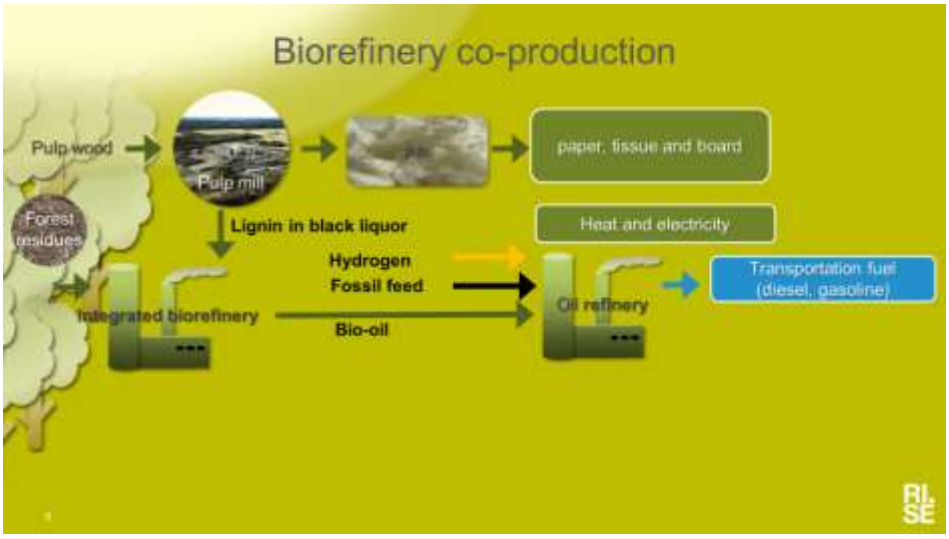
REPORT f3 2017:16

DETERMINATION OF POTENTIAL IMPROVEMENTS IN BIO-OIL PRODUCTION

PRODUCTION OF TRANSPORTATION FUEL COMPONENTS IN VALUE CHAINS INTEGRATING PULP & PAPER AND OIL REFINERY INDUSTRY

Report from a project within the collaborative research program *Renewable transportation fuels and systems*

December 2017



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THE SWEDISH KNOWLEDGE CENTRE
FOR RENEWABLE TRANSPORTATION FUELS

PREFACE

This project has been carried out within the collaborative research program *Renewable transportation fuels and systems* (Förnybara drivmedel och system), Project no. 39587-2. The project has been financed by the Swedish Energy Agency and f3 – Swedish Knowledge Centre for Renewable Transportation Fuels.

f3 Swedish Knowledge Centre for Renewable Transportation Fuels is a networking organization which focuses on development of environmentally, economically and socially sustainable renewable fuels, and

- Provides a broad, scientifically based and trustworthy source of knowledge for industry, governments and public authorities
- Carries through system oriented research related to the entire renewable fuels value chain
- Acts as national platform stimulating interaction nationally and internationally.

f3 partners include Sweden's most active universities and research institutes within the field, as well as a broad range of industry companies with high relevance. f3 has no political agenda and does not conduct lobbying activities for specific fuels or systems, nor for the f3 partners' respective areas of interest.

The f3 centre is financed jointly by the centre partners and the region of Västra Götaland. f3 also receives funding from Vinnova (Sweden's innovation agency) as a Swedish advocacy platform towards Horizon 2020. Chalmers Industriteknik (CIT) functions as the host of the f3 organization (see www.f3centre.se).

In this project, the participating partners have been RISE, ÅF Industry, Chalmers and Preem.

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SUMMARY

A potential future route for renewable transportation fuels is to produce bio-oil from forest raw material through i.e. fast pyrolysis, which is then upgraded to a transportation fuel at an existing refinery. The oxygen content of the produced bio-oil is determining the hydrogen demand to meet the required oxygen level from a refinery perspective and for use in engines. This in turn significantly affects the cost and the associated CO_2 emissions from production of the transportation fuel, as it to a large extent is a result of the used raw material and production route for hydrogen. This has been shown in an earlier project within the Swedish Energy Agency and f3 collaborative research program Renewable transportation fuels and systems: Project 39587-1, *Value chains for production of renewable transportation fuels using intermediates*, reported in Anheden et al. (2016).

The goal of this project is to investigate emerging technologies that result in a lower oxygen content of the produced bio-oil than what is currently achieved through near commercial fast pyrolysis. In addition, alternative routes for production of hydrogen with a lower climate impact than the leading technology for hydrogen production today, which commonly is based on steam reforming of natural gas, are evaluated. Value chains utilising forest residues and lignin in black liquor from kraft pulping as feedstock are simulated and analysed. The value chains combine existing know how, infrastructure and equipment within the pulp and paper and oil refinery industry, including different technology options for bio-oil production and hydrodeoxygenation and upgrading of the produced oil to fuel components. The value chains are evaluated and compared with respect to total production cost, overall value chain fossil CO_2 emissions, conversion yield and total energy efficiency.

The results show that there are emerging technologies for bio-oil production under development that has the potential to produce bio-oil with a lower oxygen content than through fast pyrolysis. In this study, hydropyrolysis and hydrothermal liquefaction (HTL) have been identified as promising emerging technologies and have been studied in more detail. Further analysis shows that there is also a good potential to reach a lower total transportation fuel production cost than through fast pyrolysis, however the data used in the analysis is still uncertain due to the lower technical maturity of both the hydropyrolysis and HTL processes. The production cost for transportation fuel components from pyrolysis is about 100 EUR/MWh (or 0.9 EUR/l), while the emerging technologies have a production cost of about 65 EUR/MWh (or 0.6 EUR/l). This means that the production cost could compete with the price on the European spot market in 2016, which was in the range of 82-100 EUR/MWh during 2016. The cost is also lower than the 122-127 EUR/MWh reported production cost for Swedish suppliers of biofuels according to data submitted in 2016 to the Swedish Energy Agency by biofuel producers.

Integration of process excess energy with the existing pulp mill and especially the refinery has a large positive impact in the economic evaluation and also on the reduction of the value chain CO_2 emissions. The calculated value chain CO_2 emissions indicate a CO_2 reduction potential of 95% or more compared to fossil fuel (excluding emissions of other climate gases). However, the allocation of CO_2 emission to the biofuel components according to the EU Renewable Energy Directive (RED) would make the transportation fuel's CO_2-footprint look very different, with resulting significantly higher footprint that might even exceed the stated reduction targets. This requires further analysis and discussion and re-evaluation of allocation rules. Further concept development might

also be required, where for instance excess energy instead could be used for hydrogen production or where alternative sources of hydrogen are used.

Using alkaline electrolysis of water based on average Nordic power production mix results in reduction of the value chain CO_2 emissions at comparable cost as use of hydrogen from natural gas steam reforming at low electricity price (20 EUR/MWh electricity). Use of gasification-based hydrogen production from forest residues results on the other hand in both higher value chain costs and higher CO_2 emissions. This is an effect of moving the location of the hydrogen production and hydrodeoxygenation from the refinery location to the pulp mill location to support the gasification plant with raw material, knowhow and infrastructure for handling of forest residues. The integration of the residual energy from the HDO process is not as favourable in the pulp mill as in the refinery location, where use of fuel oil for heating purposes could be replaced by the excess energy. It is recommended that these integration aspects are studied in more detail in a follow-up project.

Another important observation made during the project is that the consumption of add-on chemicals has quite a large influence on both cost and CO_2 emissions of the studied value chains. In one of the analysed value chains, very high production cost and CO_2 emission was found as a consequence of adding phenol during the production of bio-oil to prevent repolymerisation. The lesson learned is that the effect of chemical addition must be investigated early in the process development phase, and that chemicals preferably need to be recycled, regenerated in the process or replaced by chemicals that can be produced in a sustainable way to minimize their influence.

Finally, it is noted that the investigated emerging technologies, both for depolymerisation to bio-oil and upgrading of the oil to transportation fuel components, have a lower technical maturity level than the near commercial fast pyrolysis technology, and still require technical development and verification in one or several areas. In addition, very little experience from operation of this type of plants are available, even in pilot-scale. Accordingly, both the technical and economic performance data used in this evaluation are quite uncertain. However, the sensitivity analysis indicates that there is room for adjustments of the performance data with remaining acceptable profitability for the emerging technologies.

SAMMANFATTNING

En framtida väg att producera förnybara drivmedel är att förvätska och producera en bioolja från skogsråvara genom t ex snabb pyrolys och sedan uppgradera den till drivmedel i ett existerande oljeraffinaderi. Biooljans syreinnehåll avgör vätgasbehovet för att uppgradera biooljan enligt specifikation för integrering i oljeraffinaderi och för användning i motorer. Detta i sin tur har stark inverkan på kostnaden för att framställa drivmedel och de fossila CO_2-emissionerna som uppstår i samband med drivmedelsproduktionen, då detta till en stor del är beroende av råvara och produktionsväg för vätgas. Detta har visats i ett tidigare projekt inom samverkansprogrammet Förnybara drivmedel och system, Projekt nr 39587-1, Värdekedjor med intermediära biobränslen, som rapporterats i Anheden et al. (2016).

Syftet med detta projekt är att utvärdera nytillkomna teknologier som producerar en bioolja med lägre syreinnehåll än vad som uppnås genom den nära kommersiella teknologin för snabb pyrolys. Dessutom utvärderas alternativa vägar för vätgasproduktion med lägre klimatpåverkan än dagens ledande produktionssätt som sker via ångreformering av naturgas. Värdekedjor baserade på utnyttjande av restprodukter från skogen i form av grenar och toppar (grot) och sulfatlignin från massabrukens svartlutar som råvara simuleras och analyseras. Värdekedjorna kombinerar tillgänglig kunskap, infrastruktur och utrustning inom massa- och pappersindustrin och oljeraffinaderiindustrin och inkluderar olika teknikalternativ för produktion av bioolja samt hydrodeoxygenering (HDO) och uppgradering av biooljan till drivmedelskomponenter. Värdekedjorna jämförs med avseende på total produktionskostnad, totala utsläpp av fossilt CO_2, omvandlingsgrad och total energieffektivitet.

Resultaten visar att det finns nytillkomna teknologier för produktion av bioolja som är under utveckling och som har potential att producera bioolja med lägre syreinnehåll än vad som är fallet vid snabb pyrolys. I denna studie har hydropyrolys och hydrotermisk förvätskning (HTL) identifierats som lovande nytillkomna teknologier, och har därför studerats i mer detalj. Vidare analys visar att dessa teknologier potentiellt kan uppnå en lägre total drivmedelsproduktionskostnad än vid snabb pyrolys. Emellertid råder det en del osäkerhet kring de data som använts i analysen på grund av den lägre tekniska mognadsgraden för både hydropyrolys- och HTL-processerna. Produktionskostnaden för drivmedelskomponenter från pyrolys är i storleksordningen 100 EUR/MWh (eller 0,9 EUR/l), medan de nytillkomna teknologierna har en uppskattad produktionskostnad i området kring 65 EUR/MWh (eller 0,6 EUR/l). Detta visar att de är konkurrenskraftiga mot priset på biodrivmedel på den europeiska spot-marknaden år 2016 som var runt 82-100 EUR/MWh. De är också lägre än de rapporterade produktionskostnaderna på 122-127 EUR/MWh från svenska biodrivmedelsleverantörer enligt inskickade data från svenska drivmedelsleverantörer till Energimyndigheten år 2016.

Integrering av överskottsenergi från processerna med massabruket och särskilt raffinaderiet har en stor positiv inverkan på produktionskostnaden och också på de uppskattade reduktionerna i utsläpp av fossilt CO_2 för de studerade värdekedjorna. De beräknade CO_2-utsläppen längs värdekedjan indikerar att det finns en reduktionspotential på 95% eller mer jämfört med fossila drivmedel, när utsläpp av andra klimatpåverkande gaser än CO_2 exkluderas. Emellertid skulle allokering av CO_2-emissioner till drivmedelskomponenterna enligt de regler som finns i EUs förnybarhetsdirektiv, (RED), göra att drivmedlets CO_2-avtryck skulle se väldigt annorlunda ut, med signifikant större avtryck, som till och med kan överskrida reduktionsmål för biodrivmedel. Detta kräver vidare analys

och diskussion och omvärdering av allokeringsregler. Ytterligare konceptutveckling kan också vara nödvändigt där man t.ex. använder överskottsenergi för vätgasproduktion eller där alternativa källor för vätgas implementeras.

Att producera vätgas med hjälp av alkalisk elektrolys av vatten, där elen belastas med utsläpp motsvarande nordisk elproduktionsmix, leder till minskade utsläpp av CO_2 jämfört med vätgas från naturgas. Ungefär samma kostnadsnivå är möjlig att uppnå vid låga elpriser (20 EUR/MWh el). Förgasningsbaserad vätgasproduktion från grot ger å andra sidan både högre kostnader och högre utsläpp av CO_2. Detta är en effekt av att placeringen av vätgasproduktionen och hydrodeoxygeneringen flyttas från oljeraffinaderiet till massabruket för att utnyttja massabrukets tillgång på råmaterial, infrastruktur och kunskap för hantering av skogsråvara. Integreringen av restenergin från HDO-processen är inte lika gynnsam i massabruket som i oljeraffinaderiet, där användning av eldningsolja kan undvikas genom att ersättas med restenergi. Det rekommenderas att dessa integrationsaspekter studeras i mer detalj i ett uppföljningsprojekt.

En annan viktig observation som gjorts under projektets genomförande är att konsumtionen av tillsatskemikalier har en relativt stor inverkan på både kostnad och CO_2-emissioner för de studerade värdekedjorna. I en av de analyserade värdekedjorna så konstaterades det både väldigt höga produktionskostnader och CO_2-utsläpp som en konsekvens av att fenol tillsattes under produktionen av biooljan för att undvika återpolymerisering. Lärdomen är att effekten av kemikalietillsatser måste analyseras tidigt i processutvecklingsfasen, och att kemikalier företrädesvis bör återvinnas och återcirkuleras eller återgenereras i processen alternativt ersättas av kemikalier som kan produceras på ett hållbart sätt för att minimera deras inverkan.

Slutligen så kan det noteras att de nytillkomna teknologierna för både depolymerisering till bioolja och uppgradering av oljan till drivmedelskomponenter har en lägre teknisk mognadsgrad än den nästan kommersiella teknologin för snabb pyrolys. Vidare teknisk utveckling och verifikation är nödvändig på flera områden. Dessutom saknas driftserfarenheter, även i pilotskala. Därmed är de tekniska och ekonomiska data som använts i utvärderingen relativt osäkra. Emellertid indikerar de känslighetsanalyser som gjorts att det finns utrymme för försämrad prestanda med fortsatt acceptabel lönsamhet för de nytillkomna teknologierna.

CONTENTS

1 INTRODUCTION

The Paris agreement during COP21 in 2015 sets the way for the transformation of the world from dependent on fossil energy carriers with large emissions of fossil CO_2 to a world supported by renewable and low carbon intensity energy. Sweden has ambitions to lead the way and has stated that Sweden should be one of the first fossil energy free welfare nations. About 20% of Sweden's fossil CO_2 emissions are related to the transport sector, or about 13.5 Mton CO_2 eqv. in 2016 (SCB, 2017). Sweden has set as a goal to reduce the CO_2 emissions from domestic road transport by 70% by 2030 compared to 2010 year's level, and by 2045, Sweden should reach zero CO_2 emissions. The transformation toward no CO_2 emissions from the transport sector is planned to take place through increased efficiency in the transport system, electrification and introduction of renewable and sustainable transportation fuels. The last option is the focus of this report.

The increased demands on renewable transportation fuels requires a transformation of the petroleum industry to an industry that produces and delivers sustainable fuels with a substantial lower climate impact. The increased demands on renewable fuels will result in increased competition about the raw materials, and new types of raw material and processes are required to fill the large need and demand. Today about 90% of the biofuels used in Sweden are imported. However, residues and waste from the Swedish forest industry are very interesting raw materials due to the large availability in Sweden. This type of raw material can be processed in existing refinery processes or in stand-alone plants after transformation to bio-oil, which is the main topic of this report and can in this way contribute to domestic fuel production.

In the so called fff-investigation and the report "Fossilfrihet på väg" (SOU 2013:84), the potential for domestic production of renewable transportation fuels is estimated to about 25-30 TWh/year in 2030, of which 4-5 TWh/year is estimated to be HVO, i.e. hydrogenated vegetable oils and other bio-oils (e.g. synthetic diesel). Since then, the estimation of the potential for drop in fuel such as HVO has been considerably increased as the investments in technology to produce high blend fuels such as DME and methanol from biomass has turned out to be difficult to realise. In the more resent so-called SOFT-investigation (Energimyndigheten 2017b) and the report "Förslag till styrmedel för ökad andel biodrivmedel i bensin och diesel" (Energimyndigheten 2016), the estimated additional potential access to forest related raw material in the form of waste and residues is 20-22 TWh/year in 2030. This is equivalent to about 13-14 TWh/year transportation fuel ready for use, assuming a yield of 65%.

A challenge with the bio-oil is the high content of oxygen compared to fossil crude oil that has an oxygen content close to zero. Use of refinery hydrodeoxygenation processes where hydrogen gas is used to reduce the oxygen content or entirely remove all oxygen in the bio-oil at the same time cracking to molecular structures suitable for transportation fuels is required. The hydrogen has a big impact on the economy and the CO_2 footprint of the transportation fuel and is dependent on the source and the process for production of hydrogen. The conventional fossil feedstock has a limited demand for hydrogen and the hydrogen production capacity in present refineries is normally too low to supply the hydrogen for bio-oil upgrading and large investments are necessary to increase the hydrogen production. It is important that industry find a way to decrease the oxygen content in the bio-oil and optimise the hydrogen production based on both economy and CO_2 emissions. The oxygen content in the bio-oil and the production path for hydrogen are the most important parameters for economic and environmental sustainability in addition to the yield from raw material to transportation fuel according to a previous project within the Swedish Energy Agency and f3 collaborative research program *Renewable transportation fuels and systems* (Förnybara drivmedel och system), where value chains based on fast pyrolysis of forest raw material have been studied, (Anheden et al., 2016).

1.1 GOALS AND OBJECTIVES

The goal of this project is to estimate the economic and environmental potential for advancements in bio-oil production for transportation fuel purposes by utilising emerging technologies and sources of renewable hydrogen in value chains connecting existing pulp and paper industry with existing oil refinery industry.

The detailed objectives are:

A- Investigate alternative technologies other than fast pyrolysis for production of bio-crude with respect to:
  - Oxygen content in the produced oil in relation to oxygen in the raw material
  - The yield from raw material to bio-oil

The investigation is based on collection of data from open literature and technology developers and providers.

B- Investigate alternatives for production of hydrogen with a low CO_2 footprint that can be compared to traditional hydrogen production through steam reforming of natural gas.

C- Use the collected information to estimate and calculate the:
  - Total production cost
  - CO_2 footprint
  - Yield
  - Total energy conversion efficiency

During the work, the following additional objectives have been added:

D- Evaluation of processing path and cost to achieve a final transportation fuel component

E- Estimation of potential for integrating residual mass and energy streams in existing refineries (integration in pulp mill was included in the study from the start)

The work has not included any attempts to investigate the potential technical and economic benefits of separating side streams for chemical and material production.

A project group consisting of representatives from petroleum oil refinery (Preem), pulp and paper industry research institute (RISE), technical consultants (ÅF Industry) and academia (Chalmers University of Technology) has jointly carried out the work. The project has had as a general purpose to generate a better understanding of the possibilities of collaboration between forest industry and transportation fuel producers. The collaboration in this diverse group has resulted in exchange of experience and new ideas, and an improved comprehensive view of the entire value chain. It is the hope that this study can support development of future industrial solutions for production of renewable transportation fuels in Sweden, based on the results and conclusions.

The results from this study can be utilized for general information by companies that have an interest in production and upgrading bio-oil from forest based raw material such as forest owners, pulp mills, saw mills, petrochemical industry and refineries in addition to technology providers to those industries. The material can also be used by technical consultants and research institutes while giving services and advice to customers. Technology developers and financiers can use the information to prioritize research needs and estimate the potential for development.

The potential effect goal for the society will be a system with production of renewable transportation fuels using local forest resources with lower cost and lower CO_2-footprint that will generate work places and incomes in Sweden.

1.2 DESCRIPTION OF THE INVESTIGATED VALUE CHAIN

The value chains investigated in this report are focusing on production of transportation fuel components in the diesel and gasoline range starting from two different types of forest-based raw materials classified as raw materials for advanced biofuels according to Annex IX, Part A, point o in the so called EU Indirect Land Use Change directive (ILUC) from 2015, (amending the EU Renewable Energy Directive (RED) and the EU Fuel Quality Directive (FQD), now part of the discussion of the proposed RED2 directive), (ref. Directive EU 2015/1513). The two selected fuels are forest residues from forestry such as branches and tree tops or kraft lignin in black liquor from the pulp and paper industry. These feedstocks are in the study upgraded to an intermediate, pumpable bio-oil product in a depolymerisation process integrated with a kraft pulp mill. The intermediate product is in a next step considered to be transported to an oil refinery for further upgrading to transportation fuel components using processes commonly used in the oil industry. The target is to produce 100 000 ton/year of transportation fuel components. An overview of the formed value chain is seen in Figure 1.

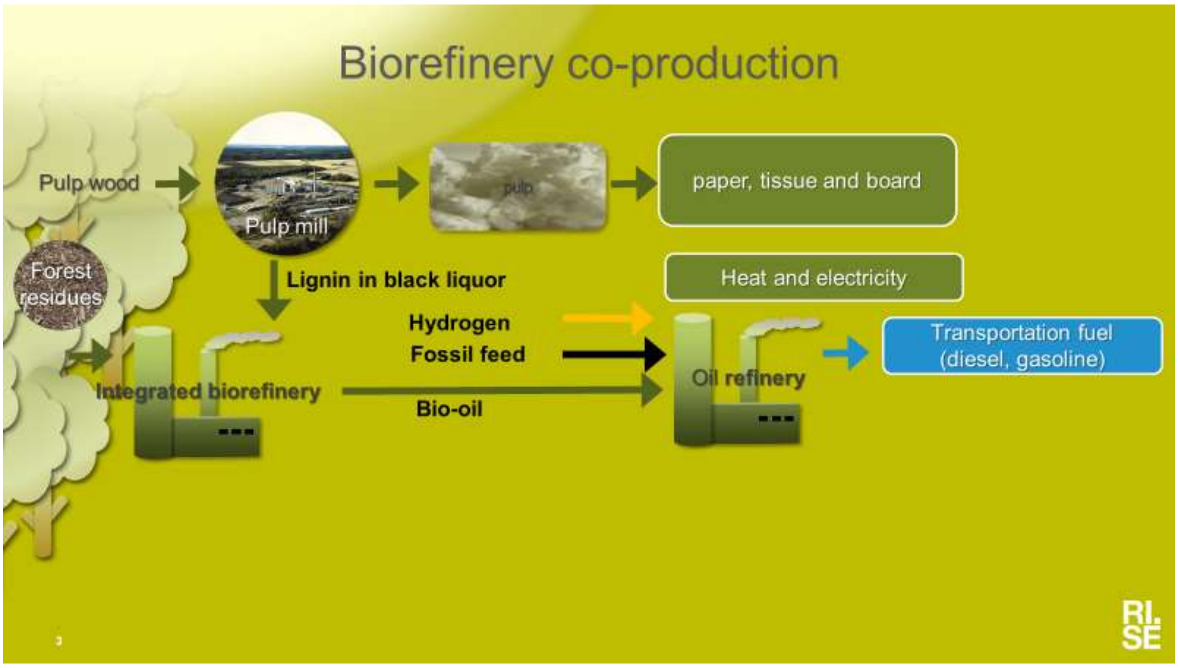


Figure 1. Value chain from forest residues or lignin to transportation fuel investigated in this study.

2 BACKGROUND AND PREVIOUS WORK

Transportation fuels mainly consists of molecules containing carbon (C) and hydrogen (H), while biological renewable raw materials consist of carbon-hydrogen-oxygen molecules. This means that the oxygen needs to be eliminated from the biomass to produce an acceptable transportation fuel that can be processed and delivered through existing infrastructure and used in conventional diesel and gasoline engines. Fast pyrolysis is the most mature production path for transportation fuels through liquefaction of biomass and has been developed over a large number of years and demonstrated in near commercial scale by several suppliers. Fast pyrolysis was assumed to be used in the bio-oil value chain study for utilisation of forest residues or kraft lignin for transportation fuel production through liquefaction in the proceeding project “Value chains for production of renewable transportation fuels using intermediates” (Anheden et al., 2016), carried out by the project partners.

2.1 STATE OF THE ART FOR FAST PYROLYSIS

In fast pyrolysis, the biomass is liquefied into a bio-oil, and in addition coke and energy containing gases are released. The process takes place in a low-oxygen environment and requires a fast heat-up period and biomass with a small particle size. The biomass is transformed to vapours that are quickly quenched to produce the condensable oil-phase that is separated from the non-condensable lighter gases. The bio-oil can next be treated with hydrogen to remove oxygen to be able to be processed in a refinery to transportation fuels such as gasoline and diesel. Near commercial plants for fast pyrolysis have been constructed. One example is Ensyn’s fast pyrolysis plant in Ontario, commissioned 2006, later enhanced 2014, using Envergent’s and Ensyn’s technology RTP®. Under construction (started 2016) is a new biocrude facility in Port Cartier, Quebec (Ensyn 2017, Envergent 2017). Other examples are Fortum Joensuu plant, by Metso/Valmet commissioned 2013 (Valmet 2013) and Empyro pyrolysis plant by BTG-BTL located in the Netherlands (BTG-BTL 2014). Yields of bio-oil in the range of 60-70 wt% have been demonstrated on stem wood. Technology to transform the pyrolysis oil to acceptable transportation fuel is under development and verification.

2.2 IMPACT OF RAW MATERIAL

From literature it is clear that not only the pyrolysis process affects the yield, but also the composition of the raw material. It is for instance known that forest residues, one of the selected raw materials in this study, has a significantly lower yield of bio-oil than when using stem wood. Data on large scale tests with pyrolysis of kraft lignin is not available in the public literature, however it is known that it is a difficult fuel for this process, with high tendency for coke formation. The bio-oil produced from pyrolysis is known to contain high levels of oxygen, up to 40 wt% and the oil is also known to be very unstable and contain high levels of water that are difficult to remove. Development work is underway to be able to upgrade the bio-oil from fast pyrolysis to refinery feed and to process into final fuel components, but the technology has not been implemented in a full commercial setting yet. Overall, the impression is that there might be other emerging technologies that could potentially be competitive to fast pyrolysis for the chosen raw materials. For this reason, it was decided to evaluate the potential improvements with respect to economy, yield and efficiency on implementing other emerging technologies for biomass liquefaction.

2.3 EMERGING TECHNOLOGIES FOR BIO-OIL PRODUCTION

Hydrothermal liquefaction is an alternative to pyrolysis that has regained increased interest the last couple of years, especially for biomass that is very wet or in a slurry, normally with water as solvent. Some calls HTL for pyrolysis in a solvent. The process is normally conducted at a temperature around 350°C and with sufficient pressure for the solvent to remain in liquid phase (i.e. 2-2.5 MPa), close to critical conditions for the solvent (water). There are a large number of process alternatives, using different chemical additions, catalysts, pH etc., resulting in different yields, different amounts of gas and different coke production. Compared to fast pyrolysis, a more viscous, more hydrofobic product is achieved, with lower content of oxygen, lower density and higher energy content. This makes it more probable that the HTL bio-oil has better possibilities for upgrading using similar processes as during upgrading of petrochemical oils (Elliott, 2016).

A version of HTL, characterised by use of a solid catalyst and alkaline process conditions, Base Catalysed Depolymerization, (BCD or also known as CATLIQ) has been originally developed in Denmark and then further in other countries, with the primary application for utilization of agricultural residues. This process has been adopted for kraft lignin as feedstock at Chalmers. Adapting this process to kraft lignin has turned out be a challenge, as the lignin has turned out to have a larger tendency to re-polymerise than other feedstocks, requiring a repolymerization capping agent. The start-up company SunCarbon is also developing its version of HTL process for processing of lignin for transportation fuel production, using the remaining cooking chemicals from the pulp mill as catalysts. SunCarbon’s process is also base-catalysed but uses milder process conditions and no additive to prevent repolymerisation. HTL is in addition also considered for upgrading of forest residues as raw material in this study, based on process development mainly taking place at Pacific North West National Laboratory in the USA.

The approach in the previous f3 project “Biofuels Intermediates” was to assume that the bio-oil was produced in an industrial plant close to a location with available forest resources and industrial know-how, infrastructure and equipment for handling of forest raw materials, while the bio-oil is further upgraded through hydrodeoxygenation and additional treatment at an oil refinery. One reason for placing the upgrading and refining at the refinery is that refineries have existing units for hydrogen production and know-how and infrastructure for handling of this explosive gas. However, an alternative development path would be to also do the hydrodeoxygenation at the same site as the bio-oil production, utilising the hydrocarbon-rich non-condensable off-gases consisting of lighter hydrocarbons for hydrogen production. An advantage with this approach would be reduced cost for transport of the bio-oil as its energy content is increased when the oxygen is eliminated. It also provides an opportunity for the forest based industry to take a step further up the value chain and produce a more valuable product. The idea to hydrodeoxygenate combined with the bio-oil production has been developed further by Gas Technology Institute (GTI) in USA in collaboration with CRI Catalyst Company. The idea is to carry out the hydrogenation during the pyrolysis reaction, so called hydropyrolysis. This can be made under favourable conditions in presence of a catalyst and also other gases than only hydrogen. This have been studied by several researchers, (Yildiz 2016, Guizani 2014, Huiyan et al. 2016). What is still missing is to investigate this technology for valorisation of forest residues and lignin and evaluate it from a system perspective.

2.4 PRODUCTION OF HYDROGEN

Technology for production and storage of hydrogen is expected to make significant advances based on the large interest for using hydrogen as energy carrier for vehicles (e.g. fuel cell cars) and as energy carrier in industrial applications and in society at large. The dominating method to produce hydrogen is through steam reforming of natural gas. This production path has a significant lower cost than producing hydrogen from electrolysis of water (depending on the price of electricity). An example of new technology for production of hydrogen is the technology combining pyrolysis and gasification of biomass developed by Cortus AB. This has the advantage that the hydrogen is produced from the same raw material as the bio-oil and has a low CO_2 footprint. There are more examples of this type of technology for hydrogen production.

Hydrogen from electrolysis could also have a low CO_2 footprint, in case the electricity production is based on renewable power, or like the Nordic electricity mix, has very low levels of electricity production from fossil fuels. Examples of new technology for hydrogen production through electrolysis is developed among others by the Norwegian company NEL in Notodden in Norway. NEL has a new technology where the electrodes are rotated so that the gas bubbles quickly can leave the cathode and anode. This is thought to reduce the size of the electrolyser and save costs. There are more technologies and development aspects related to electrolysis of water and these should be mapped in a separate study.

2.5 PROCESS INTEGRATION AND SYSTEM ASPECTS

One important aspect of the current study is to include the process integration and system aspects in the analysis of the value chains. This type of analysis has also been done by others, e.g. de Jong et al 2015, Tews et al. 2014, Zhu et al. 2011 and 2014 and Elliott et al. 2014. However, in our case we have put the analysis in a context that is relevant for Swedish conditions.

3 METHODOLOGY AND WORKING PROCEDURE

This report provides a techno-economic assessment of different technologies for production of bio-oil and further upgrading to transportation fuels. It also includes an assessment of the impact of different production pathways for hydrogen. The input data used in the calculations have been gathered from available open literature, databases, through contacts with technology developers and technology suppliers and based on discussions with technical experts from Preem.

The activities in the project has followed a step-wise procedure that can be summarized as follows:

Step 1: Screening of emerging technologies for bio-oil production and upgrading based on forest residues respective kraft lignin as feedstock

> The partners have used their own knowledge and experience to identify relevant technologies and screen and rank them based on specified criteria. For results from screening, see *appendix 1: screening of technologies for bio-oil production*. The selected technologies for production of bio-crude are described in Section 4.2. The upgrading technologies are described in section 4.2.6.

Step 2: Screening of alternative technologies for hydrogen production with a low CO_2 footprint

> The same methodology as above has been used for screening hydrogen production options. For results from screening, see *appendix 1: screening of technologies for bio-oil production*. The selected technologies are described in Section 4.3.

Step 3: Definition of studied value chains/cases and system boundaries based on selected bio-oil and hydrogen production technologies for respective feedstock.

> The definition includes identification of relevant physical location of the main equipment either in the pulp mill or at the oil refinery as well as mass- and energy integration aspects of residual process streams and necessary utilities.
>
> The studied value chains are described in Section 5.

Step 4: Definition of key performance indicators used for evaluation and comparison of the studied value chains

> Key performance indicators describing energy efficiencies, yields of products, CO_2 footprint and costs have been defined to be able to analyse and compare the studied value chains. The definitions are found in Sections 6.2, 6.3 and 6.4.

Step 5: Identification and collection of necessary data for the studied value chains to define overall mass- and energy flows and data necessary for economic analysis.

> The data has been collected from data available in literature as well as data received in confidence from technology developers. The data include input data to mass and energy balance calculations such as stream compositions, efficiencies and yields as well as investment costs and costs of consumables. When data are non-existing, own estimations have

been carried out based on own know-how, similarities with other processes and interpolations and extrapolations, always with the goal of ensuring closure of mass- and energy balances.

Data collection for mass- and energy balances has been the most time-consuming activity in the project. The main challenge has been to find relevant performance data for the two feedstocks selected, forest residues and kraft lignin in black liquor. The main part of the experimental evaluations has been made using stem wood as feedstock, and not forest residues. Data related to conversion of lignin is generally lacking or is based on experiments at low technical maturity level. Especially, data on final upgrading of bio-crude to final fuel components using refinery technologies has been lacking, and for bio crude related to lignin, some major analogies and extrapolation of data have been necessary.

Data is available in Section 4 and *appendix 4: detailed input data and assumptions*.

Step 6: Collection of additional data.

Additional input data related to the surrounding system such as energy prices etc. has been collected from official open sources. These data are found in *appendix 4: detailed input data and assumptions.*

Step 7: Calculation of production cost, CO_2 emissions and energy efficiency for the studied value chains.

The production cost based on annuity method, CO_2 emissions and energy efficiency are calculated for the studied value chains. Results are presented in Section 6. RISE in-house data and simulation models for modern kraft pulp mills have been used to estimate integration aspects with the pulp mill. The key performance indicators defined in step 4 are calculated and compared.

Step 8: Analysis of outcome from sensitivity analysis on selected parameters

To improve the overall understanding of the main influencing parameters on the studied value chains and to see the influence of parameters with major uncertainties in their estimation, a sensitivity study has been performed. The results are presented in Section 6.5. The influence of hydrogen production technology is reported in Section 6.6.

The work has to a certain extent been iterative between step 4 and 8.

4 DESCRIPTION OF SELECTED TECHNOLOGIES FOR BIO-OIL AND HYDROGEN PRODUCTION

4.1 SELECTED BIOMASS FEEDSTOCK

The raw material selected as feedstock for the bio-oil value chains are softwood kraft lignin from Scandinavian softwood pulping and forest residues. The reason for selecting these raw materials are that they are available in relatively large quantities in Sweden to a reasonable cost, they are accepted as feedstock for advanced biofuel production according to the EU Indirect Land Use Change (ILUC) directive (even though the list of raw materials is currently being debated). Lignin has the potential to be readily available at the site of the pulp mill if necessary investments in lignin separation equipment are made.

