Final report

SUNALFA

System oriented analysis of processes for renewable fuels from forest raw material

May 2021

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A project within

RENEWABLE TRANSPORTATION FUELS AND SYSTEMS 2018-2021

A collaborative research program between the Swedish Energy Agency and f3 The Swedish Knowledge Centre for Renewable Transportation Fuels



www.energimyndigheten.se



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PREFACE

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

The Swedish Energy Agency is a government agency subordinate to the Ministry of Infrastructure. The Swedish Energy Agency is leading the energy transition into a modern and sustainable, fossil free welfare society and supports research on renewable energy sources, the energy system, and future transportation fuels production and use.

f3 Swedish Knowledge Centre for Renewable Transportation Fuels is a networking organization which focuses on development of environmentally, economically and socially sustainable renewable fuels. The f3 centre is financed jointly by the centre partners and the region of Västra Götaland. Chalmers Industriteknik functions as the host of the f3 organization (see https://f3centre.se/en/about-f3/).

This report should be cited as:

Stigsson, C.C., Furusjö, E., Börjesson, P. (2021). *SunAlfa - System oriented analysis of processes for renewable fuels from forest raw material* Publ. No FDOS 14:2021. Available at https://f3centre.se/en/renewable-transportation-fuels-and-systems/

SUMMARY

The increasing average temperature in the atmosphere is in part a consequence of the present usage of fossil fuels and its associated net release of carbon dioxide. Moreover, the global fossil resources are limited and consequently, the need for alternative sources of fuel is considerable. Electrification is a solution of disputable sustainability, and energy density is a difficult parameter to increase. The aviation industry is arguably particularly complicated to electrify, although a new primary source of energy is required. The product of the chemical process researched in this project is compatible with the current technology and infrastructure, and is derived from a renewable low-value feed-stock.

In summary, the setup consists of four main integrated subprocesses: hydrothermal liquefaction (HTL), evaporation, gasification and Fischer-Tropsch synthesis (FT). A biomass feedstock, here forest residues, is mixed with recycle streams from the evaporator and the gasifier, pressurized and fed to the HTL reactor. The HTL product is concentrated by an evaporator and subsequently used as feed to an oxygen blown and pressurized entrained flow gasifier. Upon exiting the gasification process, the syngas is conditioned by water-gas shift in a mixture with reformed tail gas from the FT synthesis product separation and mixed with raw tail gas before entering the FT reactor. FT crude is separated from the reactor product and as a suggestion sent to upgrading, albeit outside the scope of the project.

The entire process was arranged and simulated computationally, and its input and output were analyzed in a life cycle assessment (LCA). The energy, mass and carbon efficiencies achieved were 34.5 %, 20.2 % and 32.3 %, respectively, which are deemed fair in a biorefinery. In the HTL product evaporator, the steam economy settled in at 3.45, also decent. The FT crude carbon chain length distribution peaked at C10 with a heavy right tail, appropriate for cracking in an upgrading unit. The LCA showed high GHG performance in the two studied cases with Swedish locations for a production plant: one in the county of Norrbotten (coastal area) and one in the county of Kalmar. Both were analyzed based on three feedstock options: logging residues, bark and sawdust. The reduction of life cycle GHG emissions, compared with the fossil fuel comparator and according to the EU Renewable Energy Directive II, amounted to 85–95 % for the FT crude produced in northern Sweden, and to 92–97 % in southern Sweden, depending on transportation distances and feedstock utilized.

Based on the attained results, the process as described is in theory a viable method to synthesize middle distillates using woody biomass as raw material. For all subprocesses, the efficiencies are in practice assumed to be typical and a combination of them in this manner uses forest residues to produce jet fuel. Nevertheless, to properly evaluate its practicability in a production plant, additional studying is required. Integration aspects need a great deal of attention, and focus should be on how the subprocesses of proven function operate as connected in entirety.

SAMMANFATTNING

Den genomsnittliga temperaturökningen i atmosfären är delvis orsakad av det nuvarande användandet av fossila bränslen och dess associerade nettoutsläpp av koldioxid. Den globala tillgången på fossila bränslen är dessutom begränsad, och därför är behovet av alternativa drivmedelskällor påtagligt. Hållbarheten hos elektrifiering är diskuterbar, och energidensitet är en svår parameter att öka. Luftfarten är troligen särskilt omständlig att elektrifiera, men en ny energikälla behövs. Produkten från den studerade kemiska processen i detta projekt är kompatibel med nuvarande infrastruktur och teknik, och härrör från förnybara råvaror av lågt värde.

Sammanfattningsvis består uppställningen av fyra huvudsakliga integrerade delprocesser: hydrotermisk förvätskning (HTL), indunstning, förgasning och Fischer-Tropsch-syntes (FT). Biomassa, som i detta projekt är skogsrester, blandas med återvinningsströmmar från indunstningen och förgasaren, trycksätts, värms upp och matas in i HTL-reaktorn. HTL-produkten indunstas och pumpas uppkoncentrerad in i en syrgasblåst och trycksatt suspensionsförgasare. Syngasen som lämnar förgasningen gaskonditioneras med vattengasskift i en blandning med reformerad tailgas från FTsyntesens produktseparering och blandas upp med rå tailgas innan FT-reaktorn. FT-oljan separeras från reaktorprodukten och går förslagsvis vidare till uppgradering, som är utanför projektets ramar.

Hela processen ställdes upp i beräkningsprogram och resultaten användes för att utföra en livscykelanalys (LCA). Energi-, mass- och kolverkningsgraderna som erhållits var 34,5 %, 20,2 % respektive 32,3 %, och dessa bedöms som rimliga i ett bioraffinaderi. I indunstningen av HTLprodukten åstadkoms en ångekonomi på 3,45, vilket också är fullt godtagbart. Fördelningen av kolkedjelängder hos FT-oljan hade sin topp vid C10 och en tung högersvans som lämpar sig för krackning. LCA:n visade på hög GHG-prestanda i de två studerade fallen med en anläggning lokaliserad i Norrbottens län (kustområde) och en i Kalmar län. Båda analyserades utifrån tre råvarualternativ: avverkningsrester, bark och sågspån. Reduktionen av växthusgaser, jämfört med fossila alternativ och baserat på EU's Renewable Energy Directive II, uppgick till mellan 85–95 % för FT-oljan producerad i norra Sverige respektive 92–97 % i södra Sverige, beroende på transportavstånd samt vilken råvara som utnyttjas.

Av resultaten att döma, är den beskrivna processen en teoretiskt genomförbar väg att gå från skogsbiomassa till syntetiserade medeltunga destillat. Verkningsgraderna antas i praktiken typiska för varje delprocess och en sekvens av dem på detta sätt nyttjar skogsrester till att producera jetbränsle, men dess praktiska implementerbarhet i en produktionsanläggning behöver vidare efterforskning för att kunna bedömas korrekt. Speciellt integrationsaspekter bör uppmärksammas, och hur de relativt beprövade delprocesserna fungerar som anslutna i helhet.

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1 INTRODUCTION

The current usage of fossil fuels with its net emission of carbon dioxide brings about an increase in the average temperature of the global atmosphere (Anderson et al., 2016; Mitchell, 1989; Stern, 2007). This global warming, in combination with a debatable peak oil, impel the energy sector to change towards producing a greater deal of renewable energy and reducing carbon dioxide foot-prints. Commonly, realization of production concepts based on renewable feedstocks would fail as a consequence of relatively low yields and, consequently, poor economic performance. External political drivers and frameworks are typically needed for their ability to compete with fossil-based equivalents.

While vehicle electrification is a solution to locally eliminating carbon dioxide emissions, it must be ensured that the electricity comes from renewable energy sources, not to mention the extraction and recycling of rare-earth metals for batteries facing many challenges from an environmental perspective. However, electric passenger cars are becoming more common, and arguably, haulage may follow. One sector which may take longer to electrify is aviation, particularly medium and longhaul flights. It is therefore of great interest to find methods on renewable grounds to obtain jet fuel.

This paper delineates the project SunAlfa, aiming to investigate, simulate and develop a large chemical process centered around hydrothermal liquefaction (HTL), using a biomass feedstock (here forest residues, which may be pretreated with e.g. steam explosion), and where the HTL product (with a hydrophobic phase referred to as bio-oil) can be (i) catalytically hydrotreated to hydrocarbons and (ii) gasified for the production of syngas to synthesize hydrocarbon fuels via the Fischer-Tropsch (FT) process. This project, that originally aimed to evaluate both pathway (i) and (ii), was during the course of the project redirected to a strong focus on the second gasificationbased conversion route. The rationale for this re-balanced focus in this project was the significant uncertainties about practical feasibility for route (i) that was revealed after the initial literature study, not least coupled to the low but important levels of ash components that will be present in the HTL-liquid and represent a large uncertainty regarding life-time (and hence practical viability) for catalytic unit operations in a refinery. Furthermore, it was deemed to be difficult to establish a generic refinery configuration and there was therefore a risk that any simulation results would have poor generic relevance. It was concluded in the project team that the second route could be simulated and evaluated with higher accuracy and represented a more thoroughly examined technological area were systems for ash handling and their performance could be more readily found in literature.

The mass and energy balances as an output from the process simulations of pathway (ii) are used as an input for a life cycle assessment (LCA), and a techno-economic assessment is carried out to estimate expenditures associated with the construction of the process and its operation.

2 BACKGROUND

Brief background descriptions on relevant subprocesses are given in this section.

2.1 HYDROTHERMAL LIQUEFACTION

Hydrothermal liquefaction (HTL) is a biomass-to-liquid method in which thermochemical decomposition of a biomass feedstock yields an oil-like product commonly referred to as bio-oil or biocrude. Possible feedstocks include algae, forestry residues, lignocellulosics, husks, vegetable wastes, manure, sewage and garbage. The approximate temperature range at which the chemical process can be deemed as HTL is 520–647 K (Ahmad 2018, 194; Elliott et al. 2015, 147). At below 520 K, hydrothermal carbonization results in hydrochar, and at above 647 K, syngas is produced since gasification reactions prevail. Typical operating pressures of HTL applications are in the range of 4–22 MPa (Elliott et al. 2015, 147; Gollakota et al. 2018, 1379). Lower pressures do not allow for properly maintaining a liquid water phase. The upper bound of the interval signifies the supercritical pressure of water, which may be exceeded.

