Research and Innovation Action

H2020-LC-SC3-2019-NZE-RES-CC

Scale-up, risks and costing of integrating the production of HTL biofuels in conventional pulp mills

WP4 - Task 4.2

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Lead Beneficiary: SINTEF-ER

Author(s): Gonzalo del Alamo (SINTEF-ER), Thomas Indlekofer (SINTEF-ER)

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Disclaimer

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Abbreviations, acronyms, and symbology

Acronym/Symbol	Description
Variables	
$\dot{M}$, $\dot{V}$, $\dot{V}_N$	Mass flow rate, volumetric flow rate, normal volumetric flow rate
Y, X, ϕ	Mass fraction, mole fraction, volume fraction
$\dot{H}$, $\dot{H}_T$, $\dot{H}_F$,	Rate of total, thermal and formation enthalpy
$\dot{Q}$, $\dot{W}_{el}$	Heat power, electric power
h, h_T, h_F	Specific total, thermal and formation enthalpy per unit mass
$\bar{h}$, $\bar{h}_T$, $\bar{h}_F$	Specific total, thermal and formation enthalpy per unit mole
ρ, k, μ	Bulk density, thermal conductivity and viscosity
c, c_P	Specific heat, specific heat at constant pressure
Subscripts	
i	Atomic composition
j	Molecular composition
k (O, W, S, G)	Phase composition (oil, aqueous, solid and gas phases)
l	Biomass constituent or molecular functional group
BL	Black liquor
O	Oil phase
S	Solids phase, insoluble fraction
A	Aqueous phase, including solubilized material
G	Gas phase
cat	Catalyst
$solv$	Solvent
h	Heating
c	cooling
el	electricity
Superscripts	
SP	Salts precipitation
IHTL	Integrated salt precipitation and hydrothermal liquefaction

HTL	Hydrothermal liquefaction
IHDO	First-stage hydrodeoxygenation
PS	Phase separation
APR	Aqueous Phase Reforming
HDTR	Hydrotreating
HDCR	Hydrocracking
HDI	Hydro-isomerization

Keywords

Black liquor, HTL biocrude, upgrading, refinery processes, aviation fuel, marine fuel, risks, scale-up, production costs

Executive Summary

This deliverable reports the scale-up and cost analysis of the complete technological route proposed in the BL2F project for production of liquid biofuel from black liquor. The overall conversion includes two main steps. The first step involves the production of an intermediate oil product, or so-called biocrude, from the black liquor in a decentralized plant fully integrated into the Kraft pulp mill. The second step involves the upgrading of the biocrude to naphtha, kerosene, and heavy distillate in a centralized refinery. The production of biocrude from black liquor includes several innovative processes: 1) integration in one reactor of the precipitation of salts followed by hydrothermal liquefaction, so called IHTL, under near critical-water conditions (under temperature and pressure conditions of 350-400 deg. C and 250-300 bar); 2) partial hydrodeoxygenation of the desalinated HTL oil phase, so called IHDO, in aqueous phase also under near critical-water conditions; 3) aqueous phase reforming (APR) of dissolved organic components present in the HTL process water to produce hydrogen required by the IHDO process. Considering the IHTL process, one main risk came from the fact that, within the temperature range investigated, the precipitation of the salts and the hydrothermal liquefaction of the organic fraction overlap. Moreover, both the salts brine and desalinated IHTL product form a quite stable solution containing oil, solid and aqueous phases. As consequence, the salts brine extracted from the IHTL reactor bottom can contain high concentration of oil and other organics dissolved in the aqueous phases or as non-dissolved solids. Separation of the oil from the salts brine and the desalinated IHTL product is quite challenging and requires, as described more in detail later, cooling, depressurization, and a multi-stage centrifugation system with intermediate mixing with acid and organic solvents. The main risks of the IHDO and the APR processes are the catalyst deactivation or poisoning and clogging of the reactor bed. Therefore, both processes have stringent requirements for the feed to be free of solids. Another risk associated to the APR process is that the hydrogen production is too low, due to either poor reforming rate or low hydrogen selectivity, to supply the local demand of hydrogen by the IHDO process. This risk can be mitigated by including a separate hydrogen production unit in the biocrude production plant or the mixing of additional organic compounds to increase the gas yield and the hydrogen selectivity in the APR process.