The lignin is separated from the pulp mill black liquor using either the LignoBoost process or using membrane filtration. In the LignoBoost process, the lignin is separated through precipitation in acidic conditions using CO_2 and H_2SO_4 and washing of the lignin (Tomani, 2010). The lignin has a moisture content of approximately 65% and a heating value of 27.7 MJ/kg dry, HHV (Higher Heating Value). The concept using membrane filtration is in this study based on use of the lignin-rich retentate as feedstock to the depolymerization step, similar to the approach described in Arkell et al. (2014). The retentate has a moisture content of approximately 60% and heating value of 17.4 MJ/kg dry, HHV when entering the process (low due to the high content of inorganics).

The forest residues are branches and tops from typical Scandinavian forestry. The residues have an average moisture content of 50%. Typical heating value is 19.4 MJ/kg, dry, LHV (Lower Heating Value) or 20.7 MJ/kg, HHV and the composition is given in *appendix 4: detailed input data and assumptions.*

4.2 TECHNOLOGIES FOR BIO-OIL PRODUCTION

Emerging technologies for production of bio-oil has been identified by the project participants. About 20 different technologies that are based on different supplier data were originally identified, see *appendix 1: screening of technologies for bio-oil production.* Four emerging technologies were selected for further investigations based on some main criteria such as access to process data, expected oil oxygen-content, process layout known, expected yield of product, information of investment cost is available and the largest scale of demonstration. The highest ranked criterion was for practical reasons the access to process data. These technologies are all at a relatively early stage of development, where tests have been carried out in small scale laboratory pilots to larger industrial pilots, however not yet in demonstration plants scale.

The selected technologies for forest residues are:

- Hydropyrolysis (data mainly based on published literature from the IH2 process by GTI and CRI)
- Hydrothermal liquefaction (data based on research and evaluations from PNNL, Pacific North West National Laboratories)

For lignin, two different approaches to hydrothermal liquefaction, HTL, were selected

- Hydrothermal liquefaction of membrane separated kraft lignin (based on data from SunCarbon, HTL process at milder process conditions)
- Base catalysed depolymerization of kraft lignin from the LignoBoost process (based on Chalmers research and published data, HTL at more severe process conditions)

Short descriptions of the technologies are available in the following sections. More detailed description with assumptions is available in *Appendix 4: Detailed Input data and assumptions*. These technologies were compared to fast pyrolysis of forest residues, which is considered as more mature technology with several near commercial plants in operation.

The following acronyms have been used in the report when describing the technologies

Pyr-FR - Fast pyrolysis of forest residues,

HydroPyr-FR - Hydropyrolysis of forest residues,

HTL-FR - Hydrothermal liquefaction of forest residues,

HTL-MSL - Hydrothermal liquefaction of membrane separated lignin,

HTL-LBL - Base catalysed depolymerization of kraft lignin from the LignoBoost process

Even though specific technologies and technology suppliers are mentioned in this text we would like to point out that the processes described here are based on technology that is still under development, and that the process could still be modified from what are describe here, and that performance data will change over time as the processes are further developed.

4.2.1 Fast pyrolysis of forest residues, Pyr-FR (reference)

Pyrolysis is a collection of thermochemical processes in which organic compounds are broken down into smaller one’s due to heating to high temperatures in the absence of oxygen. Pyrolysis of biomass results in the products char and vapour, where part of the vapour condenses to liquids, the rest are non-condensable gases, NCG such as lighter hydrocarbons and carbon dioxide. The distribution of char, NCG and liquor depends on the temperature and time in the pyrolysis. Fast pyrolysis is to give high yields of liquor and is completed in seconds. It requires very high heating and heat transfer rates which requires the biomass to be milled to mm particles (IEA Bioenergy 2007). There are several types of pyrolysis reactors and several manufacturers and technology suppliers. This study is based on a reactor with rotating cone developed and sold by BTG-BTL (Benjaminsson et al. 2013).

In the BTG-BTL pyrolysis plant, the biomass is ground to a particle size of max 6 mm before the biomass goes into a belt dryer for drying. From the dryer, biomass enters the pyrolysis reactor of the rotating cone where sand is used as heat carrier. The biomass is disintegrated and forms vapour and solids. The solids or char formed goes with the sand and are burned in a char burner. The vapour from the pyrolysis reactor are purified from larger particles in two cyclones before subjected to condensation. The NCG go on to a burner for steam production. The condensed vapour, the liquid which is the desired product pyrolysis oil, is purified by a filter before storage and transport for further upgrading.

Forest residues contains higher amount of ash compared to for example wood chips or saw mill dust where ash has shown to have a negative effect on the pyrolysis oil yield, to the favour of more char and NCG. Hence, since this study uses forest residue as feedstock a lower pyrolysis oil yield is to expect. From (Benjaminsson et al 2013) energy yields of 54% from forest residues have been used.

A block flow diagram of the fast pyrolysis plant is shown in Figure 2.

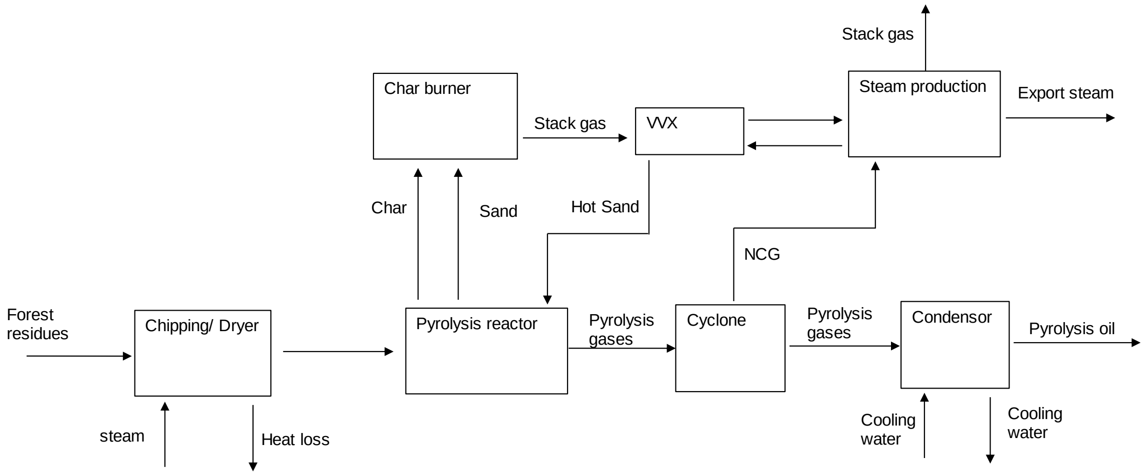


Figure 2. Illustration of the fast pyrolysis process by BTG-BTL (Benjaminsson et al 2013).

A more detailed description of the process and the assumptions is found in *appendix 4: detailed input data and assumptions.*

4.2.2 Hydropyrolysis of forest residues, HydroPyr-FR

The hydropyrolysis of forest residues based on the IH2 (Integrated Hydropyrolysis and Hydroconversion) process seen in Figure 3 was first developed by the Gas Technology Institute (GTI) in 2009 and is now being further developed together with CRI, Shell's catalyst business. The IH2 technology is a so-called hydropyrolysis process meaning that biomass is converted to liquid, gas and char in the presence of hydrogen. In the first stage, the hydropyrolysis stage, the biomass is fed into a pressurized fluid-bed pyrolysis reactor (14 bar, 340-470 ˚C) together with hydrogen. The vapours leaving the pyrolysis reactor enters a hydroconversion reactor (23 bar, 370-400 ˚C) and the char is extracted (Gas Technology Institute (GTI) 2012). In the hydroconversion reactor, oxygen is removed to produce deoxygenated diesel and gasoline as well as C1-C3 gases. The C1-C3 gases are directed to an integrated steam reformer in which the hydrogen is produced. The IH2 process is said to be able to be self-sufficient in hydrogen due to the steam reformer (Gas Technology Institute (GTI) 2012). Proprietary catalysts are used in both the hydropyrolysis and hydroconversion reactors. Both the hydropyrolysis and the hydroconversion processes are exothermal and produces excess heat that is used for steam production. The energy yield of the hydropyrolysis process from forest residue raw material to product from the hydroconversion step is about 60%, which should be considered as high, since no external H_2 has been added and the oxygen content is close to zero.

A more detailed description of the process and the assumptions is found in *appendix 4: detailed input data and assumptions.*

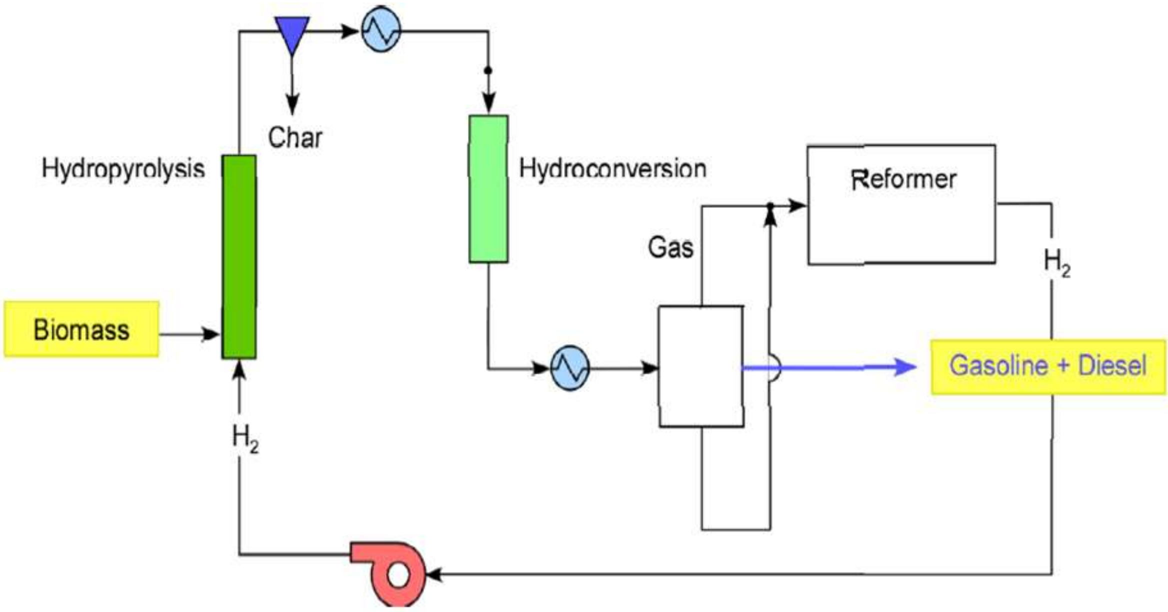


Figure 3. Overall process flow of the hydropyrolysis process based on initial design of the IH_2 process, HydroPyr-FR, (Gas Technology Institute (GTI), 2012).

4.2.3 Hydrothermal liquefaction of forest residues, HTL-FR

One of the more basic forms of HTL is to create a slurry of biomass and water and then pump it up and heat it to 300-400 C/20-25 MPa, which results in obtaining a gas phase, often two liquid phases and some solids/ashes. The variation included here is based on investigations and reports from VTT (Finland) and PNNL (USA) presented in Zhu (2014) and then further processed by de Jong et al. 2017.

In the Zhu paper (which is the basic reference for the evaluation in this report), two cases of HTL on forest residues are considered, one “State of Technology” (SOT) and one “goal case” including possible future improvements that are still not experimentally proven. The presentation here follows mainly the latter one, and this is the case used in the evaluations in this study. The process consists of 4 main process blocks as described below.

1. Biomass preparation

Finely ground wood biomass (pine forest residues including bark in the experiments) with a moisture content of about 50% is mixed with recycled water to obtain a 15% dry biomass content. No other chemicals are assumed to be added, but in the experiments, alkali in the form of Na_2CO_3 was added in an alkali to dry wood ratio of 1/8 as a buffering agent and to avoid formation of high-molecular weight compounds.

2. HTL reactor

The slurry from the preparation step is pumped to 20.4 MPa and by heat exchange with reactor outlet heated to 327 °C and then further heated to 355 °C which is the reaction temperature. The reactor is assumed to be of plug flow type with a shell-and-tube design. A fired heater is used to provide the heat to a heat transfer fluid, which then is used to heat the reactor. The hot effluent from the reactor passes a filter to separate fine particles from the stream and is then cooled by the internal heat exchange to 148°C. The cooled stream is depressurized to 0.2 MPa and separated into gas (about 17% yield), aqueous and oil phases. The gaseous product is assumed to be used to produce hydrogen by steam reforming in a hydrogen plant in the paper but in this study, it is used for electricity production integrated in the pulp mill. About 20% of the aqueous phase is purged and the remainder is recycled back to the biomass feed slurry. The purged aqueous phase is cooled and sent to a wastewater treatment process. Anaerobic digestion is assumed to be used to convert this stream of organics to a gas rich in CH_4 and CO_2. A portion of this off-gas is sent to the pulp mill recovery boiler for heat recovery in our case, however in Zhu (2014) it is sent to hydrogen production. The oil phase is sent to the downstream upgrading process. An energy yield of approximately 78% is achieved and a bio-oil oxygen content of approximately 15 wt%.

3. Upgrading

Both hydrotreating and hydrocracking are assumed to be used for the bio-oil upgrading. The purpose of the hydrocracking is to increase the yield of fuel products rich in hydrogen (i.e. diesel) from heavy hydrocarbons. The product stream is cooled and separated to form condensate and gas phases. This upgrading is in this study assumed to take place at the refinery.

4. Hydrogen plant

A conventional steam reforming process is assumed to be used in the hydrogen plant in this study.

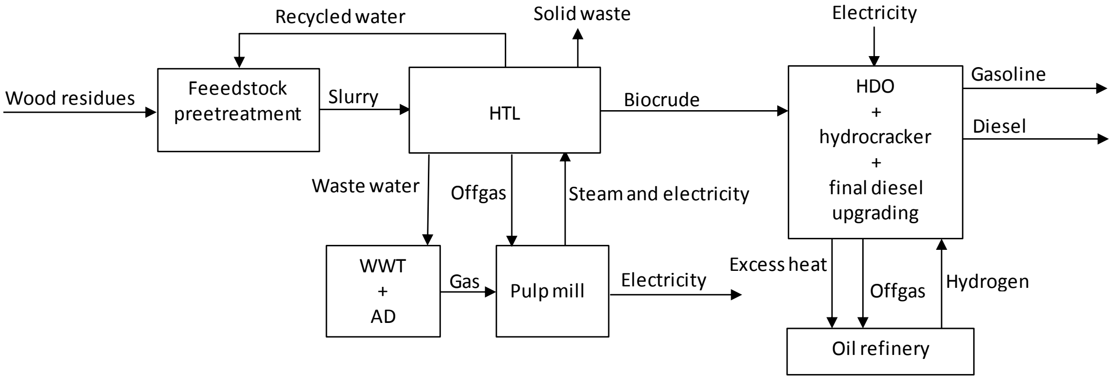


Figure 4. Process configuration for the HTL process using forest residues as feedstock based on PNNL's concept and a case with production separated between pulp mill and oil refinery as in this study, HTL-FR.

A more detailed description of the process and the assumptions is found in *appendix 4: detailed input data and assumptions.*

4.2.4 Hydrothermal liquefaction of membrane separated lignin, HTL-MSL

The HTL process using membrane separated lignin as feedstock is meant to be integrated with a pulp mill. Weak black liquor which is first filtrated is fed to the process, treated hydrothermally and purified into lignin oil that is sent to a petroleum refinery for final hydrogenation (SunCarbon, 2016). A schematic drawing of the process is seen in Figure 5.

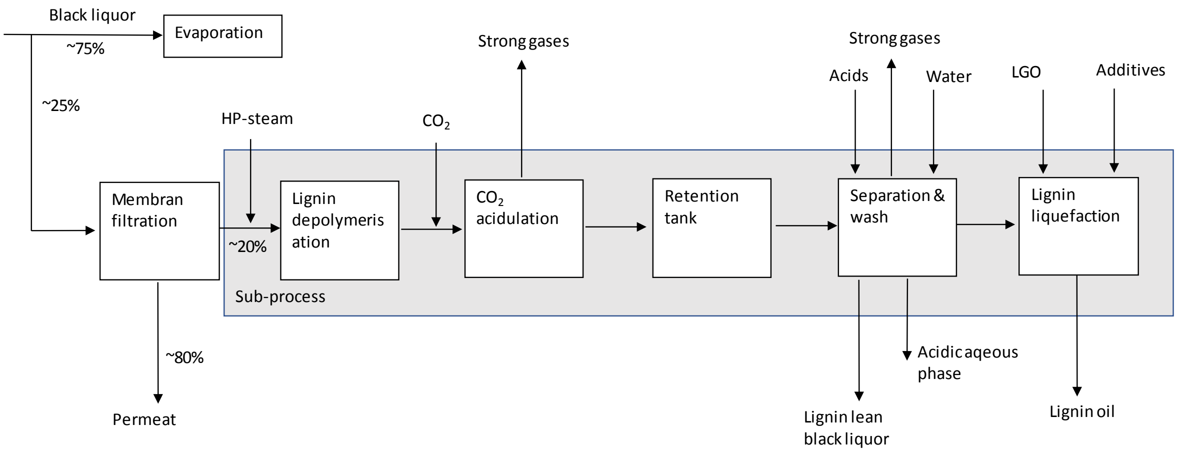


Figure 5. A simplified flow chart of the case lignin HTL-MSL depicting HTL process using membrane separated kraft as feedstock, based on initial design by SunCarbon.

In the process, around 25 wt% of the mill’s weak black liquor, prior to the evaporation units, is extracted to a membrane filter. The permeate from the membrane is said to be able to be recycled to the green liquor preparation stage of the mill, However, in this study it has been taken to the evaporation plant. The lignin rich retentate is mixed with medium pressure steam (23-56 bar) from the mill and fed into a thermal catalytic cracker at sub-critical conditions for depolymerization. A catalyst internal to the pulp and paper mill is used in the catalytic cracker. The hydrothermal treated lignin mixture is then mixed with low quality carbon dioxide to lower the pH. After spending some time in a retention tank, the lignin mixture is subjected to an acid wash (H_2SO_4) which separates it from ash, residues, lean black liquors and strong gases (mainly H_2S). The removed lean black liquor contains around 30% of the energy content of the initial lignin entering the process resulting in an energy yield of 70% of the lignin oil, which has an oxygen content of approximately 20 wt%. Lastly the remaining lignin is suspended in gas oil or tall oil pitch assisted by additives. After this step the resulting product is a lignin oil ready to transport to a petroleum refinery for hydrogenation.

Depending on pulp mill type, the integration of the HTL process on membrane separated kraft lignin will render different results. If the mill’s recovery boiler is a bottle neck the extraction of black liquor can increase the pulp production, in a similar fashion as a LignoBoost process. If the mill’s operation is already in balance with respect to Na/S before the HTL and membrane separation process is installed, the additional sulphur might upset the balance and Na_2S will have to be bled out and compensated by virgin NaOH makeup. There is also an uncertainty in the estimation of how much of the alkali is lost with the retentate from the membrane separation, which in turn is connected to the need of NaOH make-up chemicals in the pulping process. The alkali metals also have a negative impact on the expected catalyst life length, if the alkali is not removed to sufficient extent before it is transferred to the downstream refinery and upgrading process steps. In case larger amounts of lignin is removed than the mills energy excess, additional biomass to compensate for the loss of energy utilised internally in the mill will be needed.

A more detailed description of the process and the assumptions is found in *appendix 4: detailed input data and assumptions.*

4.2.5 Base catalysed depolymerization of kraft lignin from the LignoBoost process, HTL-LBL

Base catalysed depolymerization is a variation of Hydrothermal Liquefaction (HTL) characterized by adding bases, such as K_2CO_3 and KOH, to the feed to act as catalyst and to ensure that the process is carried out under basic conditions, which for several different feeds has been found to improve the bio-oil yield. To improve the catalytic strength, it has been suggested to use a solid co-catalyst. Zirconia (ZrO_2) has been suggested as a suitable co-catalyst for this, and this was the basis for the so called CATLIQ technology. This technology has at a development work at Chalmers been adopted to be used to depolymerize kraft lignin from the LignoBoost process and this adopted process will here be called HTL-LBL. A simplified flow-chart for the reactor and recycle part of this process is shown in Figure 6.

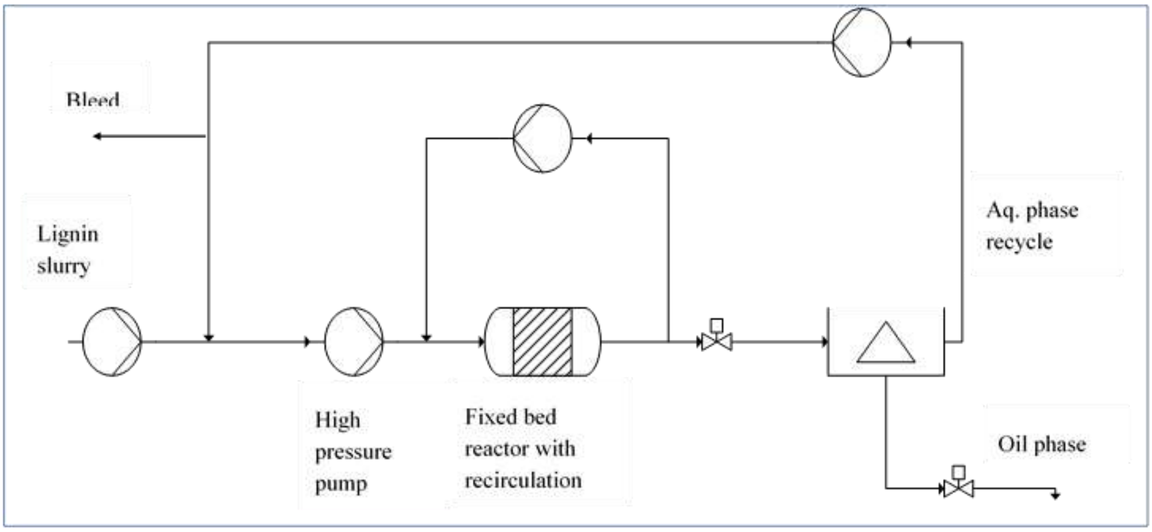


Figure 6. A simplified flow chart depicting the reactor and recycle part for the HTL-LBL process.

A special challenge when depolymerizing lignin is its tendency to repolymerize to unwanted solid products. One way to partly suppress this is to add a so called capping agent. So far, phenol and methanol have been tested at Chalmers and phenol was found to be the more effective of those.

The calculations presented in this report are based on experimental results from Chalmers lab pilot unit using phenol as capping agent.

The process can be described as follows:

1. Feed preparation

LignoBoost separated lignin is mixed with alkali (bases), phenol and water. Alkali, phenol and water is partly supplied by recycling the product aqueous phase (see below). A typical mix is 5,5% (dry) lignin, 4% phenol (a compromise between cost and oil viscosity), 1,8% K_2CO_3 and 0,2% KOH.

2. Heating and reaction

The feed stream is first pressurized with a pump to 25 MPa and heated to maximum 80 °C before it is mixed with a stream recycled from the reactor outlet, in order to rapidly heat the feed to almost reaction temperature. The reaction temperature is typically 350 °C and the pressure 25 MPa. The reactor is of tube reactor type, with the tubes filed with catalyst pellets. A dual reactor system is probably needed, with one reactor in operation while the other is regenerated by burning off coke formed. The part of the reactor outlet flow that is not recycled is depressurized and cooled, which results in two liquid phases being formed, one oil and one aqueous. After separation, the oil phase is filtered to remove insoluble parts and is then ready for further processing to transportation fuels. The yield from lignin to bio-oil is approximately 76% on energy basis is achieved for a bio-oil with an oxygen content of about 20%.

The main part of the aqueous phase is recycled and used for feed preparation. A minor part (a so-called bleed stream necessary to avoid build-up of some components) can, if integrated with a pulp mill, be handled by the chemical recovery cycle, to recover alkali and energy and avoid the need for wastewater handling.

A more detailed description of the process and the assumptions is found in *appendix 4: detailed input data and assumptions.*

4.2.6 Upgrading to final diesel and gasoline fuel components

The bio-oils produced in the different value chains using forest residues or kraft lignin as feedstock and different technologies for conversion to bio-oil are upgraded to meet final transportation fuel product specifications and standards for diesel and gasoline using typical refinery upgrading processes. The need of processing of the bio-oil is based on the structure and composition of the bio-oil. Bio-oils containing oxygen are hydrodeoxygenated, normally in two steps, to reach close to zero oxygen content using hydrogen as reactant supported by catalyst. Oils with lower oxygen content and high stability might require only one-stage hydrodeoxygenation. Bio-oil hydrotreating means processing the bio-oil with a large excess of hydrogen and produces a gas and two liquid fractions. The compounds in the gas product are excess hydrogen, light hydrocarbons and carbon dioxide. The excess hydrogen is assumed to be recovered by pressure swing absorption and recycled. The liquid products consist of hydrocarbon oil and an aqueous phase, which separate easily. The aqueous phase carbon content depends upon the degree of bio-oil deoxygenation. The hydrocarbon oil yield and quality are dependent on catalyst selection, reactor configuration and hydrotreating conditions. Fixed bed reactors similar to the ones used in conventional hydroprocessing of petroleum to final fuels can be used. Chemical reactions during bio-oil hydrotreating are very complex. Overall, the bio-oil is almost completely deoxygenated by a combination of hydrodeoxygenation and decarboxylation, with oxygen removed in the form of water and carbon oxides. A good combination of hydrotreating catalysts and optimum operating conditions can help control reaction selectivity and minimize hydrogen consumption.

An example of a process flow diagram for a 2 stage HDO process is found in Figure 7. The bio-oil is first pre-treated in a stabilization bed under relatively mild process conditions, 140 to 180 °C) and 1200 psia (83 bar), followed by processing under more severe hydrotreating conditions in the 1st and 2nd stage hydrotreating reactors (Jones 2013). The 1st stage hydrotreating reactor is designed as a single bed catalytic reactor operated at 180 to 250 °C and 2000 psia (138 bar). The 2nd stage hydrotreating reactor is operated at a higher temperature of 350 to 425 °C.

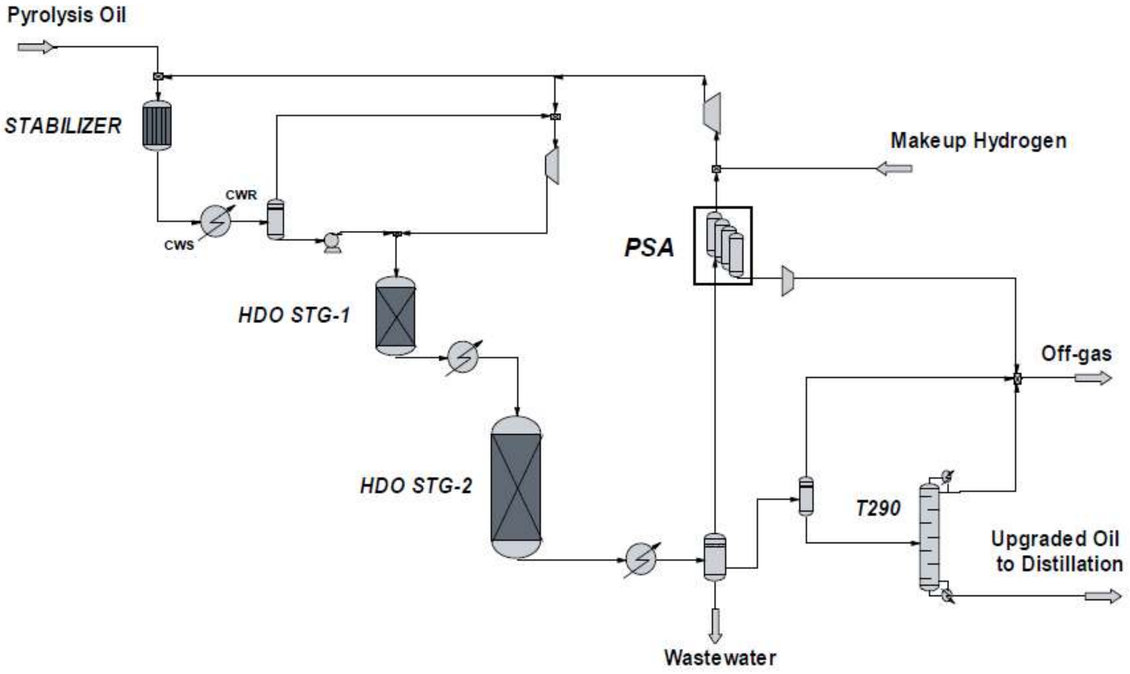


Figure 7. Bio-oil upgrading by HDO, (Jones et al., 2013). Example with pyrolysis oil.

The product is distilled in a gasoline fraction, a diesel fraction and also a heavy fraction. The heavy fraction is further cracked to a diesel fraction, utilising the existing refinery cracker. The goal of the cracking process is to reach hydrocarbon chain-lengths in the diesel range, e.g. about C18, under addition of hydrogen.

In case the H/C ratio of the diesel fraction is below 2, due to high content of aromatic compounds and unsaturated hydrocarbons, the diesel fraction is assumed to be further treated with hydrogen in a SynSat plant at the refinery to reach the targeted H/C ratio 2, which is common for diesel and gasoline. The SynSat process has been developed by Criterion/Lummus and employs several different catalyst beds within a single reactor shell with intermediate by-product gas removal and uses both co-current and counter-current gas/liquid flow contacting, see
Figure 8.

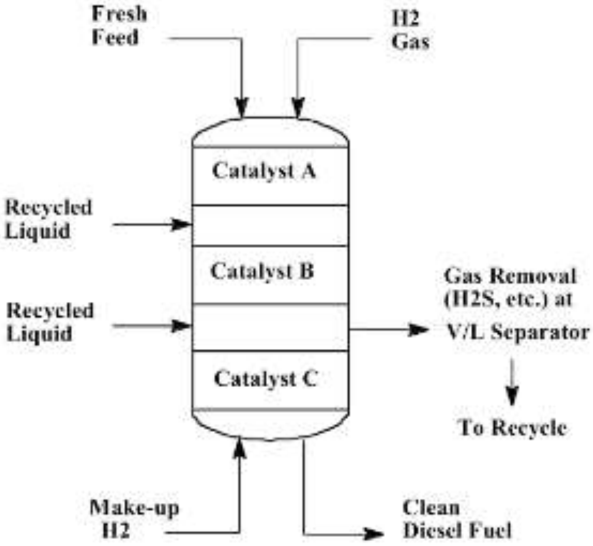


Figure 8. SynSat process reactor configuration (Hsu and Robinsson 2006).

Bed A and B are used for deep sulphur removal before the nobel catalyst bed C used for aromatic reduction. The typical pressures and temperatures in a SynSat reactor are from 315-400 °C and 30-61 atm, depending on feedstock and required product properties.

The gasoline fraction is assumed to be left as it is and mixed in with the gasoline pool. Detailed descriptions of the assumptions related to the upgrading processes is found in *appendix 4: detailed input data and assumptions.*

4.3 TECHNOLOGIES FOR HYDROGEN PRODUCTION

Different technologies for hydrogen production with low fossil CO_2 footprint as an alternative to the conventional steam reforming of natural gas are screened in this study. The objective is to improve the environmental performance while investigating the impact on the overall cost of producing transportation fuel components. In total, four technologies for hydrogen production from biomass and four technologies based on electrolysis of water were reviewed, see further information in *appendix 2: screening of alternative technologies for hydrogen* production. The main screening criteria were access to process data, commercial supplier, process layout known, yield of hydrogen, and information about size of investment.

The alternative hydrogen production technologies selected to be included in the further work in the study were:

- Atmospheric Alkaline electrolysis of water (supplier data from NEL, http://nelhydrogen.com/)
- Gasification of forest residues (supplier data from Cortus, http://www.cortus.se/)

These have been compared to steam reforming of natural gas. Process descriptions are given in the following Sections.

4.3.1 Steam reforming of natural gas (reference)

Steam reforming of natural gas is a conventional method for hydrogen generation used within the oil refining industry, Figure 9. When located by a refinery, refinery off-gases as well as off-gases from the hydrotreating and hydrocracking are used together with natural gas as reformer feed or as fuel for the reformer burners. After hydrodesulfurized (HDS) the feed is mixed with superheated steam and sent to the steam reformer to produce syngas (Jones et. al. 2013). The syngas hydrogen content is increased by high temperature shift (HTS). After condensing out the water, the hydrogen is purified by pressure swing adsorption (PSA). Off-gas from the PSA is recycled as fuel for the reformer burners. The produced hydrogen is at lower pressure than required by hydrogenation and hydrocracking and will therefore be compressed.

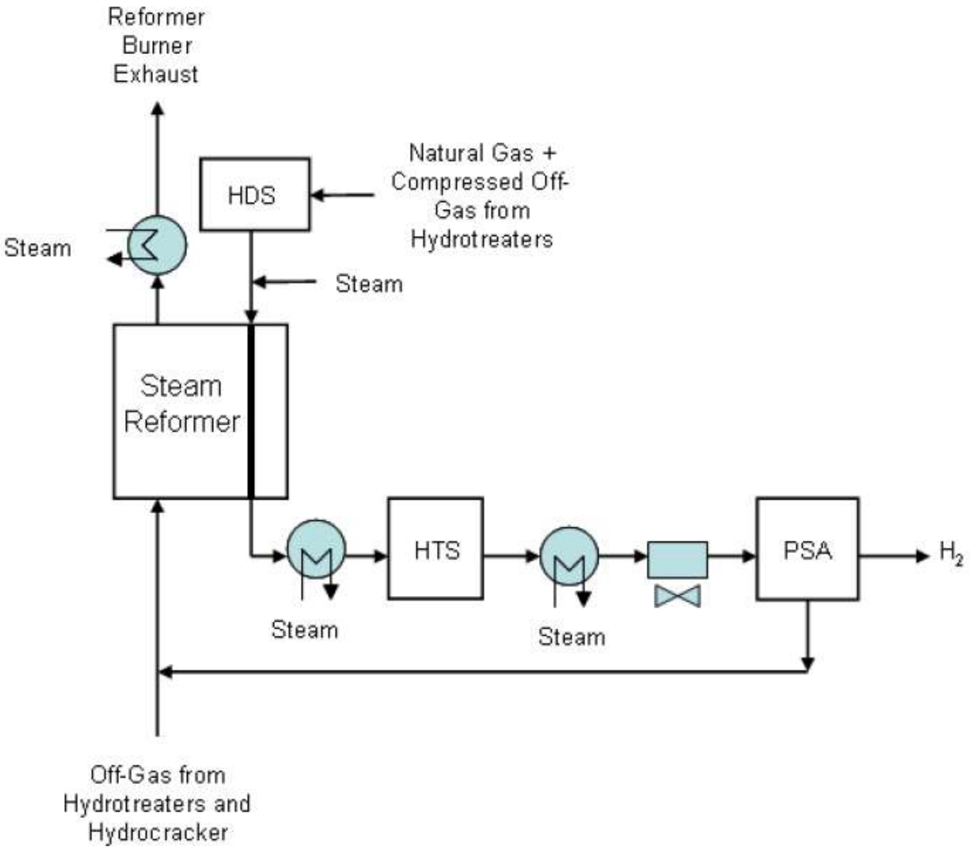


Figure 9. Steam reforming of natural gas (Jones et al. 2009)

4.3.2 Atmospheric alkaline electrolysis of water for hydrogen production

The alkaline electrolyser is a proven, robust and efficient hydrogen generator, Figure 10 and Figure 11. There is no obvious preference for a refinery or for a mill location, however by-product oxygen may possibly be used by the mill for e.g. pulp bleaching and combustion air oxygen enrichment albeit, this has not been studied in this work. The specific power requirement for electrolysis is 4.60 kW/Nm^3 H_2. The electrolyser need a supply of pure water below 5 µSwhich is provided by a reverse osmosis unit. Water requirement is approximately 1 litre per Nm^3 H_2. The plant is highly automated requiring one operator to take a routine inspection of the plant once per shift. The produced hydrogen is at atmospheric pressure and needs to be compressed to meet the pressure requirement of hydrogenation and hydrocracking. The power for final H_2 compression corresponds to 5.7% of the power required for electrolysis. Impurities in the hydrogen are on a ppm level and consist of traces of oxygen and water without any impact on hydrogenation or hydrocracking. A performance summary is provided in *appendix 5: selected hydrogen technologies*.