One advantage of liquefaction is the ability to treat wet biomass without the necessity to dry the feedstock. A disadvantage is the decreased carbon efficiency with the increased amount of organic carbon in the large water phase. Pyrolytic mechanisms are activated in HTL, and consequently, presence of a catalyst is not critical to achieve depolymerization as such. Nevertheless, the degree of depolymerization is enhanced by catalysis. The chosen catalyst in catalytic HTL is not seldom alkali salts due to the resultant tendency of the modified ionic medium to support base-catalyzed condensation reactions and thus potential formation of aromatic oil. The produced bio-oil is more deoxygenated than products from fast pyrolysis, and more hydrophobic. However, issues may arise with the coexistence of alkali catalyst and acids from the thermally treated feedstock, which potentially react and yield salts. On the other hand, non-catalytic HTL runs the risk of favoring acid-catalyzed polymerization reactions, leading to solid products (Elliott et al. 2015, 148).

From the HTL process, the liquid phases exiting are two: an aqueous phase and an oil phase. The oil phase comprises a complex mixture of different compounds and is relative to petroleum rich in oxygen. Thus, it is not to be considered a petroleum analog. The composition of the bio-oil depends on its source and the transformation route of the biomass to liquid. However, there are some general trends. First, the hydrocarbons generated are branched, and the oxygen atoms are found mainly in ketones/aldehydes, oxygen aromatics, carboxylic acids/esters, and alcohols (Seehar et al. 2021, 6). Broadly, a wide diversity of oxygenated compounds can be found in the bio-oil, such as acids, alcohols, ketones, phenols (including guaiacol in the case of softwood lignin), naphthols, furans and esters (Elliott et al. 2015, 148; Gollakota et al. 2018, 1389).

Indeed, also the fate of the heavy metals in the feedstock have been subject to investigation. The heavy metals have been found to primarily exit with the ash for compounds such as Cu, Cr, Ni and Zn, although the part that does not exit with the ash is found mainly in the bio-oil and not in the aqueous phase, with potentially as much as 20–30 % of the heavy metals (Seehar et al. 2021a, 9). When investigating other common alkali metals and alkaline earth metals in wood such as Na, K and Ca, these are found to mainly end up in the water fraction, although as much as 12% of the Na, 10 % of the K and 2 % of the Ca can be found in the bio-oil fraction from HTL at 350°C (Seehar

et al. 2021b, 15). At these conditions, approximately 57 % of the S and 12 % of the P is found in the oil (Seehar et al. 2021b, 15).

In an HTL-only application, for the product to achieve similarity with readily available light and middle distillate hydrocarbon fuels, hydrotreating is necessary (untreated bio-oil is a viable direct substitute for heavy fuel oil) (Elliott et al. 2015, 154). A fraction of the produced bio-oil may be recirculated to the bio-slurry preparation, in addition to a separated organic (including coke) aqueous recovery stream. A non-recirculated aqueous stream can be further processed with catalytic hydrothermal gasification to produce syngas, or undergo anaerobic digestion (limited owing to phenols and furfurals).

2.2 HYDROTREATING OF HTL-OILS

The biocrude leaving the hydrothermal liquefaction process (HTL) can have an oxygen content of around 10–20 wt%, making it into a non-desirable fuel source due to its lower heating value (Tzanetis et al. 2017, 1389). There could also be miscibility issues when attempting to add it to fossil fuels. Moreover, impurities such as sulfur, nitrogen and metals can cause damages to machine equipment and affect the local environment negatively due to emissions (Duarte et al. 2011, 2158–2164). This requires the oil to be modified such that it fits the specifications for both fuel standards and environment control. The most straight forward approach to do this is by treatment with hydrogen.

A great deal of literature is available on hydrotreatment of biomass pyrolysis oil derived from other processes than HTL, which is where inspiration is to be found and utilised. Although the composition of HTL-oils may differ to some extent from other biomass pyrolysis oils, identical challenges are faced, and therefore also similar upgrading procedures are applicable. Generally, the oils are treated with hydrogen in a reactor under 250–450°C and 0.75–30 MPa (Tzanetis et al. 2017, 1392). Every stage of the process can differ depending on the biocrude-oil composition and could even be performed in multiple steps with varying thermodynamic parameters.



Figure 1. Simplified flow sheet of the HTL pyrolysis oil purification process.

The simplified process flow chart describes the path of the crude oil to a valorised product, figure 1. Oxygen-rich biocrude-oil is mixed with hydrogen before being sent to a reactor were multiple hydrotreatment reactions occur. In the subsequent step, the product leaving the reactor is separated in a flash tank to a gas and liquid phase. Most of the hydrogen passes the reactor without being consumed. The hydrogen-rich stream is therefore directed to a pressure-swing adsorption unit to purify the gas before it gets recirculated (Snowden-Swan et al. 2017, 23). Make-up hydrogen is added to the recycle stream before it is mixed with the feed oil. The liquid phase leaving the flash tank is upgraded to a product with a higher heating value and increased compatibility with fossil fuels.

The upgraded oil is comprised of hydrocarbons with different lengths which need to be fractionated to sought-after specifications. The heaviest fraction might be in need of hydrocracking for shortening of the hydrocarbon chains (Snowden-Swan et al. 2017, 20).

2.2.1 Hydrotreating strategies

The major difference between bio-oils and petroleum oils are the extensively higher amount of oxygenates. However, the content of other commonly deleterious heteroatoms in petroleum oils such as nitrogen and sulphur are lower in bio-oils. Upgrading of bio-oils is derived from the substantial need of oxygen removal.

The hydroprocessing method considered to be most promising for upgrading bio-oils is catalytic hydrodeoxygenation, HDO. The technology was adapted from, and is also compatible, with existing hydrotreating processes used for conventional petroleum upgrading (Elliot, 2007; De et al., 2015; He & Wang, 2013). The process is similar to conventional heteroatom removal hydrotreatment processes, such as hydrodesulfurisation, HDS, and hydrodenitrogenation, HDN (He & Wang, 2013). HDO is unique as it provides a process for not only oxygen removal but also a possibility of depolymerisation and saturation of C=O, C=C bonds and aromatic rings concurrently (He & Wang, 2013; Salam et al., 2020). Furthermore, knowledge of hydrodemetallisation, HDM, i.e. removal of metals from residual oils can also be applied and extended to HDO of bio-oils (Arora et al., 2020). Presence of metals in the stream during hydroprocessing causes deactivation by covering of the active phase and pore blockage (Kohli et al. 2016, 269).

However, a wide variety of oxygenated compounds, including different functional groups and molecular weights is a challenge of hydrotreatment upgrading of bio-oils to liquid fuels. The reactivity of the multifunctional groups in the HDO process differs, and the operational temperature needed varies. Furthermore, compounds with low hydrodeoxygenation reactivity require relatively high hydrogen pressures. The combination of different iso-reactive temperatures and high pressures can result in complete hydrogenation of certain compounds (He & Wang, 2013). Furthermore, the unsaturated double bonds and oxy-compounds, e.g., aldehydes, ketones and olefins are reactive and are prone for condensation and polymerisation, which in turn can result in accelerated deactivation of the catalyst due to poisoning products as char and water (Wang et al., 2013; Arora et al., 2020).

A major challenge in hydrotreating bio-oils is the high oxygen content and the exothermic heat of reaction. During the processing, the reaction mixture heats up, and with more than 10-15 % oxygen in the feed, there is a significant risk of overheating the catalyst. This required dilution of the feed-stock, either by recycling or co-processing with feedstocks with lower oxygen content; or indeed an

approach for providing external cooling. To achieve a more controlled and desirable conversion of bio-oils to liquid fuels, the hydrotreatment upgrading process is commonly performed by two different technologies, namely two-step hydrotreatment of bio-oils and co-processing of bio-oils with petroleum fractions (Pinheiro Pires et al., 2019).

2.2.2 Two-step hydrotreatment

To eliminate unstable oxygenated compounds to polymerizate and complete hydrogenation of compounds with lower iso-reactive temperatures is hydrodeoxygenation of bio-oils preferable performed in a two-stage process (He & Wang, 2013; Pinheiro Pires et al., 2019).

The first stage is a mild hydrotreatment step and is performed below 300°C and 10 MPa. [T1] It serves as the stabilisation step of the reactive components. The aim is to convert ethers, methoxy-phenols and biphenols to phenols, and hence reduce polymerisation and coke formation (He & Wang, 2013).

The second stage is performed at more severe conditions, and temperature varies between 300–500 °C and with a hydrogen pressure of 13 MPa (Xu et al., 2013). This step is performed for sufficient hydrodeoxygenation and hydrocracking to gain preferable liquid fuel quality (He & Wang, 2013; Pinheiro Pires et al., 2019).

2.2.3 Co-processing of bio-oils with petroleum fraction

An alternative method of treating biocrude-oil is by mixing it with a fossil crude oil in a conventional refinery process. Depending on the biocrude oil composition there exists a limit on the amount that is possible add. Moreover, the biocrude oil must undergo a mild hydrotreatment to enhance the miscibility with fossil oil fractions and stabilise the most active oxygenated compounds. The coprocessing of oxygenated compounds has its disadvantages, such as increasing catalyst deactivation and promoting coke formation. Catalysts containing noble metal has shown to handle biocrude-oil better than the conventional hydrotreatment catalysts. Modification of existing refinery units is consequently of most importance to combat the issues of coprocessing (Graça et al., 2013).