This process design for the biocrude production has been updated from the one previously reported [1] to include mitigation of the risks described above. The analysis reported in this deliverable considers three different cases for the design of the biocrude production plant:

> Case 1. Production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and full extraction of the oil phase from both the salts brine and the desalinated HTL product. The oil phase is partially upgraded by catalytic hydrothermal deoxygenation using hydrogen as reducing gas produced by aqueous phase reforming of the process water.

> Case 2. Production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and extraction of the oil phase from the desalinated HTL product only. The salts brine is cooled and depressurized before being fed into the recovery boiler at the pulp mill. The HTL biocrude is partially upgraded by catalytic hydrothermal deoxygenation using hydrogen as reducing gas produced by aqueous phase reforming of the process water.

Case 3. Production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and full extraction of the oil phase from both the salts brine and the desalinated HTL product.

Table 1 summarizes the main mass and energy balance and cost performance of the biocrude production for plant capacities in the range of 100-600 dry ton of black liquor per day, equivalent to 10-30% of the black liquor produced in a reference pulp mill with an annual bleached pulp capacity of 500 kt/year.

Table 1: Comparison of the main mass and energy balances and cost performance indicators of the production of biocrude for process design cases 1-3.

Case	1	2	3
Oil mass yield (% wt. dry basis)	21.1	19.0	23.9
Oil energy yield (%)	50.4	45.3	55.4
Salts brine mass yield (% wt. dry basis)	24.3	32.6	24.3
Salts brine energy yield (%)	1.1	13.4	1.1
Gas mass yield (% wt. dry basis)	6.40	6.42	6.36
Gas energy yield (%)	17,4	17.1	5.4
Solid residue mass yield (% wt. dry basis)	16.2	16.2	16.2
Solid residue energy yield (%)	4.3	4.3	4.3
Dissolves in aqueous effluent mass yield (% wt. dry basis)	15.6	12.7	14.7
Dissolves in aqueous effluent energy yield (%)	20.1	16..6	18.0
Make-up hydrogen feed to IHDO (g/kg)	11.9	10.7	-
Make-up acid to phase separation (g/kg dry)	66	50	66
Make-up organic solvent to phase separation (g/kg dry)	4.8	4.3	4.8
Specific electric load (kWh/dry ton)	537 - 526	516 - 510	134 - 143
Specific permanent investment (M€/dry ton/day)	0.82 – 0.59	0.80 – 0.58	0.45 – 0.33
Specific annual operating cost (k€ / dry ton)	0.89 – 0.76	0.85 – 0.73	0.32 – 0.27
Levelized cost of biocrude (€/Liter)	5.77 – 4.25	6.11 – 5.08	2.09 – 1.70

The yields shown in this table are relative to black liquor feed into the biocrude production plant. The maximum mass and energy yields of oil produced from IHTL relative to black liquor on dry basis are 23.9 % and 55.4 % respectively, assuming that the oil is extracted from both the salts brine and the desalinated HTL product. The mass yield of the salt brine before and after extraction of the oil has been estimated to be 32.6 and 24.3%, respectively, containing approximately 96% of the total salts in the black liquor. The yields of dry solids, gas, and dissolved material in the aqueous phases from the IHTL process are, respectively, 16.2, 6.3 and 44,4% on dry mass basis, which correspond to 4.3, 6.4 and 33,7 % on energy basis. Further upgrading of the total oil extracted from IHTL by hydrodeoxygenation (IHDO) decreases the mass and energy yields of oil to 21.1% and 50.4% while it increases the gas and dissolved material in the aqueous phase by 2.4 and 0.4 % on dry mass basis, equivalent to 10.6 and 12.6 % on energy basis. This case exhibits the lowest cost of biocrude, in the range of 2.1-1.7 €/Liter for black liquor feed capacities between 100 and 600 dry ton per day. Including hydrodeoxygenation of the total amount of IHTL oil with local production of hydrogen with full treatment of the process water in APR leads to a significant increase of the cost of biocrude, to 5.8-4.7 €/Liter, mainly due to the large increase in operating costs. Discarding the oil extracted from the salts brine leads to a further increase of the cost of biocrude, which ranges between 6.1 and 5.1 €/Liter when the oil content in the salts brine is 10% of the total.

