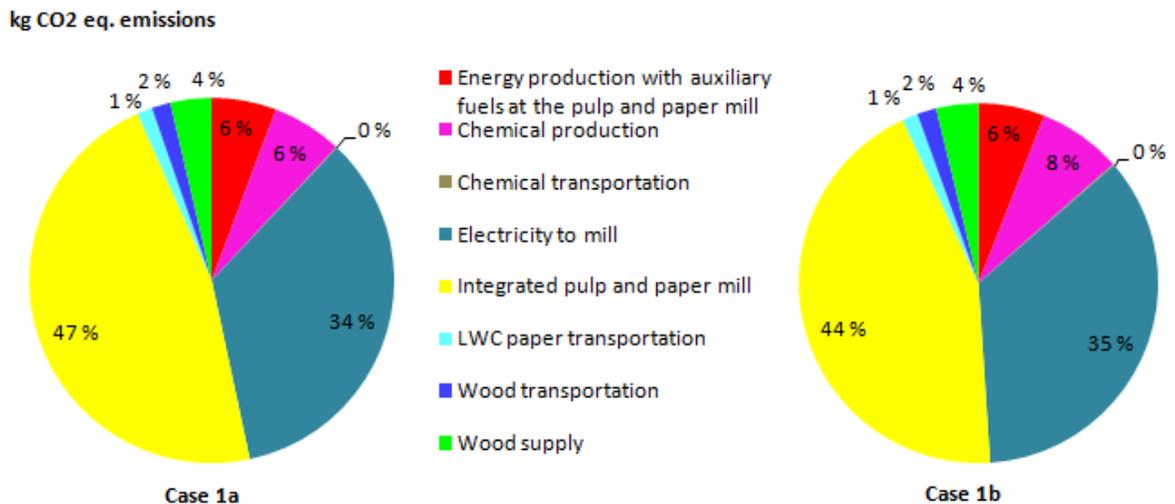


**Figure 18.** Carbon footprints in cases 1a and 1b. The CO<sub>2</sub>-equivalent emissions are reported per ton of paper.

The reduction is mainly due to heavy fuel oil replacement in the lime kiln. The reduction in electricity use from the power grid reduces emissions. The use of auxiliary fuels at the mill increases, however, and the emissions therefore also increase. The use of H<sub>2</sub>SO<sub>4</sub> and NaOH in the LignoBoost process increases emissions from the chemical production but because flue gases from the lime kiln are used as a CO<sub>2</sub> source, the total CO<sub>2</sub> emissions can be reduced a little in this way. Based on these facts, the GHG emissions also decrease in the cradle-to-customer approach by about 30 kg. Figure 19 presents the CO<sub>2</sub>-equivalent emissions in a cradle-to-customer approach. The GHG emission comparison between cases 1a and 1b is presented in Figure 20.



**Figure 19.** CO<sub>2</sub>-equivalent emissions per ton of paper in case 1b in the cradle-to-customer approach.



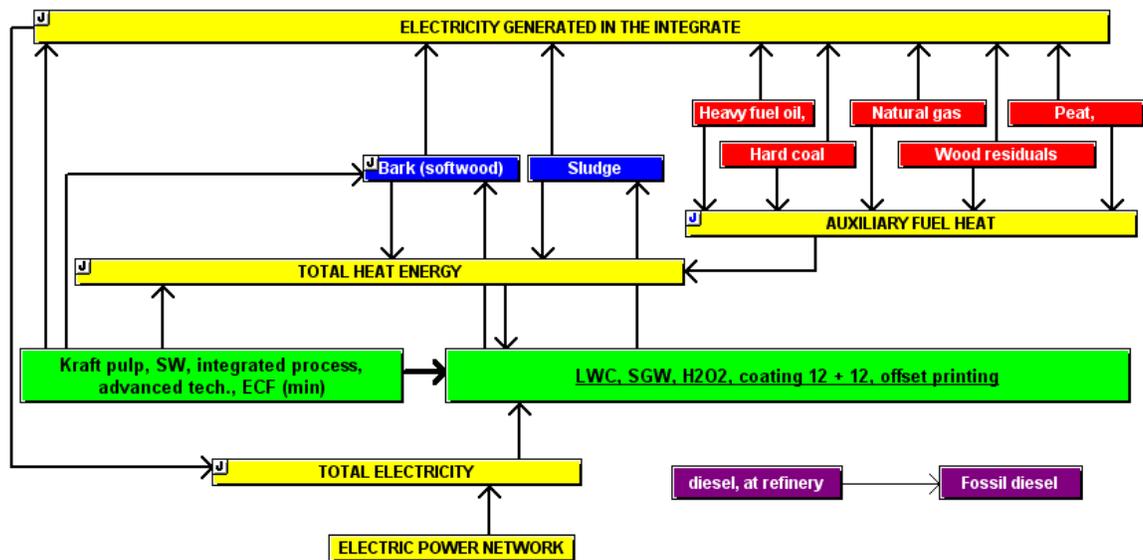
**Figure 20.** The percentage shares of CO<sub>2</sub>-equivalent emissions per ton of paper in cases 1a and 1b in different life cycle phases.

As Figures 16, 18 and 19 show, the CO<sub>2</sub>-equivalent emission reduction is not remarkable when the effect of the LignoBoost is examined. The electricity and chemical production phases even increase the CO<sub>2</sub>-equivalent emissions. Thus as a conclusion, it can be said that the CO<sub>2</sub> reduction cannot be the only reason to integrate the LignoBoost process into pulp

production. However, as mentioned in Section 3.2, the recovery boiler is often a bottleneck in the kraft pulp mill, and with lignin removal the flow of organics to the recovery boiler decreases. The lime kiln is the only part of the pulp mill that uses fossil fuels (if supporting fuels are not taken into account). If lime kiln fuels can also be replaced with bioenergy, it is possible to achieve a fossil-fuel-free pulp mill. In this case, when the paper mill is also taken into account, the CO<sub>2</sub> reduction is not significant. As an example, a paper mill that produces 700,000 t/a can reduce its CO<sub>2</sub> emissions by 17,500 t/a, which corresponds to the emissions of about 23 tons of produced paper with reference technology.

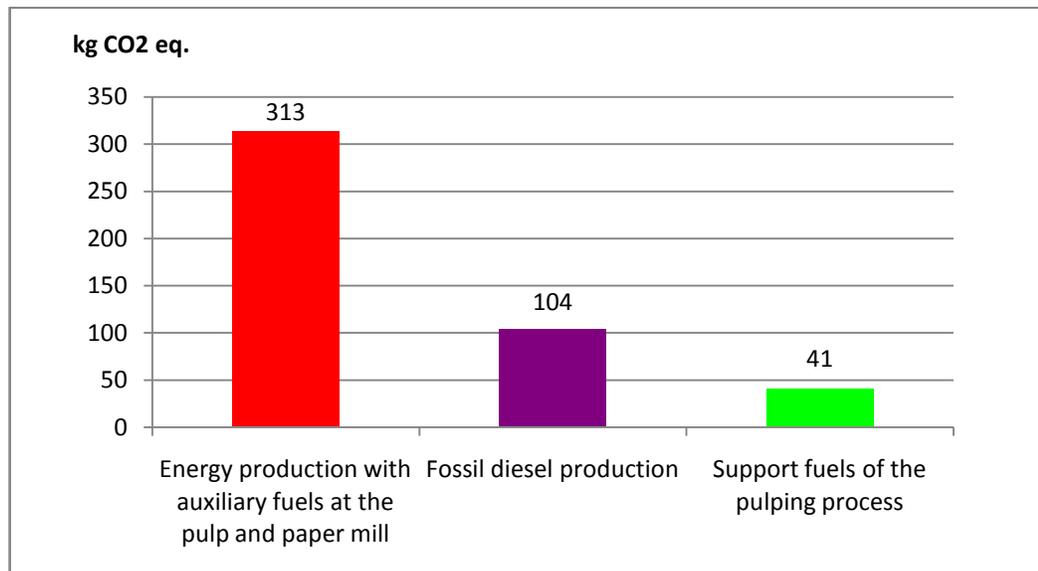
### **5.3 Case 2a: Integrated LWC mill and fossil diesel**

Fossil diesel production and Fischer-Tropsch diesel production are examined in cases 2a, 2b and 2c. Case 2a is a reference case for cases 2b and 2c. It is similar to case 1a, but the fossil diesel production is added alongside the integrated pulp and paper mill. Therefore, the total carbon footprint calculated is not the carbon footprint of integrated pulp and paper mill only. It includes the CO<sub>2</sub>-equivalent emissions both from the integrated pulp and paper mill and from the fossil diesel production even though the production sites of paper and fossil diesel would be unconnected in reality. LCI data of fossil diesel production is from the Ecoinvent database. In this reference case, fossil diesel production does not benefit the integration of pulp and paper production. The agglomerate of case 2a combines two different production lines into one module. The final products in this case are 1000 kg LWC paper and 214 kg fossil diesel. The amount of fossil diesel is defined based on the amount of FT diesel produced. The amount of FT diesel is calculated in Section 5.4. This allows the cases to be compared. Figure 21 shows the system boundaries on production sites level of paper and fossil diesel examination.