2.2.4 Catalysts used

A crucial aspect during HDO process is the catalyst being used. The activity and stability of the catalyst is decisive for the upgrading of the bio-oils and the quality of the converted liquid-fuel. The catalyst deactivation is a major challenge during HDO processes of bio-oils (He & Wang, 2013). The physiochemical properties of the catalyst and operational conditions determine the product distribution (He & Wang, 2013; Salam et al., 2020). Furthermore, as previously discussed, the composition of bio-oils varies widely dependently on source and transformation route and hence contains extensively different compounds and functional groups. Different compounds result in different reaction mechanisms. The catalyst-support system activity and design affect in turn the reaction mechanism. The combination of a variety of compounds and reactive functional groups with affecting catalyst-support system makes the upgrading of bio-oils complex. Different amounts of heteroatoms and impurities result in different reaction mechanisms, and the deactivation of the catalyst is differently affected (Arora et al., 2020; Mu et al., 2013). A lot of development work in

this field is done by using model substances for gaining better knowledge about the reaction mechanism and deactivation of the catalyst.

Catalysts that have gained focus in utilising and in development of active catalysts in hydrotreatment and hydrodeoxygenation processes are sulfided transition metals, noble metals, metal oxides, non-precious metals, phosphides, nitrides, carbides and oxynitrides (He & Wang, 2013; Mu et al., 2013).

Sulfided CoMo and NiMo on alumina are commonly used catalysts in the industry for hydrodeoxygenation of bio-oils. Sulfided catalyst is the most reported catalysts used for hydrotreatment in literature (He & Wang, 2013). These catalysts are traditionally used in conventional hydrotreatment process for removal of sulphur and nitrogen in oil refineries, in which the reaction mechanisms are well defined. Sulfided CoMo/NiMo/Al₂O₃ catalysts have been proven to remove oxygen in bio-oils (Arora et al., 2020; Mu et al., 2013). However, a challenging aspect during HDO using sulfided transition metals are their poor stability due to their sensitivity towards H₂O. H₂O is the main product in HDO (Arora et al., 2020), and can lead to accelerated deactivation of sulfided transition metals due to the poisoning effect of H₂O (He & Wang, 2013; Mu et al., 2013). Another aspect of using sulfided catalysts for hydrotreatment is their need for a certain concentration of sulphur in the feed in order to maintain the conversion (He & Wang, 2013). Due to the essential difference between the content of bio-oils and petroleum oil, further development within this area is necessary (Arora et al., 2020; Mu et al., 2013). Bio-oil upgrading with sulphur-free catalysts would from an economically and environmentally perspective be preferable (He & Wang, 2013).

Catalysts with platinum, ruthenium, rhodium and palladium are other commonly used catalysts in hydrotreatment research (only Pt is commercialised to the knowledge of the authors) and have gained focus in development work within this area. These catalysts are tolerant towards H₂O and other solvent used in HDO. Noble metal catalyst-support systems have shown high activity for the hydrogenation processes. However, these catalysts are more expensive (He & Wang, 2013; Mu et al., 2013). A possible recycle technique for reuse of these catalysts could also be challenging (Mu et al., 2013). Furthermore, impurities such as sulphur and iron have a greater poisoning effect towards noble metal catalysts than sulfided catalysts (He & Wang, 2013).

2.3 GASIFICATION

Gasification is the process in which carbonaceous materials, for example biomass, coal or various petroleum fractions, can be transformed into synthesis gas using controlled (sub-stoichiometric) amounts of steam, oxygen or a combination of them at high temprature (Higman & van den Burgt, 2008). Synthesis gas, or syngas, is primarily a mixture of carbon monixide (CO) and hydrogen (H₂) but can also contain additional gaseous compounds at various concentrations, such as carbon dio-xide (CO₂), hydrogen sulfide, tars and light hydrocarbons. Syngas with such contaminants present is usually called raw syngas, indicating that it is not yet cleaned and conditioned to facilitate upgrading. The concentrations of such contaminants depend on the gasification technology used and the process conditions, as discussed further below.

A gasification process can be implemented in many types of equipment, the most important being fixed beds, fluidized beds and entrained flow reactors. Fluidized beds and entrained flow reactors are considered most relevant for large scale biomass gasification (Molino et al., 2018). Fluidized

bed processes are either so-called indirect or indirect gasification. In indirect gasification only steam is used as the gasification agent and the heat for the endothermal process needs to be supplied externally. This is usually solved by a dual fluidized bed setup, in which a combustion reactor supplies the heat through transfer of bed material between the reactors. This is the technology utilized in the GoBiGas demonstration plant (Larsson et al., 2018). In direct gasification, which can be implemented in both bubbling and circulating fluidized bed setups, oxygen and steam are used in combination. Fluidized bed gasification typically gives significant concentrations of methane and tars in the produced syngas, with potentially lower concentrations for direct gasification due to the oxygen supply, which, however, of course comes at a cost. Direct fluidized bed gasification has been implemented in demonstration scale at Gas Technology Institute in Chicago, IL and Skive CHP in Denmark.

Oxygen-blown entrained flow gasification of biomass does not use a fluidized bed. Instead the biomass feedstock is fed together with sub-stoichiometric oxygen in a burner nozzle, forming a local high temperature flame. A main development effort for this technology has been related to black liquor gasification (BLG), which has been extensively demonstrated in Piteå, Sweden (Landälv et al., 2014; Furusjö et al., 2014). There have also been significant development efforts for pulverized wood (Weiland, 2015) and a demonstration is currently underway for torrefied biomass in the BioTFuel project in France. Using a liquid feedstock, such as black liquor, in entrained flow gasification means pressurization can be accomplished, leading to a number of potential advantages. For black liquor, the combination of entrained flow technology and the catalytic effect of alkali in the black liquor has been shown to lead to low concentrations of methane and very low concentrations of tar (Landälv et al., 2014; Bach-Oller et al., 2015, 2017a, 2017b).

The process concept investigated, including a combination of liquefaction and gasification, aims at utilizing the addition of alkali salts to provide benefits in both the liquefaction step (see above) and the gasification step. In gasification, the positive effect of alkali in carbon conversion, tar formation and soot formation has been demonstrated for both fluidized bed (Thunman et al., 2018; Larsson et al., 2021) and entrained flow (Bach-Oller et al., 2015, 2017a, 2017b, 2019; Umeki et al., 2017). However, presence of alkali implicates the need for durable masonry in the gasifier. Due to the similarity of the liquefied biomass gasification feedstock to black liquor, the entrained flow BLG technology has been used as a model for the gasification process evaluated in this work.

2.4 GAS CLEANING

2.4.1 Sulphur removal

Sulphur is the most important catalyst poison as all natural organic materials contain sulphur. For natural gas, sulphur is removed as the first step in processing as sulphur is a poison for the reforming catalyst. For oil, coal and biomass, sulphur is removed from the raw syngas generated by the gasifier (or steam-cracker). Sulphur bonds irreversibly (at reaction conditions) to the active metal sites on the catalyst, disabling and blocking the active sites from CO adsorption.

Removal of sulphur containing gases is often referred to as acid gas treatment, as the compounds containing sulphur are acidic in contact with water. Sulphur in raw syngas is predominantly in the form of H₂S, but organic sulphur compounds are usually also present, such as COS and CS₂. For

biomass and coal gasification, larger sulphur containing compounds such as thiophene and dibenzothiophene are common, but not in high concentration. Organic sulphur components increase the challenges of sulphur removal as the physical properties of the organic sulphur differ from that of H_2S .

Sulphur can be removed either with physisorption or chemisorption and the choice of which technique to use depends on raw syngas composition, economy, etc. Common physisorption techniques Rectisol, Selexol and Purisol, use methanol (Rectisol), dimethyl ethers of polyethylene glycol (Selexol) or N-methyl-2-pyrrolidone (Purisol). These processes also have the benefit of removing CO_2 from the raw gas and to separate H₂S from CO₂.

Chemisorption of sulphur is performed using fixed bed adsorption with usually ZnO as the active material. Metallic zinc, copper, iron, nickel etc. in catalysts react with sulphur to form sulphided metal sites. Plants using a physisorption step usually have a small fixed bed of ZnO prior to synthesis, known as a guard bed, to capture the last of the sulphur. Most synthesis catalysts are susceptible to sulphur poisoning, which may be mitigated by using spent synthesis catalyst in a sulphur guard bed.

2.5 GAS CONDITIONING

Two major gas conditioning subprocesses are employed: reforming to minimize methane in the FT crude and water-gas shift to increase the H_2 :CO ratio to 2,05 (slight excess of H_2 in relation to the ideal ratio of 2 since the FT reactor conversion is limited by the amount of CO).

2.5.1 Reforming

Raw gas from most gasifiers contains hydrocarbons such as methane, ethylene, benzene etc. Most of the hydrocarbon part is methane. The hydrocarbons in raw gas from gasifiers can account for a large part of the total heating value of the gas. However, in the considered gasifier, methane formation is limited to 1%, but there is a significant amount in the tail gas from the synthesis. To utilize the energy, a reformer is employed to form more syngas, according to reaction (1).

$$C_{x}H_{y} + x H_{2}O \leftrightarrow x CO + (x + 0.5y)H_{2}$$
(1)

The reforming reactions are endothermic and heat for the reactions can be supplied either direct, by adding oxygen to the gas and thereby partially oxidizing the methane to generate heat, or indirect, by burning fuel on the outside of the reactor tubes. Direct heated reforming reactors are termed auto-thermal reforming (ATR) and operates around 1000 °C, with higher temperatures in the flame caused by the injection of oxygen. Indirect heated reactors are called steam reforming (SREF) reactors because steam is used to break down the hydrocarbons as opposed to a steam/oxygen mixture used by ATR. Both types are operated with a catalyst, usually nickel-based, which is deactivated by sulphur. A third reforming reaction is available that operates at even higher temperatures (>1300 °C) without a catalyst. This is known as partial oxidation (POX). The drawback of POX is a lower thermal efficiency, but the strength of POX is a chemically stable reactor.

A pre-reformer is necessary to have when compounds such as ethylene or benzene in the reformer system feed are present as these form coke and soot on the reformer catalysts (ATR and SREF).