The overall upgrading process considered naphtha and kerosene as the main products, the process design including hydrotreating of the biocrude for reduction of S and O heteroatoms, catalytic hydrocracking of the distillate residue separated during fractionation of the hydrotreating and hydrocracking organic liquid products to naphtha and kerosene, and treatment of the hydrogen-rich sour gas for separation of CO2 and H2S and recycling of the remaining hydrogen back to the catalytic processes. The overall process design for the biocrude upgrading [1] is defined based on conventional refinery processes. The biocrude feed is treated by catalytic hydrotreating for complete hydrodeoxygenation followed by separation of light gases and process water. The remaining hydrotreated organic liquid is fractionated in a multi-stage distillation to naphtha and kerosene. The remaining heavy distillate from distillation can be used directly as marine fuel or treated in a catalytic hydrocracking unit for further production of naphtha and kerosene. The naphtha and kerosene ranges are assumed to have boiling points in the range 90-180 deg. C and 180-250 deg. C respectively. Naphtha can be further treated by hydro-isomerization to produce aviation fuel, although this is not included in the analysis. The main risks (based on measured oil composition) associated to the biocrude upgrading process are the high concentration of organic-oxygen heteroatoms and other inorganics in the oil. The main and energy balance and cost performance of the overall biocrude upgrading process is summarized in Table 2.

Table 2: Comparison of the main mass and energy balances and cost performance indicators of the production of biocrude for process design cases 1-3.

Naphtha mass yield (% wt. biocrude feed)	52.5
Naphtha energy yields (%)	47.2
Kerosene mass yield (% wt. biocrude feed)	13.4
Kerosene energy yields (%)	15.0
Distillate residue mass yield (%)	24.0
Heavy distillate energy yield (%)	27.3
Light hydrocarbons mass yield (%)	42.5
Light hydrocarbons energy yield (%)	47.2
Specific electric load (kWh/dry ton)	6,22 – 3.46
Specific permanent investment (M€/dry ton/day)	0,319 – 0,077
Specific annual operating cost (€/dry-ton), with biocrude cost of 1.3 €/Liter	1.29 – 1.20
Minimum fuel Selling Price Naphtha (€/Liter)· , with biocrude cost of 1.3 €/Liter	2.3 – 2.1
Minimum fuel Selling Price Kerosene (€/Liter) , with biocrude cost of 1.3 €/Liter	1.5 – 1.3
Specific annual operating cost (€/dry-ton), with biocrude cost of 6 €/Liter	5.89 – 5.86
Minimum fuel Selling Price Naphtha (€/Liter)· , with biocrude cost of 6 €/Liter	7.1 – 6.9
Minimum fuel Selling Price Kerosene (€/Liter) , with biocrude cost of 1.3 €/Liter	6.2 – 6.1

Assuming that the whole distillate residue from the kerosene column is further treated by hydrocracking, the total yields of naphtha and kerosene products, relative to the biocrude feed, are estimated to be 24.2% and 54.4% on dry mass basis and 29.9 and 66% on energy basis. Under this assumption, the overall upgrading process requires approximately 13 kg of make-up hydrogen per ton of biocrude, 80% of which is consumed by the hydrotreating process and the remaining by hydrocracking. Scale-up and cost analysis of the upgrading of biocrude considers a capacity range at refinery of 100-2500 ton of biocrude per day, equivalent to the maximum biocrude production rate from 5-25 reference pulp mills all with an annual bleached pulp capacity of 500 kt/year. Considering production of naphtha and kerosene only and assuming the kerosene has a market value 1.5 higher than naphtha on energy basis, the minimum fuel selling prices (MFSP) are in the range 10.9-2.1 €/Liter for kerosene and 7.1 and 1.3 €/Liter with biocrude feed capacities considered in the analysis, based on a specific biocrude cost varying between 1 and 6 €/Liter.

1. Introduction

1.1 Purpose of this deliverable

The primary objective of this deliverable is to perform a realistic estimation of the cost of producing marine and aviation biofuels from black liquor based on the integration of hydrothermal liquefaction into a representative kraft pulp mill.

The specific objectives are:

- To identify and quantify technological risks and specify concrete technical measures to mitigate the risks.
- To perform scale-up analysis of the equipment design included in the overall HTL route under a representative capacity range of the kraft pulp mill.
- To calculate capital and operating costs of the HTL plant integrated into the pulp mill, and the upgrading of biocrude to biofuels.

Experimental data of the main processes involved in the conversion route using representative feed samples were not available. Therefore, evaluation of the process has been performed using reference empirical parametric models and assumptions to be able to scale-up the equipment design and calculate the costs. An update of the mass and energy flows of the overall process will be included in the last deliverable of WP4 (D.4.3).