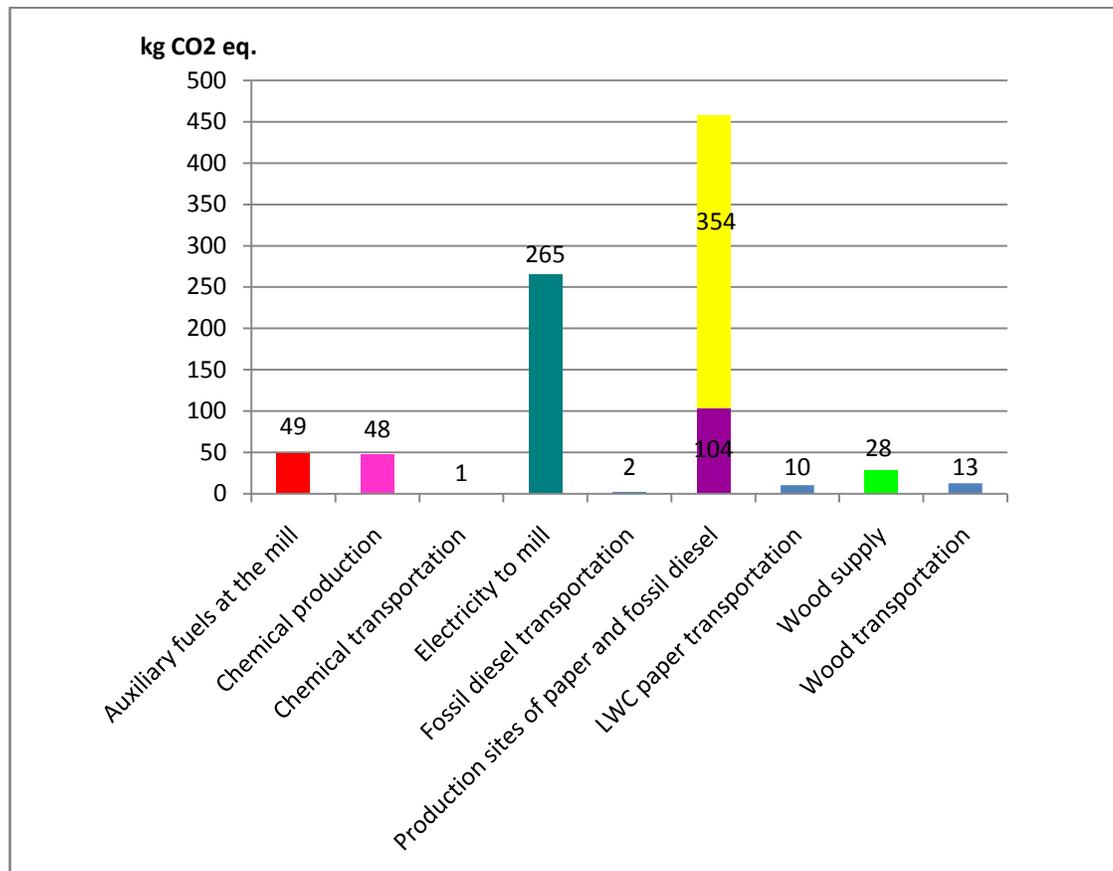
**Figure 21.** The unit processes on production sites level of paper and fossil diesel -examination in case 2a. There are two different production lines where the final products are 1000 kg of LWC paper and 214 kg of fossil diesel. The amount of fossil diesel is defined based on the amount of FT diesel produced which is calculated in Section 5.4.

When examining the production sites of paper and fossil diesel, the CO<sub>2</sub>-equivalent emissions caused by integrated pulp and paper mill are same as in case 1a but when also the CO<sub>2</sub>-equivalent emissions from fossil diesel production are taken into account, the total CO<sub>2</sub>-equivalent emissions in the mill-level approach are approximately 455 kg. The share of fossil diesel production is approximately 105 kg of these emissions, which is about 23% of the total CO<sub>2</sub>-equivalent emissions. Figure 22 shows the sources of CO<sub>2</sub> equivalent emissions and their amounts in case 2a in the production sites of paper and fossil diesel -examination.



**Figure 22.** CO<sub>2</sub> equivalent emissions per ton of paper and 214 kg fossil diesel in case 2a in the production sites of paper and fossil diesel -examination. The emissions from the production of paper are approximately 355 kg and the emissions from the fossil diesel production approximately 105 kg. The emissions from the pulp and paper mill are caused by fossil fuel burning in a lime kiln. Auxiliary fuel combustion produces heat and electricity to the pulp and paper mill.

In the cradle-to-customer approach in case 2a, the CO<sub>2</sub> equivalent emissions are about 875 kg. The emissions from fossil diesel production are included in the total CO<sub>2</sub> equivalent emissions. Figure 23 shows the CO<sub>2</sub>-equivalent emissions by sources. The integrated pulp and paper mill and fossil diesel production cause over half (52%) of the total CO<sub>2</sub>-equivalent emissions. The share of electricity production is about 30%. The share of transportation is insignificant (26 kg or 3%) compared with other emissions. The emissions from the chemical, fossil diesel, LWC paper and wood transportation are therefore examined in one number in cases 2b and 2c. The flowsheet of the cradle-to-customer approach is presented in Appendix V.

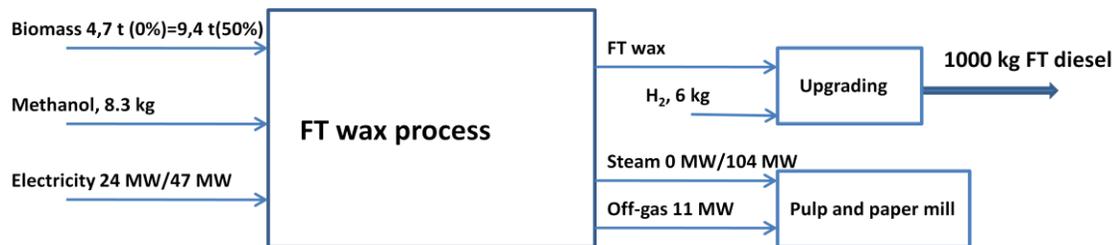


**Figure 23.** CO<sub>2</sub>-equivalent emissions per ton of paper and 214kg fossil diesel in case 2a in the production sites of paper and fossil diesel -examination by emissions sources. In case 2a paper and fossil diesel are produced separately. The share of emissions from fossil diesel production is 104 kg and from pulp and paper production 354 kg. LCI data include all emissions from fossil diesel life cycle; therefore all the CO<sub>2</sub>-equivalent emissions related to fossil diesel production are included in the column of the production sites of paper and fossil diesel.

#### 5.4 Cases 2b and 2c: Integrated LWC mill and Fischer-Tropsch diesel

These cases examine the effect of Fischer-Tropsch diesel production on the CO<sub>2</sub>-equivalent emissions when the separately produced paper and fossil diesel is compared with the situation when the FT diesel production is integrated with pulp and paper mill. Fossil diesel production, case 2a, is used as a reference case. Data for these FT diesel cases were obtained via personal communication with Steven Gust of Neste Oil. Two different FT diesel production cases are examined and presented in Figure 24. Figure 24 represents a situation in which 1000 kg of FT diesel are produced and inputs are defined for that amount. In the

FT diesel process, the FT primary product is an intermediate product of the FT synthesis and must be upgraded to satisfy the requirements of a traffic fuel specification. It may be upgraded to an FT diesel component. The cases differ from each other in the relative amounts of electricity and steam that are produced. In case 2b, electricity is produced in a condensing power plant. The process does not produce any steam and the purchased electricity is 24 MW. In case 2c, the electricity is produced in a back-pressure steam turbine power plant. The process produces approximately 104 MW of low-pressure steam, which is used as a heating source in a pulp and paper mill. The heat produced in a bark boiler can be replaced with this heat and bark can thus be used as raw material in the FT process. In both cases, 11 MW of off-gas is generated during the FT process. This off-gas can be burnt in a lime kiln to replace heavy fuel oil. Methanol is used as a solvent in the FT process to remove CO<sub>2</sub> from syngas. The amount of methanol is 8.3 kg/t<sub>FT diesel</sub>. Hydrogen is used in the upgrading process to produce FT diesel. The amount of H<sub>2</sub> is 6 kg/t<sub>FT diesel</sub>. The mill-level and the cradle-to-customer approach are both examined. The benefit from the carbon footprint reduction cannot be transferred directly to the pulp and paper mill because two different products, paper and FT diesel, are produced in integration. Therefore the emission allocation for paper and FT diesel is not examined because the share of CO<sub>2</sub>-equivalent emissions which are caused only by pulp and paper mill and which are from FT diesel production is very complex. Appendix IX shows the calculation procedure used in these cases.



**Figure 24.** FT diesel production cases. In Case 2b the purchased energy is 24 MW and the process does not produce any steam. In Case 2c the purchased energy is 47 MW and the steam production is 104 MW. In both Cases 11 MW of off-gas is produced.

First, the size of the FT diesel production unit is examined. Biomass is used as a raw material. Its availability defines the plant size. The European Forest Institute (EFI) has calculated the amount of forest chips that is harvestable during pulp wood harvesting. The harvest potential for pine wood is from thinning operations 5 % and from final felling 21 % forest chips per ton of pulp wood. For spruce wood, the corresponding numbers are 8% and 29%. This means that when 448.8 kg pine wood and 480 kg spruce wood are used per ton of paper (in this thesis integrated pulp and the paper mill are used as an example), approximately 187 kg of forest chips can be harvested which means that only about 20 kg FT diesel can be produced. It can therefore be concluded that it is not profitable to define the amount of FT diesel production from the amount of forest chips harvested during the pulpwood. When defining the plant size of the FT diesel production, the potential of forest chips from the larger region has to be taken into account. According to Timo Heikka (18.2.2010), the gasification process is a very flexible process from the viewpoint of raw material. Based on this fact, it can be assumed that the FT diesel process has plenty of raw material available when forest chips as well as bark and saw dust are taken into account. The FT diesel process can be sized for an amount of raw material of 1.0-2.4 solid cubic metres per year on these grounds. (Heikka 18.2.2010.)

The minimum and maximum amounts of FT diesel production are calculated. The density of pinewood is  $411 \text{ kg/m}^3$  and spruce wood  $390 \text{ kg/m}^3$ . When the average density and biomass requirements of FT diesel production are taken into account, the minimum production capacity of FT diesel is approximately 85,000 tons per year and the maximum capacity approximately 205,000 tons per year. The FT diesel production capacity is chosen to be 150,000 tons per year. In order to size the FT diesel production into the capacity of 1000 kg LWC paper production, the annual capacity of the paper mill has to be defined. The capacity of 700,000 tons of LWC paper per year is chosen and thus FT diesel production is approximately 214 kg per ton of LWC paper. If the annual operation time is 8,300 hours, the production of FT diesel is 18 tons per hour. The electricity consumption of the FT diesel process can now be calculated. In case 2b, the electricity consumption is  $1.328 \text{ MWh/t}_{\text{FT diesel}}$  and in case 2c, the consumption is  $2.601 \text{ MWh/t}_{\text{FT diesel}}$ . Table 9 presents the electricity

consumption, low-pressure steam production and off-gas production per ton of produced paper.