For Fischer-Tropsch, reforming is preferred over flaring, as the energy bound in the light fraction that is separated out after synthesis can then be partially recovered.

2.5.2 Water-gas shift

The gas generated by the front-end in a synthesis plant is usually not suitable for synthesis without further processing. A typical gasifier product has a H_2 :CO ratio of 1:1, and that of steam reformer product is 3:1. The water-gas shift (WGS) reaction adjusts the ratio between H_2 and CO in the synthesis gas according to the equilibrium reaction (2).

$$CO + H_2O \leftrightarrow CO_2 + H_2$$
 (2)

The lower the temperature, the further towards H_2 the reaction is pushed. Shifting towards H_2 is slightly exothermic. A temperature increase of 100–150°C over the reactor is not uncommon if shifting all the way towards H_2 . Inlet temperatures, depending on process parameters, range from 200°C to 350°C. Since the reaction goes towards equilibrium, controlling the H_2 :CO ratio is achieved with a bypass of the reactor, see figure 2.



Bypass valve

Figure 2. WGS-reactor configuration with bypass.

The catalysts used in WGS reactors can be categorized as operating at high or low temperature. Commercial high temperature WGS uses Fe/Cr and operates at 300–550°C. The catalyst can handle low levels of sulphur at a loss of some activity. Commercial low temperature WGS uses Cu/Zn as a catalyst which is highly susceptible to sulphur poisoning. Low temperature shift is normally only used when the gas needs to be shifted all the way towards H₂ and is operated at 200–250°C. A third option for commercial WGS-catalysts is Co/Mo which is a sulphur activated catalyst. It requires a minimum of 300 ppm sulphur in the gas, which is usually too high for biomass feedstocks that normally have some 20–100 ppm of sulphur. Co/Mo is active in the range 200–500°C meaning it can be operated both at low and high temperature. In most cases, the water-gas shift reaction is performed to increase the concentration of H₂, as higher temperatures (>500°C) are required to significantly increase CO concentrations. However, in recent years there has been an increased interest in utilizing CO₂ in performing reverse water-gas shift, by reacting H₂, from for example electrolysis, with CO₂ to produce hydrocarbons.

2.6 FISCHER-TROPSCH SYNTHESIS

Fischer-Tropsch (FT) synthesis, developed in the early 20th century Germany, is broadly defined as a polymerization reaction of CO and H₂. The reaction is not an equilibrium reaction, unlike, for example methane and methanol synthesis. Because of this, conversion of CO and H₂ could approach 100 %, at least in theory. In practice however, the conversion is much lower at 60–80 % once-through. The reaction is highly exothermic owing to the breakage of less stable π bonds and the formation of more stable σ bonds in polymerization, which is one of the reasons why the conversion is kept low. This is usually achieved by recycling of gas over the reactor to keep concentrations of reactants lower.

The overall reaction for Fischer-Tropsch follows reaction (3), where $-CH_2$ - represents a methylene group in the hydrocarbon chain.

$$CO + 2H_2 \rightarrow -CH_2 - + H_2O \tag{3}$$

2.6.1 Fischer-Tropsch process

A clean synthesis gas $(CO + H_2)$ is required for most syntheses, be it methanol, ammonia or Fischer-Tropsch (FT). A thorough gas cleaning is required for most raw syngases generated by the front-end (gasifier, reformer etc.). Furthermore, different feedstocks yield producer gases with different levels of contaminants. After gas cleaning, depending on the composition of the producer gas, a reformer step may be necessary to increase the yield of liquid products. Figure 3 shows a flowsheet of a typical FT synthesis setup with a reformer followed by a water-gas shift reactor.



Figure 3. Typical process layout from raw synthesis gas to FT-fuels.

For systems with a gasifier that produces a raw syngas with a very low amount of hydrocarbons such as an entrained-flow gasifier, the reformer and water-gas shift reactor can be moved (see figure 4) to only handle the recycle after the FT-reactor as the methane in the raw syngas need not be reformed if the concentration of methane is low.



Figure 4. Process layout for an FT synthesis with low levels of hydrocarbons in the raw syngas.

The major advantage of the process in figure 4 is that the reformer and water-gas shift reactor can be made smaller as it is only processing the recycle stream, instead of the total flow.

2.6.2 Catalysts

The FT reaction requires a suitable catalyst and commercially two different catalysts are in use, an iron-based and a cobalt-based catalyst. The iron-based catalyst is usually denoted high temperature Fischer-Tropsch (HTFT) and the cobalt low temperature Fischer-Tropsch (LTFT). High temperature in this case refers to above 300 °C and low temperature is in the range 220–250°C.

The mechanism is believed to be a stepwise chain-growth reaction. Due to the nature of such reactions, being rather stochastic, the product composition follows a distribution curve as a function of hydrocarbon length.

The Schulz-Flory distribution (4) where α is the probability factor, adapted from the work performed independently by Schulz and Flory in the mid-1930's, gives the mole fraction (m_n) of molecules with *n*-number of carbon atoms.

$$m_n = (1 - \alpha)\alpha^{(n-1)} \tag{4}$$

If this distribution is adjusted for the molecular mass of each hydrocarbon, the expression becomes

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

The weight-based distribution (depicted in figure 5) shows the mass fraction of hydrocarbons (or a range) as a function of the probability of forming the specified hydrocarbon. The probability factor is the result of the synthesis system design i.e. temperature, catalyst, gas composition etc. An LTFT reactor would normally produce a product with a probability factor (α) around 0,9, whereas an HTFT-reactor would yield one around 0,7.

As can be seen in figure 5, an α -value of around 0,7 would be in the gasoline range. The hydrocarbons produced in Fischer-Tropsch are dominantly straight-chain paraffins and olefins, which usually have too low octane rating to be useful as gasoline. Straight-chain paraffins are more suitable for jet- and diesel fuels, even though oil refinery operations such as isomerization and aromatization can be used to increase the octane rating of such hydrocarbons.



Figure 5. Schulz-Flory distribution-based product composition for FT-products as a function of the probability of forming specified hydrocarbon.

Depending on the conditions (high H_2 concentrations, temperature etc.) termination of the chain can occur either with a H_2 or a CO, resulting in paraffins, olefins and oxygenated products.

HTFT reactor conditions yield a product containing alcohols, aldehydes, aromatics as well as the primary products – paraffins and olefins.

2.6.3 Reactor design

The first reactors in operation were of a fixed bed design, which is still used today. The fixed bed reactors are normally of a multi-tube design with the tubes full of catalyst and cooling between the tubes. Multi-tube reactors have the advantage of simple design and operating conditions. The clear downside to the reactors is the difficulty to control reactor temperature. Controlling reactor temperature in FT is more difficult than in for example methanation and methanol synthesis. The catalyst used for methanation can handle a higher and wider temperature span, while maintaining selectivity and conversion. For methanol synthesis, the conversion is restricted by the chemical equilibrium. For FT, the temperature needs to be as steady as possible to control product distribution and prevent catalyst damage due to overheating.

Fluidized bed reactors allow a better temperature control but increase the level of complexity. The disadvantage to the fluidization is an increased attrition of the catalyst particles.

The third reactor type is the slurry reactor. In the slurry reactor three phases are present – the gas phase where the reactants and light products resides, the liquid phase with heavy products and solvent and the solid phase containing the catalyst. The slurry reactor has the biggest advantage when it comes to temperature control and the ability to remove heavy products and waxes from the catalyst. The disadvantage of the slurry reactor comes from the three-phase system in itself. Three phases severely decrease the mass-transfer as the reactants must diffuse from gas to liquid and finally into the catalyst particle.

Fixed and slurry reactors are used for LTFT and the fluidised bed reactors are used for the HTFT synthesis.

3 THE SUNBTL PROCESS

Presently, the main feedstock considered for the SunBTL chemical process is forest residues. The eventual pretreatment of biomass, albeit not simulated at present, in the case of softwood feedstocks in this project consists of a steam explosion reactor to break down the lignocellulosic bonding before preparation of a bio-slurry. As can be seen in figure 6, an alkali catalyst solution produced by the gasifier and optional additives are added to the slurry, as well as a recirculated aqueous stream from the evaporator.

The bio-slurry is conveyed to a pump where it is pressurized and run through a series of three heat exchangers. The first (HEX1) is fed with decompression gas and vapor from the last effect. The second (HEX2) is feed-effluent and fed with HTL product recuperatively. The third (HEX3) is fed with hot oil, heated by an external source. Subsequently, the feed is introduced into the HTL reactor where a hydrophobic bio-oil and an organic aqueous phase is formed, in addition to solids and gas.





Evaporation is conducted in a backward feed multiple-effect evaporator, and its flowsheet is displayed in figure 7. The backward feed choice is motivated by the viscous feed and the need for a higher temperature at higher dry matter contents to maintain pumpability. The primary steam is currently generated utilizing the exothermic conditions of the FT synthesis. Prior to entering the last (vapor side) effect, the cooled HTL product exiting the hot side of HEX2 is depressurized and flashed (vessel A) where water, most of the non-condensable gases and some volatile organic compounds are assumed to evaporate. The decompression gases are mixed with the vapor from the last effect before exchanging latent heat with the bio-slurry by condensing through HEX1.

The mixture used as a heating medium in HEX1 is mixed with the other condensates (originally vapor from flash chambers) from all but the last effect. The resultant saturated mixture is flashed

(vessel B), and the major part of the liquid is recirculated to the bio-slurry to attain an appropriate water content for the HTL, while the excess is bled off. The gas from the flash drum is discharged to further processing, possibly flaring or anaerobic digestion. The HTL product is stepwise concentrated through the effects until reaching a satisfactory dry matter content for gasification. Thereafter, the concentrated product is gasified to yield syngas which after cooling is input to the FT synthesis.