1.2 Background information and input data from other tasks

The following information has been used (see also references):

1. General objectives, technological options and process performance targets from the proposal SEP-210593035, " Black Liquor to Fuel by Efficient HydroThermal Application integrated to Pulp Mill."
2. Measurements of the composition of black liquor samples. Reported in the deliverable H2020-LC-SC3-2019-NZE-RES-CC/D1.3, 2021, "Report on the feedstock characterization"
3. The description of a hardwood kraft pulp mill process model that forms the basis for the simulations studying the integration of the HTL plant into the pulp mill is presented in Kangas el at. 2014.
4. Empirical process models described in "Process design and analysis of the integration of the production of HTL biofuels in conventional pulp mills," H2020-LC-SC3-2019-NZE-RES-CC/D4.1, 2022.

2. Process design

This section describes an updated design for the biocrude production process considering the main risks identified during the experimental campaigns within the project and including, when possible, mitigating measures in the design.

2.1 Design of the biocrude production process

2.1.1. Risks and mitigations

- For the temperature range (350-400 deg. C) considered for the experimental tests, the salt precipitation and the hydrothermal liquefaction processes overlap. Moreover, the solid, oil and aqueous phases form a quite stable emulsion. Therefore, the precipitated salts brine extracted from the IHTL reactor bottom contains high concentrations of oil and aqueous phases as well as organic and inorganic solids other than the precipitated salts. Measured composition of the salt brine has shown that up to 80% and 90% of the total oil and solids products from the IHTL reactor ends up in the salt brine. A dedicated phase separation system is required to extract the oil phase from the salt brine and the desalinated IHTL product. The process design, including a multistage centrifugation aided by solvent and acid mixing, is described below.
- Salts in the IHTL reactor accumulate on the walls causing fouling. Mitigation of this risk is the integration of precipitation and separation of salts at the bottom of the IHTL reactor.
- Material corrosion in process equipment operating under near critical or supercritical water conditions. P91, a chrome moly alloy metal that contains excellent strength and temperature resistance, has been identified as a suitable material. However, assessing the corrosion of this material for long-term continuous operation is outside the scope of the BL2F project.
- Leakages during extraction of salts brine. Sealings of the piston valves considered in the design have typically a maximum operational temperature 120 deg. C). Therefore, the salts brine needs to be cooled below the maximum operating temperature.
- Gas and solid particles in the feed to the IHDO process can cause clogging and deactivation of the catalyst bed. Include separation of gas and solids from the IHTL product before IHDO. This requires separation of solids for the feed to the IHDO reactor.
- Deactivation of the IHDO and APR catalysts. Mitigation of this risk requires experimental determination of the right chemical composition and physical structure of the catalyst and catalyst support, which is still under investigation. For the APR catalyst, one cause for deactivation is due to excessive water vaporization, which typically occurs when the water content in the feed is below 85% on mass basis [2]. Mitigation of this risk is considered in the analysis by specifying a water content in the feed to APR of 90%.
- Insufficient H2 yield from APR to feed the IHDO process. The mitigation of this risk considers including an additional H2 production unit.

2.1.1 Assumptions and risk mitigation measures

- There are three proposed mitigating measures to avoid or reduced fouling: 1) modify the IHTL reactor design to include flushing of the reactor internal wall; 2) Separation of carbonates salts before IHTL reactor using Calcium. 3) increase the concentration of hydroxide and sulphite in the black liquor.
- The hydrothermal liquefaction process of 350 deg. C and 320 bar-g. Moreover, the experimental results have shown that a high fraction of phosphorous, above 85 %, remains in the solid phase after liquefaction.
- Acids and organic solvent (MEK) are supplied in liquid phase.
- Pressurized water has been considered as thermal fluid.

2.1.2 Process design scenarios

The scale-up and cost analysis reported in this deliverable considers three different cases for the design of the HTL plant, schematically represented by process block diagrams in Figures 1-3:

<u>Case 1</u>. Production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and full extraction of the oil phase from both the salts brine and the desalinated HTL product. The oil phase is partially upgraded by catalytic hydrothermal deoxygenation using hydrogen as reducing gas. by aqueous phase reforming of the process water

<u>Case 2</u>. Production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and extraction of the oil phase from the desalinated HTL product only. The salts brine is cooled and depressurized before being fed into the recovery boiler at the pulp mill. The HTL biocrude is partially upgraded by catalytic hydrothermal deoxygenation using hydrogen as reducing gas. by aqueous phase reforming of the process water

<u>Case 3</u>. Production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and full extraction of the oil phase from both the salts brine and the desalinated HTL product.