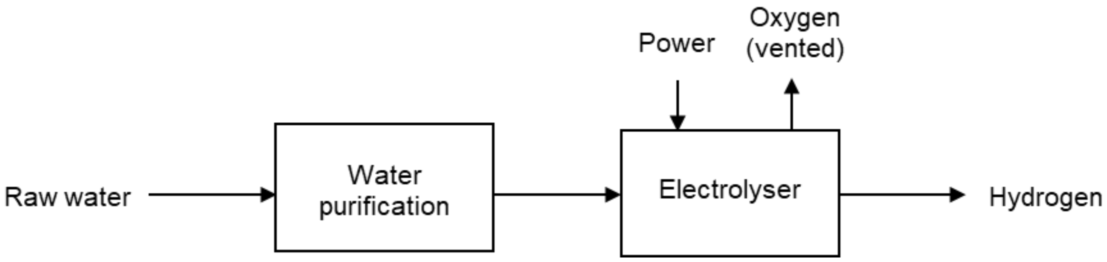


Figure 10 Block flow diagram of atmospheric alkaline electrolysis of water.

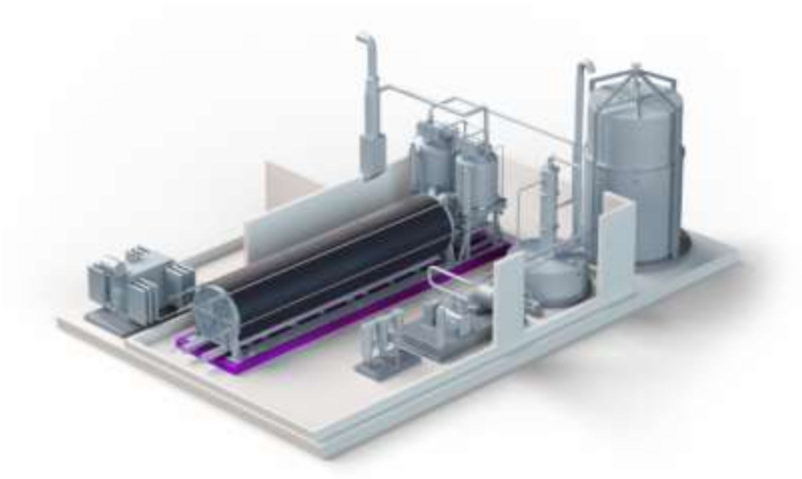


Figure 11. Electrolyser unit for H_2 production (Courtesy NEL Hydogen AS). Capacity 48 kg H_2/h (2 MW).

4.3.3 Gasification of biomass residues for hydrogen production

The motivation for selecting gasification as a separate unit for hydrogen generation, rather than having an integrated biomass depolymerisation and hydrogen production plant is that the gasification technology package is already available from commercial suppliers. The hydrogen production from depolymerisation and hydrodeoxygenation off-gases is on the other hand only available on a conceptual level, and cost estimates are uncertain. Integration of hydrogen production from off-gases also decreases the operational flexibility as off-gas production and bio-oil production ideally should be matched.

This hydrogen production by gasification plant is fit to be located by the mill because of local biomass availability. Hydrogen generation by biomass gasification, Figure 12, is made efficient in a process beginning with drying of chipped forest residues in a dryer, followed by pyrolysis in a rotating drum pyrolyser. The produced char is reacted with steam in a gasifier reactor resulting in a hydrogen rich syngas (60% H_2). The pyrolysis gas emanating from the pyrolyser fires the gasifier which has an endothermic reaction. The hot flue gas from the gasifier heats the pyrolyser by indirect heating. The flue gas from pyrolyser is finally used for chip drying by indirect heating before being vented. The process is not pressurized.

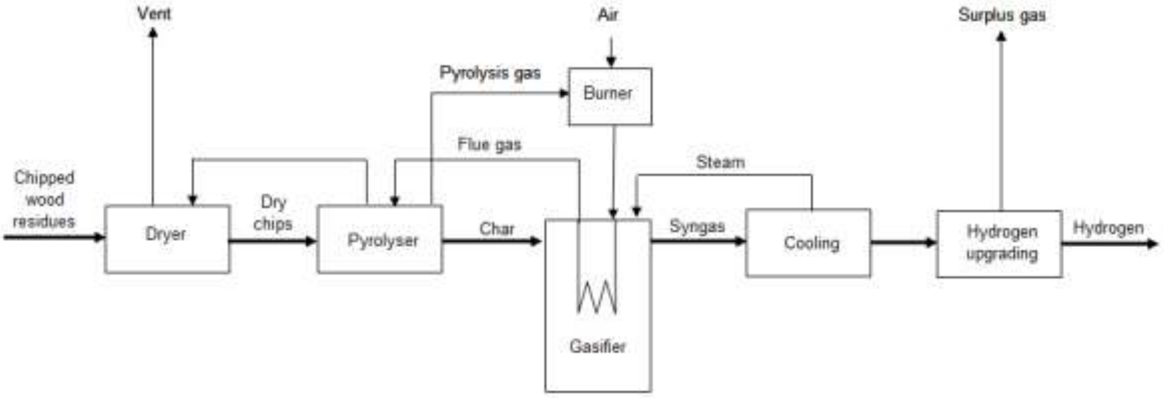


Figure 12. Gasification of forest residues for hydrogen production.

The hydrogen rich syngas is upgraded to pure hydrogen in a process comprising of a first Pressure Swing Absorption (PSA) unit. The first PSA retentate is fed to a water gas shift (WGS) followed by a second PSA. The second PSA retentate is sent for combustion in boilers at the mill. The first and second PSA permeate make up the stream of pure hydrogen. The hydrogen is finally compressed to higher pressure to meet the requirements of hydrogenation and hydrocracking. Hydrogen production is about 90 kg H_2/ton dry ash free biomass. Impurities in the hydrogen consist of traces of hydrocarbons and carbon oxides which has no impact on hydrogenation and hydrocracking. A performance summary is provided in *appendix 5: selected hydrogen technologies*. A 3D model of Cortus' Woodroll® gasification unit is found in Figure 13.

Figure 13. Woodroll® biomass gasification unit (Courtesy Cortus Energy AB). Capacity 131 kg H_2/h (6 MW).

It should be noted that bio transportation fuels also can be produced through gasification of wood followed by either synthesis of DME, methanol or FT-liquids and further processing thereof.

4.4 THE PULP MILL

The pulp mill used for evaluation in this study is a hypothetical softwood reference market pulp mill using the kraft process, using the existing, best available, commercially proven technology as of year 2010. The mill has low usage of wood, chemicals, energy and water. It is self-sufficient in steam by burning the black liquor in the recovery boiler. Therefore, the falling bark from the incoming biomass are used as fuel in the lime kiln and power boiler or could be sold. The plant has a large steam surplus which is used for power generation in a condensing turbine. For this case, the raw material used is softwood spruce and the production are set to 2000 ADt/day or ~700 000 ADt/year. This is about the size of the largest pulp mills in operation in Scandinavia today.

The power consumption for the mill is set to 726 kWh/ADt and 984 kWh is sold per ADt produced. This corresponds to 82 MW electricity exported to the grid. The power balance is based on the case where all bark is burnt in the power boiler. This surplus energy can instead by extracted in the form of separated lignin, as in the lignin cases presented, resulting in a reduction in the power export.

4.5 THE OIL REFINERY

The refinery has not been modelled in the work carried out in this report. Existing equipment for SynSat treatment and cracking is assumed to be available for the bio-oil upgrading, resulting in equivalent reduced production capacity of fossil oils on energy basis. Existing natural gas steam reforming equipment is assumed to be used for hydrogen production.

5 THE STUDIED VALUE CHAINS

Five different value chains for production of transportation fuel components has been suggested by the project team based on the feedstocks and technology options identified in Section 4. A summary of the basic structure and technologies included in the value chains are found in Table 1.

Table 1. Description of the baseline value chains studied in the project.

Value chain	Pyr-FR	HydroPyr-FR	HTL-FR	HTL-MSL	HTL-LBL
Feedstock	Forest residues	Forest residues	Forest residues	Kraft lignin retentate from membrane separation of black liquor	Kraft lignin separated from black liquor through the LignoBoost process
Depolymerisation to bio-crude	Fast pyrolysis	Hydropyrolysis	Hydrothermal liquefaction	Hydrothermal liquefaction (low T, p)	Hydrothermal liquefaction (high T, p)
Bio-crude upgrading to fuel components	HDO and SynSat of diesel fraction	Integrated catalytic upgrading, and SynSat of diesel fraction	HDO, cracking and SynSat of heavy components	HDO, cracking of heavy components	HDO, cracking of heavy components
Hydrogen production	Steam reforming of natural gas	Steam reforming of natural gas	Steam reforming of natural gas	Steam reforming of natural gas	Steam reforming of natural gas
Location of depolymerization, and number of sites	Pulp mill, 3 sites	Pulp mill, 1 site	Pulp mill, 1 site	Pulp mill, 3 sites	Pulp mill, 3 sites
Location of biocrude upgrading, and number of sites	Oil refinery, 1 site	Pulp mill, 1 site: catalytic upgrading Oil refinery, 1 site: SynSat of diesel fraction	Oil refinery, 1 site	Oil refinery, 1 site	Oil refinery, 1 site

In the HTL-MSL and HTL-LBL cases, three separate HTL depolymerisation reactors located at three separate mills are considered. This is based on the assumption that the lignin supply is limited by the lignin that can be separated from the pulp mill without impacting the recovery boiler performance to a too large extent. We have in this study assumed that about 18-20% of the lignin in the pulp mill’s black liquor is separated for the 100 000 t/y transportation fuel production.

The Pyr-FR case is also based on an assumption of three separate locations with one pyrolysis plant to fulfil the 100 000 t/y production. In this case it is due to the impact of the energy integration with the pulp mill, where the existing steam turbine is limited in additional steam that it can receive from the energy recovery from the pyrolysis plant. In addition, this also resulted in reasonable size of the pyrolysis plant of approximately 110 MW_{th}.

For the HydroPyr-FR and HTL-FR cases, a single plant located at one pulp mill site has been assumed, since these two technologies do not integrate with the pulp mill to the same extent as the fast pyrolysis plant, and therefore are not restricted in the same way by the capacity of the existing pulp mill steam turbine.

5.1 INTEGRATION OF VALUE CHAINS WITH EXISTING INDUSTRIES

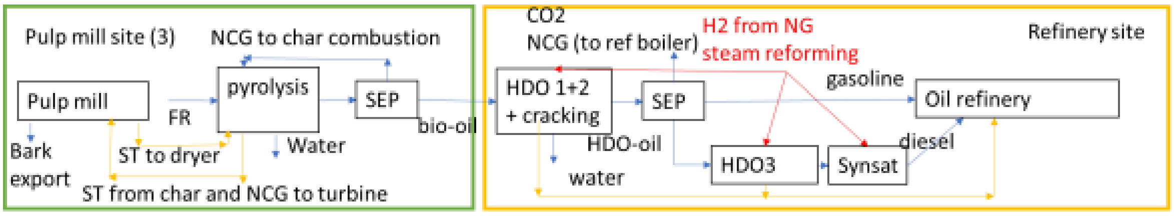
One of the study objectives is to investigate the influence of integrating the value chains in the existing industry, i.e. pulp mill and oil refinery in this study. The following general integration options were identified at an early stage of the work, see Table 2.

Table 2. Integration option included in the value chains.

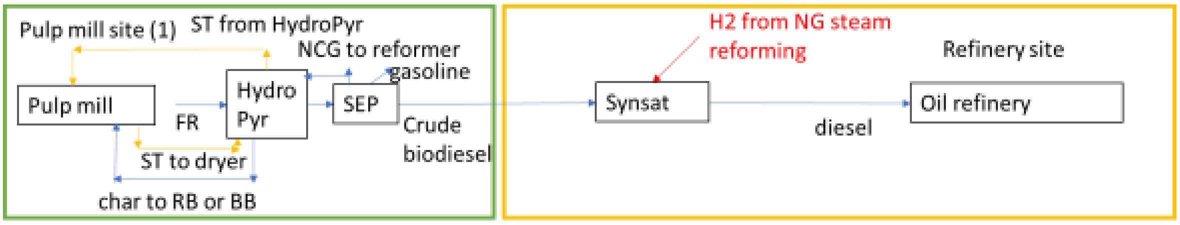
Type of stream	Pulp mill integration	Oil refinery
Energy-containing non-condensable gases from depolymerisation and upgrading, and PSA reject from hydrogen production	Combustion in recovery boiler to produce additional electricity Case: HTL-FR	Replaces fuel oil in refinery furnaces in a 1.1 ratio Case: Pyr-FR, HTL-FR, HTL-MSL, HTL-LBL
Excess heat from reactions in depolymerisation, HDO, cracking and SynSat (SynSat is only relevant for refinery integration)	Steam generation and final power generation in the steam turbine Case: Pyr-FR, HydroPyr-FR, HTL-LBL	Heat is used for refinery heating purposes and replaces steam generated through combustion of heating oil in a 0.8:1 ratio Case: All cases except HydroPyr-FR
Char, biogas and bleed stream from depolymerisation	Combusted in bark boiler or recovery boiler and results in additional electricity generation Case: HydroPyr-FR, HTL-FR, HTL-LBL	-
Black liquor permeate, acid, lean liquor from black liquor separation and lignin depolymerisate purification and separation, (potentially also waste water from depolymerisation)	To black liquor evaporation and recovery boiler for additional electricity generation from organic content and recovery of alkali Case: HTL-MSL, HTL-LBL	-
Steam from steam turbine	Drying of forest residues, heat to depolymerisation, results in loss of power production Case: Pyr-FR, HydroPyr-FR, HTL-FR, HTL-MSL,	-
Separated kraft lignin from black liquor	Loss of energy to recovery boiler and power production, supply of alkali Case: HTL-MSL, HTL-LBL	-
Bark export	Reduction of the bark that previously was combusted in the bark boiler for steam generation to ensure that utilisation of steam from the process heat recovery system and additional steam generated through combustion of by-products for power generation purposes is not limited by the steam turbine capacity. Case: Pyr_FR, HTL-FR	-
Diesel stream requiring further cracking and hydrogenation to meet the diesel fuel component specification	-	The stream is integrated in the existing cracking and SynSat equipment and reduces the equivalent amount of fossil diesel production Case: all

The integration options considered for the different specific value chains are illustrated in Figure 14. Equipment placed within the green system boundary are placed on the pulp mill site(s), while the equipment within the yellow boundary are placed on the refinery site. Blue arrows represent material flow, yellow steam and red hydrogen. The arrow that crosses the interface between the pulp mill and the refinery represents the intermediate product sent to the refinery.

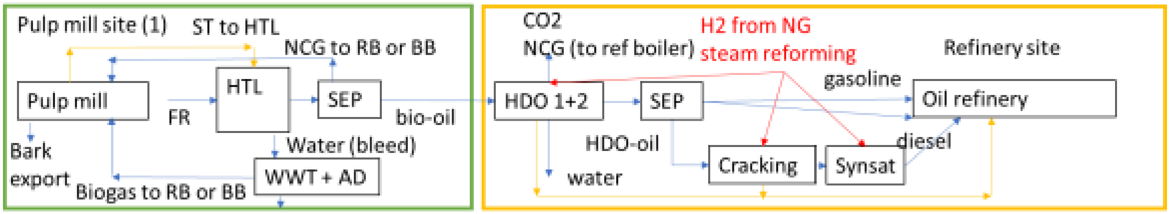
a) Pyr-FR



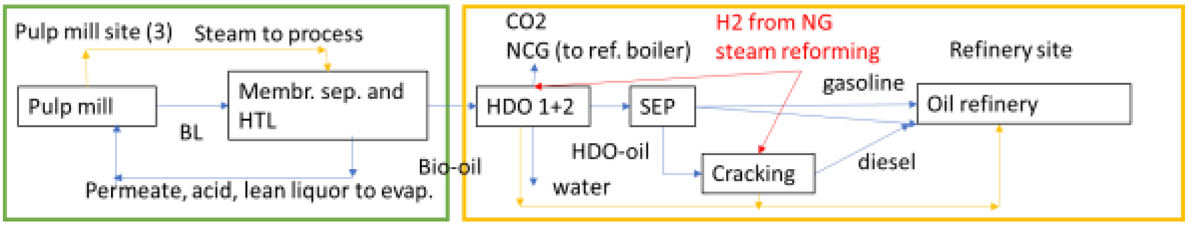
b) HydroPyr-FR



c) HTL-FR



d) HTL-MSL



e) HTL-LBL

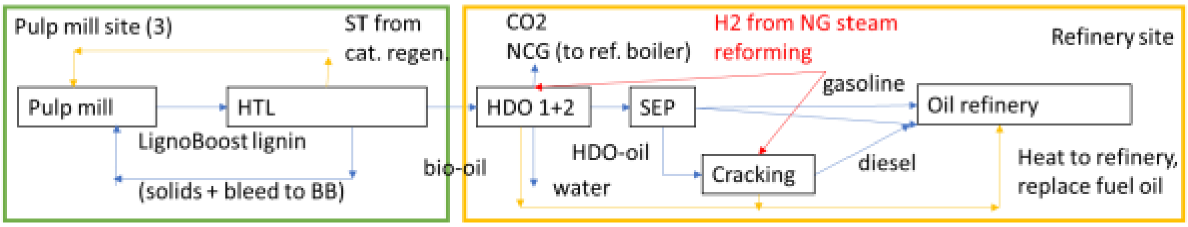


Figure 14. Integration of value chains with pulp mill and oil refinery using a) fast pyrolysis of forest residues (Pyr-FR) b) hydropyrolysis of forest residues, (HydroPyr-FR) c) HTL of forest residues (HTL-FR) d) HTL (low T, p) of kraft lignin retentate from membrane separation (HTL-MSL), Note: lignin depolymerisate is mixed with tall oil pitch or VGO to form bio-oil: e) HTL (high T, p) of Ligno-Boost lignin (HTL-LBL).

5.2 INTEGRATION OF ALTERNATIVE TECHNOLOGIES FOR LOW CARBON FOOTPRINT HYDROGEN PRODUCTION

The integration of alternative technologies for hydrogen production with a lower CO_2 footprint, replacing the conventional steam reforming of (fossil) natural gas is also included as an objective in the study. Both alkaline electrolysis of water and gasification of forest residues with hydrogen conversion have been included as two feasible alternative technologies for integration.

The location and process integration options for the alternative technologies for hydrogen production need to be defined to carry out the analysis. It was decided to locate the hydrogen production plant based on gasification of forest residues at the pulp mill site, based on the assumption it should be located at the site with easy access and most suitable infrastructure for handling of biomass. It was next decided to locate the hydrogen production plant based on electrolysis of water at the refinery, to keep the hydrogen handling at the refinery, to build on existing infrastructure and safety routines related to hydrogen handling. However, a case where the hydrogen production is located at the pulp mill could very well be considered, especially due to the availability of renewable electricity from the pulp mill.

Location of the hydrogen production at the pulp mill or the oil refinery assumes co-location with the main equipment for HDO of the bio-oil. This means that the HDO-process will be located at the pulp mill in the case with gasification of forest residues for hydrogen production, while the HDO process will remain at the refinery in the case with electrolysis. This also impacts the opportunities for process integration as described in 5.1.

For the case with gasification, the non-condensable gases and the recovered excess heat from the HDO plant will be connected to the pulp mills heat recovery system and contribute to production of additional electricity and in the case the turbine’s capacity is restricted result in additional export of bark that cannot be burnt in the bark boiler. In addition, the reject stream from the PSA unit in the plant based on gasification contains energy-rich gases that can be utilized in the recovery or bark boiler.

For the case with electrolysis of water for hydrogen production with location at the refinery, the HDO and further upgrading will be integrated in the same way as when producing hydrogen from steam reforming of natural gas. Cooling water from the electrolyser can be integrated with the refinery’s cooling water system but does not contribute with any additional energy supply.

The detailed integration options for the studied value chains are described in further detail in figures in *appendix 3: integration of hydrogen production through gasification and electrolysis in the studied value chains.*

6 RESULTS

The input data used in the calculations have been gathered from available open literature, databases, through contacts with technology developers and technology suppliers and based on discussions with technical experts from Preem. The main input data and intermediate results used for the calculations in the project are summarised in *Appendix 4: Detailed Input data and assumptions*, together with some detailed notes on the calculations for the individual technology steps.

It should be noted that the investigated emerging technologies for bio-oil production have a lower technical maturity level than the near commercial fast pyrolysis technology. Also, technology for upgrading of the produced bio-oils have a low maturity level. These technologies till require technical development and verification in one or several areas. In addition, very little experience from operation of this type of plants, even in pilot scale are available. Accordingly, both the technical and economic performance used in this evaluation is uncertain with the exception for the fast pyrolysis technology.

6.1 ENERGY BALANCE

The main energy flows in the entire value chains investigated in the project are found in Figure 15-Figure 18 below. Please observe that the figures do not give a complete energy balance but just depicts the main flows in the process.

In Figure 15 the main energy flows for fast pyrolysis of forest residues when producing 100 000 ton gasoline and diesel components a year can be seen. Calculated energy yields and energy efficiencies are shown in Section 6.2. A large part of the energy supplied with the forest residues is not transferred to the bio-oil but is transformed to excess energy in the pyrolysis process. Steam from the pulp mill steam turbine is used to dry the forest residues before pyrolysis. The excess energy in the form of char and non-condensable gases can be utilised for steam production that can be used in the pulp mill steam turbine to produce more power. For the hydrotreatment, HDO, large amounts of H_2 are needed due to the high oxygen content (37 wt%) in the bio-oil and because the oil has a H/C ration below 2. A significantly part of the total entering energy to the HDO process becomes non-condensable gases (NCG) and excess heat. At the refinery this energy can be used to replace large amounts of fuel oil used for heating purposes in the refinery.

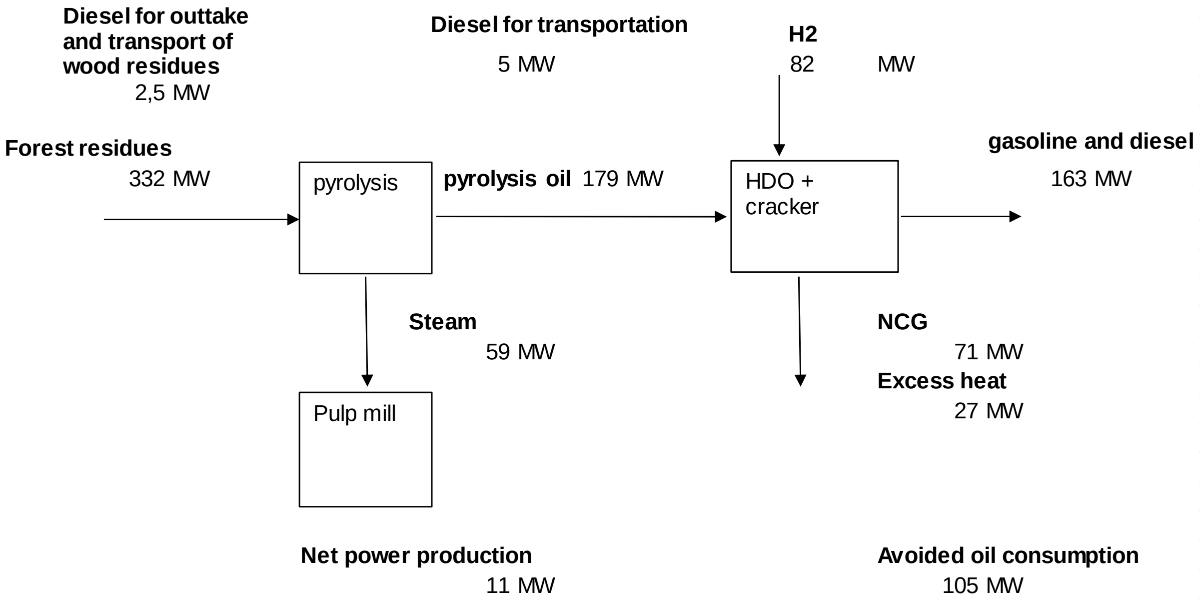


Figure 15. Overview of main energy flows (HHV basis) for fast pyrolysis of forest residues Pyr-FR. Three parallel units at different sites for conversion to bio-oil.

In the hydropyrolysis process in Figure 16, there is a high degree of internal utilisation of the energy in the feed. Due to the integration of the hydrogenation process with the pyrolysis and the utilisation of non-condensable gases for hydrogen production, the oxygen content of the bio-oil is low (just around 0.6 wt%) and the external hydrogen demand is low. Char is used for electricity production in the pulp mill, and steam extracted from the steam turbine system is used to dry the incoming wet biomass.

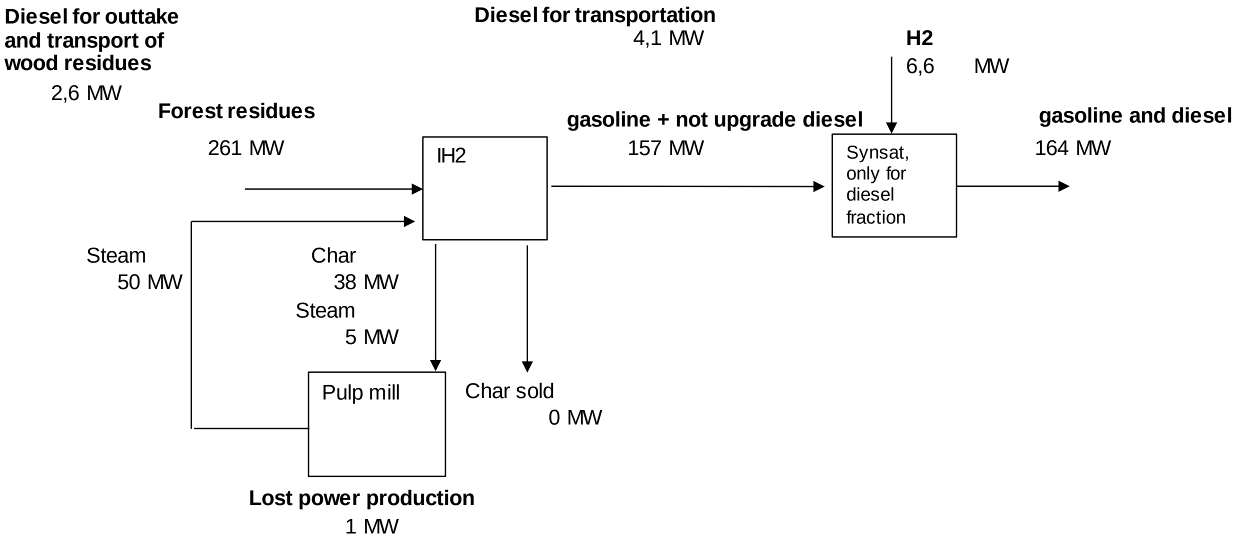


Figure 16. Overview of main energy flows (HHV basis) for hydropyrolysis of forest residues, HydroPyr-FR.

The energy flows in the value chain converting forest residues using HTL is shown below, Figure 17. As can be seen energy-rich gases from the HTL process and from anaerobic digestion of the waste water rich in organics are used to generate power in the pulp mill, while steam and power is provided from the pulp mill to heat the incoming feed and pressurise the slurry. The bio-oil contains medium level of oxygen (15 wt%), and has a H/C ration below 2, and significant amounts of

H_2 is used at the refinery, however lower than in the case of pyrolysis. Fuel oil is replaced by excess heat and NCGs from the upgrading of the oil, but again significantly less than in the pyrolysis case.

HTL bio-oil contains a significantly lower concentration of oxygen before upgrading than bio-oil from fast pyrolysis and is more thermally stable, and a conventional sulfidated NiMo catalyst can be used. However, FP bio-oil is more oxygenated and thermally reactive, therefore several stages have to be employed for stabilization and upgrading to achieve a finished product.

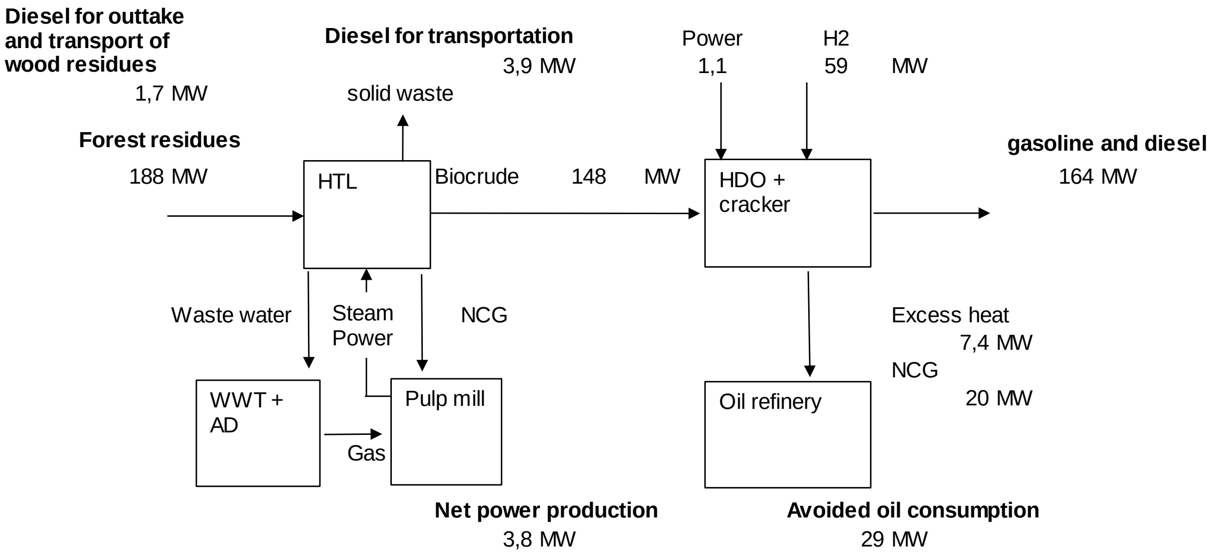


Figure 17. Overview of main energy flows (HHV basis) for hydrothermal liquefaction of forest residues, HTL-FR.

The value chain with HTL of membrane separated kraft lignin shows a relatively large retentate flow being supplied to the HTL process
Figure 18). Large amounts of energy-rich liquids are recycled to the pulp mill for energy recovery. The power loss in the pulp mill is quite significant, but lower than for the case with LignoBoost technology, see below. The oxygen level in the bio-oil is around 20%. Significant amount of refinery fuel oil is replaced also in this case.

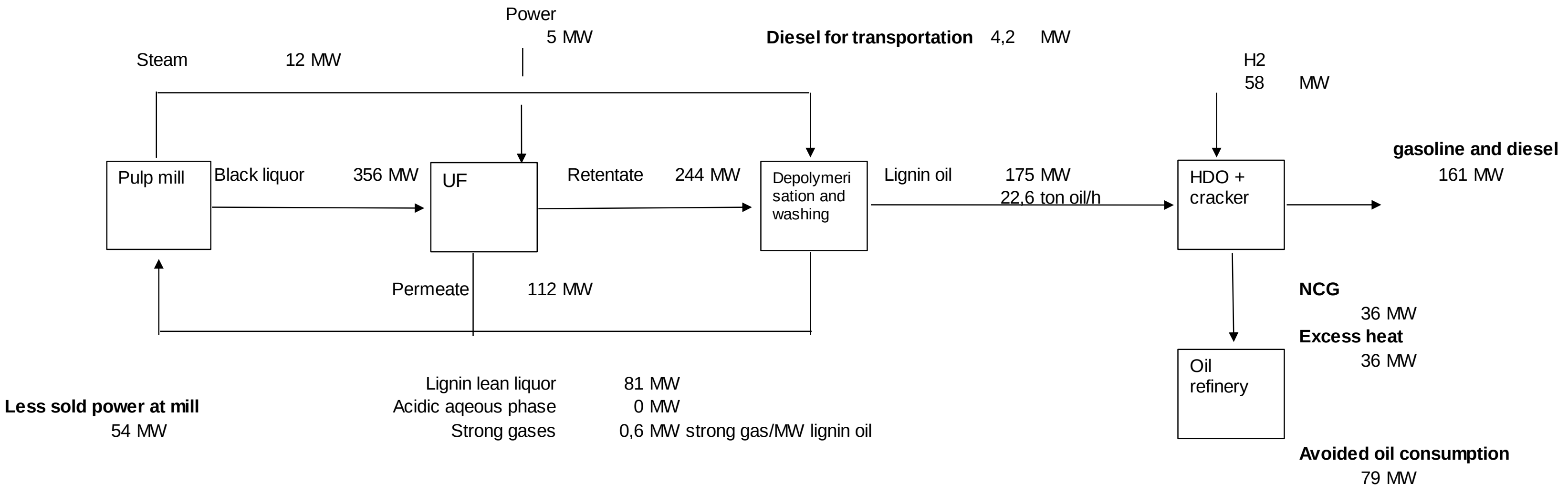


Figure 18. Overview of main energy flows (HHV basis) for hydrothermal liquefaction of membrane separated lignin, HTL-MSL. 3 parallel plants for conversion to bio-oil at 3 different sites.