As is shown in figure 8, water-gas shift (WGS) is performed on the syngas feed and reformed tail gas to shift the CO/H_2 ratio more toward H_2 . The FT reactor is calibrated to maximize production of hydrocarbons in the light to middle distillate range, and the FT crude is separated from the aqueous and gas phases. A large fraction of the carbon dioxide is expelled from the gas phase in a subsequent separator. The remaining gas enters a pre-reformer (PR) with steam which cracks carbon chains with two carbon atoms or more. A steam methane reformer (SMR) then shifts the tail gas from methane in favor of carbon monoxide and hydrogen.



Figure 8. Fischer-Tropsch subprocess flowsheet.

4 METHOD

4.1 PROCESS MODELLING

An advanced modelling software is required to simulate a chemical process with recycle loops, advanced chemical reactions and separation. Aspen Plus was employed to model and simulate the hydrothermal liquefaction subprocess, evaporation and synthesis plant from syngas to product. The property method NRTL was used unless otherwise specified. The gasification calculations were performed in Matlab.

4.1.1 Hydrothermal liquefaction and evaporation

Softwood with an assumed dry matter content of 50 wt% is used as feedstock, modelled as a set of monomers, applying composition data from Anca-Couce & Obernberger (2016, 164). The hemicellulose is assumed to be composed in accordance with data on Pinus radiata as obtained by Reyes et al. (2013, 177). Cellulose is here modelled as glucose, and lignin as guaiacol. The mass fractions of water and monomers are shown in table 1.

Substance	Mass fraction
Water	50%
Glucose	24.2%
Guaiacol	15%
Mannose	5.5%
Xylose	2.8%
Galactose	1.8%
Arabinose	0.7%

Table 1. Composition of the recusions.	Table	1. Com	position	of the	feedstock.
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The mass flow of wet biomass feedstock is set to 50 000 kg/hr. The feedstock is diluted to 4 parts of water (including the water content of the biomass) per 1 part of dry biomass, i.e. an additional 75 000 kg/hr of water. The catalyst chosen is sodium carbonate, with a mass flow of 3 257 kg/hr solved in the 50 000 kg/hr alkali catalyst solution stream from the gasifier, resulting in a sodium content per dry organic matter of approximately 7.7 % in the gasifier feed to enhance the carbon conversion. To maintain the dilution, the splitter is set to return 28 257 kg/hr water as part of the aqueous recycle stream, settling in at a total of 29 864 kg/hr. All sodium carbonate is assumed spent in the HTL reactor, with all sodium exiting only in sodium acetate, and returning to sodium carbonate in the gasifier quench. The discharge pressure of bio-slurry in the main pump is set to 200 bar (a).

Hot oil composed of 26.5 wt% diphenyl and 73.5 wt% diphenyl ether enters HEX3 at 395 °C and 12 bar, allowing the HTL feed to reach 300 °C upon exiting HEX3. Neglecting the effects on the HTL stream temperature in the reactor and assuming adiabatic transport of hot HTL product to HEX2, product enters at 300 °C and allows the feed to reach 290 °C. The sensible heat required from the hot oil to boost the feed 10 °C implies a heat exchanger duty of 2.76 MW which, while also maintaining the assumed minimum cold inlet-hot outlet temperature difference of 10 °C, renders a hot oil flow sufficient at 41 000 kg/hr.

The yields in the HTL reactor are assumed to follow experimental characterization data compiled by Tzanetis et al. (2017, 1391–1393) at 300°C with a sodium carbonate catalyst, where a reference compound is used for each significant class (aromatics, hydrophobic ketones, light aliphatic acids, heavy aliphatic acids, furans, hydrophobic phenols, hydrophilic acids, alcohols, hydrophilic phenols and hydrophilic ketones). Fixing yields in this manner gives rise to an atom balance error, which is further elaborated on in sections 5.2 and 6. The mass fractions of the HTL product substances, as recalibrated to conform to the dilution in the process, are displayed in table 2.

Substance	Mass fraction
Water	77.7%
Sodium acetate	3.88%
Graphite	1.70%
2-methyl-2-cyclopenten-1-one	0.76%
Pelargonic acid	0.21%
Trimethyl orthobenzoate	0.55%
2,5-dimethylfuran	342 ppm
Guaiacol	1.30%
Oleic acid	3.93%
Glycolic acid	4.74%
Methanol	1.20%
Phenol	0.13%
Acetone	0.60%
Carbon monoxide	0.15%
Carbon dioxide	3.10%
Hydrogen	33 ppm
Methane	329 ppm

Table 2. HTL reactor yields.

The HTL product exits HEX3 at 118°C and is flashed in vessel A at atmospheric pressure. The liquid is fed to the last (vapor side) evaporator effect, and thereafter pressurized in steps of 1 bar through each of the following effects, until leaving the first (vapor side) effect at 5 bar(a) and 181 °C, whereupon the gasifier feed pump discharges at 31 bar (a). 14,5 MW of primary steam at 12,5 bar (a) is used in the first effect.

An HTL-only setup was simulated for comparison to the extended process, excluding evaporation, gasification and FT synthesis. This setup contains a separator for the HTL product to directly produce bio-oil.

4.1.2 Gasification

The gasification process was modelled using a modified thermodynamic equilibrium calculation (TEC), based on Gibbs energy minimization. The model was originally developed for black liquor (Carlsson et al., 2010; Furusjö & Jafri, 2018a) but has subsequently been used for alkali-enhanced gasification of solid biomass (Furusjö et al., 2018b; Carvalho et al., 2016, 2017a) and for co-gasification of pyrolysis oil with black liquor (Carvalho et al., 2017b).

Feedstock compositions used as inputs to the TECs were taken from the HTL process simulations. The gasification reactor was modelled as an oxygen-blown entrained-flow reactor pressurized at 30 bar. The model solves an energy balance to calculate the amount of oxygen required to reach a reactor temperature of 1050°C. Based on past experimental studies for black liquor, the methane content was fixed at 1 mol% (Furusjö et al., 2018a). Heat loss from the reactor was set to 0.7 % of the feedstock energy input.

4.1.3 Fischer-Tropsch synthesis

Input to the model is a cleaned producer gas, assuming no tar, sulphur or other trace elements. Lower hydrocarbons such as methane, ethylene, propylene etc. are however included, as these affect efficiency calculations. For entrained-flow gasification, such as is the case for HTL-product gasification, only a few percent of the gas contain hydrocarbons, and thus, the configuration used in figure 3 but with WGS after mixing with raw syngas is modelled. A flowsheet can also be seen in figure 8.

Compression prior to FT synthesis is not needed as the gasification pressure is sufficient for a moderate FT synthesis pressure of 30 bar(a) (Schubert et al. 2001, 459–464). The model uses a multi-stage compressor with intercooling in three stages to 70 °C. The final compression stage in this compressor is without cooling to allow a temperature increase before the downstream synthesis which is operated at higher temperature.

The FT-reactor was modelled as a plug-flow reactor with a kinetic model following equation (4) in subsection 2.6.2. The α -value was set to 0,9 to optimize the trade-off in depressing formation of both methane and heavy waxes. Reactor temperature was set to 250 °C as in LTFT and pressure at 30 bar (a) to fully utilize the gasification pressure.

Separation of the FT-products was conducted in a flash-vessel at 30 °C and 10 bar(a) which gives a cut-off (where 50 % of the compound, or hydrocarbon length, is in vapor phase and 50 % in the liquid phase) around C7. 70 % of the carbon dioxide is expelled in the carbon dioxide separator.

Reforming is modelled as a Gibbs free energy minimization reactor, which gives an equilibrium concentration on the outlet of the reactor. Temperature for the equilibrium is set to 200 °C above the reactor temperature to give a more correct composition. SREF is operated at 800 °C.

Water-gas shift is also modelled as a Gibbs free energy minimization reactor. The temperature at the inlet of the reactor is set to 300° C to balance fast reaction rates with low carbon dioxide production, and a bypass of the reactor is controlled to set the desired ratio of 2,05:1 for H₂:CO.

4.2 LIFE CYCLE AND TECHNO-ECONOMIC ASSESSMENT

The life cycle assessment (LCA) of the biofuel production system investigated in this project include two parts. The first part contains an assessment of the feedstock potentials in form of logging residues from final felling, sawdust from sawmills, and bark from sawmills and pulp mills, which are estimated to be suitable feedstocks for the production of the FT-fuel in focus. Two different locations for a future biofuel plant are assessed, one in the northern part of Sweden, in the city of Luleå in the county of Norrbotten, and one in the southern part of Sweden, in the city of Mönsterås in the county of Kalmar. Based on the potential supply of logging residues, sawdust and bark in these regions, theoretical transportation distances to a large-scale biofuel production facility are calculated.

The second part of the LCA include calculations of the greenhouse gas (GHG) performance of the FT-fuel produced. These calculations include six different alternatives, namely a production facility in Mönsterås fuelled with logging residues, bark or sawdust, and a production facility in Luleå fuelled with logging residues, bark or sawdust. The life cycle GHG calculation methodology is based on the EU Renewable Energy Directive, which is described in detail in following sections. An alternative calculation method is also included based on the ISO standard of Life Cycle Assessment (ISO 140 44), which apply an expanded systems boundaries approach also covering potential changes in soil carbon content.

4.2.1 Assessment of biomass feedstock supply and transportation distances

The total yearly demand of biomass feedstock to the biofuel plant in focus is assumed to correspond to 220 000 tons dry matter (DM). The biomass potential in form of logging residues in the respective regions is based on data from the Swedish Forest Agency (2015) and include restrictions regarding long-term environmental sustainability. The biomass potential in form of sawdust and bark is based on data from the Swedish Forest Industries (2020) regarding the number, location and production capacity of sawmills and pulp mills in the respective regions. The amount of bark generated per cubic meter solid saw timber and pulp wood is estimated to, on average, 27 and 30 kg DM, respectively (Karjalainen, 2018). The amount of sawdust is estimated to, on average, 48 kg DM per cubic meter solid saw timber (Swedish Environmental Protection Agency, 2010; Novator 2019). In table 3, the gross potential of logging residues, sawdust and bark is summarised for the counties of Kalmar and Norrbotten. The area included for the Norrbotten county refer to the coastal region and thus exclude the inland mountain area (Swedish Forest Agency, 2015). A conclusion from table 3 is that the total gross potential of logging residues exceeds the estimated yearly biomass feedstock demand of 220 000 tons DM in a large-scale biofuel production facility, whereas bark almost fulfil the demand but where sawdust does not. Added together, the total gross biomass potential is almost similar in the two regions, or between 560 000-580 000 tons DM. This is equivalent to roughly 2.5 times of the total biomass feedstock demand in the biofuel production plant modelled in this study.