2.1.3 Integrated salt precipitation and liquefaction

The process design is described through the process flow diagram shown in Figure 4. The black liquor diverted from the pulp mill is stored in a stirred tank to ensure uniform mixing. The black liquor is discharged from the storage tank by a pump to feed the main HTL pump, designed as a single piston low stroke reciprocating pump, that pressurizes the black liquor to the operating conditions of the IHTL reactor. The pressurized black liquor is heated to near critical-water conditions in a jacked U-tube heat exchanger, using pressurized water as heating medium, and then fed into the IHTL reactor from the bottom. In the lower part the IHTL reactor, the dissolved salts precipitate, and the remaining desalinated slurry liquefies to form a multi-phase product containing dispersed oil, solids, gas in an aqueous solution with dissolved organic and inorganic material. The IHTL reactor has a tubular design with a heating jacket in the upper part to regulate the temperature of the hydrothermal liquefaction process. The precipitated salts are extracted from the bottom of the IHTL reactor and immediately cooled by a heat exchanger, using the return thermal fluid from the main black liquor heating. The cooled salts brine is then depressurized and stored in a buffer tank. The desalinated HTL product exits the IHTL reactor at the top,is rapidly cooled using return thermal fluid from the main black liquor heater, depressurized in a capillary system, flashed, and stored in a tank. The process design includes an additional auxiliary cooler that uses water as cooling medium.

2.1.4 Oil extraction from the salts brine

The process design for the oil extraction from the salts brine is schematically shown in Figure 4. The salts brine solution stored in the tank is mixed with citric acid and the organic solvent (MEK) to break up the emulsion and then centrifuged to separate the aqueous, oil and solid phases. The solids are transported back to the pulp mill by a screw conveyor. The aqueous phase is flashed to recover the organic solvent and light oil components, and then stored in a tank. The volatiles organic components are condensed and recirculated and mixed back with the salts brine. The oil phase from the centrifuge is directly stored in an oil tank.

2.1.5 Phase separation of the desalinated HTL product

The process design for the separation of the oil, solid and aqueous phases from the degassed desalinated HTL product is shown in Figure 4. Due to the high concentration of water, the slurry from the buffer tank is first dewatered in a centrifuge and then stored in a buffer stirred tank where it is mixed with the acid and organic solvent. This solution is then taken to a second centrifuge to separate the oil, solid and aqueous phases. Handling of these phases is the same as for the oil extraction from the salts brine described above.

2.1.6 Hydrodeoxygenation (IHDO)

The design of the IHDO system is schematically shown in Figure 6. The separated aqueous and oil phases from the storage tanks are remixed in a stirred reactor to ensure uniform mixing and then pressurized and heated, using pressurized water as heating medium, before being fed into the IHDO reactor. This reactor is designed as a cylindrical vessel with the fixed-bed catalyst supported in internal vertical tubes. The product from the IHDO process exits at the top of the reactor and is then cooled in two heat exchangers, the first one using return heating water from the IHDO feed heater and the second one using cooling water. The cooled product is then taken to a three-phase gravimetric separator where the hydrogen-rich sour gas is separated from the top and process water is partially separated from the oil phase at the bottom. The oil stream is then heated and taken to a second gravimetric separator for separation of light hydrocarbons and further separation of the remaining process water from the oil. This oil product is stored before being transported to the refinery for further upgrading. The process water from both gravimetric separators is taken to a buffer tank before further treatment in the aqueous phase reforming (APR) system.

2.1.7 Aqueous Phase Reforming (APR)

The APR design is described through the process flow diagram shown in Figure 6. The process water is pressurized, heated and fed into the APR reactor at the bottom. The design of this reactor is similar to the IHDO reactor as described above. A fraction of the APR reactor product is recirculated back to the bottom feed and the remaining is cooled and flashed for separation of the gas. The liquid from the flash tank, containing dissolved organic and inorganic material is stored in a tank before being taken back to the evaporator of the pulp mill. The gas stream from the flash tank is further treated in an amine system for separation of hydrogen, which is compressed and used in the IHDO process, and the remaining gas is transported back to the recovery boiler of the pulp mill.