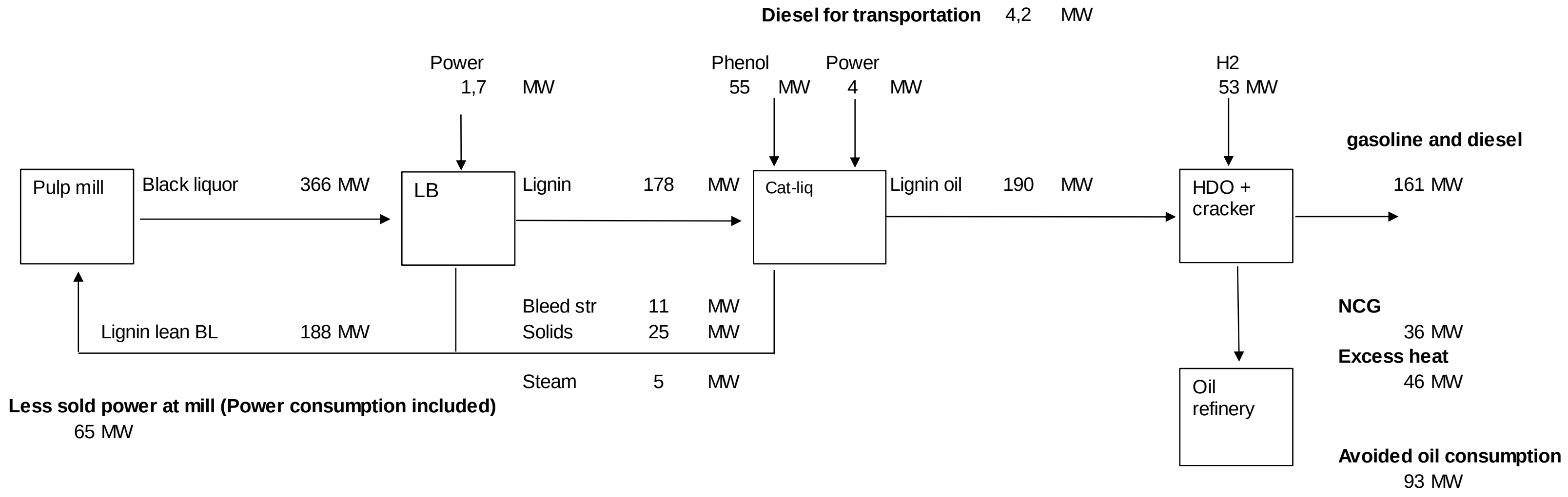


Figure 19. Overview of main energy flows (HHV basis) for hydrothermal liquefaction of Kraft lignin, HTL-LBL. 3 parallel plants for conversion to bio-oil at 3 different sites.

The hydrothermal liquefaction value chain for LignoBoost kraft lignin, see Figure 19, shows that 178 MW of energy in the form of lignin is removed from the pulp mill recovery boiler. Energy rich streams from the HTL process and from the LignoBoost process are recycled to the pulp mills energy recovery system. The reduction in the power production is quite significant, 65 MW. The lignin oil also contains remaining oxygen about the same level as in the case above with membrane separated lignin, requiring hydrogen. Excess heat and non-condensable gases replaces quite significant amount of refinery fuel oil in this case. It should also be noted that the capping agent phenol added during the HTL process contributes with quite a significant amount to the energy balance in this case.

The main observations made when comparing the value chains from an energy perspective are the following:

- Because of the relatively low yield of pyrolysis oil for fast pyrolysis process of forest residue, a large part of the energy will be in the form of steam going to the pulp mill for power production.
- For the hydrothermal liquefaction of kraft lignin, HTL-LBL, the charged phenol significantly contributes to the energy balance.
- The energy integration in the pulp mill, where residual energy streams are used to generate additional electricity has a relatively low energy conversion efficiency.
- In general, for all value chains with significant amounts of oxygen remaining in the bio-oil, i.e. all cases except the hydropyrolysis case, the HDO process requires relatively high amounts of H_2. However, a significantly part of the energy from the HDO plant are in the form of non-condensable gases and recovered excess heat. In this study it is assumed that the NCG and recovered heat replace fuel oil at the refinery with a high recovery efficiency which will have large positive impact on the cost and CO_2 emissions.
- The hydropyrolysis process has an efficient internal energy integration and does therefore not benefit to the same extent on being co-located with existing industry, perhaps with the exception of the steam used for drying the biomass.

The oxygen content in the raw material feed and the bio-oil for the different cases are shown in Table 3. As can be seen the hydropyrolysis process and the HTL processing of forest residues results in lower oxygen content than pyrolysis. HTL of forest residues also results in significantly lower oxygen content in the bio-oil than in pyrolysis oil. Interestingly, the oxygen content of the membrane separated retentate is lower than for the LignoBoost lignin. The CatLiq inspired HTL treatment of the LignoBoost lignin reduces the oxygen content in the oil quite significantly, to 18% oxygen content, while the milder HTL treatment in the case with membrane separation just reduces the oxygen content marginally in comparison. Both oils from lignin ends up with an oxygen content of around 20%, this has been observed also in other projects. It is suspected that this depends on that remaining oxygen must be removed from phenolic groups, (Anheden et al, 2017).

Table 3. Concentration of oxygen, w% in feed and bio-oil.

O_2 concentration w%	Feed	Bio-oil
Pyr-FR	40	37
HydroPyr-FR	40	0
HTL-FR	40	15*
HTL-MSL	23	21
HTL-LBL	26	18

*** Estimated from Tews et al., 2014, (report PNNL 23579), Table 2, dry basis.**

In Table 4 below, the carbon efficiency for the different value chains can be seen. The efficiencies have been calculated by taking the w% carbon in final fuel multiplied by its mass flow divided by the w% carbon in the feed multiplied with its corresponding mass flow.

Table 4. Calculated carbon efficiency by carbon in final fuel/ carbon in feed.

Carbon efficiency	Pyr-FR	HydroPyr-FR	HTL-FR	HTL-MSL	HTL-LBL
C in transport fuel/ C in wood residues	0.37	0.47	0.65	0.75	0.72

The results show a high carbon efficiency for all the HTL cases, especially for lignin, while the carbon efficiency for the fast pyrolysis option is in comparison low. The hydropyrolysis is somewhere in-between. This means that the HTL processes efficiently utilises the feedstock carbon content due to low char and gas formation in the form of light hydrocarbons and CO_2.

6.2 ENERGY CONVERSION

The energy efficiency, (fossil) CO_2 emissions and economic performance for the value chains are studied from a well-to-gate perspective, that is, from the outtake of raw material to the production of the end-product gasoline and diesel components.

There are several ways of defining energy yields and energy efficiencies. In this study we have chosen to the define energy yield and energy efficiency as follows:

$$Energy\ yield = \frac{energy\ in\ gasoline\ and\ diesel}{energy\ in\ feed\ (forest\ residue\ or\ lignin)}$$

where energy in feed have been calculated as the mass flow of forest residue or lignin multiplied with its corresponding higher heating value.

The energy efficiency for the cases where forest residues is the raw material, the energy efficiency defined below has been used. The energy efficiency is defined as the net energy output in form of energy in transportation fuel components, net power generated and replace fuel oil, divided by the energy input in form of raw material, added hydrogen and transportation fuel used for transport of raw material and intermediates.

$$Energy\ efficiency = \frac{(energy\ in\ gasoline\ and\ diesel + (generated\ power - power\ consumption) + replaced\ fuel\ oil)}{(feed\ raw\ material + H2 + fuel\ for\ transports\ of\ feedstock\ and\ intermediate\ products)}$$

In case of lignin as raw material, the energy efficiency is based on the same principle, useful energy out divided by the energy input, but clarifications are made in the two efficiencies below on how to handle the flows originating from the black liquor and that there is no net generation of electricity. In addition, it is made clear that the phenol added in HTL-LBL should be considered in the energy balance

$$Energy\ efficiency\ 1 = \frac{(energy\ in\ gasoline\ and\ diesel + lignin\ lean\ liquor + replaced\ fuel\ oil)}{\begin{gathered}(LignoBoost\ lignin\ or\ retentenate + power + steam + H2 + phenol\\ + fuel\ for\ transports\ of\ feedstock\ and\ intermediate\ products)\end{gathered}}$$

$$Energy\ efficiency\ 2 = \frac{(energy\ in\ gasoline\ and\ diesel + lignin\ lean\ liquor + permeate + \ replaced\ fuel\ oil)}{\begin{gathered}(black\ liquor + power + steam + H2 + phenol\\ + fuel\ for\ transports\ of\ feedstock\ and\ intermediate\ products)\end{gathered}}$$

In energy efficiency 1, the system boarder is set to consider LignoBoost lignin and retentate as the direct input of energy, while in energy efficiency 2, the system boarder is set so that black liquor to the LignoBoost unit and the membrane separation unit is the input, meaning that the LignoBoost and membrane separation units are included in the system. To be noted is that in both these efficiencies, the system boundary is extended to include the effect on the pulp mill electricity production.

Figure 20 below shows both energy yield from feedstock to gasoline and diesel and the energy efficiency with forest residues as feedstock. It can be seen that the hydrothermal liquefaction of forest residues has the highest energy yield and efficiency with almost 90% and 80%, however it should be remembered that this case is still a fictional case (“goal case”) that has not been experimentally proven to the same extent as the other cases. The hydropyrolysis has about 60% energy efficiency and yield. Even though the pyrolysis process has the lowest energy yield it has an energy efficiency of 68% as the energy integration of the excess heat at the HDO upgrading is very efficient. From the energy flows in Section 6, it can be noticed that the main yield loss takes place in the depolymerisation step, while the yield loss is less in the upgrading of the bio-oil, due to the assistance of hydrogen.

In Figure 21, energy yield and two different energy efficiencies are shown for the hydrothermal liquefaction of lignin and membrane separated lignin. Energy efficiency 1 uses lignin or retentate as raw material input. Energy efficiency 2 uses black liquor as raw material input instead, which could be seen to be more in line with the system boundaries defined in the project. The energy loss in the lignin cases are mainly a result of the relatively low energy conversion efficiency when the residual products from lignin separation and purification are sent back to the pulp mill for electricity production, i.e. for conversion to higher quality energy. Compared to the cases that use forest residues as raw material, the lignin cases do not produce char and gas to the same extent which will lower the energy efficiency. Comparing the lignin cases, membrane separated lignin has lower energy efficiency 1 because retentate is used as raw material hence a flow separated from the lignin oil (lignin lean liquor) going back to the pulp mill will lower the efficiency as it is converted to electric power. Comparing energy efficiency 2 the major effect is the amount of black liquor used, this in turn depends on the yield in the LignoBoost or the ultrafiltration unit. It is to be noted that the yield in the ultrafiltration unit is a parameter associated with high uncertainty.

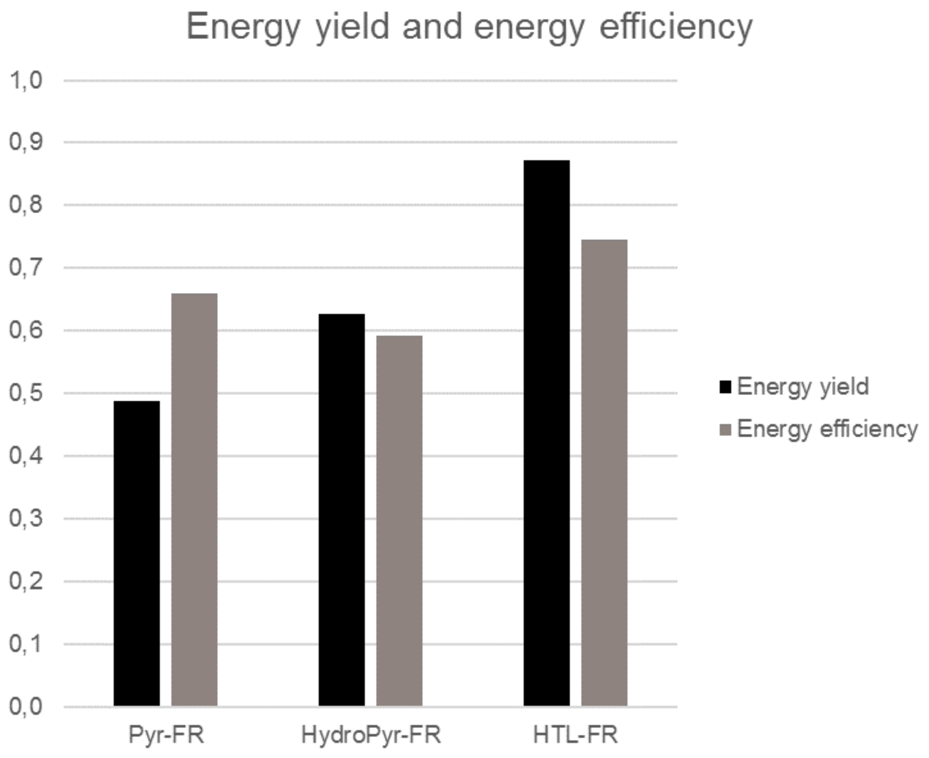


Figure 20. Energy yield from feedstock to gasoline and diesel, MWhtf/MWhf = black and the energy efficiency = grey.

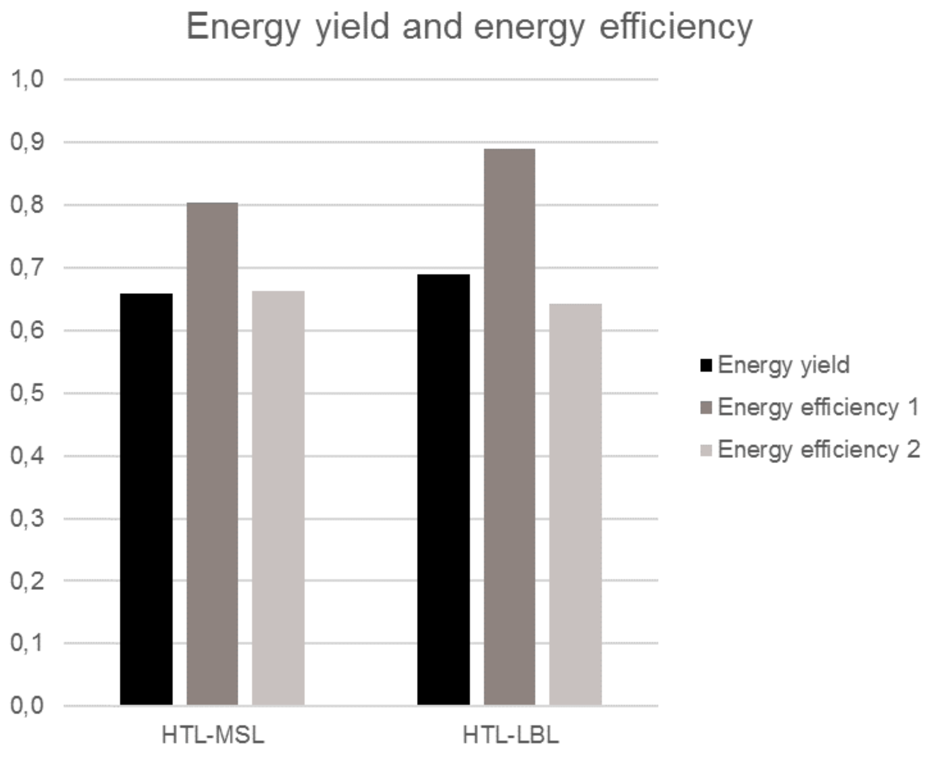


Figure 21. Energy yield from feedstock to gasoline and diesel, MWhtf/MWhf = black and the energy efficiency = grey.

6.3 ECONOMIC PERFORMANCE

Table 5 below gives an overview of the total investments required in the different value chains.

Table 5. Overview of total investment of the different value chains for production of 100 000 t/y gasoline and diesel components based on available scaled literature data from multiple sources.

		Pyr-FR	HydroPyr-FR	HydroPyr-FR	HTL-FR	HTL-MSL	HTL-LBL
Intermediate production	MEUR	200*	87	230	110	180*	230*
Intermediate conversion & upgrading	MEUR	150	0	0	98	89	94
Total investment	**MEUR**	**350**	**87**	**230**	**210**	**270**	**320**

***Three parallel plants for conversion to bio-oil.**

The following main observations are made:

- The hydropyrolysis has relatively low investment cost. Thus, two different investment cost estimations have been included. The lower one originates from GTI (2012) and the higher one from information from media (Igjerstadsidene, 2017). Comparing the investment cost of hydropyrolysis with the other value chain investment costs, it seems that the lower investment cost may be somewhat underestimated.

- The value chain with fast pyrolysis has the highest investment cost. The pyrolysis oil has a higher oxygen concentration and is thermally instable compared to the HTL oil. Therefor the pyrolysis oil requires more stages in the HDO process to upgrade the oil to the finished product. Hence the higher investment cost for this stage (Tews et al, 2014). The technical maturity is also highest for the pyrolysis process, making it more likely that the cost estimate is made with somewhat higher accuracy, that could be suspected to result in higher costs.

- One could perhaps suspect that the investment cost for the case for HTL on forest residues could be a bit underestimated compared to the other investments estimates for the HTL processes.

- Overall, the investment cost for hydropyrolysis (the high cost) and HTL on forest residues looks promising.

The estimated operational costs for the different technologies are found in Table 6.

Table 6. OPEX, EUR/MWh transportation fuel components, for the value chains divided on the main cost items.

Total value chain costs	**Pyr-FR**	**HydroPyr-FR**	**HydroPyr-FR HC**	**HTL-FR**	**HTL-MSL**	**HTL-LBL**
Forest residues	35	27	27	20	0	0
Lost/consumed power	3	3	3	3	15	17
Catalyst	8	0.4	0.4	2	8	8
Chemicals	0	0	0	4	6	80
H_2	28	2	2	14	20	19
Profit margin on lost fossil diesel	6	2	2	4	7	7
Transports	9	10	10	7	2	3
Maintenance	11	3	7	8	9	10
Benefit: Sold bark/power/ replaced fuel oil	-31	-2	-2	-12	-20	-24
Total OPEX EUR/MWh	**69**	**45**	**49**	**50**	**46**	**120**

The following observations are made:

- For value chain with forest residue as feed, the raw material is largest contribution to the operating cost, about 40-50%.
- The intermediate production cost for the HTL-LB case is high and is significantly dominated by the cost of the capping agent phenol
- The OPEX for the pyrolysis case is high, mainly due to high raw material cost due to the relatively low conversion efficiency on fast pyrolysis of forest residues and due to the high hydrogen consumption caused by the high oxygen content.
- The assumed cost for catalyst in HydroPyr-FR is significantly lower than in the other cases, based on information from CRI. This need to be experimentally verified. Also, the cost of hydrogen is low, due to the integrated process for hydrotreatment using the produced off-gases as hydrogen source.

Figure 22-Figure 24 below illustrate the cost for the total value chain in EUR/MWh transport fuel components broken down in different ways to support the analysis of the results.

The estimated production cost for suppliers of ethanol and HVO today in Sweden is 1,15 and 1,10 SEK/kWh or 127 and 122 EUR/MWh transportation fuel (Energimyndigheten 2017a). The production cost estimates in this study are in the same range or lower (see comparison with the blue area in Figure 22), indicating a potential for profitability for investments in local production capacity on the Swedish market. The average yearly European spot market price (FOB, ARA) on ethanol and biodiesel has been about 5.30 SEK/l and 6.80 SEK/l, i.e. 99.80 EUR/MWh and 82.10 EUR/MWh during 2016, see the green area in Figure 22. Compared to this, the emerging technologies still have some profit margin to the spot market price with the exception of the HTL-LBL case, while the production cost for the pyrolysis case is in the upper range of the European spot price, leaving a low margin for profit. In Figure 22, one can also clearly see that the OPEX costs are dominating the production cost for all cases. The CAPEX makes up about 10-30% of the production cost. This is expected for this type of technology based on the author’s experience. From

Figure 23 it is evident that the transportation costs for these value chains are relatively low, and that the major costs are related to raw material and production of the bio-oil intermediate. The impact from the final bio-oil upgrading step is low.

The case showing the largest transportation fuel production cost is the hydrothermal liquefaction of kraft lignin. The main cause of the relative high cost is the addition of phenol in the depolymerization step, which stands for 43% of the total cost. However, it also has a relatively high cost for preparation of feed through separation of lignin with the LignoBoost process, compared to the other process concepts. It is to be noted that different levels of data quality have been received for the different value chains. Since the technical maturity level of membrane separation of lignin is lower than for HTL-LBL, one could speculate if the other lignin value chain has other chemical cost not included in this assessment that will contribute to the total cost as well. Lowering the price of phenol by 50% decreases the total value chain cost for HTL-LBL to 100 EUR/MWh, a decrease by 30%, however, to get to the same production cost levels as the other emerging technologies would rather require an even further reduction to close to zero cost, requiring a totally recyclable capping agent or a capping agent that can be produced and recycled in the process at a very low cost.

Further on, the three value chains hydropyrolysis and hydrothermal liquefaction of forest residues and hydrothermal liquefaction of membrane separated lignin seem to result in production costs in the same range of 50-70 EUR/MWh transportation fuel. The pyrolysis ends up with a cost of almost 100 EUR/MWh, where the largest contribution to the cost is forest residue (~35%) and intermediate conversion and upgrading (~30%).

Catalyst consumption is often sited to be critical to transportation fuel production. The catalyst consumption assumed in the hydrotreating of the bio-oil from Pyr-FR, HTL-MSL and HTL-LBL stands for roughly up to about 10% of the total value chain cost. Catalyst consumption in the hydrotreating of the bio-oils have been estimated based on data from Tews et al. 2014. For the Pyr-FR and the lignin cases the following catalyst have been assumed a Ru/C catalyst in the stabilization, sulfided Ru/C in the first stage hydrotreater, and sulfided NiMo in the second hydrotreater. The uncertainty about lifetime and performance of the catalyst is quite large. In this study the catalyst life time has been assumed to be 2 years. The catalyst consumption for HydroPyr-FR stands for about 1% of the total cost. CRI Catalyst Company (CRI) has developed and provided the catalyst for the HydroPyr-FR case but no more information is given about what kind of catalyst it is and its life time (Gas Technology Institute (GTI) 2012). For the HTL-FR value chain, catalyst consumption is based on data from de Jong et al., 2017 which is based on Tews et al. 2014 and Zuh et al., 2014. Type of catalyst assumed, and life length is unknown.

Since it is noticed that the required number of HDO-stages is uncertain, as well as the catalyst material and life length, especially for the lignin cases, a sensitivity study was performed where these parameters were varied for the HTL-MSL case. Two extreme cases where investigated

a) with catalyst consumption as in the pyrolysis case and 2-stage HDO and

b) catalyst consumption similar to the experimental catalyst consumption for HTL-FR and 1 stage HDO (Tews, 2014).

The transportation fuel component production cost then varies between 62-76 EUR/MWh.

From Figure 24 it is possible to see that the lost power production in the pulp mill in the two lignin cases make up a quite significant part of the production cost in a similar way as the cost of forest residues in the other value chains. One can also see the clear positive contribution from the refinery integration of the excess heat from the HDO an upgrading steps for all cases except for the hydro-pyrolysis case.

It is also possible to distinguish the cost related to loss of profit margin for the necessary reduction in fossil diesel production due to that the bio-diesel fraction requires capacity in the common cracking and SynSat equipment. However, the contribution of the fossil diesel capacity reduction is in no way dominating over other costs.

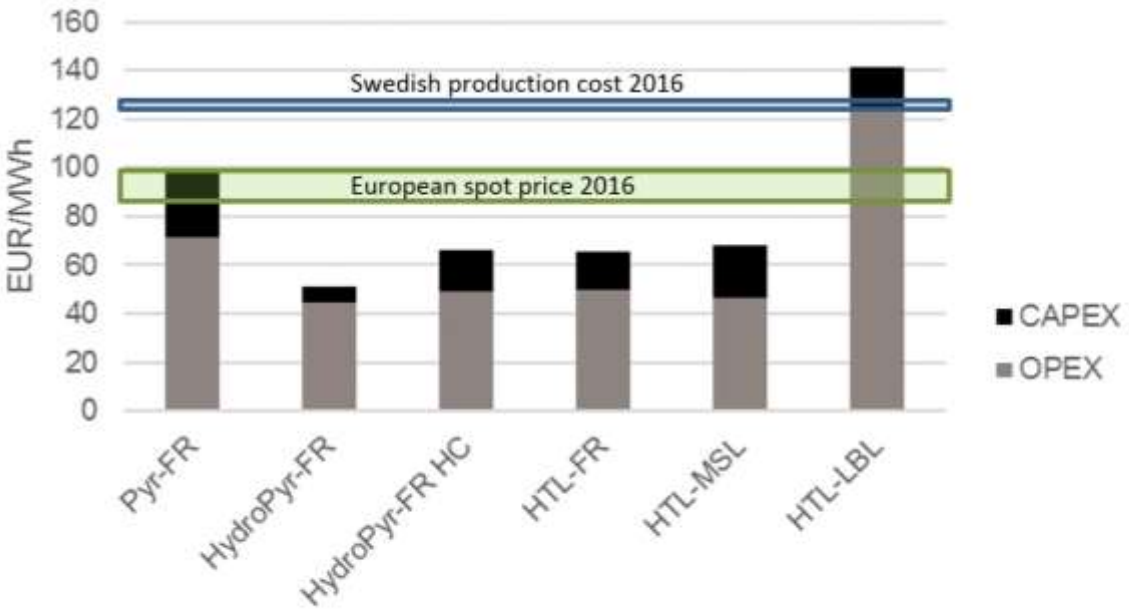


Figure 22. Total value chain production cost, including contribution of OPEX and CAPEX in EUR/MWh transportation fuel. The blue area indicates the current (2016) reported biofuel production cost for ethanol and HVO for Swedish suppliers, while the green area represents the yearly average spot prise (ARA, FOB) on the European market in 2016, (Energimyndigheten 2017a).

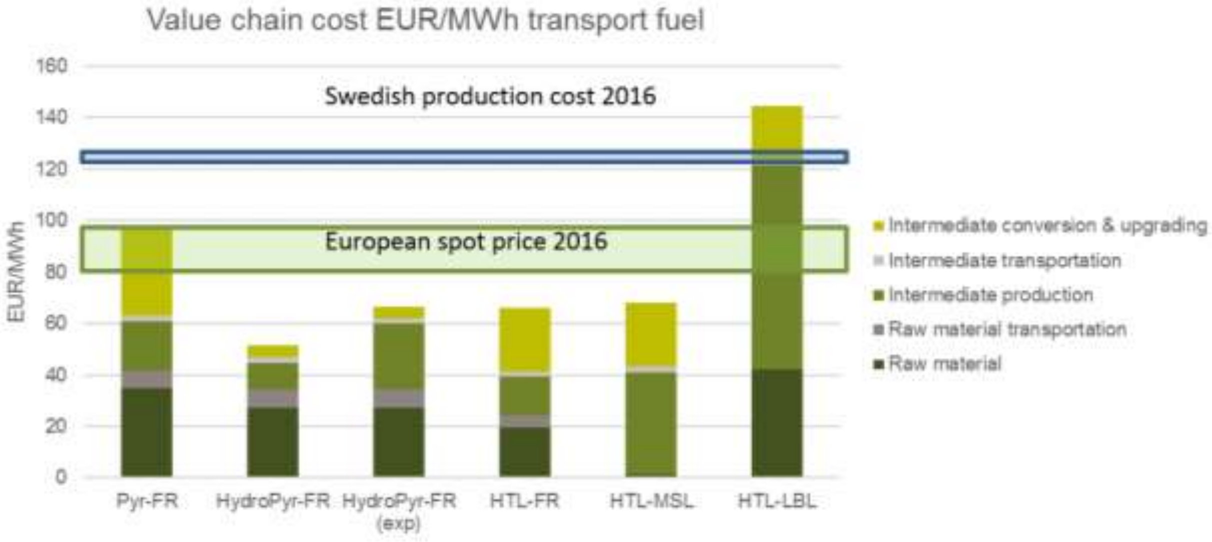


Figure 23. Total value chain production cost in EUR/MWh transport fuel.

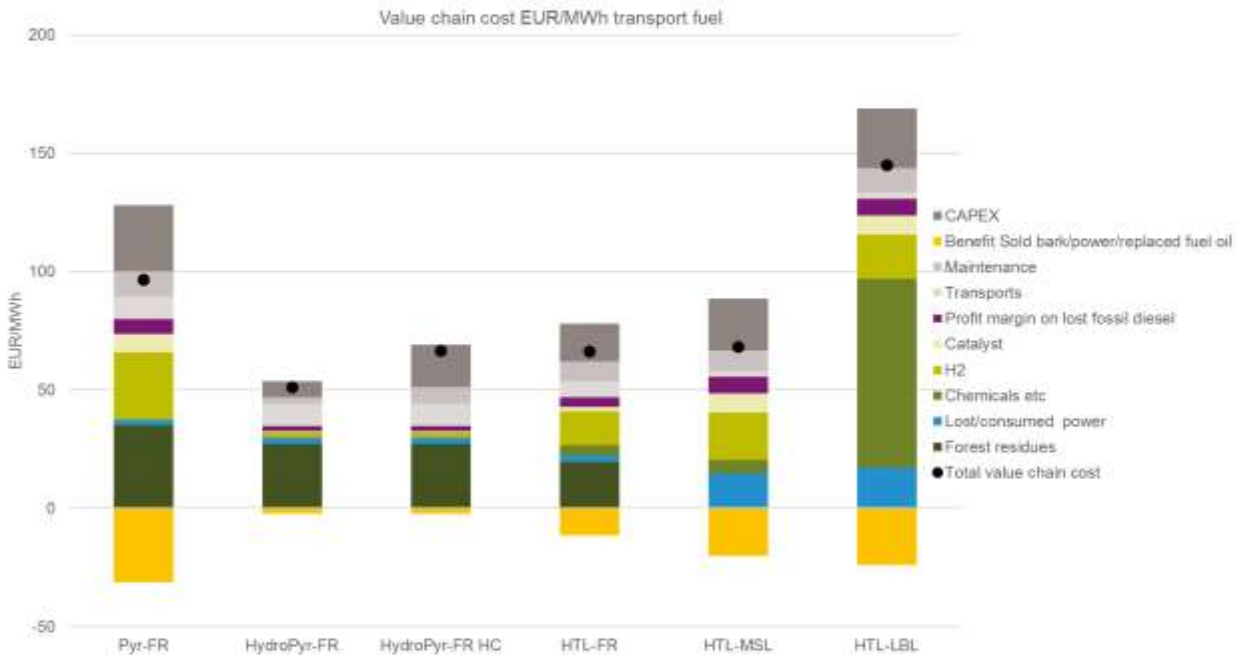


Figure 24. Total value chain production cost in EUR/MWh transportation fuel divided by cost and income items. The black dot indicates the net production cost.

Looking at just the OPEX part of the costs in Figure 22 indicate that it is quite likely that the investigated technologies will remain in operation once they are built since there is a good margin from the OPEX to the spot price, except for the HTL-LBL case. The margin is also less for the fast pyrolysis case.

6.4 CO_2 EMISSIONS

The total net CO_2 emissions for the value chains (kg CO_2/MWh transportation fuel) includes emissions related to the collection and transportation of the raw material, the bio-oil production and conversion to final product (including the hydrogen required) and transportation, either of crude bio-oil or final product depending on case. There are also credits given for e.g. net surplus of electricity and replacement of fossil fuels. Impact of soil carbon decrease is not considered since the magnitude of the impact of the soil carbon decrease is uncertain and still under investigation and discussion (Holmgren et al. 2007).

The net total emission of fossil CO_2 emission from the studied transportation fuel value chains is shown in Figure 25, based on the system boundaries defined in Figure 14. The studied system then includes important effects such as the effect on the pulp mill electricity production by separating lignin and other effects on the pulp mill caused by integration, the effects of hydrogen supply and other chemical consumption and the effects from integration with the oil refinery in the form of reduced fuel oil consumption. As can be seen, all value chains except the value chain with HTL on LignoBoost kraft lignin has a low impact on fossil CO_2 emissions, or even negative in the case of pyrolysis.

As can be seen in Figure 26, the large impact on the CO_2 emission in the HTL-LBL case is related to the intermediate production step, and more specifically related to the use of phenol as capping agent, as phenol has a large CO_2 footprint from the phenol production chain. Also, the raw material contributes, which in this case is mainly the loss of electricity production from the pulp mill when the lignin is used for production of transportation fuel. To note is also that the chemical consumption of H_2SO_4, NaOH and CO_2, due to the introduction of the LignoBoost process also has an impact.

The pyrolysis value chain receives its negative CO_2 emission mainly due to effects related to integration of residual energy, mainly in the refinery. This is due to the high oxygen content in the oil and the large amounts of energy that are released when the oxygen is removed and due to the light gases formed during the HDO process that can be used to efficiently replace fuel oil used for heating purposes, see Figure 27 and Figure 28. This integration opportunity also plays an important role for the total CO_2 emissions from the two lignin value chains and the chain with HTL of forest residues. From Figure 28 it is also clear that the integration of excess heat in the pulp mill with the objective of generating additional electricity only contributes to a small extent to reduce CO_2 emissions.

Figure 26 shows the contribution to the CO_2 emissions from the fossil-based hydrogen as the light green part of the bar, showing the contribution from hydrogen in both the intermediate conversion and upgrading steps. This is the major contribution to CO_2 emissions for all value chains except the HTL-LBL value chain.

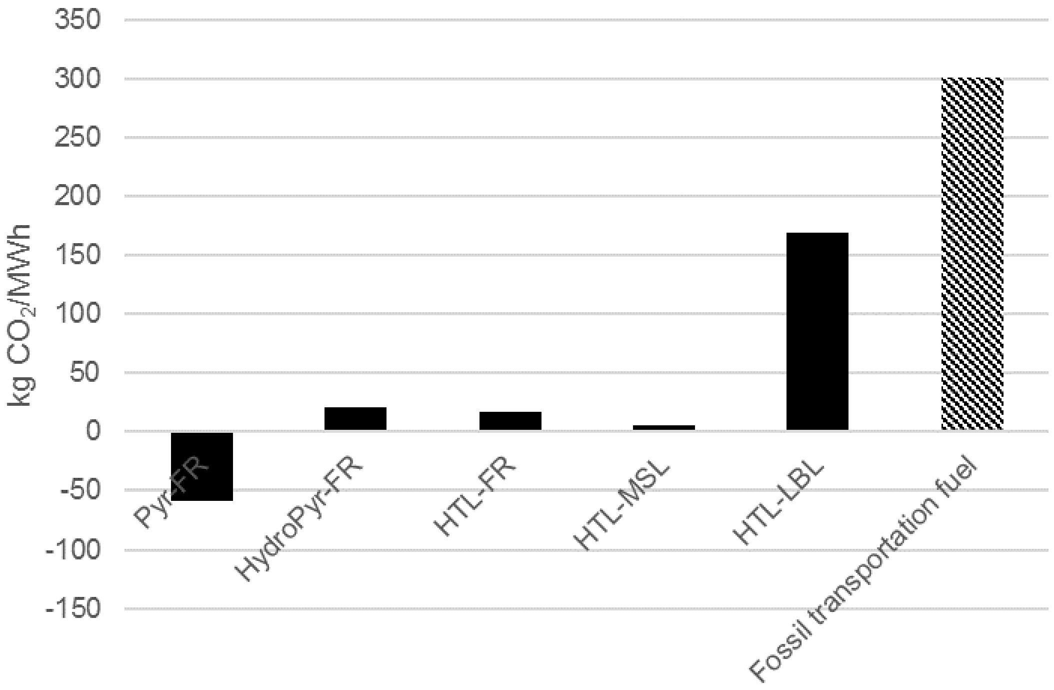


Figure 25. Total fossil CO_2 emissions of the value chains, kg/MWh transportation fuel.