Biomass feedstock	Norrbotten Cour	nty (coastal area)	Kalmar County						
	Best estimate Interval		Best estimate	Interval					
Logging residues – final felling	300	250—350	230	200–260					
Bark – sawmills	40	35–45	90	75–105					
Bark – pulp mills	150	130—170	110	95—125					
Sawdust – sawmills	70	60—80	150	125–170					
Total	560	470-650	580	495-660					

Table 3. Calculated gross biomass feedstock potentials in form of logging residues, bark and sawdust in the county of Norrbotten (coastal area) and Kalmar, expressed as 1000 ton DM.

The calculations of the theoretical transportation distances of the various biomass feedstocks to the biofuel plant is based on the potential gross supply presented in table 3, and the assumptions that the average transportation distance from the biomass recovery area to the biofuel plant is equivalent

to 70 % of the radius of the recovery area (which is shaped as a half circle since the city of Luleå and Mönsterås are located at the coast), and that the practical road transportation distance is 20 % and 30% longer than the linear distance in the county of Kalmar and Norrbotten, respectively (Börjesson and Gustavsson, 1996). In table 4, the estimated theoretical transportation distances of the biomass feedstock to the large-scale biofuel plant are shown, as well as the share of the supply compared to the total demand. A conclusion from table 4 is that the theoretical transportation distances are estimated to vary from 55 to 91 km in the county of Norrbotten, and from 17 to 75 km in the county of Kalmar, where the shortest distances refer to bark, followed by sawdust.

Table 4. Estimated theoretical transportation distances of logging residues, bark and sawdust in the county of Norrbotten (coastal area) and Kalmar to a biofuel plant located in the city of Luleå and Mönsterås, respectively, and the share of the yearly feedstock demand met by the respective feedstocks (based on gross biomass potential).

Biomass feedstock	Norrbotten Cour	nty (coastal area)	Kalmar County		
	Theoretical trans- portation distance ¹ (km)	Share of biomass supply of total demand ² (%)	Theoretical trans- portation distance ¹ (km)	Share of biomass supply of total demand ² (%)	
Logging residues	91	>100	75	>100	
Bark	55	87	17	91	
Sawdust	76	32	38	68	

¹Based on gross biomass potentials.

²The yearly biomass feedstock demand in the biofuel plant corresponds to 220 000 tons DM biomass.

4.2.2 LCA calculation methodology

The life cycle GHG emission calculations were in this study performed according to the revised European Union's Renewable Energy Directive, RED II (EU, 2018), which is to be implemented in national legislation in 2021. The reason why this specific methodology is utilized is that the Swe-dish GHG reduction obligation system in the road transportation sector, and in the aviation sector from 2021, is built upon the GHG performance of the blend in biofuels according to the calculation method defined in the EU RED.

According to the RED II (EU, 2018) the GHG performance is calculated as global warming potential (GWP) with a 100-year time frame including emissions of CO₂, CH₄ and N₂O, were 1 g of CH₄ and 1 g N₂O are valued 25 and 298 gCO₂-equivants, respectively. The emissions of the intermediate and final products are expressed in g CO₂-equivants, and the functional unit (FU) to which environmental impact is related is 1 MJ of FT-fuel (44.75 MJ/kg). According to the RED II, the default value for "the fossil fuel comparator $E_{F(t)}$ shall be 94 g CO_{2-eq}/MJ" (petrol and diesel). The equation used to calculate the total life cycle emission from the produced fuel is given in RED II (EU, 2018) as follows:

$$"E = e_{ec} + e_l + e_p + e_{td} + e_u - e_{sca} - e_{ccs} - e_{ccr}",$$

where "*E* is the total emissions from the use of the fuel, e_{ec} is emissions from the extraction or cultivation of raw materials, e_l is annualized emissions from carbon stock changes caused by land-use change, e_p is emissions from processing, e_{td} is emissions from transport and distribution, e_u is emis-

sions from the fuel in use, e_{sca} is emission savings from soil carbon accumulation via improved agricultural management, e_{ccs} is emission savings from CO₂ capture and geological storage, e_{ccr} is emission savings from CO₂ capture and replacement".

In the present assessment, "emissions from carbon stock changes caused by land-use change" is not relevant since the logging residues and sawdust are defined as "advanced feedstock" and set to zero, according to RED II (EU, 2018). The EU RED also state that the "emissions from the fuel in use shall be taken to be zero for biofuels and bioliquids", and therefore not included since we assess 100% pure biofuels with no fossil fuel blending. Furthermore, "emission savings from Soil carbon accumulation via improved agricultural management", "emission savings from CO₂ capture and geological storage", as well as "emission savings from CO₂ capture and replacement", are not relevant in this study. Thus, the parameters in the RED II calculation methodology that are included in following life cycle GHG emission analysis are " e_{ec} ", which includes the recovery and handling of the biomass feedstocks, " e_p ", covering the production of the electricity and heat needed for the biofuel process, and " e_{td} " which includes the transport operations for the logging residues, bark and sawdust from forest sites and pulp mills and sawmills to the biofuel plant, and the transport of the FT-fuel to the depot before distribution to the filling stations.

LCA calculations based on the ISO Standard is to a large extent following the RED calculation methodology described above, but with the difference that also indirect effects in form of changes in the content of soil carbon from logging residue harvest are included. The reference system is here taken to be no harvest of the logging residues and the logging residues are then left at the forest site for decomposition resulting in a minor increase in the long-term soil carbon content. This will lead to a somewhat reduced GHG saving for the biofuel, compared with when the RED calculation methodology is applied (see e.g., Olofsson et al., 2017). Regarding bark and sawdust, no potential indirect effects are included in the ISO calculations since these biomass resources are estimated to be available for biofuel production without causing shortage in alternative energy applications. This is due to the possibilities of reallocation of the various biomass resources depending on the actual and local utilisation option of the specific biomass feedstock (see also Haus et al., 2020).

4.2.3 Life cycle inventory data

The inventory data regarding the collection of logging residues at logging sites, chipping along the roadside, and transport of wood chips by truck to the biofuel plant, are based on De La Fuente (2016) and Lindholm et al. (2010; 2011). The date regarding transportation distances is based on table 4. LCA data for the transportation of bark and sawdust (by truck) are also based on De La Fuente (2016) and table 4. The calculated energy input in the handling and transport of the biomass feedstocks are shown in table 5, and the corresponding GHG emissions in table 6. A conclusion from table 5 is that the energy input in the recovery and transport of logging residues is equivalent to some 3 to 5 % of the energy content of the biomass. The corresponding energy input in the transport of bark and sawdust is 1 to 2 %. According to table 6, the GHG emissions associated with the handling and transport of logging residues amount to some 3 to 4 g CO₂-equivants per MJ biomass.

Table 5. Energy input in the collection, chipping and transport of logging residues, and transport of bark and sawdust, expressed as kJ per MJ biomass, in the county of Norrbotten and Kalmar to a biofuel production plant located in Luleå and Mönsterås, respectively.

Biomass feedstock	Norrbotten County (coastal area)				Kalmar County	
	Collection & chipping	Transport ¹	Total	Collection & chipping	Transport ¹	Total
Logging residues	24	20	44	17	17	34
Bark	-	17	17	-	5	5
Sawdust	-	21	21	-	11	11

¹Including empty return transport.

Table 6. Emission of GHG associated with the collection, chipping and transport of logging residues, and transport of bark and sawdust, expressed as gram carbon dioxide equivalents (g CO₂-eq.) per MJ biomass, in the county of Norrbotten and Kalmar to a biofuel production plant located in Luleå and Mönsterås, respectively.

Biomass feedstock	Norrbotten County (coastal area)				Kalmar County	
	Collection & chipping	Transport ¹	Total	Collection & chipping	Transport ¹	Total
Logging residues	1.68	1.41	3.09	1.20	1.16	2.36
Bark	-	1.18	1.18	-	0.37	0.37
Sawdust	-	1.52	1.52	-	0.76	0.76

¹Including empty return transport.

Life cycle inventory data regarding the biofuel production process are based on the process simulations performed in this study. The input of electricity in the process is assumed to be Swedish electricity mix, according to the RED (and ISO) calculation methodology, and the input of external process heat (for hot oil) is assumed to be based on forest fuels produced in a stand-alone heat boiler with a conversion efficiency of 95 %. GHG emission factors for electricity and external process heat are taken to be 13.1 and 3.4 g CO₂-equivalents per MJ, respectively (Haus et al., 2020). The input data for the transport and distribution of the FT-fuel is based on Haus et al. (2020), assuming a total transportation distance of 300 km, including a fuel consumption of 0.26 MJ per kg FT-fuel and an electricity input of 35 kJ per MJ fuel. Other external inputs and consumables in the process are estimated to be neglectable, such as catalysts, and therefore not included. Changes in the long-term soil carbon content after logging residue harvest, included in the ISO calculations, are based on data from Lindholm et al. (2011). These changes are equivalent to 6.8 g CO₂-equivalents per MJ logging residues regarding two or three rotations.

4.2.4 Techno-economic assessment

Capital cost estimations of process equipment in subprocesses hydrothermal liquefaction, evaporation and Fischer-Tropsch synthesis were performed in Aspen Process Economical Evaluator (APEA), through open source data bases, and through vendor quotations. Factorial methods were used to estimate the capital cost of these subprocesses which gives a ± 25 % accuracy of the project cost for these subprocesses. For those process steps that where cost estimated with APEA a comparison was made between the cost factors selected for the evaluation and the cost factors used in APEA which was found to agree well on a plant level.