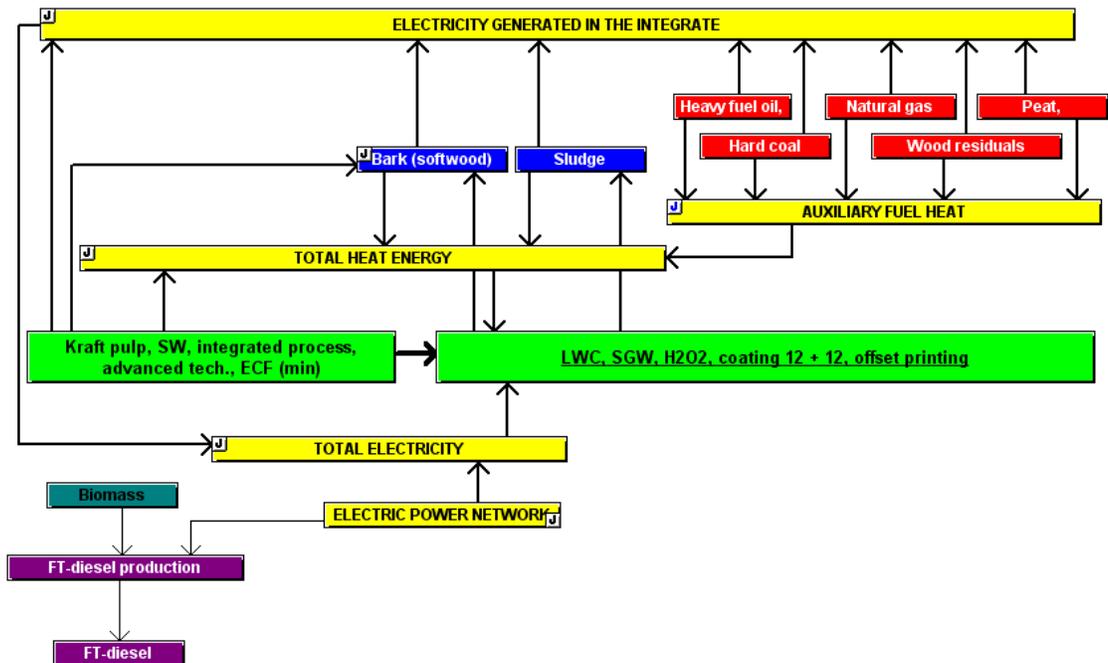
**Table 9.** Comparing the energy flows of cases 2b and 2c. The numbers are calculated per ton of produced paper.

|  | <b>Case 2b</b> | <b>Case 2c</b> |
|--|----------------|----------------|
| <b>Electricity consumption [MWh/t<sub>paper</sub>]</b> | 0.284          | 0.557          |
| <b>Steam production [GJ/t<sub>paper</sub>]</b>         | 0              | 4.43           |
| <b>Off-gas production [MJ/t<sub>paper</sub>]</b>       | 470            | 470            |

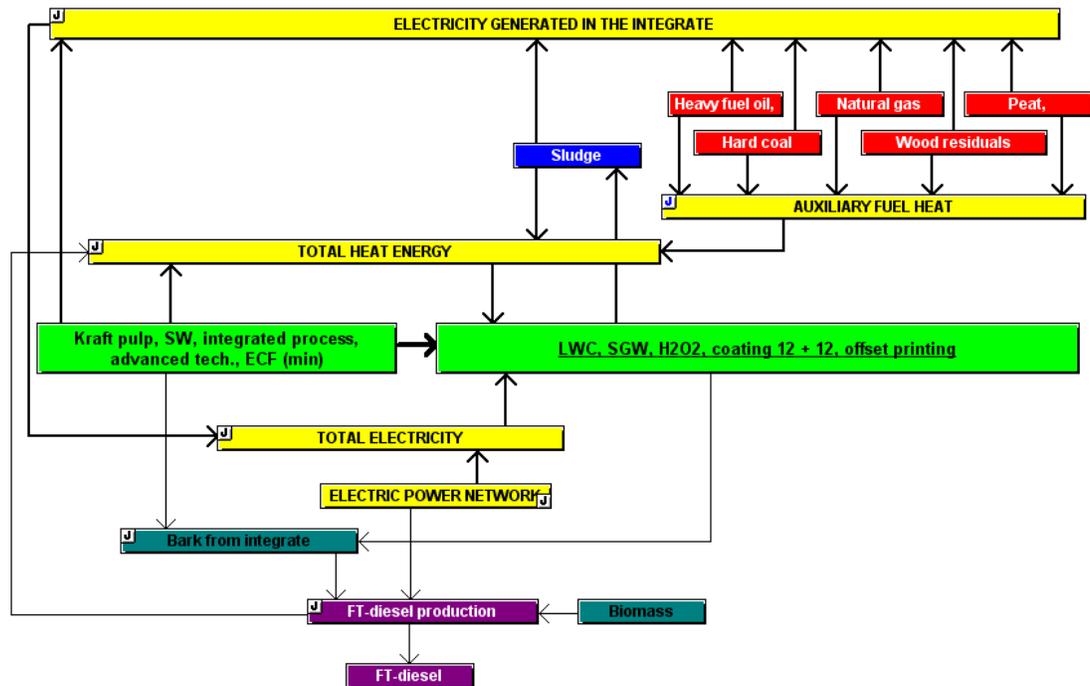
In both cases, off-gas can be burnt in a lime kiln to replace heavy fuel oil. The amount of heavy fuel oil used per ton of paper is 12.96 kg. If the lower heating value of heavy fuel oil is 41 MJ/kg, the energy content of 12.96 kg of heavy fuel oil is approximately 531 MJ. This means that although off-gas is burnt in a lime kiln, 62 MJ/t<sub>paper</sub> (1.5 kg/t<sub>paper</sub>) of heavy fuel oil are still needed. This is about 6 kg/t<sub>pulp</sub> of heavy fuel oil. In case 2c, the FT diesel production process produces enough steam for the bark boiler to be removed, and the bark can be used as a raw material in the FT diesel process. Without the FT diesel process, the amount of bark burnt in a bark boiler is 120 kg/t<sub>paper</sub>. The electricity production in a bark boiler is 0.089 MWh/t<sub>paper</sub> and the heat production is 1.683 GJ/t<sub>paper</sub>. The total energy produced with bark is about 2 GJ/t<sub>paper</sub>. When this is compared with the steam production in the FT diesel process, it can be concluded that the bark boiler can be removed. Figure 25 and Figure 26 present a flowsheet, and the differences of Cases 2b and 2c. The flowsheets with flow values are presented in Appendices VI and VII. Table 10 shows the energy flows with the most significant changes when comparing cases 2a, 2b and 2c.

**Table 10.** Energy flow comparison between the cases 2a, 2b and 2c. In case 2a the energy flows do not transfer between the integrated pulp and paper mill and fossil diesel production site.

| Energy flow per ton of paper                     | Case 2a | Case 2b | Case 2c |
|--|---------|---------|---------|
| Electricity generated in the integrate [MWh]     | 0.448   | 0.448   | 0.080   |
| Auxiliary fuel electricity [MWh]                 | 0.316   | 0.316   | 0.035   |
| Auxiliary fuel heat [GJ]                         | 3.109   | 3.109   | 0.358   |
| Heat from FT diesel production [GJ]              | -       | -       | 4.434   |
| Electricity requirement from power network [MWh] | 1.052   | 1.336   | 1.977   |

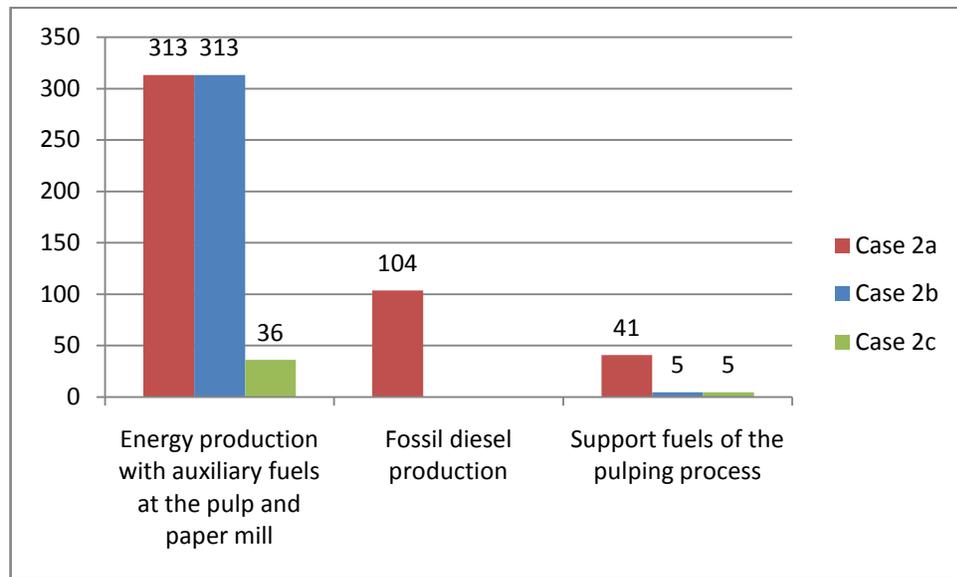


**Figure 25.** System boundaries of production site of integrated paper and FT diesel production in case 2b. In this case the off-gas can be used as fuel in a lime kiln to replace fossil fuels but the FT diesel process does not produce any steam to pulp and paper mill.