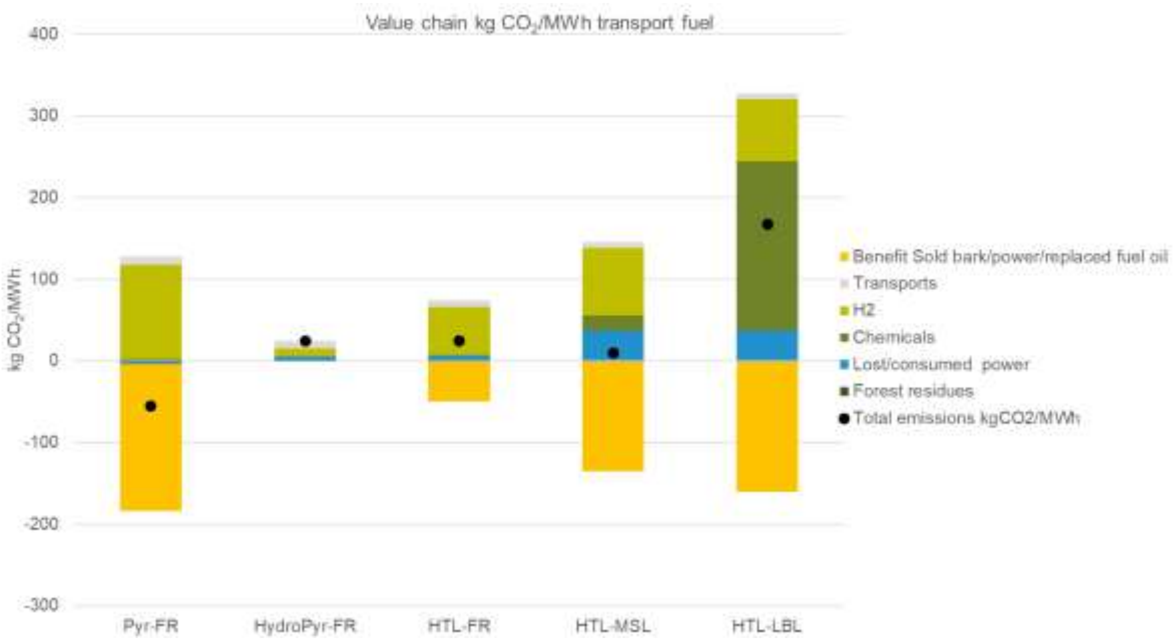


Figure 26. Total value chain CO_2 emissions, divided on the main contributions in the value chain kg CO_2/MWh transportation fuel.

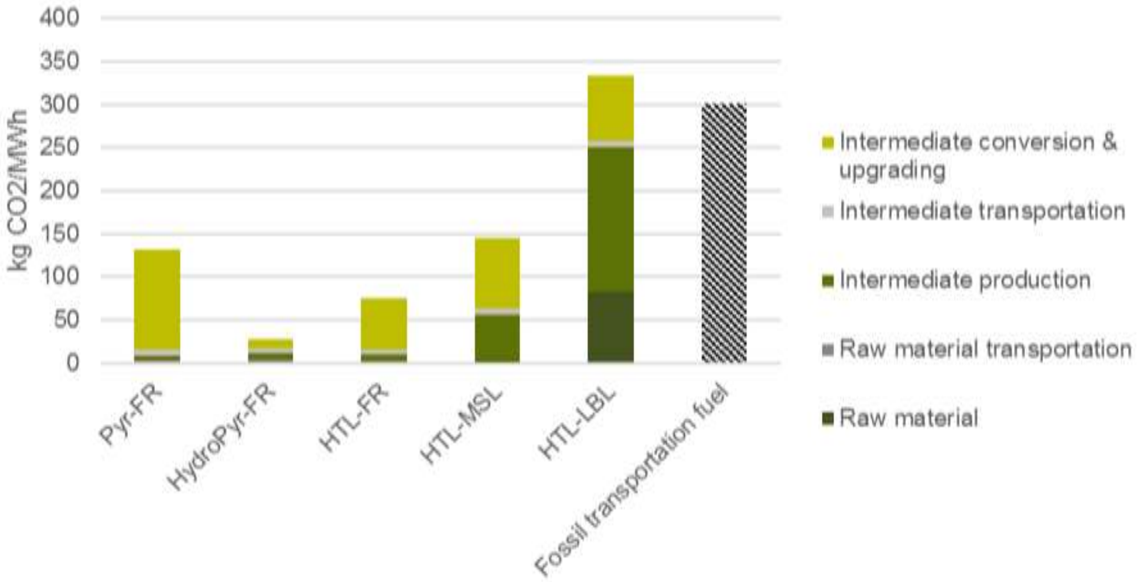


Figure 27. CO_2 footprint of transportation fuel excluding effects related to recovery of residual energy streams, kg CO_2/MWh transportation fuel.

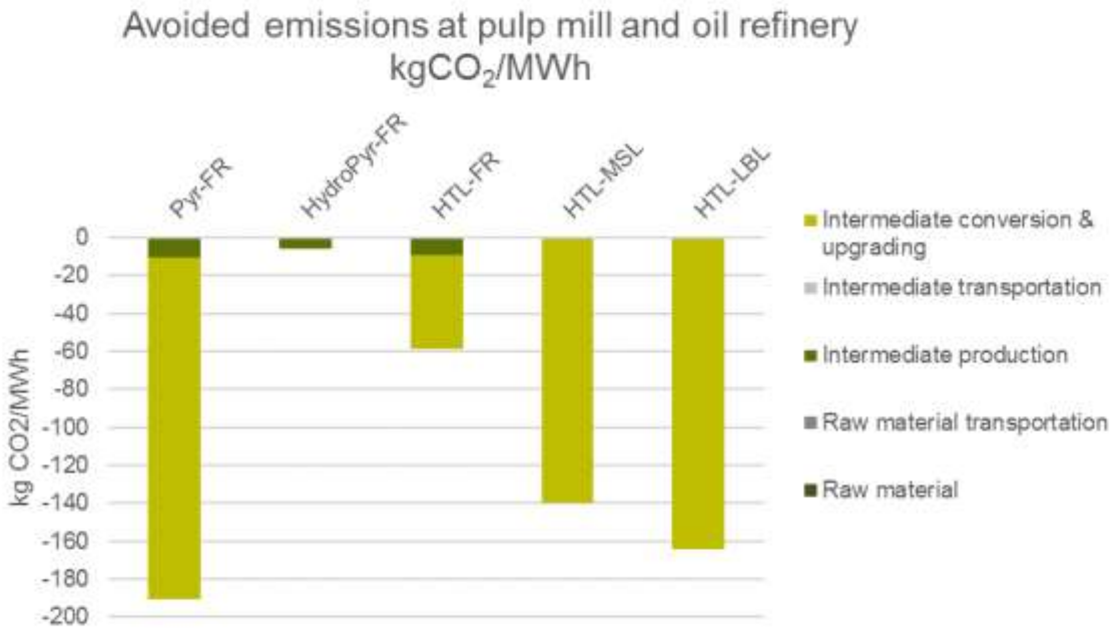


Figure 28. Avoided emissions through integration of residual energy in the pulp mill (dark green) and the refinery (light green), kg/MWh transportation fuel.

The analysis presented so far has analysed the CO_2 emissions from implementing the studied value chains. However, the reporting of the CO_2 footprint related to a certain biofuel must follow the rules stated in the EU Renewable Energy Directive (RED) and the so called Indirect Land Use Change (ILUC) directive that is now being implemented in Swedish law. It is noted that the actual fuel CO_2 footprint will look quite different from the value chain CO_2 emissions since:

a) Both forest residues and lignin are considered to be residues in the law and should therefore not be burdened with any emissions that have arisen before the residue leaves the process where it was created. For lignin it means that the CO_2 emissions caused by loss of electricity production in the pulp mill due to removal of lignin and from the usage of chemicals for the lignin separation should not be assigned to the transportation fuel.

b) Utilisation of residual heat from the value chain cannot contribute to reduce the transportation fuels CO_2 footprint. It means in this case that the significant reduction in fuel oil usage due to utilisation of process heat cannot be accredited the transportation fuel. Accordingly, a large share of the negative emissions in Figure 28 cannot be accounted for.

c) In the directive, there is a possibility to allocate part of the emissions on co-products. It is possible that the energy rich gases from the bio-oil production and the HDO and upgrading steps can be considered as co-products. The same could be argued for the electricity generated from excess heat. If this is the case, they should bear a share of the CO_2 emissions in proportion to the energy content.

This would mean that the value chains that have a high degree of industrial integration, i.e. all value chains except the hydropyrolysis value chain, would have significantly higher CO_2 footprint on the transportation fuel, especially the pyrolysis value chain. This would make it difficult for many of the investigated technologies to achieve to 60-70% reduction level requirement discussed for new-built plats.

It should be noted that since the plant will be in operation for one or more decennium and new EC directives and laws will be implemented during that time, the described way (point a-c) of allocating CO_2 emissions may not be representative in the plant life perspective.

It is also important to note that from a climate perspective it is the value chain’s entire emissions that are of importance, i.e. the values reported in Figure 25, and not how the transportation fuels are declared based on rules in a reporting tool. However, from a commercial perspective, this will have a large impact, especially considering the system to be implemented in Sweden with binding emission reduction targets.

6.5 SENSITIVITY ANALYSIS WITH RESPECT TO COST

A sensitivity analysis for the most dominant parameters has been made to evaluate how these parameters will affect the final result, see Figure 29 - Figure 33. The sensitivity analysis is made by varying the price for power, forest residues, phenol and investment cost by ± 50%.

As expected, the value chains based on use of forest residues are very sensitive to the cost of the forest residues. It is therefore important to ensure a stable price on this raw material and to focus on improving the raw material to transportation fuel yield. The lignin value chains show a larger sensitivity to the investment cost than the electricity cost, which is the “raw material cost” in this case. Finding opportunities for reduced investment cost is important for all cases.

All value chains except the fast pyrolysis value chain and the HTL-LBL value chain are still below the average European spot price on ethanol and biodiesel throughout the sensitivity study, which prove that these cases are relatively robust.

The fast pyrolysis case will have a higher production cost than the spot price at high raw material costs and investment costs. However, the production cost is still below the production cost for Swedish suppliers of ethanol and HVO in Sweden during 2016.

The HTL-LBL case is above both the spot price and the production cost for Swedish suppliers throughout the sensitivity study. In order for this case to be interesting, the cost for the capping agent has to be reduced almost to zero, indicating a solution where almost all the capping agent is recycled or generated during the process, as noted in previous section.

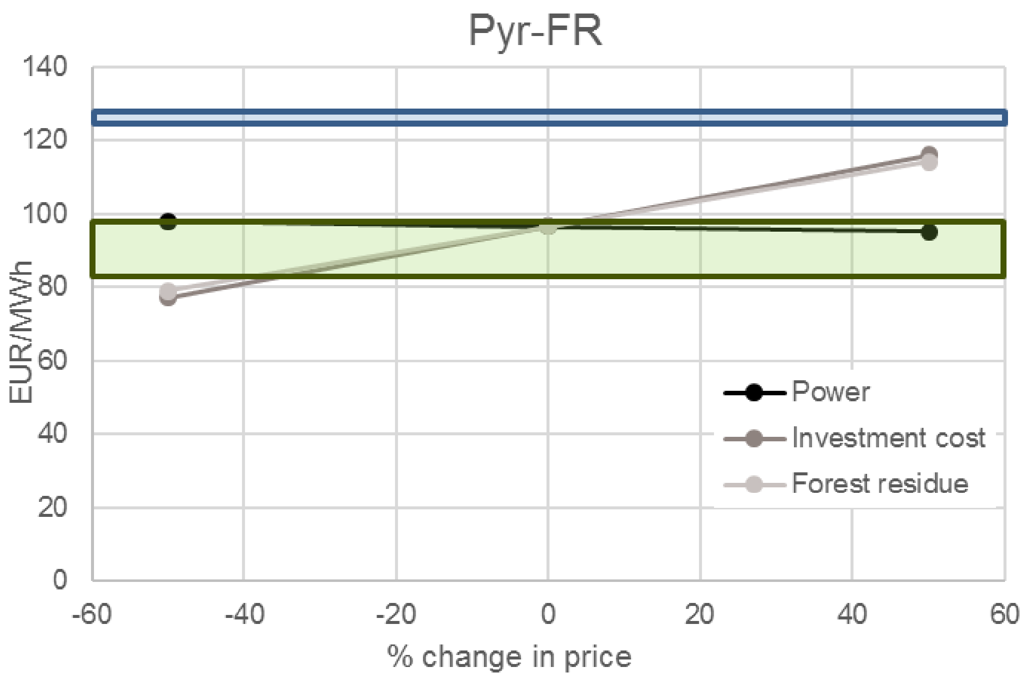


Figure 29. Sensitivity analysis for pyrolysis of forest residues by varying the prices for power, forest residues and investment cost ± 50%.

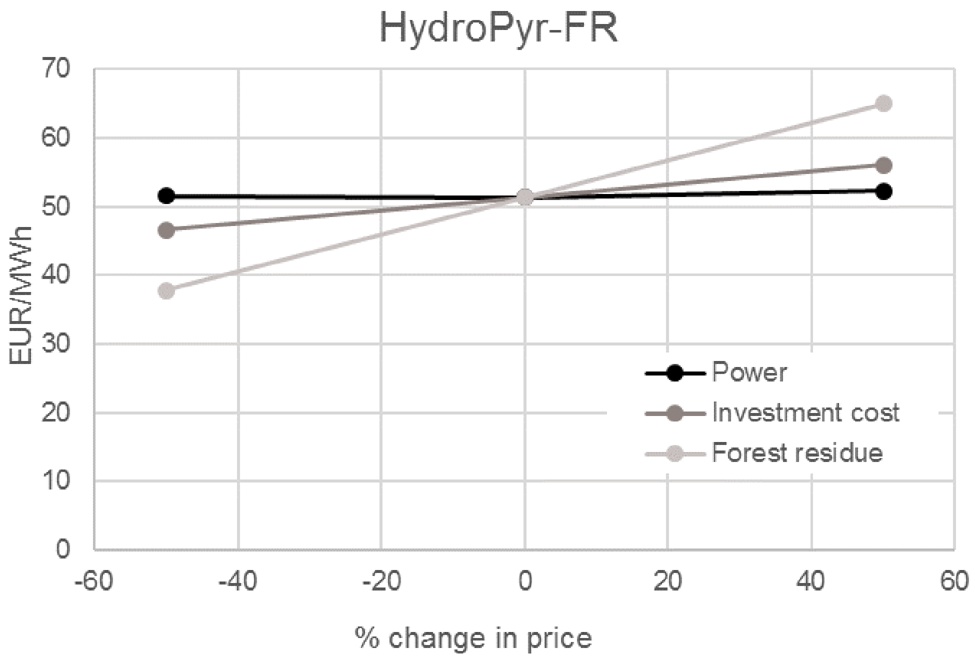


Figure 30. Sensitivity analysis for hydropyrolysis of forest residues by varying the prices for power, forest residues and investment cost ± 50%.

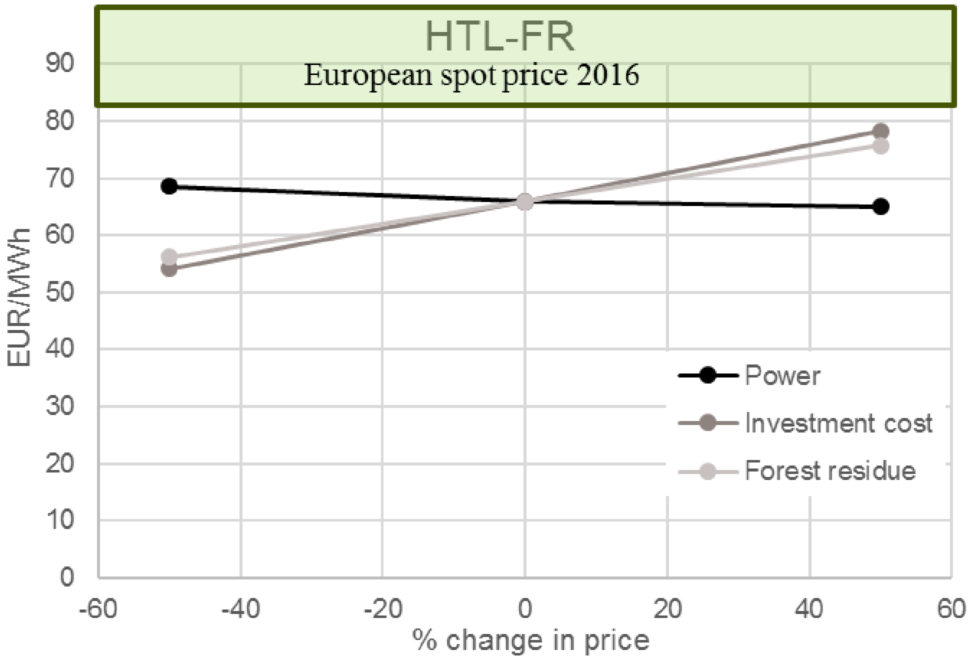


Figure 31. Sensitivity analysis for hydrothermal liquefaction of forest residues by varying the prices for power, forest residues and investment cost ± 50%.

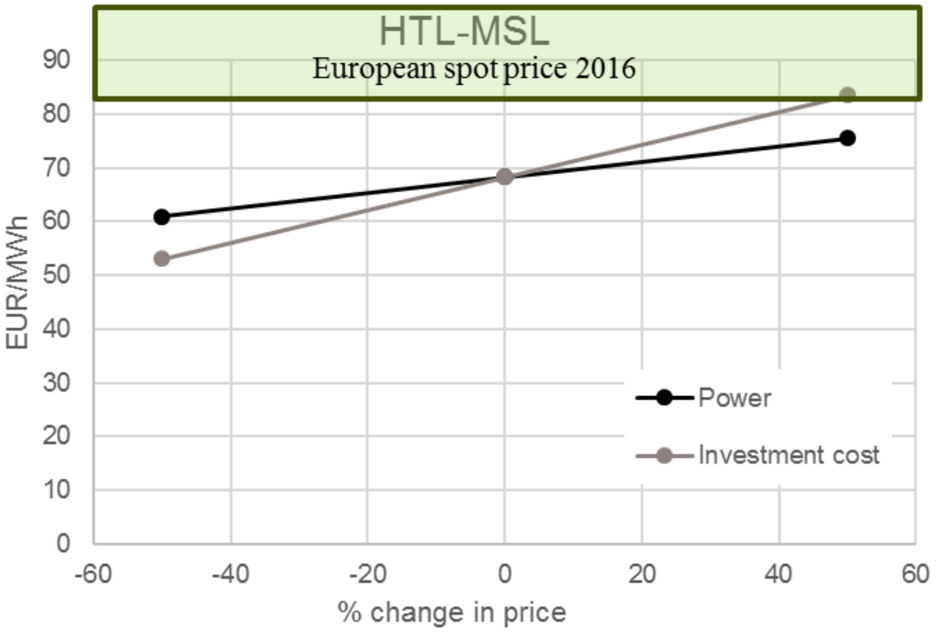


Figure 32. Sensitivity analysis for hydrothermal liquefaction of membrane separated lignin by varying the prices for power and investment cost ± 50%.

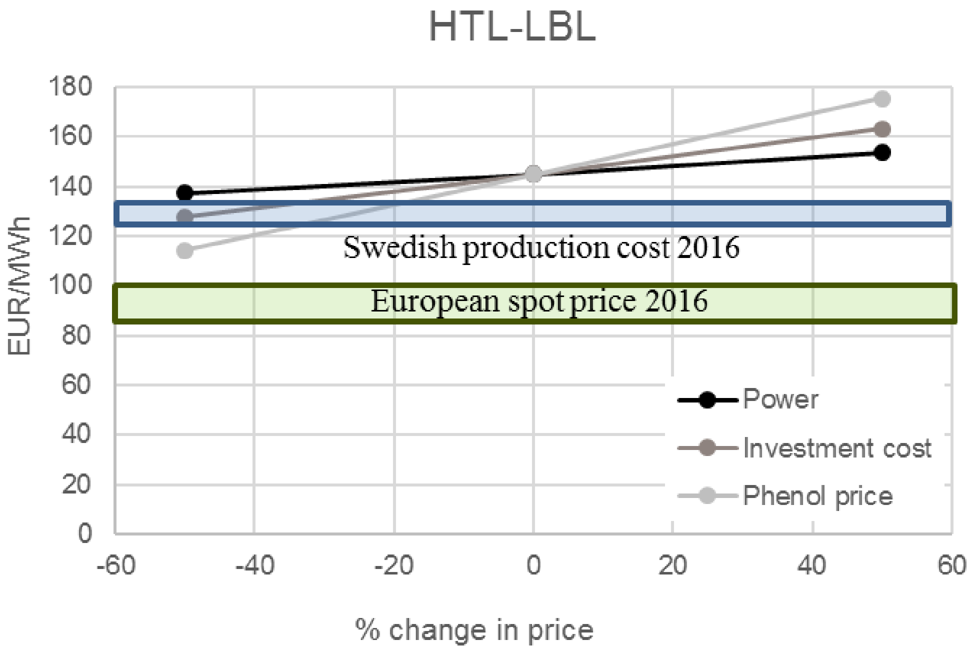


Figure 33. Sensitivity analysis for hydrothermal liquefaction of kraft lignin by varying the prices for power, phenol and investment cost ± 50%.

6.6 ALTERNATIVE PRODUCTION PATHS FOR HYDROGEN

As described earlier, the CO_2 footprint and the production cost are dependent on the raw material and production path used for making hydrogen. A sensitivity study looking at alternative means to produce the hydrogen is therefore included here. The chosen technologies are alkaline electrolysis of water assuming a Nordic electricity mix (based on technology from NEL) and gasification of forest residues (based on technology from Cortus).

Use of Nordic average power production mix for calculation of emission factor for electricity imported from the grid is stipulated in Swedish law on reporting sustainability criteria for bio transportation fuels and liquid biofuels (STEMFS 2011:2, Chapter 7, §6). However, this is currently being re-evaluated and in the future, use of emission factor representative of Swedish average electricity production mix might be possible. The emission factor would then be reduced from 102 g/kWh to about 25 g CO_2/kWh.

Table 7 shows the production cost and CO_2 footprint, and Figure 34-Figure 37 show the breakdown of costs and CO_2 emission for the options based on data in *appendix 5: selected hydrogen technologies* and Table 8 gives the hydrogen demand. The results are shown with two different power prices, 20 EUR/MWh and 40 EUR/MWh.

Table 7 Production cost and CO_2 footprint for hydrogen from different raw materials and production paths. Values in brackets are for increased power price of 40 EUR/MWh instead of 20 EUR/MWh.

H_2 production	**EUR/kg H_2**	**EUR/MWh H_2**	**kg CO_2/kg H_2**
H_2 steam reforming Base Case	2.2	56	9.0
H_2 gasification	3.4 (3.5)	86 (89)	0.6
H_2 electrolysis	2.2 (3.2)	56 (81)	5.3

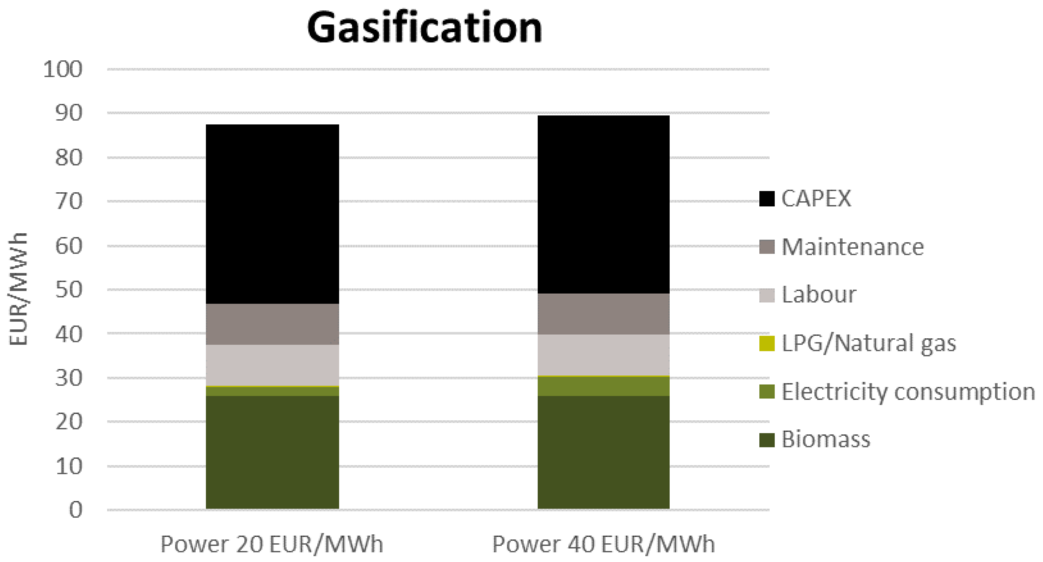


Figure 34. Total production cost in EUR per MWh H_2 for H_2 production by gasification.

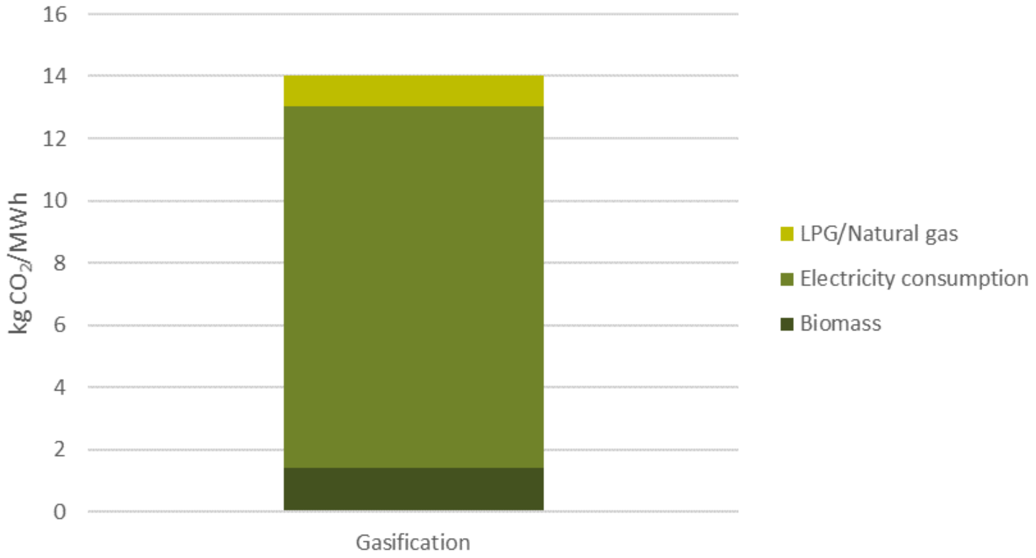


Figure 35. CO_2 emission in EUR per MWh H_2 for H_2 production by gasification assuming Nordic electricity mix.

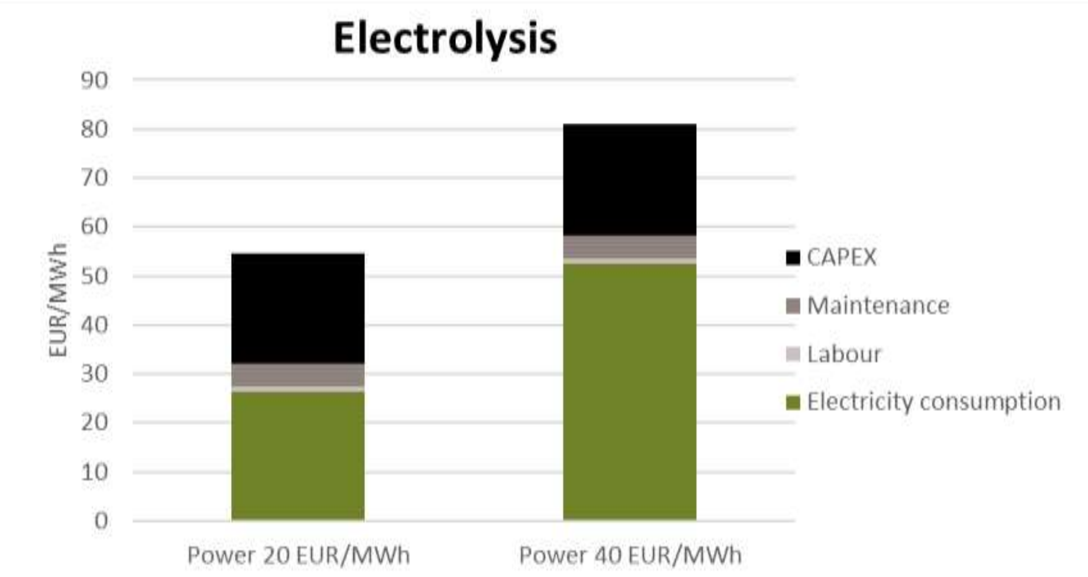


Figure 36. Total production cost in EUR per MWh H_2 for H_2 production by electrolysis.

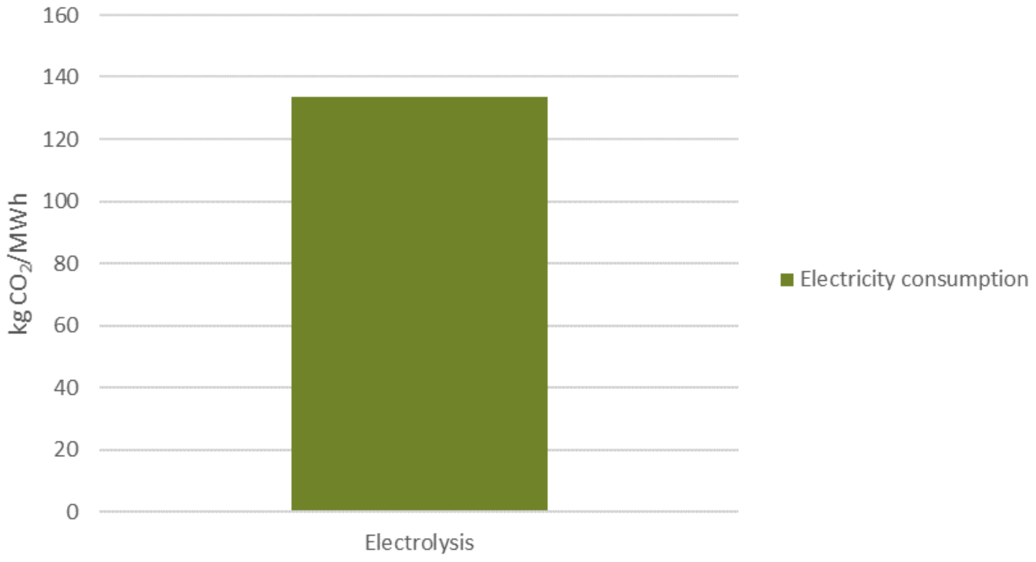


Figure 37. CO_2 emission in kg CO_2 per MWh H_2 for H_2 production by electrolysis assuming Nordic electricity mix.

Table 8. Hydrogen requirements to meet the transportation fuel component requirements.

		Pyr-FR	HydroPyr-FR	HTL-FR	HTL-MSL	HTL-LBL
H_2 required	**kg/MWh**	12.8	1.0	6.5	8.7	8.0

The total value chain cost for production of transportation fuel is found in Figure 38 and Figure 39.

The highest production costs are found for the value chains using electrolysis and gasification for hydrogen production since the production cost of H_2 is higher compared to steam reforming when applying a power price of 40 EUR/MWh. When applying a lower power price of 20 EUR/MWh, H_2 production cost by electrolysis become in the same range as for the steam reforming.

The highest production cost is found for the value chains with hydrogen production through gasification since the HDO plant is placed at the pulp mill instead of the refinery. Hence no avoided oil consumption from NCG and excess heat can be made in the refinery. Instead there is increased power production, but the income is lower compared to avoided oil consumption when the HDO Is located at the refinery. Please note that there is no gasification H_2 production case for hydropyrolysis, since no H2 is needed more than for the SynSat treatment at the refinery.

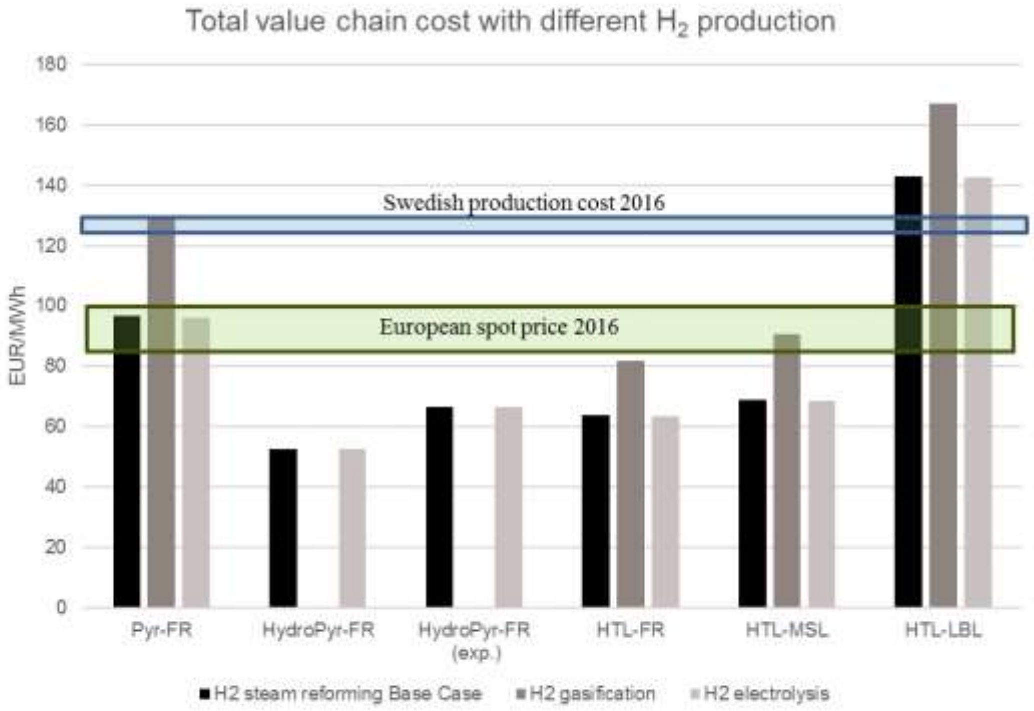


Figure 38. Power price 20 EUR/MWh. Total value chain cost with different production processes for H_2 (EUR per MWh transportation fuel), steam reforming = black, gasification = dark grey, electrolysis = light grey.