The gasification capital expenditure was obtained from Arvos (personal communication, Mar 2, 2021), and is dependent on the extent of support systems included. To fairly estimate the capital expenditure, the subprocess is assumed subject to a set of conditions similar to that of black liquor gasification. The feedstock must prove to have a viscosity that is low enough to allow a burner and preheating system of the same type as for a black liquor gasifier. The slag in the gasifier must have a low enough eutecticum to not compromise the situation in the reactor. The reactivity of the feedstock must be on par with what is expected for the black liquor type of alkaline catalysed entrained flow gasification.

5 RESULTS

5.1 PROCESS SIMULATIONS

With the given process parameters, the gasifier feed flow converged at 23 792 kg/s and is composed as according to table 7.

Substance	Mass fraction
Water	22.5 %
Sodium acetate	21.2 %
Graphite	9.26 %
2-methyl-2-cyclopenten-1-one	1.88 %
Pelargonic acid	1.09 %
Trimethyl orthobenzoate	2.61%
2,5-dimethylfuran	117 ppm
Guaiacol	5.51 %
Oleic acid	21.5 %
Glycolic acid	14.3 %
Methanol	1.20 %
Phenol	0.14 %
Acetone	293 ppb

Table 7. Gasifier feed composition.

The outgoing syngas from the gasification flows with 22 547 kg/hr at 30 bar and 40°C and its composition is given in table 8.

Table 8. Gasifier feed composition.

Substance	H ₂	CO	CO ₂	H ₂ O	CH ₄	N ₂
Mass fraction	4.6 %	54.5 %	39.7 %	0.23 %	0.82 %	0.18 %
Molar fraction	43.9 %	37.4 %	17.4 %	0.25 %	0.99 %	0.12 %

Table 9 is an excerpt from the mass and energy balances, where the relevant obtained process variables are presented. A baseline energy demand of 150 kWh/ton for cryogenic oxygen production is assumed.

Table 9	. Mass	and	energy	balances	excerpt.
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Input	
Raw material	50 000 kg/hr (133 MW)
Oxygen 99.5 %	9 281 kg/hr
Ambient air for combustion	3 880 kg/hr
Utilities	
Electricity (Nordic el-mix incl. oxygen production ~1.39 MW)	2,51 MW
Heating hot oil for HTL-feed	2,76 MW
Hydrolysis (excl. from el-mix)	1,74 MW
Heating 3-phase-separator	5,90 MW
Output	
Bled off aqueous solution	14 060 kg/hr
Gases for flaring/digestion	19 395 kg/hr (14 MW)
FT crude gross	5 040 kg/hr (57 MW)
Dirty water after FT	3 309 kg/hr
CO ₂ from CO ₂ -separator	13 497 kg/hr
Flue gas for heating reformer	4 450 kg/hr
Excess heat FT-reactor (excl. internal steam generation)	0,44 MW
Quench cooling, EF gasifier, LP Steam	16,54 MW
Cooling after EF-gasifier, Hot Water	2,10 MW

Cooling after FT-reactor (see figure 9)

Under these conditions, the carbon efficiency from biomass feed for the bio-slurry, over the whole process, to FT crude (i.e. carbon atom flow in the FT crude stream over carbon atom flow in the biomass feed stream) is 32 %. This carbon efficiency does not account for the discrepancy in the HTL reactor atom balances that is introduced when enforcing the assumed yields, a deficiency at the entrance of approximately 9,5 % in relation to the exit, i.e. 10,5 % excess at the outlet as compared to the inlet. The carbon efficiency of HTL and evaporation combined is 87 %, and the carbon efficiency of the compared HTL-only setup is 61 %. The energy efficiency, in terms of gross heating value mass flow of FT crude over that of biomass plus utilities, is 39 %. A major internal item is the duty of HEX2 at 36 MW. The mass efficiency (mass flow of FT crude over mass flow of dry biomass) is 20 %. The achieved steam economy in the evaporator (flow of total evaporated H₂O in the effects over flow of primary steam) is 3,5. Cooling after the FT-reactor releases both latent and sensible heat. In Figure 9, the recoverable heat is given as a function of the stream temperature. It can be seen that most of the energy is available at rather low temperatures and that merely 25 % of the energy from cooling of this stream can be captured in the form of LP-steam.



Figure 9. Recoverable heat from the stream leaving the FT reactor as a function of the stream temperature.

Approximately 13 wt% of the FT crude is CO₂. The mass fractions (w_n , ordinate) of the hydrocarbons in the FT crude are plotted versus corresponding carbon chain length (n, abscissa) of alkanes and alkenes in figure 10. Due to databank limitations on alkenes, only icosene (C20) and smaller are present, and thus at C21 and larger, alkanes cover the alkene absence. Furthermore, at larger than triacontane (C30), only alkanes dotriacontane (C32), hexatriacontane (C36) and tetracontane (C40) are present. As interpolation at larger than C30, groups A, B, C and D contain masses of several carbon chain lengths as described in the figure. C30 and half the fraction of C31 constitute A. C32, C33, half the fraction of C31 and half the fraction of C34 constitute B. C35, C36, C37, half the fraction of C34 and half the fraction of C38 constitute C. C39, C40 and half the fraction of C38 constitute D.



Figure 10. FT crude, individual HC mass fractions vs. corresponding carbon chain length.

In figure 11 is a cumulative graph of hydrocarbon mass fractions in the FT crude vs. corresponding carbon chain length.



Figure 11. FT crude, cumulative HC mass fractions vs. corresponding carbon chain length.

5.2 LCA ANALYSIS

The GHG performance, according to the RED calculation methodology, of the FT-fuel produced from logging residues, bark and sawdust is in this assessment calculated to vary from 4.5 to 9 g CO_2 -equivalents per MJ of biofuel in the county of Norrbotten (coastal area), and from 2.5 to 4.5 g CO_2 -equivalents per MJ of biofuel in the county of Kalmar (see figure 12). The lowest GHG emissions refers to production systems based on sawdust, and the highest to systems based on logging

residues. In comparison to a fossil fuel comparator, the FT-fuel will lead to a reduction of the life cycle GHG emissions by 90 to 95 % referring to production systems in the county of Norrbotten, and by 95 to 97 % in the county of Kalmar.

When the GHG performance is instead based on the ISO calculation methodology, the GHG emissions will increase by approximately 14 g CO₂-equivalents per MJ of biofuel for the logging residue-based systems, due to the minor reduction in the long-term soil carbon content (see figure 13). This, in turn, will lead to a reduction of the life cycle GHG emission by 77 % and 81 % regarding the logging residue-based FT-fuel in the county of Norrbotten and Kalmar, respectively.

As described in the previous section and in table 4, the transportation distances are based on the assumption that all of the gross potential of logging residues, bark and sawdust is available for biofuel production. In reality, this will probably not be the case since these feedstocks are utilised for other energy purposes today, such as district heat production, industrial process heat production, pellets production etc. Furthermore, table 4 shows that when, for example, sawdust is utilised as feedstock, only 30 to 70 % of the feedstock demand can be met by sawdust generated within the county, and additional sawdust thus need to be "imported" from surrounding counties. This, in turn, will lead to longer transportation distances for the biomass feedstocks. A sensitivity analysis is therefore performed assuming that 50 % of the gross biomass potentials are available for largescale biofuel production and that also surrounding counties are included to fulfil the total yearly demand of respective biomass feedstock in the biofuel plant modelled in this study (equivalent to 220 000 tons DM biomass per year).



Figure 12. Life cycle GHG performance of FT-fuel (expressed as gram CO2-equivalents per MJ biofuel), according to the RED calculation method logy, produced from logging residues, bark and sawdust in the county of Norrbotten (coastal area), in the city of Luleå, and in the county of Kalmar, in the city of Mönsterås.



Figure 13. Life cycle GHG performance of FT-fuel (expressed as gram CO2-equivalents per MJ biofuel), according to the ISO calculation methodology, produced from logging residues, bark and saw dust in the county of Norrbotten (coastal area), in the city of Luleå, and in the county of Kalmar, in the city of Mönsterås.

The results from the sensitivity analysis show that the life cycle GHG emissions will increase due to longer transportation distances, especially for the FT-fuel produced in Norbotten county and when bark and sawdust are used as biomass feedstock (see figure 14). The GHG emissions for sawdust-based FT-fuel will be some 14 g CO_2 -equivalents per MJ of biofuel which is equivalent to a GHG reduction of 85 % compared with the fossil fuel comparator. The main reason for this increase is that major part of the sawdust is "imported" from surrounding counties and that the density of sawmills in the county of Norrbotten is rather low in comparison. The transportation distances are estimated to increase equivalent to 130, 150 and 260 km to a biofuel plant in the county of Norrbotten (Luleå) considering logging residues, bark and sawdust, respectively. The corresponding transportation distances to a biofuel plant in the county of Kalmar (Mönsterås) are 105, 40 and 70 km, respectively. The revised GHG performance of the FT-fuel produced from logging residues, bark and sawdust is thus calculated to vary from 9 to 14 g CO₂-equivalents per MJ of biofuel in the county of Norrbotten, and from 4 to 7 g CO₂-equivalents per MJ of biofuel in the county of Kalmar (see figure 14). In comparison to a fossil fuel comparator, the FT-fuel will lead to a reduction of the life cycle GHG emissions by 85 to 90 % referring to production systems in the county of Norrbotten, and by 92 to 96 % in the county of Kalmar. According to the revised RED, new biofuel plants should produce biofuels leading to at least a 60 % GHG reduction compared to a fossil fuel comparator, and his requirement will be fulfilled with a large marginal regarding the FTfuels assessed here. Furthermore, the existing Swedish GHG emission reduction obligation system, which is now also introduced for aviation fuels, promotes biofuels will large GHG savings, such as lignocellulosic biofuels. According to previous studies, reduction levels of 85 to 96 % will significantly improve the competitiveness of lignocellulosic biofuels compared with the current use of mainly imported crop-based biofuels used on the Swedish road transport sector (Haus et al., 2020).