2.2 Design of the biocrude upgrading

2.2.1 Assumptions

- The operational pressure for the hydrotreating and hydrocracking processes are all the same. The same compressor supplies hydrogen to these processes.
- Make-up hydrogen is assumed to be available at the refinery and produced from natural gas reforming.
- The heat and cooling demand of the overall process have been minimized by heat integration so that hot process streams that require cooling are used to heat other process streams when heating is necessary.
- The off-gas stream containing light hydrocarbons obtained after gas separation, can be used to partially satisfy the internal heat demand of the process.
- Process water from phase separation of the products from hydrotreating and hydrocracking processes is disposed to the refinery process water system.
- Catalytic hydrotreating of the biocrude for reduction of oxygen and sulfur with hydrogen.
- Fractionation of the liquid organic by distillation with production of naphtha, kerosene, and heavy distillate.
- Catalytic hydrocracking of the heavier fraction from distillation for further production of naphtha and kerosene. The product from hydrocracking is recirculated to the separation process after hydrotreating.
- Treatment of the gas product from separation by removal of CO_2 and H_2S Dand recirculation of purified hydrogen back to hydrotreater and hydrocracker.
- The main products of the overall upgrading process considered in the analysis are kerosene, used as jet-fuel, and heavy distillate used as marine fuel. Naphtha is as an intermediate for producing jet fuel via hydro-isomerization before being blended again in the kerosene pool at the refinery.

2.2.2 Detailed process design

The process design of the biocrude upgrading is graphically represented Figure 7. The raw biocrude, initially stored in a tank, is pumped, heated, mixed with hydrogen, and fed into the hydrotreating reactor. The feed is heated first in a heat exchanger with recovery of thermal energy from the hydrotreating product, and then in a fired heater using light hydrocarbons separated during fractionation of the organic liquid product from hydrotreating. The product from the hydrotreating reactor contains an oil phase, process water and light gases rich with non-reacted hydrogen. Separation of these phases from the hydrotreating product is performed first in a low temperature and high-pressure three-phase gravimetric separator where a light sour gas and a fraction of the process water are separated from the oil phase. The oil phase from the gravimetric separator is then depressurized, reheated and taken to a stripper where light hydrocarbons and water vapor are separated from the remaining organic liquid. The stripped vapor is cooled for condensation and separation of the process water. The liquid organic stream from the stripper is fractionated in two distillation columns. In the first distillation, naphtha with a boiling point in the range of 80 to180 deg. C. The bottom product from the naphtha distillation column is re-boiled and fed into a second distillation column for separation of kerosene, with a boiling point in the range of 180 to 220 deg. C. The residue from the kerosene distillation column contemplates two options depending on the physical properties and chemical composition. The first option is the direct use as marine fuel. The second is further treatment by hydrocracking for further reduction of remaining S and O heteroatoms and cracking of the distillate organic residue to increase the naphtha and kerosene products. In this case, the distillate residue from the kerosene column is pumped, heated, and mixed with hydrogen before being fed into the hydrocracking reactor. The product from the hydrocracking is cooled and mixed with the hydrotreating product for phase separation. The sour gas from the high-pressure gravimetric separation is further treated by amine absorption for separation of hydrogen by solubilization of CO_2 and H_2S. The H_2 enriched gas stream from the amine system is mixed with make-up hydrogen, compressed, and recirculated back to the hydrotreating and hydrocracking reactors.