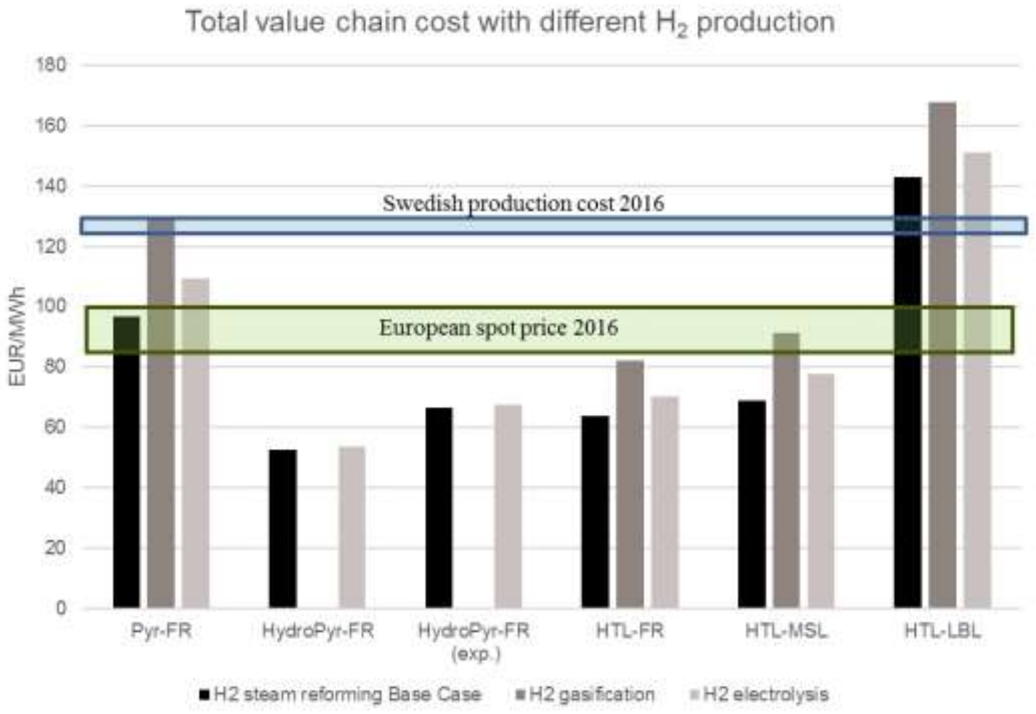


Figure 39. Power price 40 EUR/MWh. Total value chain cost with different production processes for H_2 (EUR per MWh transportation fuel), steam reforming = black, gasification = dark grey, electrolysis = light grey.

The total value chain CO_2 emissions for the hydrogen production technologies are shown in Figure 40.

The electrolysis case has the lowest CO_2 emission since it is placed at refinery and is therefore able to take credit for the avoided fuel oil consumption at the refinery though integration of excess energy, and in addition the H_2 production itself has lower CO_2 emissions than the steam reforming process assuming a Nordic electricity production mix. Using CO_2 emissions representative of Swedish electricity production would reduce the CO_2 emissions from H_2 production through electrolysis to 25% of that of Nordic electricity production mix which would reduce the CO_2 emission per kg H_2 produced by 75%. This would have a significant effect on the total environmental CO_2 emission for the total value chain (30-50 kg CO_2/MWh_{tf} further decrease).

A bit to our surprise, the hydrogen production through gasification showed the highest CO_2 emissions. This is due to the process integration and location effects again, where we have selected to place the gasification plant at the pulp mill due to the larger experience with handling and processing biomass than at the refinery. This also moves the location of the HDO upgrading to the pulp mill, and therefore the CO_2 savings made when utilising the excess heat from the HDO to replace fuel oil cannot be utilised. The utilisation of the excess heat from the HDO for power generation in the pulp mill has significantly lower benefits related to CO_2 emissions.

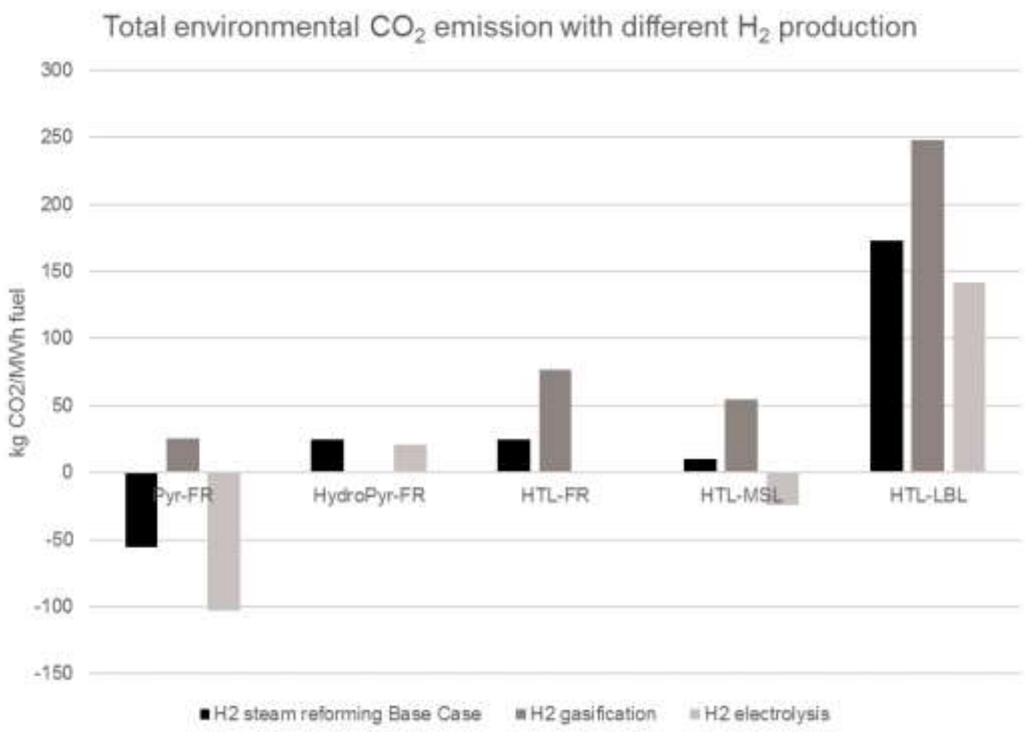


Figure 40. Total value chain emissions kg CO_2/MWh with different production processes for H_2, steam reforming = black, gasification = dark grey, electrolysis = light grey.

7 DISCUSSION AND CONCLUSIONS

This study investigates performance of emerging technologies for production of bio-based transportation fuel components through liquefaction of forest residues and kraft lignin in black liquor. The focus has been on technologies that produce bio-oils with low oxygen content compared to bio-oils from fast pyrolysis. The technologies are evaluated and compared based on some key characteristics that are important for the performance of the value chains such as cost and environmental impact. The main conclusions and identified risks and opportunities for discussion are summarised below.

7.1 OXYGEN CONTENT IN BIO-OIL AND YIELD OF TRANSPORTATION FUEL COMPONENTS

Hydrothermal liquefaction (HTL) and hydropyrolysis have been identified as promising technologies, with bio-oil oxygen content between 15-21% w for HTL, depending on raw material and HTL process conditions, and 0% for hydropyrolysis with integrated hydroprocessing. This is to be compared with the oxygen content of pyrolysis oil, which is around 40%.

The yield of bio-oil and transportation fuel components from the emerging technologies are high, around 60-70% up to 87% for HTL on forest residues compared to around 50-55% on fast pyrolysis of forest residues on energy basis. This indicates a high degree of utilisation of the forest raw material in transformation to the end-product transportation fuel for the emerging technologies. The carbon efficiency is also high for the emerging technologies, especially for HTL. For hydropyrolysis, the yield and carbon efficiency are negatively affected by the integrated hydrotreatment, as the off-gas formation is set to match the hydrogen demand.

The yield of bio-oil in the pyrolysis process is negatively affected by the high content of alkali metal content in forest residues. The pyrolysis process would be more competitive to the emerging technologies if a way to reduce the feedstock alkali content was found, or if the feedstock was changed to saw dust or other wood-based feedstock with low alkali.

7.2 PRODUCTION COST AND PROFIT POTENTIAL

It is concluded that there is an economic potential for establishing integrated value chains utilising forest residues or kraft lignin as raw material for production of transportation fuel components in the diesel and gasoline range. The sensitivity study performed with raw material cost and investment cost varying with +-50% confirms that this conclusion is stable.

The main costs of producing transportation fuel components from the biomass are related to the raw material cost and the cost of producing the depolymerised lignin-oil.

There is not a single emerging technology that is obviously more favourable than the other technology in converting forest residues into transportation fuels, however the emerging technologies based on hydrothermal liquefaction (HTL) and hydropyrolysis show lower production cost compared to pyrolysis. The transportation fuel component production cost is about 100 EUR/MWh (about 0.9 EUR/l) for fast pyrolysis, while the emerging technologies have a production cost in the range of 65 EUR/MWh (about 0.6 EUR/l). The margin to the Swedish bio-fuel production cost of 122-127 EUR/MWh transportation fuel and the European spot market price of 82-100 EUR/MWh transportation fuel is acceptable for the emerging technologies, but critical for fast pyrolysis. On the other hand, HTL and hydropyrolysis have a lower technical maturity than pyrolysis and the estimates for these technologies are accordingly more uncertain in many aspects.

There is also an economic potential in using lignin as feedstock for transportation fuel production that seems to be in the same order of magnitude as for the emerging technologies for forest residues. The transportation fuel component production cost can also here be in the range of 65 EUR/MWh. However, the technical maturity is also here considered as low, which results in a large uncertainty range. The sensitivity study performed indicates that there is still a margin for increased investment costs and loss of product yield without removing the entire profit margin. The main uncertainty is then technical show-stoppers that might not be properly addressed yet.

7.3 CO_2 EMISSION EVALUATION OF VALUE CHAIN

The total value chain CO_2 emissions from these value chains are lower than the CO_2 footprint of fossil transportation fuels. Reductions of 95% or more are found when integration aspects with pulp mill and refinery are included, with value chain emission levels of 10-25 kg CO_2/MWh_{tf} compared to 300 kg CO_2/MWh_{tf} for fossil alternatives. Even negative emissions are achieved, thanks to energy integration aspects with the oil refinery that have been accounted for. This analysis is only considering CO_2 emissions and not emissions of other greenhouse gases.

A risk that has been identified in this work is tendency for repolymerisation and the associated need of a chemical capping agent, which has been found to significantly increase costs and the CO_2 impact of the lignin value chains. This is demonstrated in the value chain with HTL of LignoBoost lignin, where the use of phenol as capping agent results in high associated emissions of CO_2, resulting in a CO_2 reduction of only about 45% compared to fossil transportation fuels and production costs that are not competitive.

In general, addition of chemicals has been identified to contribute with high costs and also significant addition to the value chain CO_2 emissions. Use of green chemicals would be beneficial for the process environmental impact.

7.4 EFFECT OF REFINERY INTEGRATION ON COST AND CO_2 EMISSIONS

An aspect identified as of great importance for cost and CO_2 emission is the integration of the bio-oil production process and the hydrodeoxygenation and upgrading processes in the pulp mill and in the oil refinery respectively. Especially the integration of excess energy in the form of heat and energy rich non-condensable gases from HDO has a positive contribution to reducing costs and CO_2 emissions through its potential to replace fuel oil for heating purposes at the refinery. However, the true potential for energy integration in the refinery need to be verified with higher degree of certainty.

Even though the heat integration has turned out to give a large positive contribution to CO_2 reductions in the value chains investigated, this contribution would not be accounted for when determining the fuel components CO_2 footprint according to the rules set out in the current RED and ILUC directives. Additional rules related to the reporting according to the above directives will also make the transportation fuel’s CO_2 footprint look significantly different than that reported for the value chain. This should be further investigated and discussed as it has a large impact on the commercial aspects of bio transportation fuel production.

7.5 ALTERNATIVE HYDROGEN PRODUCTION TECHNOLOGIES

Cost for hydrogen is significant (10-30% of total cost). Selecting the emerging technologies that produce a bio-oil with a significantly lower O-content, 20% or lower, compared with almost 40% for pyrolysis, significantly reduces the need of hydrogen and cost.

The source of hydrogen for bio-oil upgrading has a large impact on both costs and CO_2 emissions. Changing from natural gas steam reforming for hydrogen production to electrolysis of water or gasification of forest residues resulted in almost the same hydrogen production cost, however with significantly lower CO_2 footprint.

Integrating hydrogen production through electrolysis in the value chain results in a quite similar transportation fuel production cost as when using natural gas steam reforming, when assuming a relatively low electricity price of 20 EUR/MWh, while the CO_2 emissions are significantly reduced (assuming CO_2 emissions representative of an average Nordic electricity production mix). Assuming a higher electricity price of 40 EUR/MWh increases the total costs in the order of a few percent.

Integrating hydrogen production from gasification increases the costs and the CO_2 emissions compared to natural gas reforming. This is due to effects caused by moving the location of the hydrogen production and HDO plants to the pulp mill location, where there is easy access to raw material and knowhow on biomass handling for the gasification plant. The integration opportunities for the excess heat are not as favourable as at the refinery when it comes to generating a value from the excess heat and CO_2 emission reductions.

7.6 IDENTIFIED RISKS AND OPPORTUNITIES

The current CO_2 allocation and reporting rules according to RED and iLUC directives result in high CO_2 footprints for some of the production paths evaluated in this report, i.e. fast pyrolysis and paths based on kraft lignin as feed. This is an effect of not acknowledging the positive effect of heat integration on value chain CO_2 emissions. The footprint of these technologies could be improved by implementing hydrogen production with low CO_2 footprint and ensuring that the emissions from the Swedish electricity production mix can be used in the evaluation. Another opportunity is to use the hydrocarbon containing NCGs for hydrogen production rather than for heating purposes.

There is a risk associated with integration of the studied value chains in conventional refinery processes related to the inorganic content in both forest residues and lignin. The alkali metals could cause deactivation of downstream catalysts with associated need for regeneration and low availability as consequence.

The selected alternative hydrogen technologies, alkaline electrolysis of water and gasification of biomass residues, are based on today's commercially available technologies which may not be representative for low cost hydrogen in the future. Significant efforts are ongoing today in developing more efficient hydrogen production technologies, especially related to electrolysis.

The investigated emerging technologies for bio-oil production (e.g. HTL and hydropyrolysis) and upgrading to transportation fuel components have a relatively low technical maturity for the assumed feedstock and will require further technical and economic development and verification, including more operational experience in both pilot and demonstration scale.

8 SUGGESTIONS FOR CONTINUED WORK

While performing this study, some areas of interest for continued investigations have been identified. The suggested areas are described in the text below.

- It would be beneficial if more cases of emerging technologies for depolymerisation and upgrading of forest residues and lignin could be included for comparison. It is currently very difficult to gain access to sufficient data on some of the identified technologies in *appendix 1: screening of technologies for bio-oil production* to carry out comparable analyses. It is recommended that the research funding agencies use their influence to promote a certain level of openness with data in funded projects to allow this type of analysis.

- Additional raw materials should be added to broaden the analysis of options.

- The described HTL process developed by Chalmers should be further developed to significantly reduce or eliminate the cost of the depolymerisation capping agent. For instance, one should investigate the effect on cost and CO_2 emissions of omitting the capping agent and accept a higher coke formation and find way to cope with the associated operational difficulties.

- The HTL, hydropyrolysis and bio-oil hydrodeoxygenation and upgrading process need to be studied experimentally in more detail to provide better quality data for performance estimations. It is known that work is ongoing in this area today, and as results are made public, the estimations in this report should be updated. Further verification and operational experience from pilot and demonstration scale are needed.

- Detailed investigation of the heat integration opportunities at the refinery should be made, considering several integration aspects such as available temperature levels on heat sources and sinks, geographic and cost restrictions for integration. In addition, integration aspects should be investigated based on the rules set out in the renewable energy directive on the calculation of the CO_2 footprint of the produced transportation fuel.

- Using off-gases and NCG for integrated production of hydrogen, potentially combined with other hydrogen production technology) should be investigated to identify potential cost benefits and provide a processing path that should be more beneficial from the point of view of reporting CO_2-footprint according to the RED.

- An option to reduce the CO_2 impact from steam natural gas reforming would be to combine it with capture and storage or utilisation of the emitted CO_2, (so called CCUS). Engineering studies of this option is ongoing in Norway by the fertiliser producer Yara AS.

- The technology development within the field of electrolysis should be followed to evaluate how well it could compete with steam methane reforming. This relates to ongoing research at e.g. Umeå University in the field of new electrodes for electrolysis expected to significantly lower the production costs for hydrogen, (Pham et al., 2017).

- Pre-treatment and fractionation of the raw material should be considered, especially for forest residues. The fractions containing high levels of inorganics and oxygen could be mechanically separated through removal of needles, small twigs and bark. Oxygen removal could be considered through various options for carbonisation.

- The produced bio-oils contain high levels of oxygen containing components, components with aromatic structures and unsaturated hydrocarbons that require a significant amount of hydrogen to transform to transportation fuel components. It should be investigated if it is economically beneficial to separate some of these components for chemical production. This would both save hydrogen and provide a platform for chemical production and increased economic profitability.

- The importance of a high yield of transportation fuel components and the feasibility of using biomass raw material for hydrogen production should be investigated from a perspective where the limitations of the availability of biomass is considered.

NOMENCLATURE AND ABBREVIATIONS

ABBREVIATIONS	
AD	Anaerobic Digestion
ADt	Air Dried ton
ARA	Amsterdam, Rotterdam, Antwerp
BB	Bark Boiler
BCD	Base catalyzed depolymerisation
CAPEX	Capital Expences
CCUS	CO_2 Capture, Storage and Utilisation
CATLIQ	Catalytic Liquefaction, technology being commercialised by SCF Technologies A/S
COP	UNFCCC (United Nations Framework Convention on Climate Change) Conference of the Parties
CRI	CRI Catalyst company (CRICC), part of CRI/Criterion Inc., the global catalyst technology company of the Shell Group
IH2	Integrated hydropyrolysis and hydrotreating, technology being commercialised by GTI and CRI
ILUC	Indirect Land Use Change directive from 2015, regulations in Directive 2015/1513/EC
EC	Equipment Cost
ESP	ElectroStatic Precipitator
FOB	Free on Board
FR	Forest Residues
FQD	Fuel Quality Directive (FQD) (Directive 2009/30/EC)
GTI	Gas Technology Institute, American independent technology organization involved in development of new energy technologies, technical insight and training.
HDO	Hydrodeoxygenation
HDS	Hydrodesulfurized
HHV	Higher Heating Value
HP	High Pressure
HTL	Hydrothermal Liquefaction
HTS	High Temperature Shift
HVO	Hydrogenated Vegetable Oils
HydroPyr	Hydropyrolysis with integrated hydrogenation
LBL	LignoBoost Lignin
LF	Lang Factor
LGO	Light Gas Oil
LHV	Lower heating Value
LPG	Light Petroleum Gas
LSL	Membrane Separated Lignin
ME	Missing Equipment
NCG	Non-condensable Gases
OPEX	Operational Expences
PNNL	Pacific Northwest National Laboratory, one of the United States Department of Energy national laboratories, managed by the Department of Energy's (DOE) Office of Science.
p	Pressure
PSA	Pressure Swing Absorption
RB	Recovery Boiler
RED	Renewable Energy Directive (RED) (Directive 2009/28/EC)
SEP	Separation, fractionation
SMR	Steam Methane Reforming
SOT	State of Technology
ST	Steam
T	Temperature
TCI	Total Capital Investment
TOFA	Tall Oil FattyAcid

TPCE	Total Purchased Equipment Cost
VVX	Heat Echanger
WGS	Water Gas Shift
WWT	Waste Water Treatment
SUBSCRIPTS	
tf	End product transportation fuels diesel and gasoline
f	Biomass feedstock used as basis for transportation fuel production, i.e. forest residues or kraft lignin
th	Thermal
CASE NAMES	
Pyr-FR	Fast pyrolysis of forest residues
HydroPyr-FR	Hydropyrolysis of forest residues
HTL-FR	Hydrothermal liquefaction of forest residues
HTL-MSL	Hydrothermal liquefaction of membrane separated lignin
HTL-LBL	Base catalysed depolymerization of kraft lignin from the LignoBoost process

REFERENCES

Anheden M, Ehn C, Lundberg V, Pettersson P, Fugelsang M, Hjerpe C-J, Håkansson Å, Gunnarsson I, 2016, *Value chains for production of renewable transportation fuels using intermediates – Case studies for bio-oils in refinery applications and Bio-SNG*, Report No 2016:05, f3 The Swedish Knowledge Centre for Renewable Transportation Fuels, Sweden. Available at www.f3centre.se.

Anheden M, Ulin A, Wolf J, Hedberg M, Berg R, Ankner T, Berglin N, von Schenck A, Larsson A L, Guimaraes M, Fiskerud M, Andersson S ”Value Chain for production of bio-oil from craft lignin for use as biojet fuel, Nordic Wood Biorefinery Conference, March 28-30, 2017.

Arkell A, Olsson J, Wallberg O ”Process Performance in lignin separation from softwood black liquor by membrane filtration, Chem. Eng. Res. & Design, 92 (2014) 1792-1800.

Belkheiri T, Andersson S-I, Mattsson C, Olausson L, Theliander H, Vamling L (2018a) *Hydrothermal liquefaction of kraft lignin in sub-critical water: influence of phenol as capping agent*. Submitted for publication jan 2018 to Energy and fuels.

Belkheiri T, (2018b) *LIGNIN HYDROTHERMAL LIQUEFACTION IN SUBCRITICAL WATER TO PRODUCE BIOFUEL AND CHEMICALS,* PhD Thesis, to be presented in April 2018. Chalmers University of Technology, Department of Energy and Environment.

Benjaminsson G, Benjaminsson J, Bengtsson N (2013) ”Decentraliserad produktion av pyrolysolja för transport till storskaliga kraftvärmeverk och förgasningsanläggningar” Gasefuels AB, Energimyndigheten.

Berglin N L (2011) The 2010 reference mill for kraft market pulp. *TAPPI Peers Conference.* Portland, Oregon.

BTG-BTL (2014). *The Empyro pyrolysis plant*. Empyro project brochure. Available: https://www.btg-btl.com/media/cms_block/leafletempyro.pdf

Börjesson P and Gustavsson L “Regional production and utilization of biomass in Sweden”. Energy, 1996, 21(9): p. 747-764.

CEPI. Annual Statistics 2007 -European Pulp and Paper Industry, 2008, Confederation of European Paper Industries: Brussels.

Collodi G (2010). *Hydrogen Production via Steam Reforming with* CO_2 *Capture*. http://www.aidic.it/CISAP4/webpapers/7Collodi.pdf. Retrieved 2016-03-30.

Cortus: see company information at http://www.cortusenergy.com/

Directive EU 2015/1513, DIRECTIVE (EU) 2015/1513 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 9 September 2015 amending Directive 98/70/EC relating to the quality of petrol and diesel fuels and amending Directive 2009/28/EC on the promotion of the use of energy from renewable sources, http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32015L1513&from=SV

de Jong S, Hoefnagels R, Faaij A, Slade R, Mawhood B and Junginger M "The feasibility of short-term production strategies for renewable jet fuels – a comprehensive techno-economic comparison". Biofpr 9:778–800 (2015).

de Jong S, Hoefnagels R, Wetterlund E, Pettersson K, Faaij A, Junginger M "Cost optimization of biofuel production – The impact of scale, integration, transport and supply chain configurations". Applied Energy 2017; 195: 1055-1070.

Ekonomifakta. http://www.ekonomifakta.se/Fakta/Energi/Styrmedel/Konsumtionsskatter-pa-diesel/ 2016 [cited 2016-01-28].

Eliasson et al. ”Flisning av bränsleved och delkvistad energived med en stor trumhugg–CBI 6400”. Skogforsk 2014.

Elliott D C, Biller P, Ross A B, Schmidt A J and Jones S B "Hydrothermal liquefaction of biomass: Developments from batch to continuous process". [Online]. Available at: http://linkinghub.elsevier.com/retrieve/pii/S0960852414013911Bioresour Technol [Internet] Elsevier Ltd 178:147–156 (2015) [October 28 2014].

Elliott, D. (2016). *Hydrothermal liquefaction of biomass, revisited. Doug Elliott of Pacific Northwest National Laboratory, USA, revisits his earlier article.* In IEA Bioenergy Newsletter 38. October 2016. Available: http://task34.ieabioenergy.com/wp-content/uploads/2016/10/Pyne-Newsletter-38-FINAL.pdf

Energimyndigheten (2015). *Trädbränsle- och torvpriser*, Eskilstuna, Energimyndigheten och Örebro, Statistiska centralbyrån.

Energimyndigheten (2016). *Förslag till styrmedel för ökad andel biodrivmedel i bensin och diesel. En rapport inom uppdraget Samordning för energiomställning i transportsektorn.* Rapport ER 2016:30. Statens Energimyndighet.

Energimyndigheten (2017a) "Övervakningsrapport avseende skattebefrielse för flytande biodrivmedel". 2017-02-27, Dnr 2016-012009.

Energimyndigheten (2017b). *Strategisk plan för omställning av transportsektorn till fossilfrihet*. Available: http://www.energimyndigheten.se/klimat--miljo/fossilfria-transporter/nationell-strategi-for-att-stalla-om-till-en-fossilfri-transportsektor/ (2017-06-30).

Ensyn (2017). Available: www.ensyn.com, accessed 2017-12-01

Envergent (2017). Available: www.envergenttech.com, accessed 2017-12-01

EUROPAPARLAMENTETS OCH RÅDETS DIREKTIV (2009) EUROPAPARLAMENTETS OCH RÅDETS DIREKTIV 2009/28/EG av den 23 april 2009, Europeiska unionens officiella tidning 5.6.2009.

Folkelid, M. N. (2017), Cortus, personal communication.

Fredén J "Analysis and calculations of emission factors for green house gases" in Department of Energy and Technology, 2010, Swedish University of Agricultural Sciences, Uppsala, Sweden.

Gode J, Martinsson F, Hagberg L, Öman A, Höglund J, and Palm D. Miljöfaktaboken 2011. ”Uppskattade emissionsfaktorer för bränslen, el, värme och transporter” 2011, Värmeforsk, Stockholm.

GTI, Gas Technology Institute (2009). Integrated Hydropyrolysis and Hydroconversion Process for Production of Gasoline and Diesel Fuel from Biomass, Extended Abstract, AICHE November 8-13, 2009.

Gas Technology Institute (GTI) (2012). Biomass to Gasoline and Diesel Using Integrated Hydropyrolysis and Hydroconversion. U.S. DOE Award DE-EE-0002873 Technical report. Illinois: Gas Technology Institute.

Guizani C et al. “Effects of CO_2 on biomass fast pyrolysis: Reaction rate, gas yields and char reactive properties”. C. Fuel 116 (2014) 310–320.

Holmgren K, Eriksson E, Olsson O, Olsson M, Hillring B and Parikka M “Biofuels and climate neutrality - system analysis of production and utilization”. 2007, Elforsk, Stockholm, Sweden.

Huiyan Zhang et al. “Biomass fast pyrolysis in a fluidized bed reactor under N_2, CO_2, CO, CH_4 and H_2 atmospheres”. Bioresource Technology 102 (2011) 4258–4264.

Hsu C S, Robinson P R, 2006, “Practical Advances in Petroleum processing”. Vol. 1, Springer Science + Business Media Inc.

Håkansson Å (2015), Preem AB, personal communication

IAE Bioenergy; 2007:1 Biomass Pyrolysis

Igjerstadsidene (2017) “Utsatte sak om investering av 400 000 kroner i Åmli”. R Hagestrand, 2017-03-24. Available: http://gjerstadsidene.no/utsatte-sak-om-investering-av-400-000-i-amli

Iversen S B, Larsen T, Lüthje V, Felsvang K, Nielsen P R, Galla U and Boukis N ”CATLIQ – a disruptive technology for biomass conversion”. 14th European Biomass Conference, 2005, Paris, p. 1450.

Jones S, Tan E, Jacobson J, Meyer P, Dutta A, Cafferty K, Snowden-Swan L, Padmaperuma A, 2013, “Process Design and Economics for the Conversion of Lignocellulosic Biomass to Hydrocarbon Fuels. Fast Pyrolysis and Hydrotreating Bio-Oil pathway”, PNNL-23053, NREL/TP-5100-61178.

Jones S, Valkenburg C, Walton C W, Elliott D C, Holladay J E, Stevens D J, Kinchin C, Czernik S, PNNL-18284, “Production of Gasoline and Diesel from Biomass via Fast Pyrolysis, Hydrotreating and Hydrocracking: A Design Case”, February 2009.

Langås, H. G., 2017, NEL, personal communication

Löfstedt, J, Dahlstrand C, Orebom A, Meuzelaar G, Sawadjoon S, Galkin M V, Agback P, Wimby M, Corresa E, Mathieu Y, Sauvanaud L, Eriksson S, Corma A, Samec J S M, *Green Diesel from Kraft Lignin in Three Steps.* ChemSusChem 2016, 9, 1392-1396 and supporting documentation.

Network for Transport and Environment,NTM, *Road transport*, version 2010-06-17 (100603 NTM INT ROAD) (Source: HBEFA 3.1, processed by NTM.)

Network for Transportation and Environment, NTM, *Rail transport*, version 2008-03-14 (080314 NTM INT RAIL)

Nordpool 2015. https://www.nordpoolgroup.com/Market-data1/#/nordic/table

Peters M S, Timmerhaus K D, West R E (2003), "Plant Design and Economics for Chemical Engineers". McGraw-Hill, New York, Fifth Edition.

Pham, T., N., Sharifi, T., Sandström, R., Siljebo, W., Shukarev, A., Kordas, K., Wågberg, T., Mikkola, J.-P., 2017, " Robust hierarchical 3D carbon foam electrode for efficient water electrolysis", Scientific Reports 7, Article number: 6112, http://www.nature.com/articles/s41598-017-05215-1

SCB, 2017, Utsläpp till luft, Utsläpp och intensitet per år 2008-2016, preliminär statistik, http://www.scb.se/hitta-statistik/statistik-efter-amne/miljo/miljoekonomi-och-hallbar-utveckling/miljorakenskaper/

SKM. El-certificate price history, 2013. Svensk Kraftmäkling (SKM).

STEMFS 2011:2, Statens energimyndighets föreskrifter om hållbarhetskriterier för biodrivmedel och flytande biobränslen, ISSN 1650-7703.

SOU 2013:84 Fossilfrihet på väg – Betänkande av Utredningen om fossilfri fordonstrafik, Stockholm 2013.

SunCarbon (2016). SunCarbon Power point presentation 2016-09-28. SunCarbon.

Tews I J, Zhu Y, Drennan C V, Elliott D, Snowden-Swan L J, Onarheim K et al. "Biomass direct liquefaction options: technoeconomic and life cycle assessment". Pacific Northwest National Laboratory, PNNL 23579, (2014).

Tomani P,"The LignoBoost Process" Cellulose Chem. Technol., 44 (1-3), 53-58 (2010).

TU, Teknisk Ukeblad (2016). «Coca-Cola-oppskrift» kan gjøre hydrogen til nytt norsk industrieventyr. Stensvold, T., 26 January 2016. Available: https://www.tu.no/artikler/coca-cola-oppskrift-kan-gjore-hydrogen-til-nytt-norsk-industrieventyr/276348

Valmet (2013). *Fortum's bio-oil plant commissioned in Joensuu.* Article, published online 12 December 2013. Available: http://www.valmet.com/media/articles/all-articles/fortums-bio-oil-plant-commissioned-in-joensuu/

Yildiz et al. "Challenges in the design and operation of processes for catalytic fast pyrolysis of woody biomass". Renewable and Sustainable Energy Reviews 57 (2016) 1596–1610.

Ycharts 2015. European Union Natural Gas Import Price. Available at https://ycharts.com/indicators/europe_natural_gas_price . Accessed: 2016-03-16

Zhu Y, Tjokro Rahardjo S, Valkenburg C, Snowden-Swan L, Jones S and Machinal M "Techno-economic Analysis for the Thermochemical Conversion of Biomass to Liquid Fuels". Pacific Northwest National Laboratory, Richland, US (2011).

Zhu Y, Biddy M J, Jones S B, Elliott D C and Schmidt A J "Techno-economic analysis of liquid fuel production from woody biomass via hydrothermal liquefaction (HTL) and upgrading". Appl Energy 129:384–394 (2014).

APPENDIX 1: SCREENING OF TECHNOLOGIES FOR BIO-OIL PRODUCTION

The following technologies were included in the list of depolymerisation technologies considered to be included in this study:

Forest residues:

- Fast catalytic pyrolysis (Royal Institute of Technology)
- CATLIQ (Altaca)
- Hydrothermal Liquefaction – Hydrofraction (Steeper Energy)
- Hydrothermal Liquefaction (Pacific North West National Laboratory, PNNL) – state of the art and goal case
- Hydropyrolysis (IH2)
- Bio-oil through Auger reactor
- AlphaKat (Kemiinformation)

Kraft lignin:

- Hydrothermal Liquefaction (near supercritical, based on work by Chalmers)
- Three-step procedure (based on Renfuel)
- Catalytic Hydrothermal reactor, Cat – HTR (Licella)
- Black Liquor pyrolysis (Huntsman)
- SunCarbon HTL cracking (SunCarbon)

Of these following two cases were selected for the study on forest residues:

- Hydropyrolysis (IH2)
  Motivation: Plenty of published data required for estimations, there is an experimental basis for the published data, there is a commercial actor involved in promoting the technology, the technology results in very low oxygen level in the produced bio-oil, investment cost estimate is available (however a bit uncertain and seems low)
- Hydrothermal Liquefaction (Pacific North West National laboratory)
  Motivation: Very good access to data, investment estimates are available, based on experimental data but in small scale, goal case is based on targeted data that has not been proven

The two selected cases using lignin as raw material are:

- Hydrothermal Liquefaction (near supercritical) (Chalmers)
  Motivation: Good access to data, data is based on experiments in lab-scale, rough investment estimate is available (however some remaining development work around recycle of process fluids are required)

- SunCarbon HTL cracking (SunCarbon)
  Motivation: SunCarbon is willing to collaborate and provide data for process evaluation and investment estimate

The absolute dominating determining factor in the selection of process options were the ability to get access to data for process evaluation. Many of the emerging technologies lack in this respect, which makes the type of analysis as presented in this report impossible.

APPENDIX 2: SCREENING OF ALTERNATIVE TECHNOLOGIES FOR HYDROGEN PRODUCTION

The following technologies were considered in the selection process for alternative technologies to natural gas steam reforming for hydrogen production:

Based on use of forest residues and other biomass:

- Hydrogen-rich syngas (or in the report “Gasification”) (Cortus)
- Hydrogen rich syngas by oxygen blown gasification of black liquor (Chemrec)
- Hydrogen containing syngas by air blown gasification of black liquor (Chemrec)

Based on electrolysis:

- Atmospheric alkaline electrolyser (NEL)
- Rotolyzer (NEL)
- PEM electrolyser (Hydrogenics)
- Solid oxide electrolysis (DTU Energy)

Of these, the following two technologies were selected:

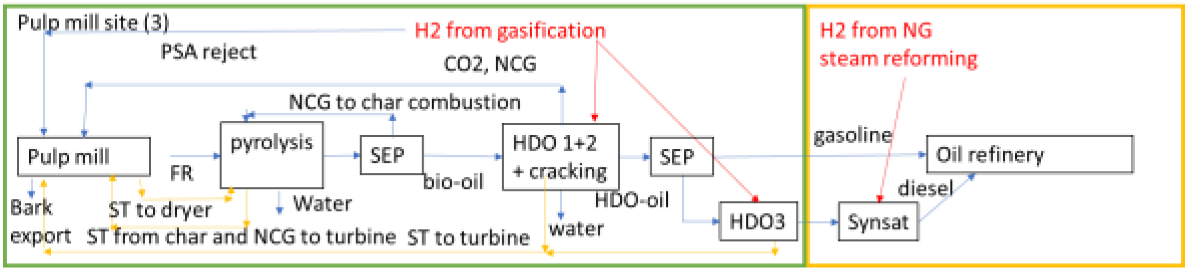
- Gasification (Cortus)
  Motivation: Is promoted by a commercial actor, good access to data
- Atmospheric alkaline electrolyser (NEL)
  Motivation: Good access to data, commercial process and actor, access to data relevant for Swedish conditions

Again, the data access has been the determining factor, in combination with that the technologies are supported by commercial actors and the technical maturity.