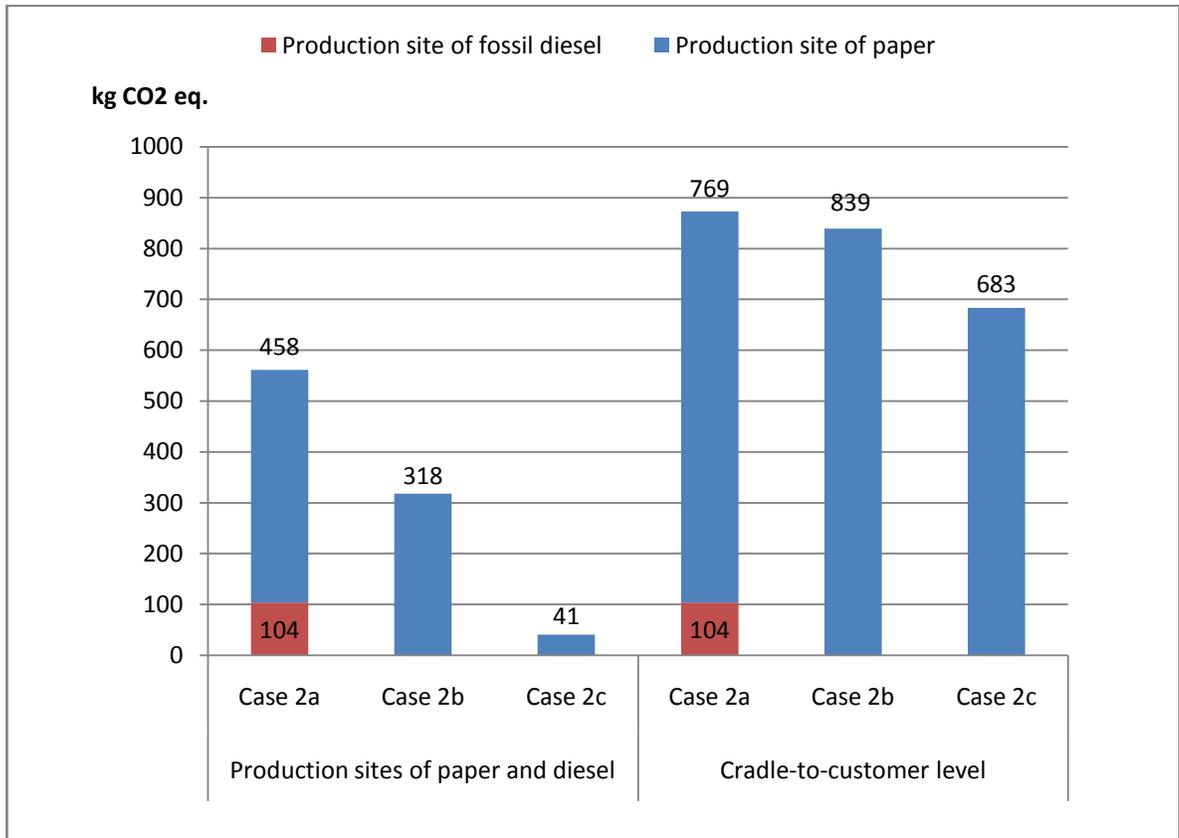
**Figure 26.** System boundaries of production site of paper and FT diesel production in case 2c. The FT diesel process produces excess heat which can be used in a pulp mill and the bark boiler can be replaced. Also the off-gas from process can be utilized in a lime kiln to replace fossil fuels.

The CO<sub>2</sub>-equivalent emissions of cases 2b and 2c differ significantly from each other in the production site level of paper and FT diesel -examination. The carbon footprint in case 2b is about 320 kg and in case 2c about 40 kg. This is because in case 2b, only the heavy fuel oil in the lime kiln can be replaced while in case 2c, the amount of auxiliary fuels can also be reduced. When the carbon footprints are compared with the carbon footprint of case 2a, the CO<sub>2</sub>-equivalent emissions in case 2b can be reduced by about 30% and in case 2c by about 90%. Figure 27 shows the production sites of paper and fossil diesel or FT diesel - examination comparison of CO<sub>2</sub>-equivalent emissions in cases 2a, 2b and 2c.



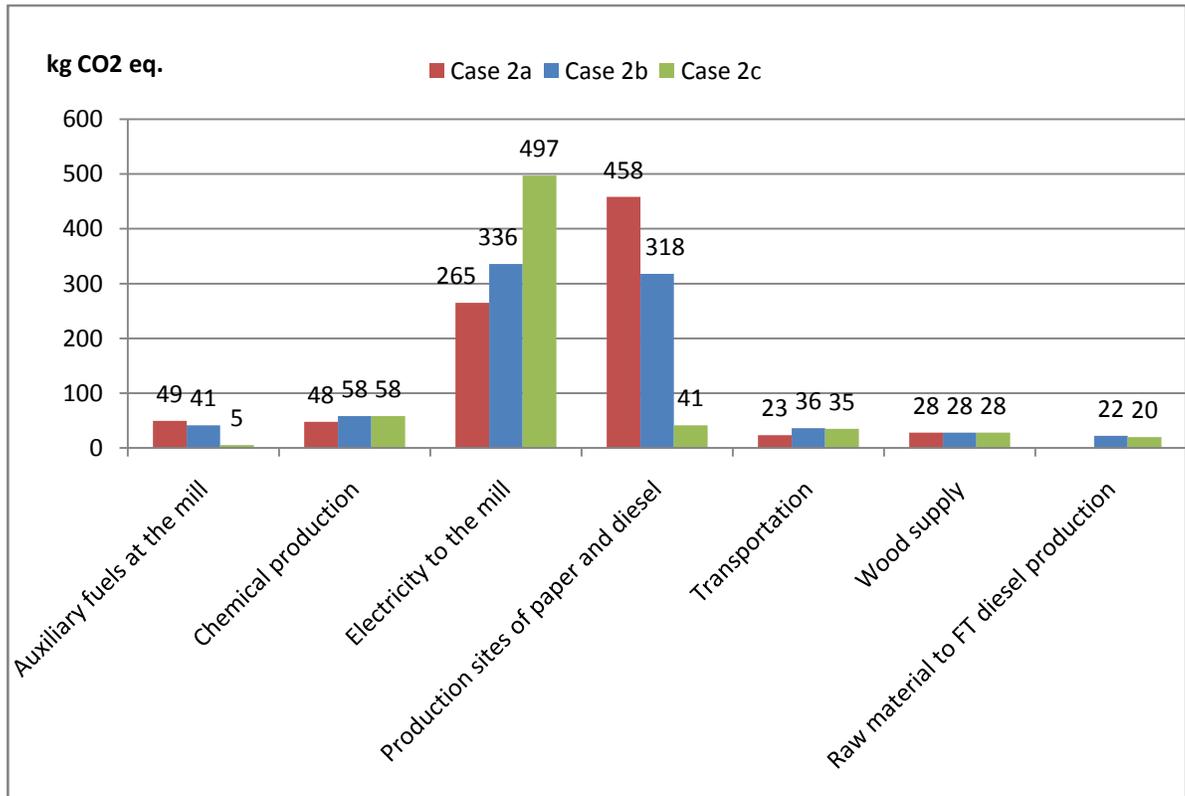
**Figure 27.** Comparing CO<sub>2</sub>-equivalent emissions in cases 2a, 2b and 2c. CO<sub>2</sub>-equivalent emissions in case 2a are from the unconnected production sites of paper and fossil diesel and in cases 2b and 2c from the integrated production site of paper and FT diesel. In cases 2b and 2c the emissions from the pulp and paper mill reduce because the heat from FT diesel production can be utilized. CO<sub>2</sub>-equivalent emissions are presented per ton of paper and 214kg diesel.

When comparing the CO<sub>2</sub>-equivalent emissions of the cradle-to-customer approaches in cases 2b and 2c, it is noticeable that the relative difference is not as significant as in the mill-level approaches. In case 2b, the CO<sub>2</sub>-equivalent emissions are about 840 kg and in case 2c about 685 kg. The reduction in CO<sub>2</sub>-equivalent emissions in case 2b compared with case 2a is therefore about 3.5% and in case 2c about 21%. It has to be taken into account that these emissions include both the emissions related to pulp and paper mill processes and also related to fossil diesel or FT diesel production. It can be concluded that when the fossil diesel production is compared with the FT diesel process which is integrated in the pulp and paper mill integration, the carbon footprint of pulp and paper mill decreases but the calculated reduction is due to both pulp and paper mill processes and FT diesel process. Figure 28 presents the CO<sub>2</sub>-equivalent emission comparison in the production sites level and the cradle-to-customer level of paper and fossil diesel or FT diesel –examinations.



**Figure 28.** Comparison of the CO<sub>2</sub>-equivalent emissions in the production sites level and the cradle-to-customer level of paper and fossil diesel or FT diesel -examinations in case 2a, 2b and 2c. The share of the emissions from fossil diesel production is separated in the columns of case 2a. The emissions in cases 2b and 2c cannot be allocated into emissions from FT diesel and paper production and that is why they are presented in congruent columns. The CO<sub>2</sub>-equivalent emissions are presented per ton of paper and 214 kg diesel.

Figure 29 shows the cradle-to-customer comparison between cases 2a, 2b and 2c divided into emission sources. Although case 2c has the lowest total CO<sub>2</sub>-equivalent emissions, it has to be taken into account that in this case the purchased electricity usage is almost one and a half times higher than in case 2a and about a third higher than in case 2b. Electricity to the mill causes about 73% of the total CO<sub>2</sub>-equivalent emissions in case 2c. When comparing the production sites -level and cradle-to-customer level of paper and FT diesel in case 2c, it can therefore be said that although the reduction in CO<sub>2</sub>-equivalent emissions of the production site of paper and FT diesel -examination was significant, the effects are not as significant when examining the cradle-to-customer approach. The electricity production methods therefore have a remarkable effect on the total CO<sub>2</sub> equivalent emissions in the cradle-to-customer approach.



**Figure 29.** Cradle-to-customer comparison of the CO<sub>2</sub>-equivalent emissions from different stages of life cycle in cases 2a, 2b and 2c. In case 2a the 104 kg emissions from fossil diesel production are included in the column of the production sites of paper and diesel. The emissions in cases 2b and 2c cannot be allocated into emissions from FT diesel and paper production and that is why they are presented in congruent columns. The CO<sub>2</sub>-equivalent emissions are presented per ton of paper and 214kg diesel.

## 6 DISCUSSION

As the case studies have shown, the GHG reduction can be achieved with both LignoBoost and FT diesel production. When the lignin is removed, the heat load on the recovery boiler also decreases and more pulp can be produced. It was also mentioned that modern pulp mills have an energy surplus and if the lignin is separated this energy surplus can be exported to other users in the form of biofuel. In this thesis, the pulp mill is integrated into the paper mill. The extra energy is therefore used directly in the paper mill processes. The heat load of the recovery boiler causes a reduction in electricity production because the energy

content of black liquor decreases. That is why extra electricity has to be purchased from the electricity power network. In this thesis, the LignoBoost process was examined based on the carbon footprint. As the bark boiler is more efficient than the recovery boiler, the alternative is to examine it from the energy perspective. From an energy perspective, the fuel in the lime kiln is not replaced with lignin but all the lignin is burnt in a bark boiler. The other interesting modification when examining the energy surplus in this case is to examine only the pulp mill without the paper mill integration. The effect of auxiliary fuel burning is then excluded and it is possible to find the energy production advantages of lignin removal when the lignin is burnt in a bark boiler instead of a recovery boiler.