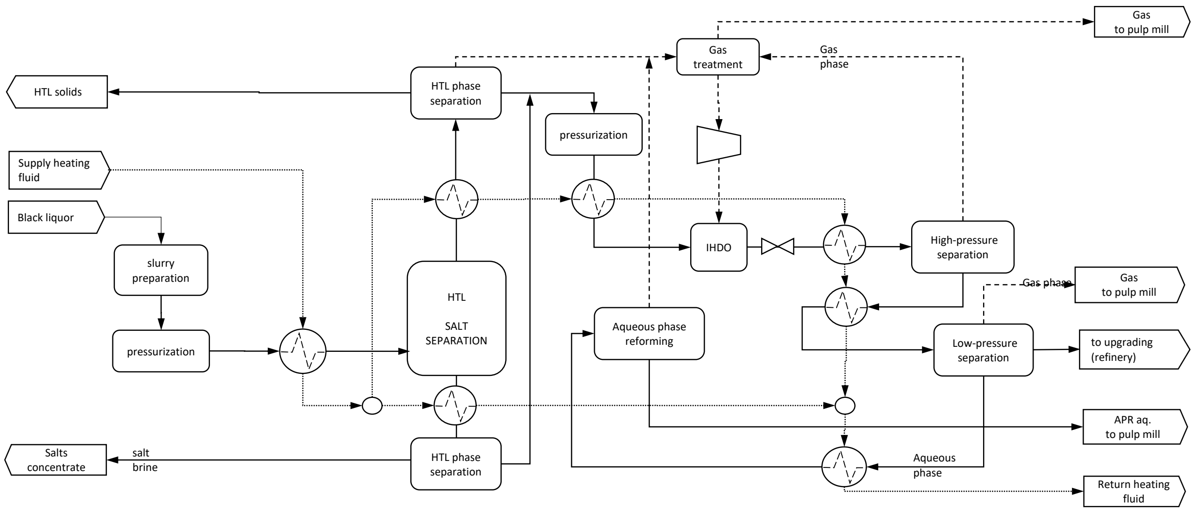

Figure 1: Process block diagram representing an updated process design of the HTL plant for production of bio-oil from black liquor, including risk mitigating measures.

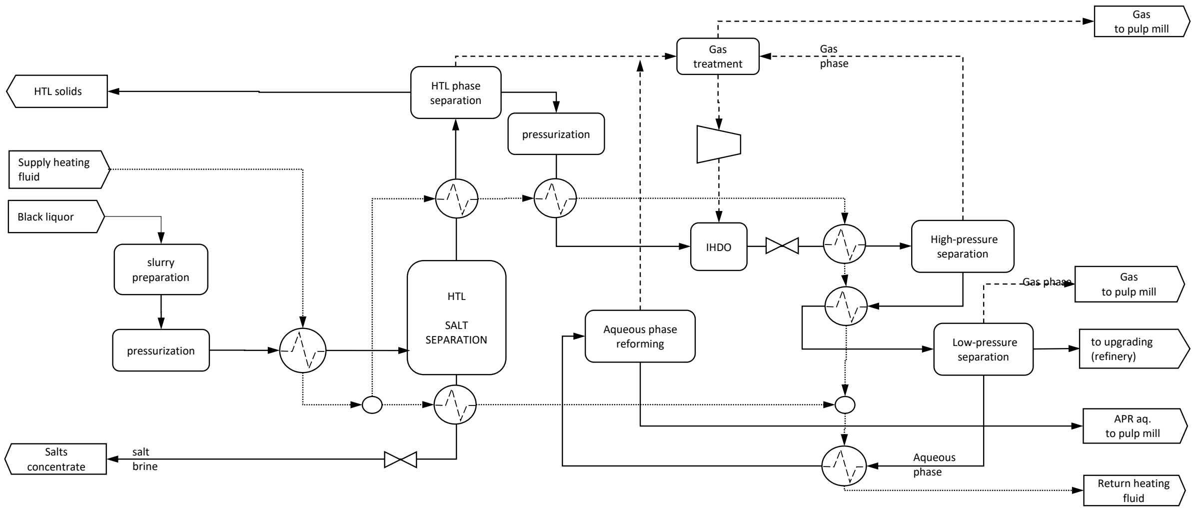

Figure 2: Process block diagram representing an updated process design of the HTL plant for production of bio-oil from black liquor, including risk mitigating measures.

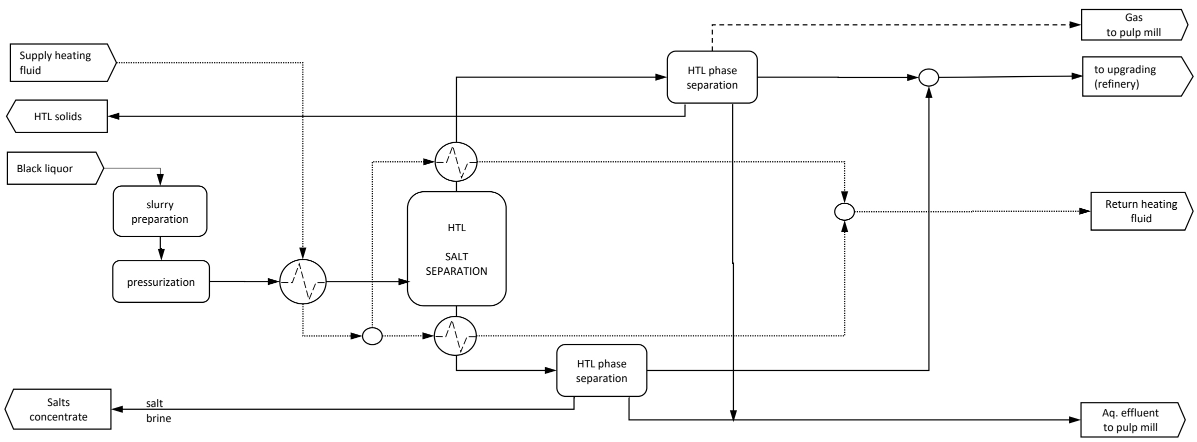

Figure 3: Process block diagram representing an updated process design of the HTL plant for production of bio-oil from black liquor, including risk mitigating measures.