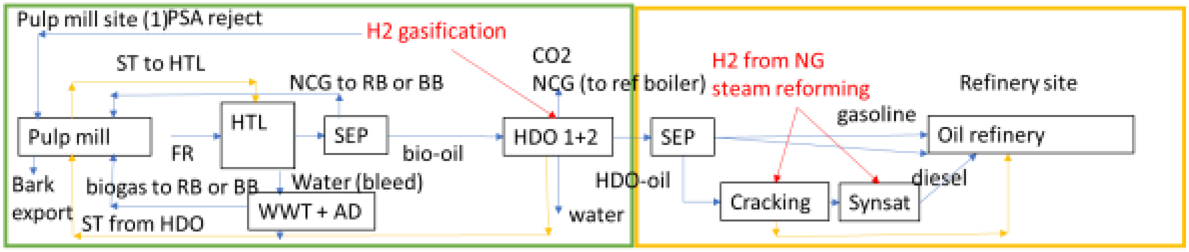
APPENDIX 3: INTEGRATION OF HYDROGEN PRODUCTION THROUGH GASIFICATION AND ELECTROLYSIS IN THE STUDIED VALUE CHAINS

HYDROGEN FROM GASIFICATION OF FOREST RESIDUES

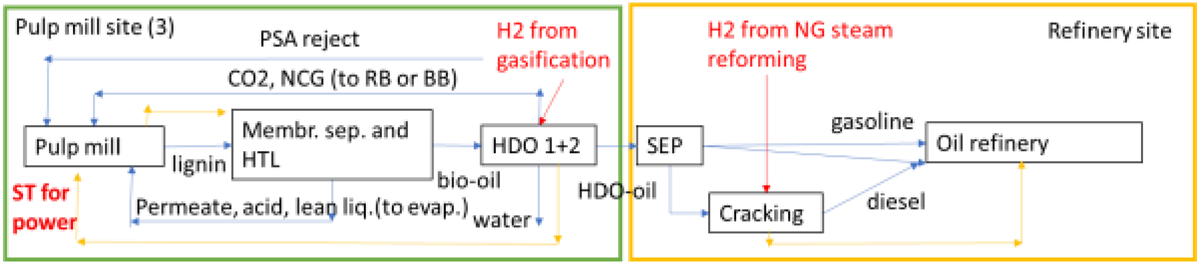
Pyr-FR



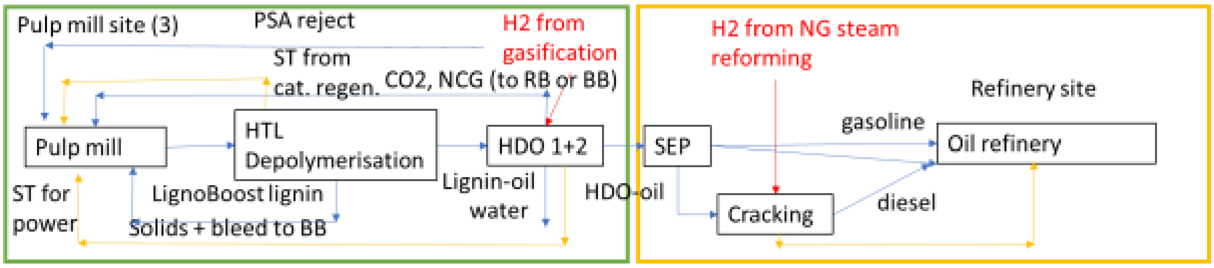
HTL-FR



HTL-MSL

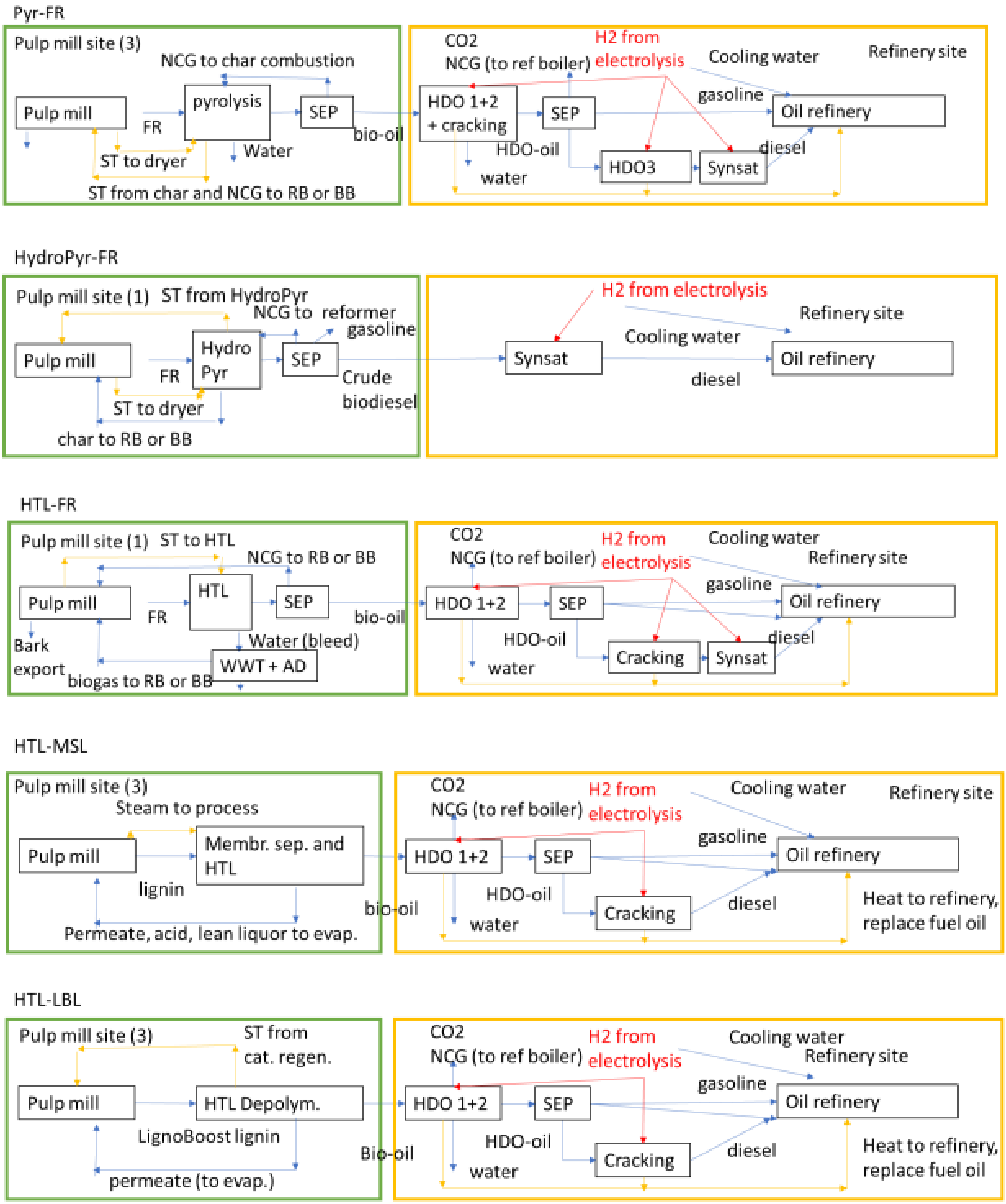


HTL-LBL



Integration of value chains with pulp mill and oil refinery using fast pyrolysis of forest residues (Pyr-FR),HTL of forest residues (HTL-FR), HTL (low T, p) of kraft lignin retentate from membrane separation (HTL-MSL) Note: lignin depolymerisate is mixed with tall oil pitch or VGO to form bio-oil HTL (high T, p) of LignoBoost lignin, (HTL-LBL). Note: The value chain with hydropyrolysis of forest residues is not relevant for integration with gasification since hydrogen is supplied internally through reforming of off-gases.

HYDROGEN FROM ALKALINE ELECTROLYSIS OF WATER, ASSUMING NORDIC ELECTRICITY MIX FROM THE GRID



Integration of value chains with pulp mill and oil refinery using fast pyrolysis of forest residues (Pyr-FR) hydropyrolysis of forest residues (HydroPyr FR) HTL of forest residues (HTL-FR) HTL (low T, p) of kraft lignin retentate from membrane separation (HTL-MSL) Note: lignin depolymerisate is mixed with tall oil pitch or VGO to form bio-oil e) HTL (high T, p) of LignoBoost lignin, (HTL-LBL).

APPENDIX 4: DETAILED INPUT DATA AND ASSUMPTIONS

This section presents the input data and assumptions for the studied value chains.

GENERAL ASSUMPTIONS

All flows of biomass and other energy have been converted to energy units, based on higher heating value (HHV). The operating time is assumed to be 7840 h/y for all studied value chains.

All investment costs are in year 2015 money value. Adjustments have been made using Chemical Engineering Plant Cost Index (CEPCI). The annuity factor is set to 0.1, which for example corresponds to an interest rate of 8% and a life time of 20 years.

The data used in this study is generally based on today’s situation. However, for relevant parameters a sensitivity analysis is performed to show how the results are influenced.

RAW MATERIAL

Forest residues

The composition and heating value of forest residues assumed as feedstock in the Pyr-FR, Hydro-Pyr-FR and HTL-FR value chains are given in the table below.

Table 9. Composition and heating value of forest residue used as feedstock.

Heating value (MJ/kg)	Moisture content (wt%)	C (wt%)	H (wt%)	N (wt%)	O (wt%)
19.4 (dry, LHV) 8.57 (wet, LHV) 20.7 HHV dry	50	51	6.1	0.4	40

In this study, outtake of forest residues (branches and tops) in Sweden is assumed to be 20 TWh/y, which is in line with the potential presented in Wetterlund et al. (2014). It is assumed that 75% of this potential is available for usage in the applications studied here. The pulp mills are assumed to be located in more forest rich regions than the refinery and the district heating system.

Estimations of future potential for forest biomass indicate a significantly larger potential than what is currently used. According to scenarios presented in (Wetterlund et al. 2014), the potential for forest residues 2030 are estimated to 28-31 TWh/y. In addition, the potential for stumps 2030 is estimated to 23-25 TWh/y. Table 10 summaries the data for the forest residues outtake and usage.

Table 10. Data for forest residues outtake and usage.

		Base	**Sensitivity analysis**
Forest residues outtake	TWh/y	20	29
Usage of total outtake	%	75	-
Area Sweden	km^2	449964	-
Usage around pulp mill[1]	MWh/y/km^2	36	54
Usage around refinery and DH system[2]	MWh/y/km^2	18	27

[1] 14% above assumed average outtake.

[2] 43% below assumed average outtake.

The energy demand (in the form of diesel fuel) associated with the outtake of forest residues is estimated to 3 kWh/MWh (Eliasson 2014).

Black liquor lignin

Two different methods are considered for separation of lignin from black liquor, the LignoBoost process and membrane separation of lignin.

LignoBoost lignin

It is assumed that lignin separation is done according to the “LignoBoost” concept, in which lignin is precipitated from black liquor by injecting CO_2, thereby lowering the pH and causing an agglomeration of lignin molecules. The precipitated lignin is separated and then washed with acidified condensate from the evaporation plant (H_2SO_4 is used as the acidifier). The filtrates from the lignin separation plant are recirculated to the evaporation plant. Details about the LignoBoost process can be found in Tomani (2010).

The depleted black liquor is returned to the mill for combustion in the recovery boiler. It is assumed that each pulp mill has the capacity to extract a maximum of 75 000 t_{lignin}/y i.e. approximately 110 kg/ton pulp (ADt) without affecting the operability of the recovery boiler. This corresponds to ~20% of the lignin in black liquor. Lignin extraction at three pulp mills is required to obtain sufficient raw material for the production of 100 000 t/y transportation fuel components.

Lignin extraction affects the energy balance of the pulp mill. First, less organic material is sent to the recovery boiler and therefore less steam and thereby also power is produced. Second, the lignin extraction process demands electricity.

The effects of lignin extraction on the mill have been simulated in WinGEMS. All the direct (e.g. chemicals) and indirect (e.g. power loss) costs of lignin extraction are taken into account as the “cost” of lignin.

Membrane separation of lignin

The lignin separation process based on membrane separation described here is based on the process claimed to be used by SunCarbon. It should be noted that other membrane separation concepts for lignin separation are being developed. In the process considered in this report, weak black liquor is taken out before the evaporation plant, utilising about 25 wt% of the mill’s weak black liquor. It is led to a membrane filtration unit that will separate the weak black liquor into two streams, a retentate and permeate. The retentate stream contains what has been retained by the membrane and the permeate what has been permitted by the membrane. In this process it is the retentate stream that is wanted, concentrated lignin (15-25 w% lignin and less than 10% of the total black liquor alkali), that is further processed in the depolymerisation stage. The permeate from the membrane is said to be possible to recycle to the green liquor preparation stage of the mill. However, in this study it has been taken to the evaporation plant.

TRANSPORT

The transportation of raw material, intermediates and products have been elaborated in the earlier study "Value chains for production of renewable transportation fuels using intermediates"

(Anheden et al. 2016). As geographical location of plants and processes are similar to the earlier study, these results are used. Results are summarized below.

For bio-oil produced from forest residues, the area needed for outtake of forest residues is determined by the usage of forest residues (presented in Table 18 in reference study). From the area needed for outtake, the average transportation distance can then be determined as 0.71 of the radius of the outtake area. Truck is the only reasonable way for the transportation of chipped forest residues from the forest.

The bio-oil produced at the pulp mill(s) is transported 500 km by ship to a refinery. Hydrogen is not transported as it is consumed at the point where it is produced.

Transport costs

Transport costs for transport by truck have been derived from Benjaminsson et al. (2013) while costs for ship transport have been calculated based on information from Preem (Håkansson 2015) and costs in Börjesson and Gustavsson (1996). Table 11 presents the transport costs used in this study.

Table 11. Transport costs.

Type of transport	Transported feedstock	Energy density (GJ/m^3)	Fixed cost (EUR/GWh)	Variable cost (EUR/GWh km)
Truck	Chipped forest residues, 50% MC	2.96	866	30.4
Ship	Bio-oil	19.2	2553	0.84
Ship	Lignin-oil, 21% MC	23	2129	0.7

CO_2 emission from transport

CO_2 emissions for truck were calculated using data from the Network for Transport and Environment (NTM INT Road, 2010; NTM INT Rail, 2008). Ship CO_2 emissions were calculated using data from Gode et al. (2011). The fuel for truck transport is assumed to be diesel MK1 including 5% RME while ships are assumed to be fuelled with heavy fuel oil. The emissions presented only include direct emissions during transport but include empty trips (50% of the distance). Table 12 summarizes the CO_2 emissions for the transport of the different feedstock.

Table 12. CO_2 emissions from transport.

Type of transport	Transported feedstock	Energy density (GJ/m^3)	CO_2 emissions (gCO_2/MWh, km)
Truck	Chipped forest residues, 50% MC	2.96	12.86
Ship	Bio-oil	19.2	3.32
Ship	Lignin-oil, 21% MC	23	2.77

Energy for transport

For calculating the energy used for transport, the same sources and methodology as above have been used. Instead of looking at the CO_2 emissions associated with the fuel consumption of the transport, the energy content of the consumed fuel was used. Similar as for the CO_2 emissions, the energy use includes empty trips (50% of the distance).

The energy content in the fuel consumed for the transport of the different feedstocks is presented in Table 13.

Table 13. Energy for transport.

Type of transport	Transported feedstock	Energy density (GJ/m^3)	Energy content in consumed fuel (kWh/MWh, km)
Truck	Chipped forest residues, 50% MC	2.96	0.05
Ship	Bio-oil	19.2	0.01
Ship	Lignin-oil, 21% MC	23	0.009

REFERENCE PULP MILL

For this evaluation, RISE Innventia reference market pulp mill model has been used. The model is built in the process simulation program WinGEMS (WinGEMS 2007). WinGEMS 5.0 is a process simulations program developed to calculate mass and energy balances for the pulp and paper industry. The pulp mill model is a hypothetical softwood reference market pulp mill using the kraft process, with the existing, best available, commercially proven technology used, BAT 2010. The model was first developed in the 2005 FRAM project and later updated in 2010 in the Energy & Biofuels cluster 2009-2011 (Berglin et al. 2011). The reference model has low usage of wood, chemicals, energy and water. It is self-sufficient in steam by burning the black liquor in the recovery boiler. Therefore, the falling bark from the incoming biomass are used as fuel in the lime kiln and power boiler or could be sold. The plant has a large steam surplus which is used for power generation in a condensing turbine.

It also has an energy surplus which is used to produce electricity in a condensing turbine. For this case the raw material used is softwood spruce and the production are set to 2000 ADt/day or ~700 000 ADt/year. This is about the size of the largest pulp mills in operation in Scandinavia today (2015). The tendency in the pulp and paper industry is fewer mills with larger production capacity (CEPI 2008). This means that some mills will be closed down, while the remaining mills will increase their production capacity. Therefore, this size could be representative for future Swedish kraft pulp mills.

The power consumption for the mill is set to 726 kWh/ADt and the resulting power balance is presented in

Table 14. As can be seen 984 kWh is sold per ADt produced. This corresponds to 82 MW electricity exported to the grid. The power balance is based on the case where all bark is burnt in the power boiler.

Table 14. Power balance for the reference mill, kWh/ADt.

Power balance	kWh/ADt	MW
Back-pressure part of the turbine	840	70
Condensing part of the turbine	871	73
Sum	1710	143
Consumption	kWh/ADt	MW
Process	726	61
Sold	984	82

It has been assumed that the mill´s turbine can handle extra steam coming from the value chain, this extra capacity has been set to 10% of the condensate flow, which correspond to 0.23 t steam/ADt or 19 t/h.

Different steam to power efficiencies have been used depending on the pressure of the steam see Table 15.

Table 15. Reference mill turbine efficiencies, %.

Steam P, bar	Turbine efficiency, %
100	35
40	29
9	23

REFINERY

The refinery has not been modelled in the work carried out in this report. It has been assumed that the fraction of the hydrodeoxygenated bio-oils that still require some cracking and SynSat treatment to be within the diesel range, e.g. chain length of about C16 and molar H/C-ratio of 2 are being treated in the existing refinery cracker and SynSat equipment. This has the consequence that the equivalent amount of fossil oil production is squeezed out, resulting in an economic loss equivalent to the estimated gross margin of diesel fuel of 1,07 SEK/litre (Energimyndigheten 2017).

In addition, it is assumed that the reaction heat generated from HDO2 step is recovered for heating purposes and replaces fuel oil in the refinery at a ratio of 0:8:1, since the heat losses with combustion exhaust gases are removed.

Light energy rich gases resulting from the HDO treatment (NCG, Non-condensable gases) are also assumed to be recovered in the refinery for heating purposes and replaces fuel oil in a 1:1 ratio.

Assumed price for the fuel oil is 40 EUR/MWh and CO_2 footprint of the replaced fuel oil is 277 kg CO_2 eqv/MWh.

BIO-OIL AND TRANSPORTATION FUEL PRODUCTION

Data developed during the value chain analysis of the technologies included in this study is found in this Section. The tables Table 16 - Table 21 provide a summary of calculations of cost and energy and CO_2 emissions for the different value chains.

Table 16. Summary data for all cases evaluated.

Case		**Pyr-FR**	**HydroPyr-FR**	**HTL-FR**	**HTL-MSL**	**HTL-LBL**
Raw material		**Forest residues**	**Forest residues**	**Forest residues**	**Black liqour**	**LignoBoost Lignin**
Final product		**Transport fuel**	**Transport fuel**	**Transport fuel**	**Transport fuel**	**Transport fuel**
Sites included		3	1	1	3	3
Energy yield biomass-biocrude	%	54	60	79	72	107
Energy yield biocrude-final product	%	91	104	110	92	85
Energy yield biomass-final product	%	49	63	87	66	90
Biocrude	MWh/year	1 404 694	1 233 496	1 164 117	1 373 348	1 490 548
Biocrude	MW	179	157	148	175	190
Final product	MWh/year	1 274 150	1 284 872	1 284 021	1 261 111	1 259 750
Final product	ton/year	100 000	100 000	100 000	100 000	100 000
Final product	MW	163	164	164	161	161

Table 17. Summary data of energy, cost and CO_2 emissions for Pyr-FR.

Pyr-FR				
OPEX	**EUR/MWh$_{tf}$**	**t or MWh/MWh$_{tf}$**		**kg CO_2/MWh$_{tf}$**
Forest residues	35	2,0	MWh/MWh$_{tf}$	1,9
Lost power	0,9	0,02	MWh/MWh$_{tf}$	2,4
Power production Benefit	-4,2	-0,10	MWh/MWh$_{tf}$	-11
Power use	1,6	0,04	MWh/MWh$_{tf}$	4,0
Sold bark	-1,2	-0,01	t/MWh$_{tf}$	0
H_2	28	13	kg/MWh$_{tf}$	116
Production of reduced fossil fuels	6,2			
Catalysts	8,0			
Avoided oil consumption	-26	-0,6	MWh/MWh$_{tf}$	-179
Transports	9,3			11
Maintenance	11			
Total OPEX	**69**			-55
CAPEX				
Pyrolysis MEUR	198			
HDO process MEUR	154			
Annuity factor	0,1			
Total CAPEX	**28**			
Total value chain cost	**97**			

Table 18. Summary data of energy, cost and CO_2 emissions for HydroPyr-FR.

HydroPyr-FR	HydroPyr-FR	HydroPyr-FR (exp)			
OPEX	**EUR/MWh$_{tf}$**	**EUR/MWh$_{tf}$**	**t or MWh/MWh$_{tf}$**		**kg CO_2/MWh$_{tf}$**
Forest residues	27	27,2	1,6	MWh/MWh$_{tf}$	1,5
Power consumption	1,4	1,37	0,03	MWh/MWh$_{tf}$	3,5
Lost power due to drying	2,6	2,56	0,06	MWh/MWh$_{tf}$	6,5
Power production from heat and char	-2,3	-2,3	-0,06	MWh/MWh$_{tf}$	-5,9
Diesel fuel	0,03	0,03	0,0004	MWh/MWh$_{tf}$	0,0
waste water treatment	0,02	0,02			
Ammonia slurry	-0,1	-0,1			
H_2	2,2	2,2	1,02	kg/MWh$_{tf}$	9,2
Production of reduced fossil fuels	2,2	2,2			
Catalysts	0,4	0,4			
Avoided oil consumption	0	0			
Transports	9,6	9,6			10
Maintenance + labour	2,7	7,0			
Total OPEX	**46**	**50**			25
CAPEX					
IH2 MEUR	87	226			
Annuity factor	0,1				
Total CAPEX	7	18			
Total value chain cost	**53**	**68**			

Table 19. Summary data of energy, cost and CO_2 emissions for HTL-FR.

HTL-FR				
OPEX	**EUR/MWh_{tf}**	**t or MWh/MWh_{tf}**		**kg CO_2/MWh_{tf}**
Forest residues	20	1,1	MWh/MWh_{tf}	1,1
Power production	-0,5	-0,01	MWh/MWh_{tf}	-1,3
Power use	2,9	0,07	MWh/MWh_{tf}	7,4
Sold bark	-3,9	-0,04	MWh/MWh_{tf}	0
waste water treatment	4,4			
Catalysts HTL	0,8			
H_2	14	6,5	kg/MWh_{tf}	59
Production of reduced fossil fuels	4,3			
Catalysts HDO	1,0			
Avoided oil consumption	-7,2	-0,2	MWh/MWh_{tf}	-50
Transports	6,7			8,5
Maintenance + labour	8,2			
Total OPEX	**51**			24
CAPEX				
HTL MEUR	107			
HDO MEUR	98			
Annuity factor	**0,1**			
Total CAPEX	**16**			
Total value chain cost	**67**			

Table 20. Summary data of energy, cost and CO_2 emissions for HTL-MSL.

HTL-MSL				
	EUR/MWh_{tf}	**t or MWh/MWh_{tf}**		**kg CO_2/MWh_{tf}**
OPEX				
Lost power	13	0,3	MWh/MWh	34
Power production Benefit	0	0,00	MWh/MWh	0,0
Power use	1,3	0,03	MWh/MWh	3,2
H_2SO_4	0,6	0,01	t/MWh	0,9
NaOH	2,5	0,01	t/MWh	8,3
CO_2	1,8	0,02	t/MWh	9,5
Solid waste	0,9	0,01	t/MWh	
H_2	20	9,1	kg/MWh	82
Production of reduced fossil fuels	7			
Catalysts	8			
Avoided oil consumption	-20	-0,5	MWh/MWh	-135
Transports	2,4			7,4
Maintenance	9			
Total OPEX	**47**			10
CAPEX				
HTL MEUR	185			
HDO process MEUR	89			
Annuity factor	0,1			
Total CAPEX	**22**			
Total value chain cost	**69**			

Table 21. Summary data of energy, cost and CO_2 emissions for HTL-LBL.

HTL-LBL				
	EUR/MWh$_{tf}$	**t or MWh/MWh$_{tf}$**		**kg CO_2/MWh$_{tf}$**
OPEX				
Lost power	16	0,4	MWh/MWh$_{tf}$	40
Power production Benefit	-2,59	-0,06	MWh/MWh$_{tf}$	-6,6
Power use	1,4	0,03	MWh/MWh$_{tf}$	3,5
Catalysts HTL	0,2			
H_2SO_4	2,1	0,03	t/MWh$_{tf}$	2,8
NaOH	7,5	0,02	t/MWh$_{tf}$	25
CO_2	2,6	0,03	t/MWh$_{tf}$	14
Solid waste	3,0	0,030	t/MWh$_{tf}$	0,0
Phenol	61	0,04	t/MWh$_{tf}$	159,0
KOH	3,4	0,003	t/MWh$_{tf}$	6,2
H_2	19	8,4	kg/MWh$_{tf}$	76
Production of reduced fossil fuels	7			
Catalysts	8			
Avoided oil consumption	-23	-0,6	MWh/MWh$_{tf}$	-160
Transports	2,6			7,4
Maintenance	10			
Total OPEX	**118**			168
CAPEX				
LignoBoost MEUR	103			
HTL MEUR	123			
HDO process MEUR	94			
Annuity factor	0,1			
Total CAPEX	**25**			
Total value chain cost	**143**			

Fast pyrolysis of forest residues

Pyrolysis is a collection of thermochemical processes in which organic compounds are broken down into smaller one’s due to heating to high temperatures in the absence of oxygen. Pyrolysis of biomass results in the products char and vapour, where part of the vapour condensate to liquor, the rest are non-condensable gases, NCG. The distribution of char, NCG and liquor depends on the temperature and time in the pyrolysis. Fast pyrolysis is to give high yields of liquor and is completed in seconds. It requires very high heating and heat transfer rates which requires the biomass to be milled to mm particles (IAE Bioenergy 2007). There are several types of pyrolysis reactors and several manufacturers for each reactor. This study is based on a reactor with rotating cone delivered by BTG-BTL (Benjaminsson et al. 2013).

A block flow diagram of the fast pyrolysis plant by BTG-BTL is shown in Figure 41.

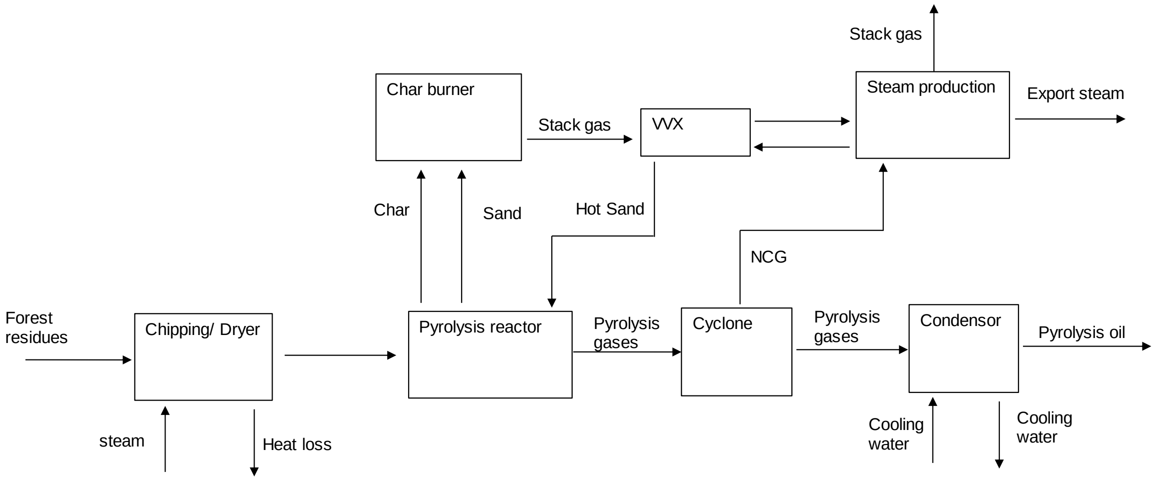


Figure 41. Illustration of the fast pyrolysis process by BTG-BTL (Benjaminsson et al. 2013).

In the BTG-BTL pyrolysis plant, the biomass is ground to a particle size of max 6 mm before the forest residues goes into a belt dryer for drying. The forest residues enter with a moisture content of 50% and a temperature of 25 ˚C. Own produced steam is used to dry the wood chips to a moisture content of 6%. The heat consumption of the belt dryer is 1.1 MWh/ton evaporated water (Benjaminsson et al. 2013). From the dryer, biomass enters the pyrolysis reactor of the rotating cone where sand is used as heat carrier. The biomass is disintegrated and forms vapor and solids. The solids or char formed goes with the sand and are burned in a char burner. The vapour from the pyrolysis reactor are purified from larger particles in two cyclones before subjected to condensation. The NCG go on to a burner for steam production. Steam from char and NCG is produced with an efficiency of 87.5%. Part of the steam goes to drying of the feed the rest goes to the pulp mill´s steam turbine to generate power. The condensed vapour, the liquid which is the desired product pyrolysis oil, is purified by a filter before storage and transport for further upgrading.

Energy yields from (Benjaminsson et al. 2013) have been used in this study. Forest residues contains higher amount of ash compared to for example wood chips or saw mill dust where ash has shown to have a negative effect on the pyrolysis oil yield, to the favour of more char and NCG. Not taken into consideration in this study is the possible increase of ash in the bio-oil and its effect in the next coming hydrotreatment stage i.e. increased catalyst maintenance. Outside of the scope of this study is the possible pre-treatments that could be applied to reduce the ash content in order to avoid the ash effect.

Table 22 shows higher heating values for pyrolysis oil, NCG and char used in this study which have been estimated based on (Benjaminsson et al 2013) and (Jones et al 2013).

Table 22. Higher heating values of pyrolysis oil, NCG and Char MJ/kg.

	MJ/kg HHV
Pyrolysis oil	17
NCG	11
Char	32

Hydropyrolysis of forest residues

The IH2 process in this work has integrations with a softwood pulp mill as described by Figure 42.

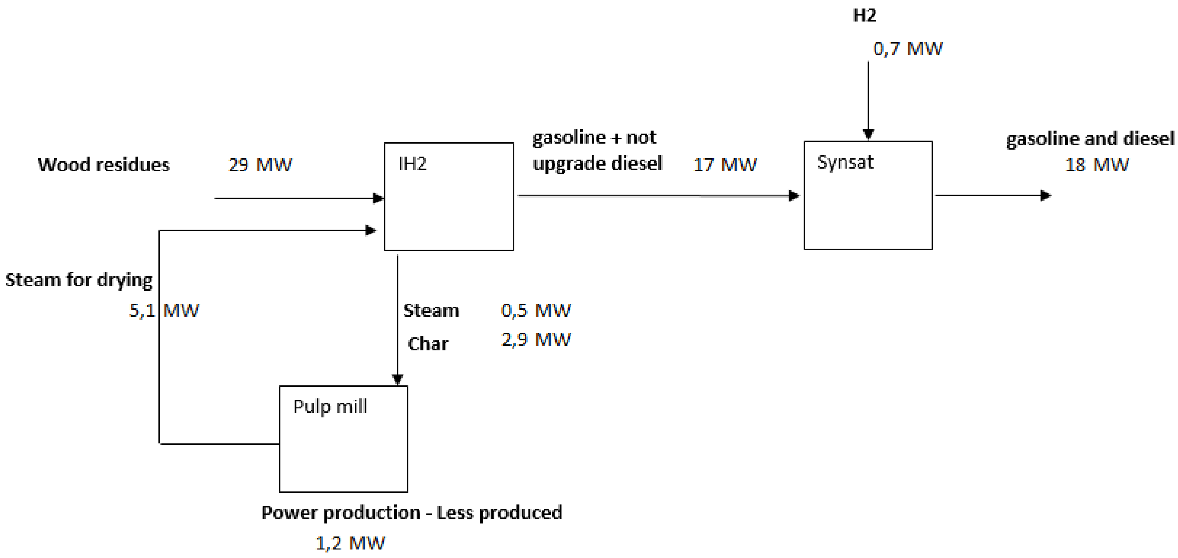


Figure 42. Schematic layout of integration of the IH2 process with the pulp mill. The energies marked in the process are based on a feed of wet biomass 10000 kg/h.

The forest residues enter the belt dryer with a moisture content of 50% and a temperature of 25 ˚C. Low pressure steam (4.5 bar, 150 ˚C) from the pulp mill is used to dry the wood chips to a moisture content of 6%. The heat consumption of the belt dryer is 1.1 MWh/ton evaporated water (Benjaminsson 2013). The use of low pressure steam from the pulp mill results in a loss of power production from the mill, see Table 23. The loss of power was quantified with a simulation of a softwood pulp mill model built in process simulations program WinGEMS. The efficiency of the low-pressure turbine in the model was 20.2%. The pulp mill model has been developed and improved for years by RISE Innventia (Berglin, 2011).

The higher heating value (HHV) of the forest residue is 20.71 MJ/kg dry. The mass yield for the liquid products of the IH2 process is 0.27 kg/kg dry biomass and the energy yield is 0.60 MWh/MWh dry biomass. It is assumed that the steam reformer produces enough hydrogen to supply the IH2 process and that no external hydrogen will be needed.

The excess heat produced from the exothermic reactions in the hydropyrolysis and the hydroconversion reactors is used to generate steam (40 bar, 370 ˚C) in a boiler internal to the IH2 process. It is assumed that the efficiency of the boiler is 80%. The resulting steam is sent to the pulp mill. That steam is not run through a condensing turbine but instead its pressure is lowered in an expansion valve to 13.5 bar and used for internal purposes (soot blowing, air preheating, feed water interheating etc.). This allowed a lesser amount of MP steam (30 bar) to be extracted from the turbine for this purpose and it could instead be used for additional electricity generation in the turbine. The increase in power production resulting from this integration was modelled in RISE Innventia’s pulp mill model (Berglin, 2011) and it is seen in Table 23. The efficiency of the middle pressure turbine was 29%.