The advantages of integrating FT diesel production into the pulp and paper mill are cost reduction and the utilization of by-product energy from FT diesel production in the pulp and paper mill processes. These advantages are realized in case 2c. The FT diesel process produces enough heat for the bark boiler to be removed. The power plant with auxiliary fuels produces the small amount of heat still needed in the paper mill. As the heat demand of the integrate determines the amount of auxiliary fuels used, the electricity production of the integrate decreases when the bark boiler is removed and the amount of auxiliary fuels is reduced. The use of electricity purchased from the power network increases and thus by favouring the electricity produced from renewable energy sources has a high effect on GHG emissions. The integration of FT diesel production into the pulp and paper mill also enables the integration of purchasing and handling of raw material, decreasing the costs of storage. The waste heat can also be utilized in a raw material drying effectively.

In this thesis, the objective was to examine the potential of the integrated pulp and paper mill to reduce the carbon footprint. In the FT diesel case, it is a question of which one benefits from the reduction in the carbon footprint: the paper producer or the FT diesel producer. That is why the allocation of CO<sub>2</sub>-equivalent emissions to paper and FT diesel production is very difficult. Both benefit from integration, but two very different products are produced. This question can be considered from, for example, the viewpoint that the FT diesel production plant is probably constructed in connection with an existing pulp and paper mill, when it can be assumed that it is constructed in order to benefit primarily the pulp and pa-

per mill. Naturally, it also benefits the FT diesel plant, as discussed in the previous chapter, but if the FT diesel plant is examined as a separate unit, it is noticeable that in this thesis the only significant benefit for the FT diesel plant of the pulp and paper integration is the bark, which is burnt in a bark boiler without integration. In this situation, the bark from the pulp and paper mill constitutes about 12% of the total raw materials used in the FT process. This decreases the energy consumption of raw material manufacturing for the FT diesel process. The energy consumption used for biomass drying also decreases but is not taken into account in this examination.

This thesis only takes into account the production of FT diesel in the cradle-to-customer approach. Possible land use changes are not considered. It is also possible to examine the use of FT diesel, but then factors such as the rules for calculating the GHG impact of biofuels, bioliquid and their fossil fuel comparators set by the European Parliament and the Council have to be taken into account. In cases 1 and 2 the bio-based carbon is excluded from the examination. When the methods to calculate the forest carbon balance are developed further, it will be interesting to examine the cases from a perspective that also includes the bio-carbon and the forest carbon balance. The economic aspects were excluded from this thesis. These aspects are however interesting from the viewpoint of companies which consider these technologies. That is why the worth of value is the examination of the profitability of these technologies with the relation to the GHG reduction received.

## **7 CONCLUSIONS**

The effects of two different bioenergy production technologies on the carbon footprint of the integrated LWC mill were studied in the thesis. The first technology was the LignoBoost process. Case 1a was the reference case: a typical Finnish integrated LWC mill. In the LignoBoost process, the lignin is separated from black liquor in a precipitation process by lowering the pH with CO<sub>2</sub>. After precipitation, the lignin precipitate is redispersed and acidified in order to avoid plugging problems. After that, the new slurry is filtered and

washed using displacement washing. The use of lignin as a biofuel in a lime kiln to replace fossil fuel and its burning in a bark boiler was also studied in the thesis.

The other bioenergy production technology was Fischer-Tropsch diesel production. Case 2a was the reference case: a typical Finnish integrated LWC mill with fossil diesel production alongside the mill. Biomass such as, for example, forest chips, bark and sawdust, was used as a raw material in the FT diesel process. Two different case studies were examined. In case 2b, the process did not produce extra steam, but in case 2c extra steam was generated that could be used as a heat source in the LWC mill. In both cases, off-gas was generated that could be used in a lime kiln to replace most of the heavy fuel oil.

The calculation of the carbon footprints was made by KCL-ECO. The carbon footprints were calculated as CO<sub>2</sub>-equivalent emissions, taking into account fossil CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O. With both of the bioenergy production technologies, LignoBoost and FT diesel production, the carbon footprints could be reduced compared with their reference cases. Table 11 shows the carbon footprints and their reductions compared with the reference cases. It has to be taken into account that case 1 cannot be compared with case 2.

**Table 11.** Carbon footprints per functional unit and their reductions compared with reference cases. Case 1 cannot be compared with case 2 since their functional units are different, i.e. 1000kg of paper in case 1, and 1000kg paper and 214kg diesel in case 2. Mill-level carbon footprint takes into account the CO<sub>2</sub>-equivalent emissions which are directly from mill processes. In cradle-to-customer approach the carbon footprint takes into account CO<sub>2</sub>-equivalent emissions from chemical and auxiliary fuel production, wood supply and all transportation. Mill-level emissions are included in cradle-to-customer approach. In case 2a the carbon footprint include the emission both from unconnected fossil diesel and paper production and in cases 2b and 2c the emissions from integrated FT diesel and paper production.

|                | <b>Mill-level</b> | <b>Change compared with case 1a</b> | <b>Cradle-to-customer</b> | <b>Change compared with case 1a</b> |
|----------------|-------------------|-------------------------------------|---------------------------|-------------------------------------|
| <b>Case 1a</b> | 355 kg            | -                                   | 760 kg                    | -                                   |
| <b>Case 1b</b> | 325 kg            | -8.5%                               | 735 kg                    | -3.3%                               |
|                | <b>Mill-level</b> | <b>Change compared with case 2a</b> | <b>Cradle-to-customer</b> | <b>Change compared with case 2a</b> |
| <b>Case 2a</b> | 455 kg            | -                                   | 875 kg                    | -                                   |
| <b>Case 2b</b> | 320 kg            | -30%                                | 840 kg                    | -4%                                 |
| <b>Case 2c</b> | 40 kg             | -91%                                | 685 kg                    | -22%                                |

Although the CO<sub>2</sub>-equivalent emission reduction is not as significant in case 1b as in case 1a, it shows that when the heavy fuel oil can be replaced with lignin in a lime kiln, it is possible to achieve a fossil-fuel-free pulp mill. Other possibilities to reduce the carbon footprint further are the choices of auxiliary fuels used in missing heat production and the production methods of purchased electricity. In these calculations was used the share of auxiliary fuel mix defined by the Finnish Forest Industry Federation in 1997. This is the average share of auxiliary fuels used in paper mill heat production in 1997. The electricity production method was the Finnish five-year average electricity production mix. In cases 2b and 2c the carbon footprint takes into account the CO<sub>2</sub>-equivalent emissions both from the FT diesel production and from paper production, therefore the carbon footprint reduction compared with case 2a in cases 2b and 2c is not only the reduction in the carbon footprint of the integrated pulp and paper mill. But because it is difficult to allocate the emissions to paper and FT diesel, the carbon footprint is examined as one entirety. The reduction in CO<sub>2</sub>-

equivalent emissions in cases 2b and 2c compared with case 2a was more significant in the production sites level of paper and FT diesel -examination than in the cradle-to-customer approach. This was due to the increased use of purchased electricity from the power network compared with case 2a. In addition of integration of FT diesel production alongside with the pulp and paper mill in cases 2b and 2c, the carbon footprint can be affected further with the methods of electricity production.

The results of the thesis contain some uncertainty. The processes and modules from the KCL-ECO database can be assumed to be accurate. The age of the data from the KCL-ECO database also varies, which could cause some uncertainty. Some assumptions were made in the calculations of transport distances and some chemical and raw material amounts. The calculation results showed that these emission sources did not have a significant effect on the carbon footprint. The error is therefore relatively small. The LignoBoost and FT diesel processes were simplified in the calculations, which could have caused some uncertainty in the results. The data for the processes were obtained from Metso and Neste Oil, however, and the data from these sources can be considered reliable.

From the perspective of the carbon footprint, the LignoBoost process can be said to be an interesting option when the main purpose is to increase the pulp production capacity and lower the operating costs of the recovery boiler. FT diesel production is one option to achieve the target of 10% renewable energy use in transportation. From the carbon footprint perspective the FT diesel process is challenging when it is integrated with the pulp and paper mill because the allocation of emissions is not simple. However, it can be said that the FT diesel process integrated with the pulp and paper mill reduces also the carbon footprint when only the integrated pulp and paper mill is examined.

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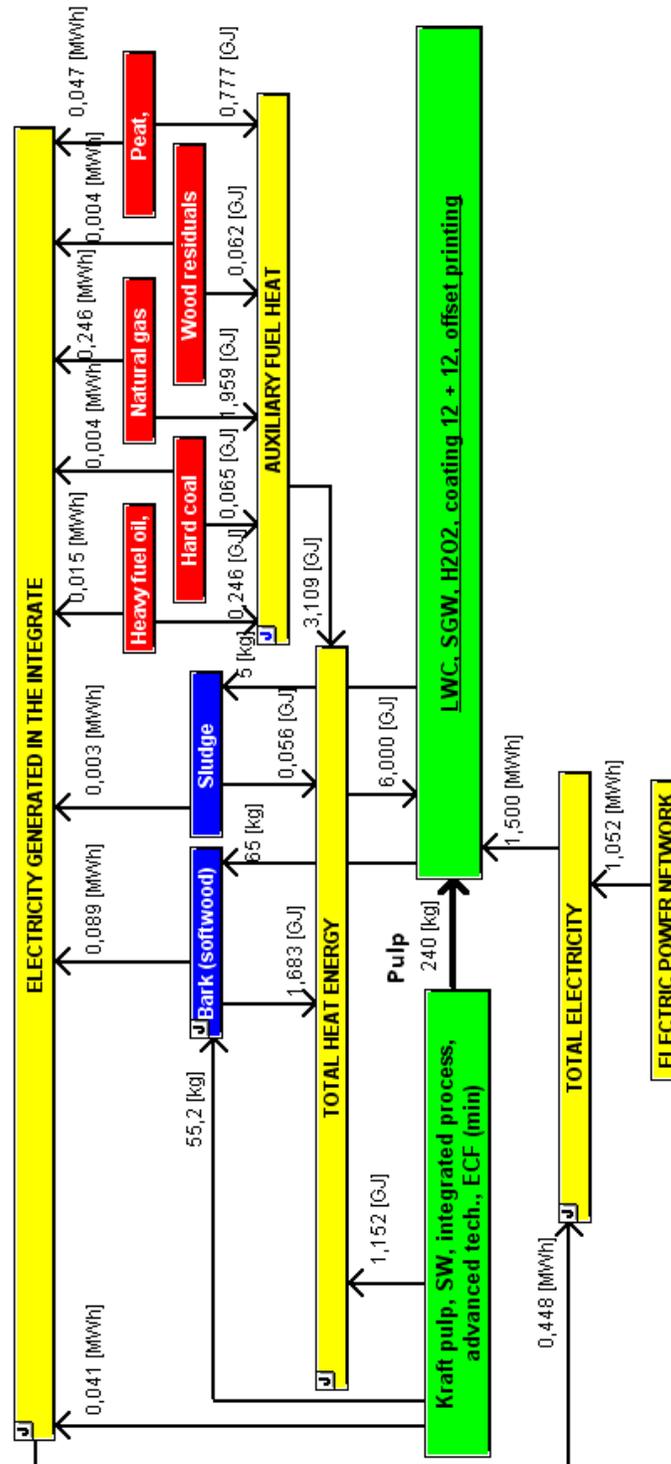
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### The effects of energy wood usage and harvesting