The sensitivity analysis is based on the assumption that the individual feedstocks will be responsible for the complete demand of biomass in the biofuel plant equivalent to 220 000 tons DM biomass per year. However, if a mix of logging residues, bark and sawdust is utilised, only about 40 % of the total biomass supply potential in the respective county is needed to fulfil the yearly demand. This, in turn, will lead to transportation distances similar to those in the base case, or, on average, 74 and 43 km for a biofuel plant located in Luleå and Mönsterås, respectively. The corresponding life cycle GHG emissions for the FT-fuel produced in the county of Norrbotten and Kalmar will then be 6.4 and 3.5 g CO₂-equivalents per MJ of biofuel, respectively.



Figure 14. Life cycle GHG performance of FT-fuel (expressed as g CO₂-equivalents per MJ biofuel), according to the RED calculation methodology, produced from logging residues, bark and sawdust in the city of Luleå, Norrbotten county, and in the city of Mönsterås, Kalmar county, considering the base case and when 50 % of the gross biomass potentials are available as feedstock and when surrounding counties are included to fulfil the total yearly demand of respective biomass feedstock in the biofuel plant (equivalent to 220 000 tons DM biomass per year).

5.3 CAPEX AND OPEX ESTIMATE

Estimated investment- and operational costs for the examined process are presented in Table 10, where also exchange rates and other assumptions for economic calculations are given. The total investment cost is estimated to 1 694 MSEK, incl. 20 % contingency but excluding biomass supply system. With an economic lifetime of 15 years and an interest of 8 % the cost of capital is almost 40 % of the total production cost. Most important operational costs are biomass and oxygen. The latter is estimated at 65 EUR/t. Integration with water electrolysis for hydrogen production may be an interesting option where oxygen arises as a by-product.

The calculated specific cost for FT-crude is in line with the current cost for bio-based FT-liquids as reported by Brown et al. (2020), thereby confirming the economic relevance of the studied concept.

САРЕХ		Exchange rates			
HTL+evaporator	28 480 000	USD		8,5	SEK/USD
Equipment cost & installation	10 500 000	USD		10,2	SEK/EUR
Piping	3 800 000	USD			
Electrical & instrumentation	3 280 000	USD		Biomass	
Civil	2 600 000	USD		200	SEK/MWh
Other direct cost	8 300 000	USD			
Gasification system	50 000 000	EUR		Operating hours	
Gas cleaning and conditioning	11 500 000	EUR		7500	h/a
Equipment cost & installation	4 000 000	EUR			
Other direct cost	7 500 000	EUR		Availability	
FT subprocess	16 400 000	USD		95%	
Equipment cost	5 000 000	USD			
Other direct cost	11 400 000	USD		Interest	
Total direct costs	1 008 780 000	SEK		8%	
BOP, engineering & services	40%				
Contingencies	20%			Economic lifetime	
Total investment cost	1 694 750 400	SEK			
				15	years
OPEX				CAPEX-factor	
Personnel	24 200 000	SEK/a		11,7%	
Catalysts	18 000 000	SEK/a			
Maintenance	35 000 000	SEK/a			
Electrical power	7 437 500	SEK/a			
Oxygen	55 000 000	SEK/a			
Other OPEX costs	10 000 000	SEK/a			
Biomass cost	189 525 000	SEK/a			
Total OPEX	339 162 500	SEK/a			
Capital cost	197 996 918	SEK/a			
Total production cost	537 159 418	SEK/a			
Output	57	MW			
	406 125	MWh/a			
Specific cost for FT crude	1 323	SEK/MWh			

Table 10. Estimated capex and opex for the HTL + FT process.

6 DISCUSSION

The development and implementation of the proposed chemical processes are not without obstacles. One primary issue is product yields. Due to high oxygen contents of biomass in relation to e.g. fossil feedstocks, the deoxygenation commonly brings about a significant loss of carbon and hydrogen via by-products. In this extensive process, a relatively large loss of carbon is necessary to obtain the upgraded product. Another issue is the selection of feedstock. A readily available feedstock such as forestry residues was here deemed superior to materials processed for the purpose of being used as a feedstock, e.g. algae, because of factors such as price and technical advancement. Nevertheless, rheology is of great concern regarding pumpability, and this property with the bio-slurry must be empirically studied. The reason for specifically considering forestry residues is the vast forest areas and sustainable forestry in Sweden where the project is taking place.

Hydrothermal liquefaction is a developing field of study and the available data at hand is limited. The choice of methodology to assess the HTL reaction is therefore slightly constrained. Tzanetis et al. (2017) is fairly recent and most importantly comprises a multitude of references to determine the process variables. Using reference compounds for the product is coarse but necessary. Arguably coarser is as in this project assuming that the recycled organic matter reacts identically to the feed-stock. Nevertheless, it is reasonable to assume that decomposition and deoxygenation is occurring in general in the HTL reactor. It should be noted that an unphysical discrepancy in the atom balance through the HTL reactor is introduced when enforcing the assumed yields. There is a carbon atom flow deficiency in the reactor feed of approximately 9.5 % in relation to the product, i.e. a carbon atom flow excess of approximately 10,5 % in the reactor outlet as compared to the inlet. The presented carbon efficiencies does not compensate for this imbalance, and are still defined as carbon atom flow in the corresponding product streams over that in the biomass.

There are several challenges with hydrotreating the bio-oil fraction based on its composition. First, the high oxygen level creates a highly exothermic reaction, calling for a solution with recycling or external cooling. Secondly, the high degree of unsaturation of the feed will cause carbon formation on the catalyst, impairing the performance. Thirdly, the oxygen reaction product water and the low sulfur content of the feedstock makes using standard CoMo and NiMo catalysts difficult. Finally, the presence of P, Cu, Cr, Zn, Ni, Na, Ca, K and other metals in the bio-oil will inevitably lead to deactivation of the catalysts used and require extensive workup of the oil before processing. All of the listed problems above are possible to solve,but will inevitably lead to a much more complicated process flow sheet than the one suggested in figure 1.

The α -value of 0.9 in the FT synthesis was intended as a compromise between depressing methane formation and heavy waxes, while maximizing products in the middle distillate range. The carbon chain length distribution of the FT crude displays a heavy right tail suitable for cracking in an upgrading process. However, a somewhat lower α -value might be desirable. With the database available and within the compass of the simulation work package in the project, at above C20, FT products were confined to alkanes. Furthermore, at above C30, only C32, C36 and C40 was available for usage. This explains the discontinuities in the FT crude composition. Weighting was implemented to account for the unavailable alkanes.

A major challenge is the design and operation of the recuperative HTL feed heat exchanger HEX2. Solely the duty of 36 MW entails a considerable capital expenditure, but the practical problem at

hand is the present phases. The multiple-phase flow is likely to cause fouling, albeit at an unknown rate, and disruption to the heat exchange. A full-scale heat exchanger is perhaps unrealistic to construct for examination, although the typical advantages of scaling up can be used to motivate an investigation into a pilot for baseline reference.

As can be seen from Table 9 there is a net heat excess from the HTL+FT process. Based on the assumption in this report, that the evaporation plant is driven by MP steam, there is only a small (0,44 MW) surplus of MP steam. However, there is a significant surplus of LP steam (16,5 MW) generated in the EF-gasifier quench. This steam can be utilized in a pulp mill, replacing LP-steam from the back-pressure turbine. Final gas cooling after the gasifier generates hot water (2,1 MW) that due to its high temperature (90 °C), in many cases also can be utilized in a pulp mill replacing LP steam. Cooling of the product stream from the FT-reactor (Figure 9) enables a significant amount of energy to be recovered. However, most of this is capturable at a low temperature, meaning there is less potential for heat integration with a pulp mill than for a district heating plant. All-in-all, the production of 57 MW FT-crude as examined in this report generates a heat-excess of approximately 25 MW that can be utilized in a pulp mill, thereby greatly enhancing the resource-efficiency for the production concept.

The SunAlfa-process where alkali-assisted HTL are used as pre-treatment for a subsequent hydrocarbon-production by means of FT-synthesis is by the time of this report analyzed in a separate project co-funded by the Swedish Energy agency (EM 47484-1). In this project, that is planned to be finalized in May 2021, yields in both HTL- and gasification processes as well as HTL-liquid properties are experimentally examined with varying operational parameters. The final outcome of the experimental tests in EM 47484-1 will yield important insights regarding the practical feasibility of the SunAlfa process and may justify a renewed simulation of the total process as carried out in this project.

7 CONCLUSION

In simulating an integrated hydrothermal liquefaction, gasification and Fischer-Tropsch synthesis process, fair energy and mass efficiencies were achieved in consideration of the biorefinery context. In this extensive process, the presented carbon efficiency of 32 % is reasonable. An adequate composition of the resultant FT crude was attained, peaking at C10, in which the carbon chain lengths were distributed with a heavy right tail suitable for cracking in an upgrading unit. Life cycle assessment revealed very high GHG performances in a Norrbotten coastal area case and a Kalmar county case, both with feedstock options logging residues, bark and sawdust.

From the scope of the findings, the described process is a theoretically applicable route from woody biomass to synthesized middle distillates. The efficiencies in practice are likely typical for all the subprocesses and a sequence of them in this manner utilizes forestry byproducts for production of renewable jet fuel, although its practical feasibility in a production plant needs further research to be accurately determined. Special attention should be paid to integration aspects and assessing how the developed subprocesses cofunction in an unexplored combination.

8 FURTHER WORK

Suggestions for further work include studying the recuperative heat exchanger (currently dubbed HEX2) in the HTL subprocess. Focus may be on quantifying the heat transfer of the multiphase flows of HTL product hot side and bio-slurry cold side and calculate heat exchanger dimensions. Another suggestion is to analyze the potential treatments of the gas remainder from the evaporator, possibly flaring or anaerobic digestion. A third suggestion is to examine how an upgrading plant would handle the incoming FT crude for the production of jet fuel. A most general suggestion is to investigate a small-scale experimental setup empirically to compare to the simulations.

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