All char produced in the pyrolysis reactor is burned in the pulp mill's recovery boiler to produce high pressure steam used for electricity generation in the condensing turbine. The pulp mill's recovery boiler produces high pressure steam with an efficiency of 72.5% and the condensing turbine's efficiency is 35.2%. It was assumed that the condensing turbine can handle an increased condensate flow of 10%. This limit was not exceeded. In fact, due to the large use of low pressure steam for drying, there was a decrease of the condensate flow with 20%.

The integrations between the IH2 process and the pulp mill result in a small net loss of power production from the pulp mill. In the power productions resulting from the integrations between the pulp mill and the IH2 process are listed.

Table 23. Lists the power consumption and production resulting from the integrations between the pulp mill and the IH2 process.

Stream	**MWh power/MWh upgraded product**
Resulting power produced from IH2 heat excess	0.008
Resulting power produced from char	0.049
Resulting power loss due to drying of biomass	-0.064
Sum	**-0.007**

The liquid phase leaving the IH2 process consist of one diesel fraction (24%) and a gasoline fraction (76%). The diesel fraction leaving the IH2 process has a H/C ratio of 1.3 (Gas Technology Institute (GTI), 2012) which is below the H/C specification for commercial diesel which is 2.0, based on information from Preem. Therefore, the diesel fraction needs to be treated with hydrogen in a SynSat-reactor at the refinery. The calculated amount of hydrogen needed to upgrade each mass unit of the IH2 diesel fraction is 5 wt%. The gasoline fraction leaving the IH2 process is here assumed to not require any further processing. It is assumed that the weakly endothermic behaviour of the SynSat process will not require any external heat.

The overall energy yield for the final fuel mix is 0.63 MWh/MWh dry biomass.

In the original paper, an indirect method has been used for the process calculations. Based on chemical analysis from lab/pilot measurements a typical set of reaction products were identified. The quantity of these were then adjusted to fit elementary analysis and the distribution between gas and liquid from the measurements. This composition was then used in calculations in a process design program (Aspen plus).

Hydrothermal liquefaction of forest residues

The HTL-FR case is based on data from de Jong et al. (2017), which in turn base their calculations on Zhu et al. (2014). Drop-in biofuels (diesel and gasoline) is produced through hydrothermal liquefaction (HTL) and hydroprocessing. Compared to pyrolysis oil, HTL produces a biocrude of higher quality in terms of heating value, moisture content, oxygen content, and stability (Tews et al. 2014, Elliot et al. 2015).

In Zhu et al. (2014) a bench-scale HTL and hydrotreating tests were conducted. In addition, a techno-economic analysis was conducted for the HTL and upgrading systems. Two different cases were evaluated: a *state-of-technology (SOT) case* based on the best available test data and a *goal case* considering potential process improvements. In de Jong et al., and in this study, data based on the goal case is used.

The major improvements for the goal case include:

1) lower HTL reactor pressure;
2) less organics loss to the water phase;
3) adding a hydrocracking process for heavy oil treatment; and
4) using a single reactor hydrotreating process.

In Zhu et al. (2014), the annual production rate for the final hydrocarbon product was estimated to be 42.9 and 69.9 million gallon gasoline-equivalent (GGE) for the SOT and goal cases, respectively, i.e. 63% higher yield is assumed for the goal case compared to the SOT case. The minimum fuel selling price (MFSP) was estimated to be $4.44/GGE for the SOT case and $2.52/GGE for the goal case, i.e. the goal case resulted in 43% lower production cost. The assumption of reducing the organics loss to the water phase led to the largest reduction in the production cost. Figure 43 shows the cost effect of improvements of moving from SOT to goal case.

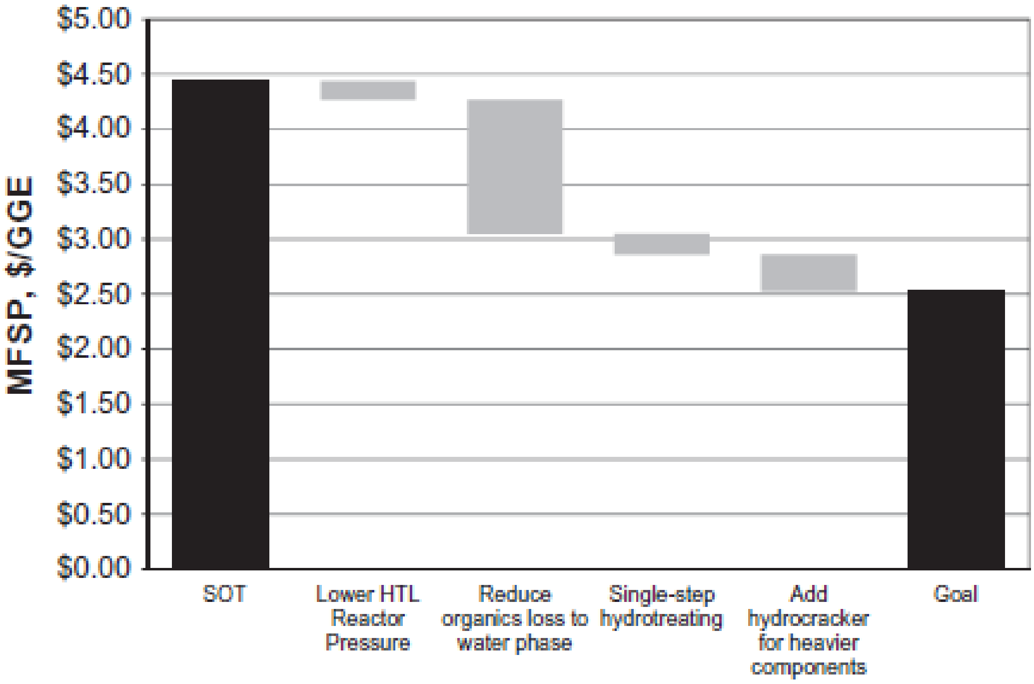


Figure 43. The cost effect of improvements of moving from SOT to goal case (Zhu et al. 2014).

In de Jong et al, different supply chain configurations are considered including different integration alternatives for the HTL unit and upgrading of the biocrude. In this study, data is based on the distributed supply chain configuration where the HTL conversion is integrated with a pulp mill and the upgrading takes place at a refinery. Also, other alternatives are considered in de Jong et al. for integration of the HTL unit as well as the upgrading of the biocrude. Figure 44 shows the general process configurations for distributed production in de Jong et al (2017).

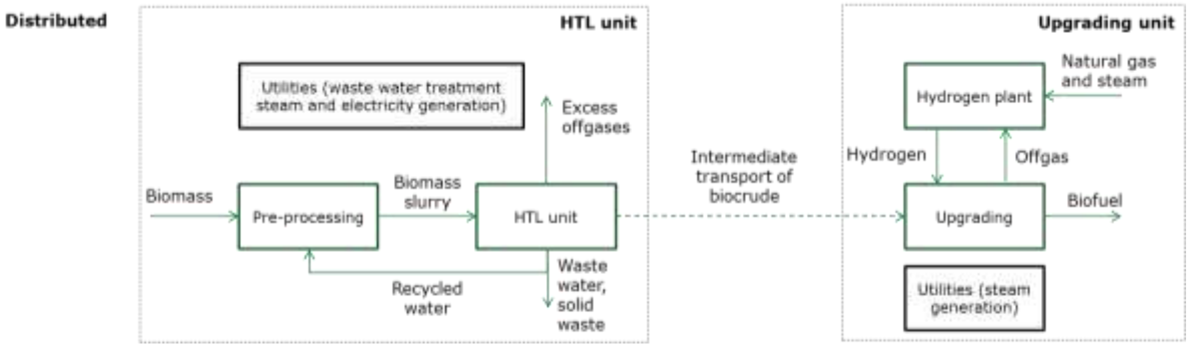


Figure 44. Process configurations for distributed production in de Jong et al. (de Jong et al. 2017).

Figure 45 shows a simplified block diagram of the HTL-FR case. The feedstock is first ground and mixed with recycled hot water to produce a slurry (15% dry biomass content). After preheating (by the hot effluent from the HTL reactor) and pressurization the slurry is pumped into the HTL reactor (16.6 MPa, 336 °C), which is of plug flow type based on a shell-and-tube design with hot fluid on the shell side and the slurry inside the tubes. Hot biomass slurry is converted in the HTL reactor. Solid waste, mostly inorganic solids, such as ash, is separated from the hot HTL effluent. The filtered effluent is cooled and separated into a gas phase (off-gas) and then two liquid phases: bio-oil and an aqueous phase with predominately water and some dissolved organics.

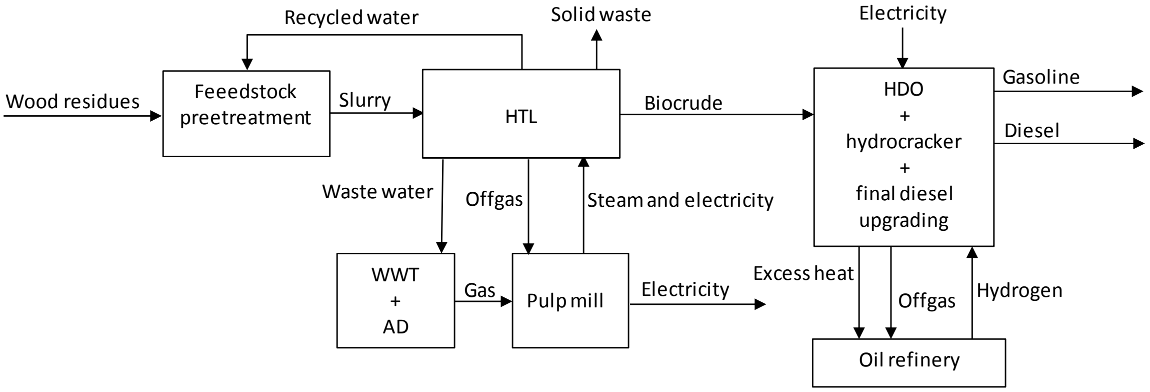


Figure 45. A simplified block diagram of the HTL-FR case.

Most of the aqueous phase is recycled to the feedstock preparation step while the remaining portion is purged to wastewater treatment (WWT) which uses anaerobic digestion (AD) to convert the dissolved organics into a gas rich in methane and carbon dioxide. The HTL off-gas contains hydrogen, carbon dioxide and methane and other hydrocarbons. In this study, the HTL off-gas and the gas from anaerobic digestion are used as fuel at the mill to produce steam, partly used for internal heating of the HTL process and partly used for electricity generation.

The bio-oil is sent to the refinery for upgrading to biofuels. For the goal case, both hydrotreating and hydrocracking are assumed to be used for bio-oil upgrading to remove most of oxygen in the bio-oil. The hydrotreating process involves contacting the bio-oil with hydrogen under elevated pressure and high temperature to remove oxygen and reduce the molecular weight of the bio-oil via hydrodeoxygenation (HDO) reactions. The upgraded hydrocarbon product is stabilized by cooling and distillation to produce liquid fuels consisting of gasoline, diesel, and heavy oil fractions based on their boiling point ranges. Hydrocracking is used to increase the yield of fuel products rich in hydrogen (i.e. diesel) from heavy hydrocarbons. By adding hydrogen, the heavy oil from the hydrotreating product separation process is decomposed and rearranges to produce a mixture of liquids spanning the gasoline and diesel range and some light gases. The diesel fraction considered in the concept studied by the Jong et al. does not meet the requirements. Therefore, additional upgrading is required to saturate the oil to meet the specified H/C ratio of 2.0. The upgrading is associated with additional hydrogen usage and a cost due to loss of fossil fuel production.

Table 24 presents the energy input data for the HTL-FR case.

Table 24. Energy input data for the HTL-FR case.

Intermediate production (at pulp mill)		
Yield	MWh biocrude / MWh biomass	0.79
Electricity use	MWh / MWh biocrude	0.07
Electricity production (from fuel by-products)	MWh / MWh biocrude	0.10
Intermediate conversion (at refinery)		
Yield	MWh biofuel / MWh biocrude	1.10
Electricity use	MWh / MWh biofuel	0.01
Hydrogen use[1]	MWh / MWh biofuel	0.26
Offgases for oil replacement[2]	MWh / MWh biofuel	0.12
Excess heat for oil replacement[3]	MWh / MWh biofuel	0.05

[1] Of which 0.1 is used for the final diesel upgrading.

[2] Estimated based on Tews et al. (2014).

[3] Of which 0.03 is from the final diesel upgrading and 0.02 from the HDO and hydrocracking units. The later number is based on Tews et al. (2014).

Table 25 presents economic input data for the HTL-FR case for calculation of different equipment costs.

Table 25. Economic input data for the HTL-FR case for calculation of different equipment costs.

	a	**b**	**cap (MW)**
Feedstock handling	0.009	0.77	FR
Biomass conditioning	0.16	0.70	FR
HTL reactor	0.30	0.70	FR
Hydrotreater	0.70	0.60	Bio-oil
Hydrocracker	0.26	0.60	Bio-oil

The equipment costs are calculated according to:

$$\textit{Equipment cost (EC)} = a * cap^{b}$$

The total purchased equipment cost is then calculated according to:

$$\textit{Total purchased equipment cost (TPEC)} = \sum EC * \textit{Missing Equipment (ME)}$$

Finally, the total capital investment is then calculated according to:

$$\textit{Total capital investment (TCI)} = TPEC * \textit{Lang Factor (LF)}$$

where ME=10% and LF=4,6. The HTL-FR biocrude production is located at one mill site, thus multiple equipment is not needed. However, previous studies indicate a maximum capacity around 90 MW feedstock for the HTL reactor (de Jong et al. 2017). Therefore, in this study two reactors are considered where each reactor has a feedstock intake of 94 MW.

Table 26 presents economic input data for the HTL-FR case for calculation of different operation and maintenance costs.

Table 26. Economic input data for the HTL-FR case for calculation of different operation and maintenance costs.

Intermediate production (at pulp mill)		
CAPEX-dependent OPEX	% of TCI	4.0
Catalysts	EUR/MWh$_{in}$	0.7
Waste disposal	EUR/MWh$_{in}$	3.8
Intermediate conversion (at refinery)		
CAPEX-dependent OPEX	% of TCI	4
Catalysts	EUR/MWh$_{in}$	1.1
Lost fossil production	EUR/MWh$_{in}$	3.7

Hydrothermal liquefaction of membrane separated kraft lignin

The SunCarbon process is meant to be integrated with a pulp mill. Weak black liquor is fed to the process which is first filtrated, treated hydrothermally and purified into lignin oil that is sent to a petroleum refinery for final hydrogenation (SunCarbon, 2016). A schematic drawing of SunCarbon's process is seen in Figure 46.

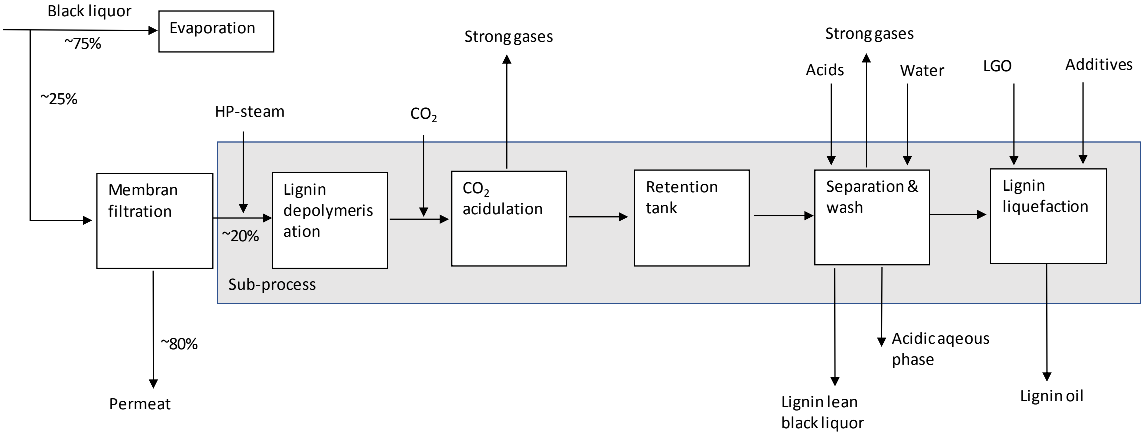


Figure 46. A simplified flow chart depicting SunCarbon's process.

The hydrothermal liquefaction of membrane separated lignin is simulated in the RISE Innventia's Reference pulp mill model. The effects on the whole mill by integrating this process can hence be observed. The weak black liquor is taken out before the evaporation plant, about 25 wt% of the mill's weak black liquor. It is led to a membrane filtration unit that will separate the weak black liquor into two streams, retentate and permeate. The retentate stream contains what has been retained by the membrane and the permeate what has been permitted by the membrane. In this process it is the retentate stream that is wanted, concentrated lignin (15-25 wt% lignin and less than

10 % black liquor alkali), that goes on the depolymerisation stage. The permeate from the membrane is said to be able to be recycled to the green liquor preparation stage of the mill, However, in this study it has been taken to the evaporation plant. The lignin rich retentate is mixed with medium pressure steam (23-56 bar) from the mill and fed into a thermal catalytic cracker at sub-critical conditions for depolymerization. A catalyst internal to the pulp and paper mill is used in the catalytic cracker. A catalyst internal to the pulp and paper mill is used in the catalytic cracker. The hydrothermal treated lignin mixture is then mixed with carbon dioxide which lowers the pH. After spending some time in a retention tank, the lignin mixture is subjected to an acid wash (H_2SO_4) which separates it from ash, residues, lean black liquor and strong gases (mainly H_2S). The removed lean black liquor contains around 30% of the energy content of the initial lignin entering the process resulting in an energy yield of ~70% of the lignin oil. Lastly the remaining lignin is liquefied with gas oil or tall-oil pitch and additives. After this step the resulting product is a lignin oil ready to transport to a petroleum refinery for hydrogenation. To be noted is that this process is in its early development phase and the final concept has not yet been formulated. For example, additional purification stages may be needed which is not included in this assessment.

Extracting part of the organic phase from the black liquor will decrease the steam production in the mills recovery boiler. For the reference pulp mill, this results in less sold power, since it has a steam surplus. Not all mills have excess steam and net power production. Another option to keep the steam production is to burn additional biomass in the bark boiler. The additional sulphur from the washing stage will affect the mill’s Na/S balance. For the reference mill, this result in a higher purge of ESP dust (mainly Na_2SO_4/Na_2CO_3) from the recovery boiler where the Na loss needs to be compensated by virgin NaOH makeup. In this study it has been assumed that the lignin oil contains some S and Na as well and depending on how much S is removed and how much Na is lost with the lignin oil it will also affect the amount of NaOH makeup for the pulp mill.

Further on if the mill’s recovery boiler is a bottle neck the extraction of black liquor can increase the pulp production, in a similar fashion as a LignoBoost.

In Table 27 below heating values, MJ/kg dry solids, of main flows in the thermal liquefaction of membrane separated lignin can be seen. The heating values has been estimated based on simulations results from the reference mill model and discussions with the supplier of the concept.

Table 27 Used heating values for thermal liquefaction of membrane separated lignin MJ/kg dry solids.

	MJ/kg dry, HHV
Weak black liquor	15
Retentate	20
Lignin oil	28

Base catalyzed depolymerization of kraft lignin from the LignoBoost process, HTL-LBL

The mass balances for the reactor step are taken from not yet published experimental data from Tallal Belkheiris PhD project at Chalmers (Belkheiri et al 2018a, Belkheiri 2018b). These data are representative for the current state-of-art for lignin depolymerisation using phenol as capping agent and are obtained in a small but continuous pilot plant reactor, which makes the data more reliable for scale-up than if obtained in e.g. a batch reactor. In the pilot plant there is, however, no recirculation of the separated aqueous phase, so for the calculations here it is assumed that 90% of this phase can be recirculated. This is a slightly conservative estimate, it is not unusual that 95% can be recirculated, but it has first to be tested that the bleed stream (the part of the aqueous phase that is not recirculated) is large enough to avoid unwanted accumulation in the process.

It is assumed that the bleed stream can be integrated with the pulp mill, i.e. it can be taken to the chemical recovery cycle to recover energy and alkali. It is also assumed that the suspended solids in the oil can be separated and used as a fuel.

It is also assumed that all (identified) phenolic-type substances in the recirculation stream are effective as capping agents and thus can reduce the need of phenol feed. This is probably true for most but not for all of these substances. The need for fresh alkali is also reduced by the recirculation. The recirculation might also affect the equilibrium between the aqueous and oil-phase increasing the light-oil yield, but this effect has not been taken into account.

The investment costs are based on estimates from a report in a previous EU project (Iversen et al. 2005) focused on the use of HTL for another type of feed (waste) but for conditions, layout and catalyst very similar to the process studied here. The only significant difference is that this other feed resulted in considerably lower coke formation. To account for that, it is here assumed that a dual reactor system is needed, resulting in a 30% higher investment cost.

Hydrodeoxygenation of crude bio-oil to HDO-oil and upgrading of HDO-oil to gasoline and diesel

Treatment through 2-step hydrodeoxygenation has been assumed for all the produced bio-oils using dedicated new process equipment.

Upgrading of bio-oil from fast pyrolysis of forest residues

In the case of HDO of pyrolysis oil, the analysis has included use of data from report PNNL-23053 from 2013, assuming the exact same composition of the pyrolysis oil as assumed in this report. Based on the decomposed data from the report, the hydrogen demand in HDO step 1 and 2 has been estimated from the stream data as well as additional hydrogen used in the step to crack and finish the heavy fraction. The distribution of components in the stream of non-condensable gases from the HDO and cracking treatment has also been collected from the same report. A rough energy balance over these process steps have been estimated based on stream flow data and heating values of the components.

It has also been discovered that the reported final diesel fraction in the used PNNL report does not meet the specified requirements of 0 oxygen content and H/C ratio of 2. An evaluation was made of the C, H, O, N, S content of the reported diesel fraction in stream 326 in the report, which resulted in the conclusion that this stream contains 0.92 wt% oxygen and had a H/C-ratio of 1.6. Based on this, a theoretical hydrogen demand was calculated based on simple stoichiometry for oxygen removal and the reactions in the SynSat treatment.

These estimations resulted in the following estimated hydrogen demand, see Table 28.

Table 28. Hydrogen demand for upgrading of pyrolysis oil.

H_2 for HDO	0,057614	kg H_2/kg dry pyr oil
H_2 for cracking	0,008551	kg H_2/kg dry pyr oil
H_2 for HDO3, diesel	0,000573	kg H_2/kg dry pyr oil

H_2 for SynSat, diesel	0,006758	kg H_2/kg dry pyr oil
Total H_2	0,073496	kg H_2/kg dry pyr oil

In addition, 0.25 kg HDO3 diesel/kg dry pyrolysis oil was estimated to have to go through the SynSat treatment. 3,4 MJ/kg dry pyrolysis oil of process heat was estimated to be recovered to replace use of fuel oil for heating purposes, as well as 9.0 MJ/kg dry pyrolysis oil of non-condensable gases.

Upgrading of bio-oil from HTL of forest residues

The hydrogen consumption for the HDO upgrading in case of HTL of forest residues are based on Tews et al. (2014). The HDO-treatment and upgrading of the bio-oil from HTL of forest residues is based on the technology and data available in de Jong et al. (2017), which in turn base their calculations on Zhu et al. (2014).

The heat recovered from the HDO treatment is calculated from information given in Tews et al (2014, p. 51-53). The heat is estimated from the heat removed from stream 310-312 in the flow sheet or 311-312A in the Table minus the heat recovered in stream 318-319 (ibid.). The excess heat is estimated to 1,73 MW/(kg/hr) gasoline production. The NCG production was estimated to 1256 kg/h/(kg/hr gasoline production) based on stream 322 in the table or 326 in the flow sheet. Based on the composition, it was estimated that this is equivalent to 20 MW/(kg/hr) gasoline production.

It was found out that the reported diesel fraction did not meet the specifications. Based on Tews et al (2014, p. 9 and 53), the more exact atomic composition of the reported diesel fraction, stream 325, was estimated. It was found that it contains no oxygen but has a H/C ratio of 0.88. 0.39 kg crude diesel /kg fuel fraction including heavy of the product from the fractionation column has to go through the SynSat treatment and pushing out fossil oil diesel production. Based on stoichiometry it was found that 0.034 kg H_2/kg fuel fraction (including heavy fraction) of the product from the fractionation column was required for the SynSat treatment, and 4.8 MJ of heat from the SynSat can replace fuel oil usage.

Upgrading of bio-oil from hydropyrolysis of forest residues

The hydropyrolysis process included in the current report is assumed to use the off-gases formed during the integrated hydropyrolysis and upgrading reaction are used for internal hydrogen production. Based on the reported diesel composition for wood in GTI (2012, Table 8), it is concluded that an H/C-ratio of 1.3 is only achieved for the diesel fraction. Based on stoichiometry it is found that additional 0.053 kg/s of hydrogen is required for each kg/s of crude diesel.

Upgrading of bio-oil from HTL of membrane separated kraft lignin and LignoBoost kraft lignin

The knowledge base on hydrodeoxygenation and upgrading of bio-oils from kraft lignin depolymerisation is so far very limited. Work is currently ongoing in the BioInnovation project BioLi2.0, but results are not public yet. The closest experimental data available is the data provided by Renfuel AB in Löfsted et al. (2016) and associated supporting documentation.

The basic assumption made is that the HTL depolymerised lignins used in this report behave similarly during HDO and upgrading as the lignin in run 3 in Löfstedt et al. (2016). This is a very uncertain assumption since the depolymerisation process in Löfstedt et al. is based on mild Ni-catalysed transfer hydrogenation using 2-propanol. In addition, the depolymerised lignin is esterified with tall oil fatty acid anhydride, to get it soluble in light gas oil (LGO). The esterified depolymerised lignin was co-processed with LGO during hydrotreatment. In the calculations, the reported run 1 was used to calculate the hydrogen consumption related to the tall oil (0.016 g H_2/g TOFA) assuming that the LGO does not consume any H_2, and based on this information it was estimated that the H_2 consumption for the depolymerised lignin is 0,05 g/g lignin fragments from run 3. Based on run 1 and 3, the distribution of products (light gases, fuel components, heavy components and CO_2 and H_2O was calculated.

Next, data on the elemental composition of the depolymerised lignins from the two cases HLT-MSL and HTL-LBL was used as input. The same oxygen reduction as reported in Löfstedt et al. (2016) was used as input in the calculations on HDO1, and a stoichiometric hydrogen consumption assuming direct reduction of oxygen in the oil to HVO. The hydrogen consumption in HDO2 was calculated assuming the same specific hydrogen consumption, resulting in a hydrogen consumption of 0.046 and 0.045 kg H_2/kg depolymerised lignin feed for HTL-LBL and HTL-MSL.

The yield from the depolymerised lignin to the different fractions from the fractionation column is calculated from run 3 assuming LGO only contribute to the heavy phase containing gasoline, diesel and heavy fraction, and that the TOFA and LGO contributes to gasoline, diesel and heavy fraction to the same extent as in run 1 and that also the TOFA contribution to light gases is the same as in run 1.

The H/C-ratio of the produced diesel is assumed to be the same as in reported by Löfstedt et al. (2016) in Table S4, which means that it is 2.1, and no further treatment is required. The heavy fraction it is assumed that C36 is cracked to C18 requiring 1 mole H_2 per C36.

The heat from the HDO that could be used to replace fuel oil is calculated based on an energy balance. Table 29 show the main data.

Table 29. Main data related to HDO and further upgrading of depolymerized lignin.

	H_2 consumption (kg/kg depolymerised lignin)	**Excess heat to recovery (MJ/kg depolymerised lignin)**	**NCG to recovery (MJ/kg depolymerised lignin)**	**Diesel (kg/kg depolymerised lignin)**	**Gasoline (kg/kg depolymerised lignin)**
HTL-MSL	0.065	5.9	5.7	0.47	0.10
HTL-LBL	0.063	8.0	5.9	0.48	0.10

INVESTMENT COSTS

The investment cost in this assessment is based on the main equipment required for the different main process steps, i.e. fast pyrolysis, hydrothermal liquefaction (HTL) and hydrotreatment of the biocrude to gasoline and diesel (HDO). When investment cost is not available for the specific size that is needed predictions can be made to scale the investment costs. If cost of a given unit b at one capacity is known, the cost of a similar unit a with X times the capacity of first is X^f times the cost of the initial unit, according to equation 1 below (Peters et al. 2003).

1) $$Cost\ of\ new\ equipment\ a = Cost\ of\ base\ equipment\ b * (X)^f$$

$$where\ X = \frac{Capacity\ new\ equipment\ a}{Capacity\ base\ equipment\ b}$$

$$f = typical\ scaling\ factor\ 0.6 - 0.7$$

The investment cost has been scaled this way to estimate the certain investment cost for the specific capacity needed. In Table 30 below, a summary of original investment cost data and it's scaling factor can be seen. All investment has been recalculated to cost in 2015 money value. Adjustments have been made using Chemical Engineering Plant Cost Index (CEPCI).

In Table 30 below, a summary of assumed investments and references are found.

Table 30. Summary of investment assumptions and references.

Investment cost	Cost	Unit	Capacity	Unit	f	Reference
Pyr-FR	607	MSEK	60	MW oil	0.7	Benjaminsson et al. 2013
HydroPyr-FR	233	MUSD 2007	2000	ton product/day	0.7	Gas Technology Institute (GTI) 2012
HTL-FR						
- feedstock	0.29	MEUR 2015	87	MW forest residues	0.77	De jong et al. 2017
- biomass conditioning	3.6	MEUR 2015	87	MW forest residues	0.7	De jong et al. 2017
- HTL reactor	6.8	MEUR 2015	87	MW forest residues	0.7	De jong et al. 2017
HTL-LBL	17	MEUR	30 000	ton biodiesel/year	0.7	Iversen et al. 2005
HTL-MSL	400	MSEK 2017	40 000	ton lignin oil/year	0.7	Personal communication SunCarbon
HDO Pyr-FR	36	MUSD 2014	280	MW Bio-oil	0.6	Tews et al. 2014
HDO HTL (Lignin cases)	21	MUSD 2014	278	MW Bio-oil	0.6	Tews et al. 2014
HDO HTL-FR	12	MEUR 2015	69	MW Bio-oil	0.6	De jong et al. 2017
LignoBoost	30	MEUR	50 000	t/y	0.7	Inhouse knowledge and contact with supplier

For the HTL and HDO investment cost uninstalled cost are given. 10% for missing equipment has been added further to this cost and to estimate the installed cost a Lang factor of 4.62 has been used.

OPERATION AND MAINTENANCE COSTS

Usually a share of the investment is taken for the annual cost of maintenance. In this study we have added 4% of the investment cost as maintenance cost and labour cost.

PRICES OF ENERGY CARRIERS

Table 31 shows the prices of different energy carriers used. The prices used as base values are chosen to represent the current situation in Sweden.

Table 31. Prices of raw material, electricity, electricity certificates, diesel, natural gas, fuel oil and hydrogen [EUR/MWh].

	Based on	Base
Forest residues HHV	Energimyndigheten (2015)	17
Bark		17
Electricity	Nordpool (2015)	20
Electricity certificates	SKM (2013)	20
Diesel	Ekonomifakta (2016)	114
Hydrogen (estimated as LNG price*3)	Ycharts (2015)	53
Fuel Oil		40
Value of reduced fossil fuel production	Energimyndigheten (2017a)	1,07 SEK/l

Renewable electricity production is assumed to be entitled to support from the electricity certificate system[1]. The used price of electricity certificates is included in Table 31. Consumed electricity is assumed to be purchased for the same price as of the renewable electricity.

It is assumed that the market price of hydrogen is approximately three times the price of natural gas, based on information from Preem.

PRICES OF CHEMICALS

The prices are a combination of prices used in earlier research and from different suppliers.

Table 32. Specific prices of chemicals used in this study.

Specific prices	**EUR/ton**
CO_2	90
H_2SO_4	80
NaOH	400
Na_2CO_3	250
K_2CO_3	1 200
KOH	1 000
Phenol, C_6H_5OH	1 600
Ash disposal	100

CO_2 EMISSIONS FACTORS

Table 33 and Table 34 presents the CO_2 emission factors used. For electricity, the emissions are for the Nordic electricity mix. Currently, the majority of hydrogen is produced from fossil fuels by steam reforming or partial oxidation of natural gas. The emissions presented in Table 33 represent this method of hydrogen production.

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[1] Policy instrument incentive scheme promoting the production of renewable electricity, where electricity producers receive one certificate per MWh produced renewable electricity. The certificates are traded between the suppliers and consumers. A quota obligation for consumers creates a demand for the certificates and thus provides them with an economic value.

Avoided CO_2 emissions when replacing fuel oil in the refinery with process heat and NCG from bio-oil upgrading in the refinery is estimated to 277 g/MWh avoided fuel oil.

Table 33. CO_2 emission factors (kg CO_{2eq}/MWh) used.

	Emission factor	Data from
Electricity	102	Fredén (2010)
Hydrogen	270	Collodi (2010)
Diesel	284	Gode et al. (2011)
Fuel oil	277	EUROPAPARLAMENTETS OCH RÅDETS DIREKTIV (2009)
Gasoline and diesel	302	EUROPAPARLAMENTETS OCH RÅDETS DIREKTIV (2009)

In Table 34 CO_2 emission factors in kg CO_{2eq}/kg of chemicals used in the processes can be seen.

Table 34. CO_2 emission factors kg CO_{2eq}/kg of chemicals used in the different processes.

	Emission factor	Data from
Sulphuric acid	0,1	Ecoinvent database 3.1 CML 2001 - Jan. 2016
Sodium hydroxide	1,4	Ecoinvent database 3.1 CML 2001 - Jan. 2016
Phenol	4,2	Ecoinvent database 3.1 CML 2001 - Jan. 2016
CO_2	0,5	Ecoinvent database 3.1 CML 2001 - Jan. 2017
K_2CO_3	2,7	Ecoinvent database 3.1 CML 2001 - Jan. 2018
KOH	1,8	Ecoinvent database 3.1 CML 2001 - Jan. 2018

APPENDIX 5: SELECTED HYDROGEN TECHNOLOGIES

Performance summary of Electrolysis of water and Gasification of biomass as used in this study is summarized in Table 35.

Table 35. Process data for the alternative hydrogen production technologies. Alkaline electrolysis of water based on information from NEL, (Langås, 2017 and TU, 2016) and gasification of biomass based on information from Cortus, (Folkelid, 2017).

	Electrolysis of water	**Gasification of biomass**
Technology	Atmospheric Alkaline Electrolyser	Hydrogen separation and up-grading of syngas
Hydrogen capacity	1455 Nm3/h H_2	1610 Nm3/h H_2
	131 kg/h H_2	145 kg/h H_2
Hydrogen purity	>99%	>99%
Hydrogen production pressure	atmospheric	14 bar
Power consumption excl. final H_2 compressor	6 402 kW	770 kW
Process water	1,45 m3/h	1,0 m3/h
Process cooling water	146 m3/h	none
LPG	none	1 Nm3/h
Surplus gas produced	oxygen (vented)	0.84 MW off-gas
Final H_2 compressor	3 stage compression	3 stage compression
Final hydrogen pressure	90 bar	90 bar
Final H_2 compressor power	365 kW	149 kW

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