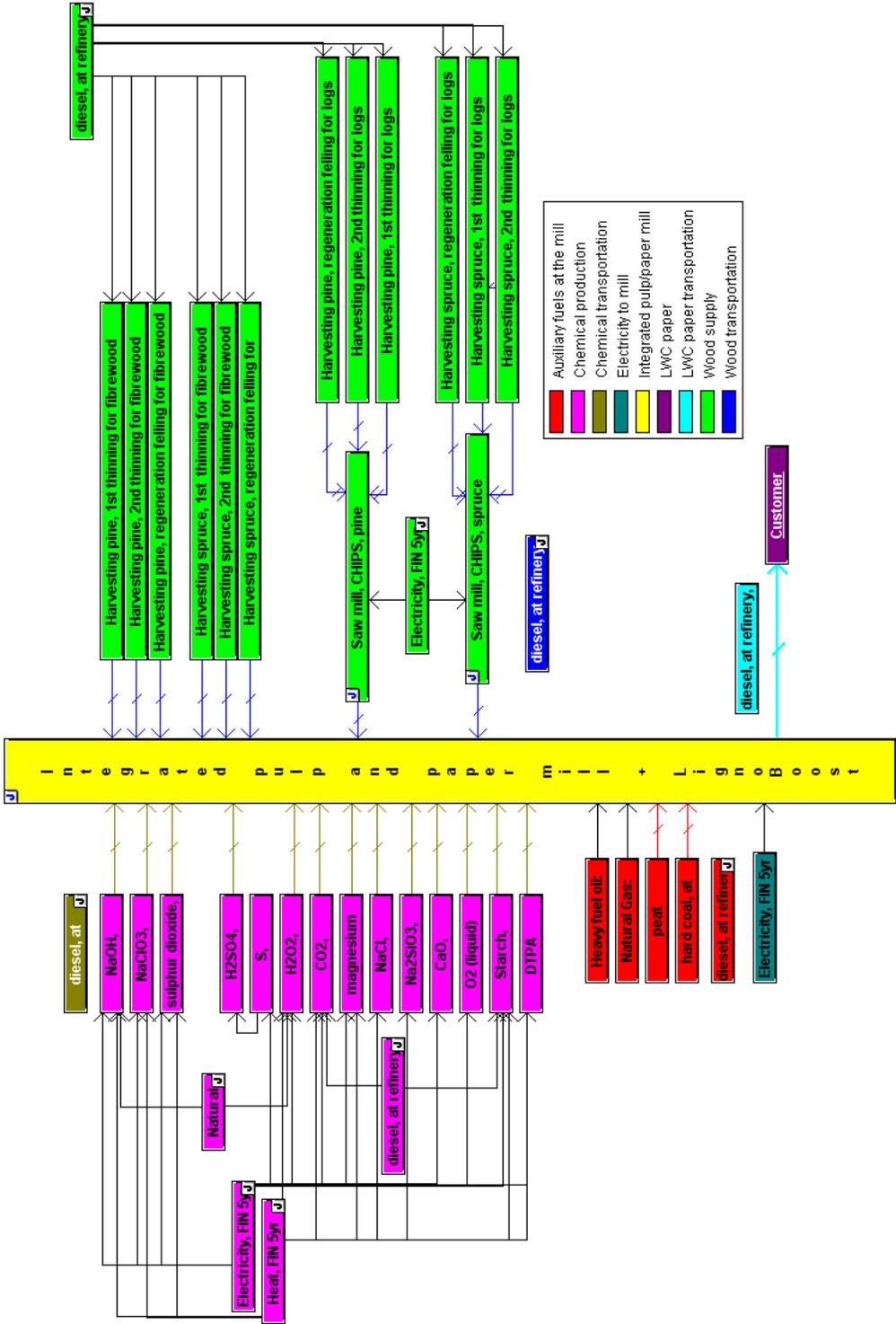
|  |  |
|--|--|
| Fossil fuel replacement (Kareinen et al. 2008, 14)                       | If the use of forest chips were to increase 9 million m <sup>3</sup> per year, 5 Tg CO <sub>2</sub> emissions could be replaced in the 2005 fuel distribution.   |
| Acidity of forest land (Helmisaari et al. 2008, 18)                      | The collection of forest residues may reduce the pH of the organic deposit of the soil.  |
| Increasing machine work in the forest (Helmisaari et al. 2008, 18)       | 3-5 machines are needed to collect wood fuel from the forest compared with 2 machines in traditional wood harvesting.  |
| Physical changes in the forest (Helmisaari et al. 2008, 18)              | Machines cause deep tracks and seal the molten soil.   |
| Changes in the nutrient content of the soil (Helmisaari et al. 2008, 18) | Enough nutrients in convenient concentrations and amounts are important for the undisturbed growth of the wood. When significant amounts of organic matter and nutrients are left, however, the alluviation of the nutrient into the water system and ground water may decrease. Wood ash can be used as a fertilizer to replace the nutrient content that is lost.  |
| Possible changes in forest biodiversity (Siitonen 2008, 30)              | <ul style="list-style-type: none"> <li>• Increase in the endangerment of threatened species</li> <li>• Significant degradation or endangerment of other species or living environments</li> <li>• Harmful effects of the long-term productivity and power of regeneration of forest land</li> <li>• Significant harmful effects on the other yielded material or immaterial commodity or services of ecosystems (e.g., mushrooms, carbon storage, quality of water, recreational use)</li> </ul> |

|   |  |
|---|--|
| <p>Biomass mass harvesting has effects on forest regeneration operations (Saksa 2008, 36)</p> | <ul style="list-style-type: none"><li>• The collection of crowns and branches facilitates wood planting.</li><li>• The collection of stumps cultivates the soil.</li><li>• According to research results, plants do better when biomass is harvested.</li><li>• The number of deciduous trees increases when the costs of clearing increase.</li><li>• The average length of wood and the development of basal decelerate, especially, in spruce planting.</li></ul> |
|---|--|

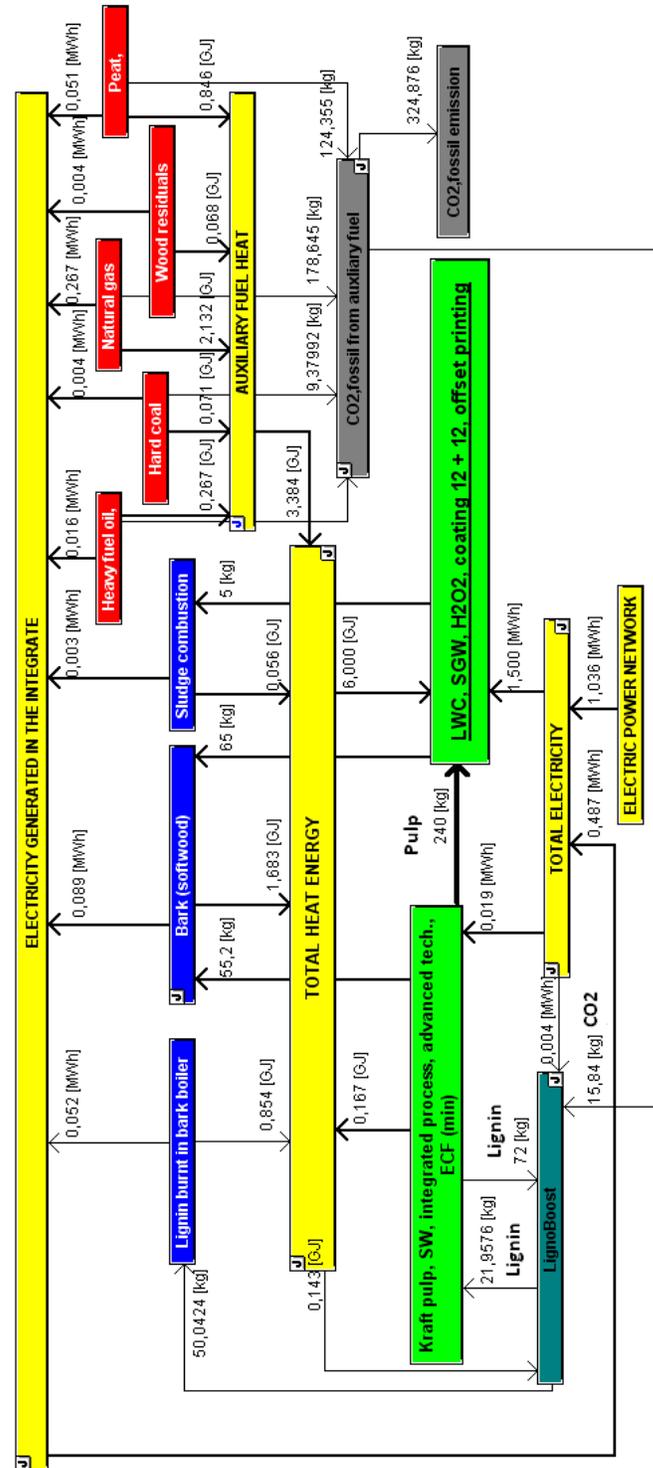
Case 1a: Reference pulp and paper mill integration with flow values



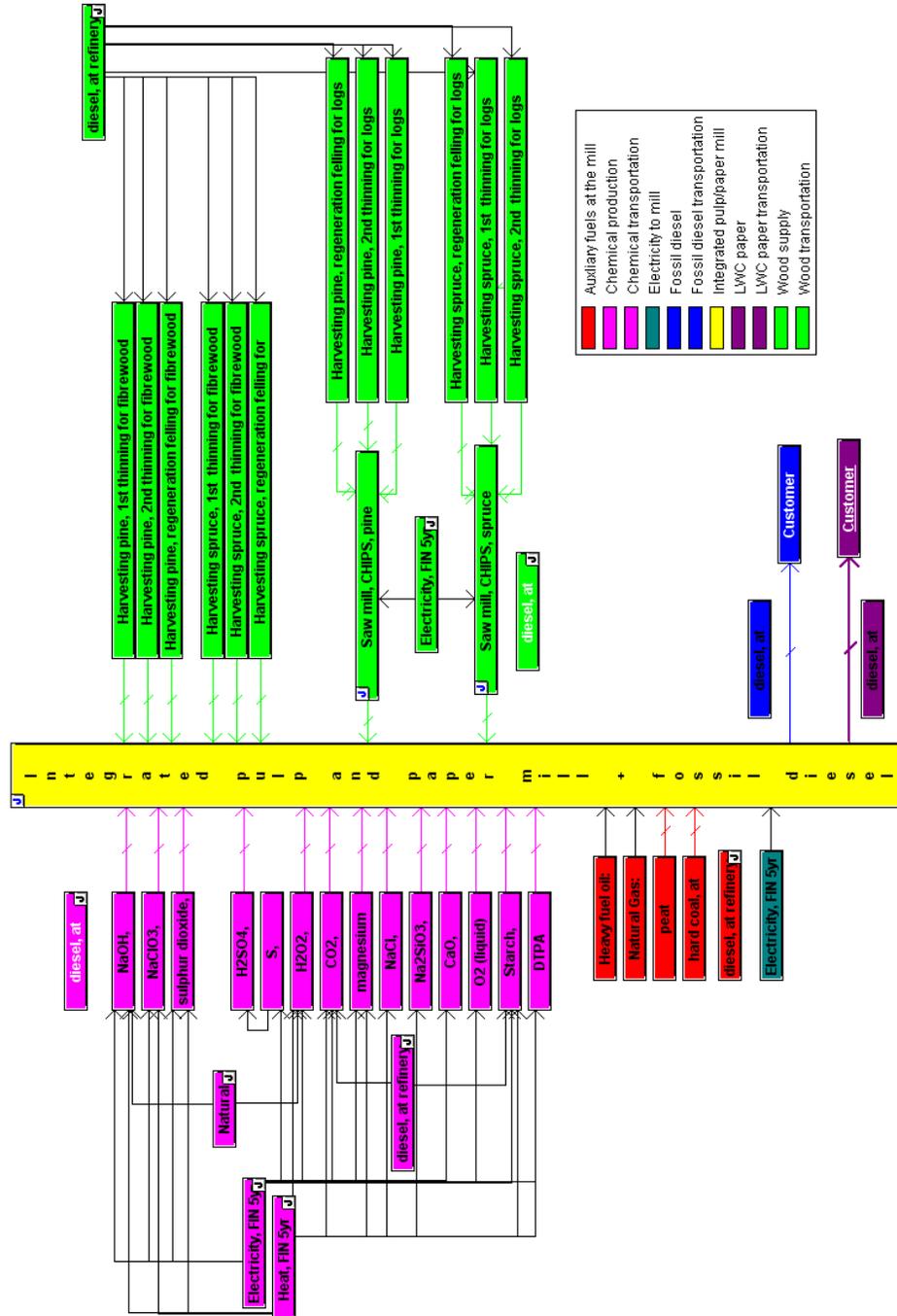
Case 1b: The flowsheet of cradle-to-customer approach



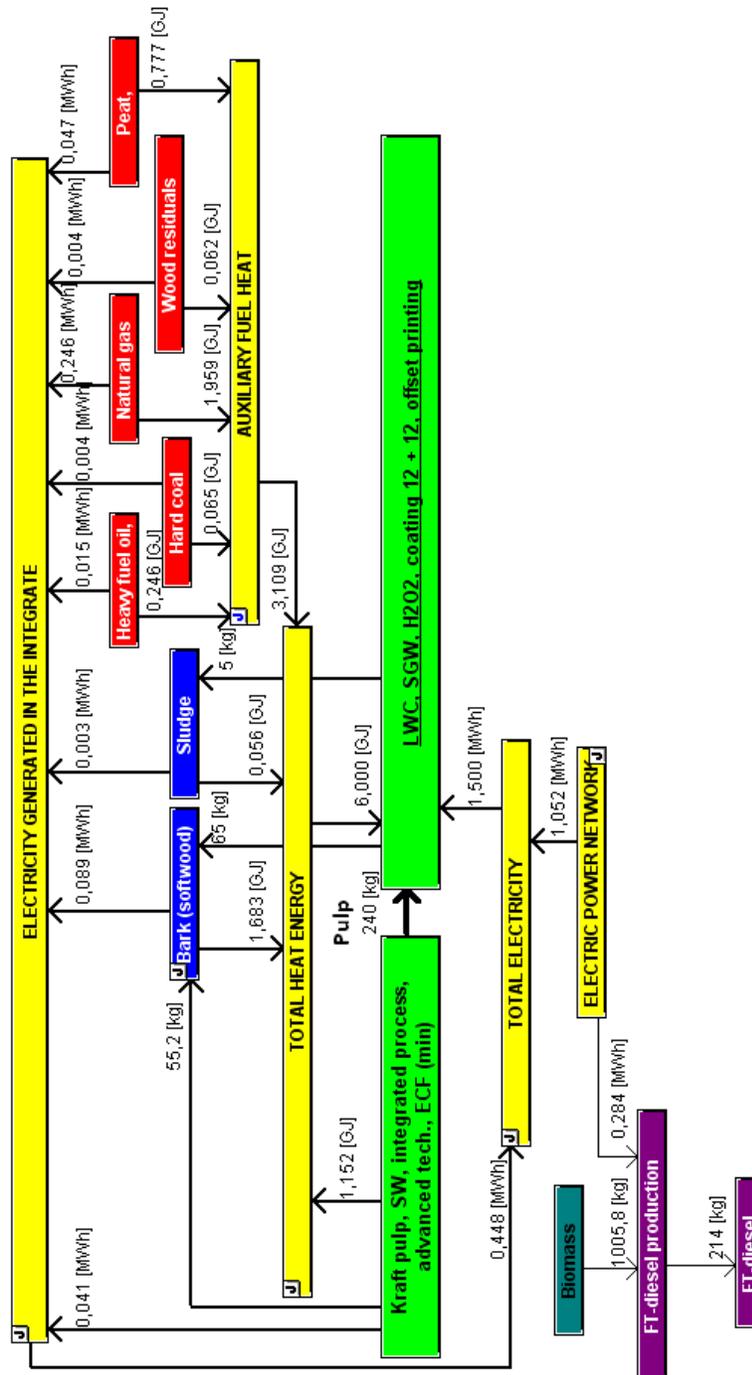
Case 1b: The flowsheet of mill-level approach with flow values



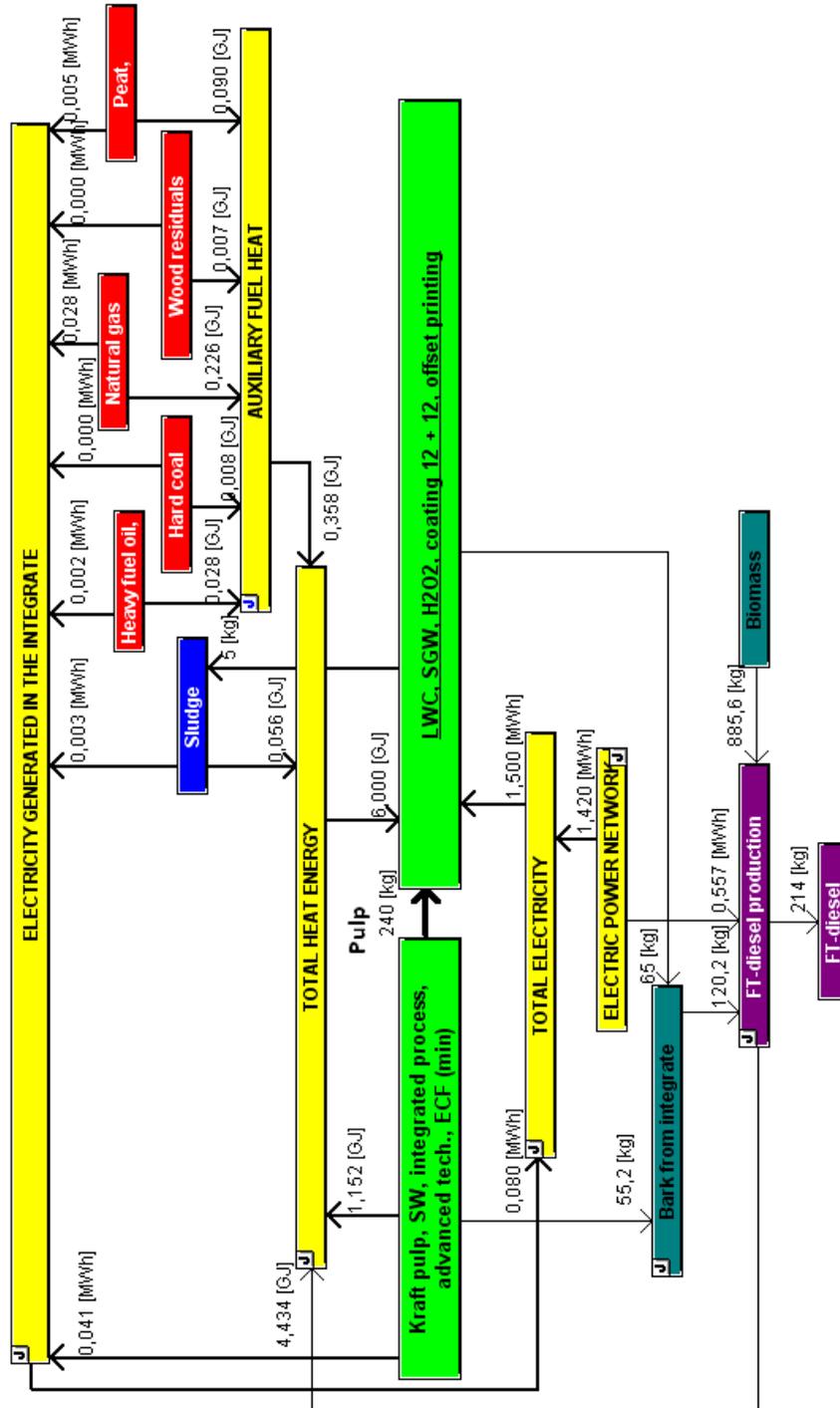
**Case 2a: The flowsheet of cradle-to-customer approach when fossil diesel and paper are produced separately**



**Case 2b: The flowsheet of production sites of paper and FT diesel -examination with flow values**



Case 2c: The flowsheet of production sites of paper and FT diesel -examination with flow values



### The calculation and assumptions used in LignoBoost case

#### Assumptions:

|                                       |   |
|---------------------------------------|---|
| Separated lignin                      | 300 kg <sub>lignin</sub> /t <sub>pulp</sub> |
| Energy for lignin drying              | 300 kWh/t <sub>DS,lignin</sub>              |
| Heavy fuel oil in a lime kiln         | 54 kg                                       |
| Lower heating value of heavy fuel oil | 41 MJ/kg <sub>oil</sub>                     |
| Lower heating value of lignin         | 24.2 kJ/kg                                  |
| Bark boiler total efficiency          | 0.86  |
| Recovery boiler total efficiency      | 0.69  |
| Back pressure construction factory    | 0.22  |
| Additional water from LignoBoost      | 2 m <sup>3</sup> /t <sub>lignin</sub>       |
| Steam consumption in a BL evaporator  | 450 MJ/t <sub>H<sub>2</sub>O</sub>          |

| Consumable                     | Consumption per ton lignin |
|--------------------------------|----------------------------|
| CO <sub>2</sub>                | 220 kg                     |
| H <sub>2</sub> SO <sub>4</sub> | 190 kg                     |
| NaOH                           | 65-130 kg                  |
| Electricity                    | 50 kWh                     |

Extra heat consumption due to LignoBoost process:

$$2 \frac{m^3}{t_{lignin}} \cdot 450 \frac{MJ}{t_{H_2O}} = 900 \frac{MJ}{t_{lignin}} \quad (1)$$

Energy consumption in lignin drying and pulverizing processes:

$$0.3 MWh \cdot 3600 = 1080 \frac{MJ}{t_{lignin}} \quad (2)$$

Total heat consumption in LignoBoost process:

$$900 \frac{MJ}{t_{lignin}} + 1080 \frac{MJ}{t_{lignin}} = 1980 \frac{MJ}{t_{lignin}} \quad (3)$$

Energy content of lignin after drying:

$$24.2 \frac{MJ}{kg_{lignin}} \cdot 300 kg_{lignin} = 7260 MJ \quad (4)$$

Energy content of 54 kg heavy fuel oil corresponds to amount of lignin which is needed to replace heavy fuel oil in a lime kiln:

$$54 kg_{oil} \cdot 41 \frac{MJ}{kg_{oil}} = 2214 MJ \quad (5)$$

Lignin separation reduces energy production in a recovery boiler:

$$7260 MJ \cdot 0.69 = 5009.4 \frac{MJ}{t_{pulp}} \quad (6)$$

This reduced energy is divided into heat and electricity. The share of heat and energy can be calculated with pair of equation when the construction factory is known. Electricity is known as  $E$  and heat as  $\emptyset$ .

$$\begin{cases} E + \emptyset = 5009.4 \frac{MJ}{t_{pulp}} \\ \frac{E}{\emptyset} = 0.22 \end{cases} \quad (7)$$

Thus

$$\emptyset = \frac{5009.4 \frac{MJ}{t_{pulp}}}{1.22} = 4106 \frac{MJ}{t_{pulp}} = 4.11 \frac{GJ}{t_{pulp}} \quad (8)$$

$$E = 0.22 \cdot \phi = 903.3 \frac{MJ}{t_{pulp}} = 0.251 \frac{MWh}{t_{pulp}} \quad (9)$$

Remaining lignin is burnt in a bark boiler. The amount of lignin burnt in a bark boiler can be calculated as follows:

$$7260 MJ - 2214 MJ = 5046 MJ \quad (10)$$

Energy from bark boiler can be calculated with the total efficiency of recovery boiler:

$$5046 MJ \cdot 0.86 = 4339.45 MJ \quad (11)$$

This is divided into heat and electricity:

$$\phi = \frac{4339.45 \frac{MJ}{t_{pulp}}}{1.22} = 3557 \frac{MJ}{t_{pulp}} = 3.56 \frac{GJ}{t_{pulp}} \quad (12)$$

$$E = 0.22 \cdot \phi = 782.54 \frac{MJ}{t_{pulp}} = 0.217 \frac{MWh}{t_{pulp}} \quad (13)$$

### Calculations and assumptions used in FT diesel cases

#### Assumptions

|                                       |                                    |
|---------------------------------------|------------------------------------|
| Case 2b, electricity consumption      | 24 MW/t <sub>FT diesel</sub>       |
| Case 2c, electricity consumption      | 47 MW/t <sub>FT diesel</sub>       |
| Case 2c, steam production             | 104 MW/t <sub>FT diesel</sub>      |
| Case 2b, 2c, off-gas production       | 11 MW/t <sub>FT diesel</sub>       |
| Biomass usage                         | 4.7 t/t <sub>FT diesel</sub> (0 %) |
| Methanol consumption                  | 8.3 kg/t <sub>FT diesel</sub>      |
| Hydrogen consumption                  | 6 kg/t <sub>FT diesel</sub>        |
| LWC paper mill capacity               | 700 000 t/a                        |
| Annual operation time                 | 8300 h                             |
| Heavy oil used in a lime kiln         | 12.96 kg/t <sub>paper</sub>        |
| Lower heating value of heavy fuel oil | 41 MJ/kg                           |

Defining of the size of the FT diesel production unit. EFI has defined the harvestable potential for forest chips during pulp wood harvesting.

Pine wood: 5 % from thinning operations per the amount of pulp wood harvested  
 21 % from final fellings per the amount of pulp wood harvested  
 Spruce wood: 8 % from thinning operations per the amount of pulp wood harvested  
 29 % from final fellings per the amount of pulp wood harvested

448.8 kg pine wood and 480 kg spruce wood is harvested. The share of pulp wood harvested from thinning operations is 27 % and from final fellings 73 % (KCL-Eco database). Thus the amount of forest chips can be calculated as follows.

Pine wood:

$$0.27 \cdot 448.8 \text{ kg} \cdot 0.05 + 0.73 \cdot 448.8 \text{ kg} \cdot 0.21 = 74.9 \text{ kg} \quad (14)$$

Spruce wood:

$$0.27 \cdot 480 \text{ kg} \cdot 0.08 + 0.73 \cdot 480 \text{ kg} \cdot 0.29 = 112 \text{ kg} \quad (15)$$

Thus the total forest chips amount is about 187 kg.

If the FT diesel process used only 187 kg forest chips as raw material, the amount of produced FT diesel would be

$$\frac{187 \text{ kg biomass}}{4700 \frac{\text{kg biomass}}{\text{t}_{FTdiesel}}} \cdot 1000 = 40 \text{ kg}_{FTdiesel} \quad (16)$$

The amount is so small that is profitable to examine the raw material potential when forest chips, bark and saw dust is included. The raw material potential is then 1.0-2.4 solid cubic meters per year. The density of pine wood is 411 kg/m<sup>3</sup> and spruce wood 390 kg/m<sup>3</sup>, thus the average density is 400.5 kg/m<sup>3</sup>. The equations 17 and 18 show the raw material potential for minimum and maximum.

Minimum raw material potential:

$$\frac{400.5 \frac{\text{kg}}{\text{m}^3} \cdot 1.0 \cdot 10^6 \text{ m}^3}{1000} = 400\,500 \text{ t} \quad (17)$$

Maximum raw material potential:

$$\frac{400.5 \frac{\text{kg}}{\text{m}^3} \cdot 2.4 \cdot 10^6 \text{ m}^3}{1000} = 916\,200 \text{ t} \quad (18)$$

The average amount of FT diesel production, calculated with these potential is

$$\frac{\frac{400\,500 \text{ t}}{4.7 \frac{\text{t}}{\text{t}_{FTdiesel}}} + \frac{916\,200 \text{ t}}{4.7 \frac{\text{t}}{\text{t}_{FTdiesel}}}}{2} = 144\,862 \text{ kg} \quad (19)$$

The amount of FT diesel production is chosen to be 150 000 kg per year.

Thus the FT diesel production per ton of LWC paper can be calculated with equations 20 and 21.

$$\frac{150\,000\ t_{FTdiesel}}{700\,000\ t_{LWC\ paper}} = 214\ \frac{t_{FTdiesel}}{t_{LWC\ paper}} \quad (20)$$

Which is

$$\frac{150\,000\ t_{FTdiesel}}{8\,300\ h} = 18\ \frac{t_{FTdiesel}}{h} \quad (21)$$

When the production per hour is known, electricity consumption and steam and off-gas production can be calculated per ton of paper. As an example is calculated the electricity consumption for 24 MW mill. The other results are presented in the table below.

$$\frac{24\ MW}{18\ \frac{t_{FTdiesel}}{h}} \cdot 0.214\ \frac{t_{FTdiesel}}{t_{paper}} = 0.284\ \frac{MWh}{t_{paper}} \quad (22)$$

|  | <b>Case 2b</b> | <b>Case 2c</b> |
|--|----------------|----------------|
| <b>Electricity consumption [MWh/t<sub>paper</sub>]</b> | 0.284          | 0.557          |
| <b>Steam production [GJ/t<sub>paper</sub>]</b>         | 0              | 4.43           |
| <b>Off-gas production [MJ/t<sub>paper</sub>]</b>       | 470            | 470            |

Off-gas to lime kiln:

Energy content of 12.96 kg heavy oil can be calculated as follows:

$$12.96 \frac{kg_{oil}}{t_{paper}} \cdot 41 \frac{MJ}{kg_{oil}} = 531 \frac{MJ}{t_{paper}} \quad (23)$$

The amount of replaced heavy fuel oil in a lime kiln can be calculated as follows:

$$\frac{(531-470) \frac{MJ}{t_{paper}}}{41 \frac{MJ}{kg_{oil}}} = 1.5 \frac{kg_{oil}}{t_{paper}} = 6 \frac{kg}{t_{pulp}} \quad (24)$$