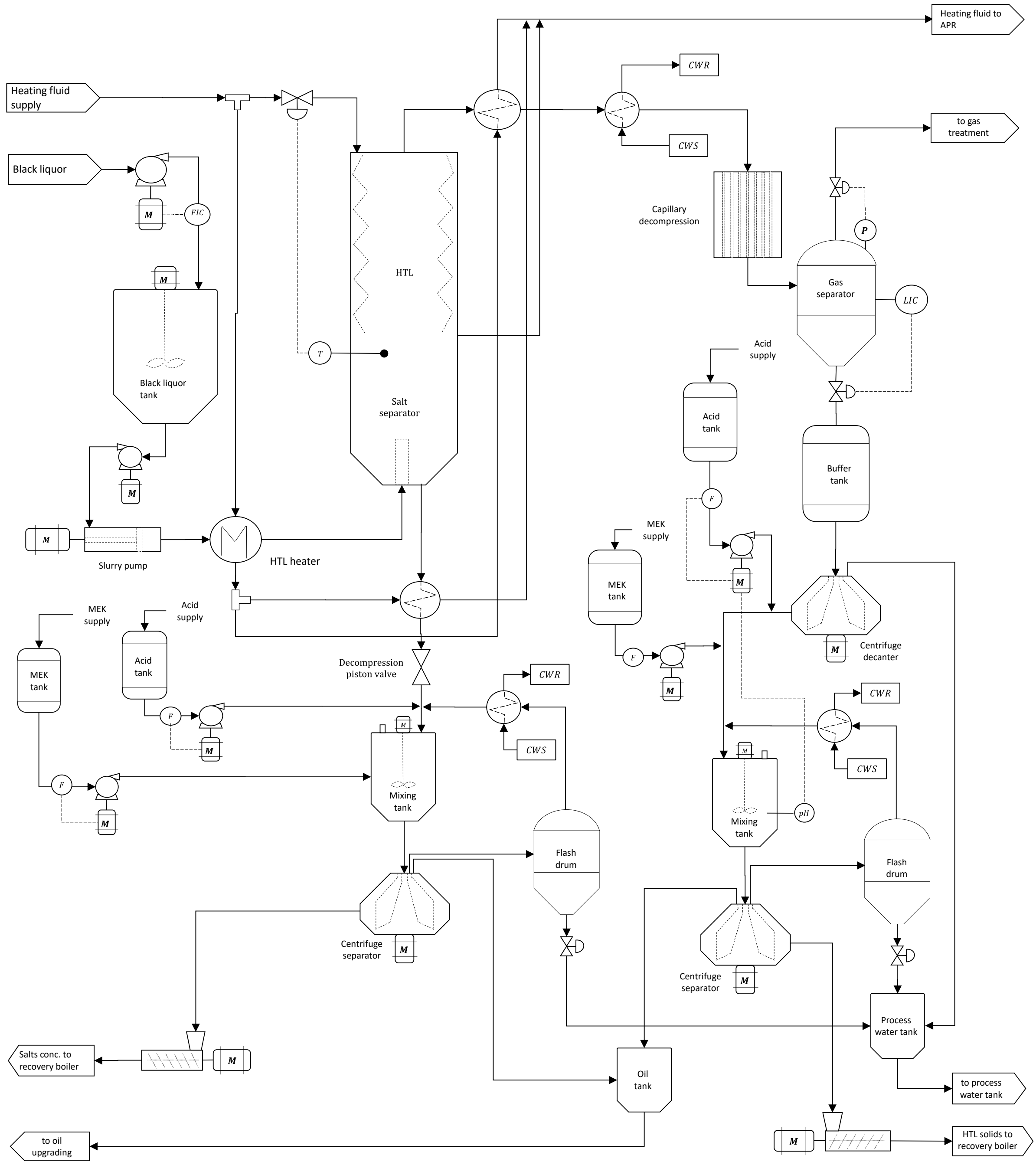

Figure 4: Process flow diagram representing the production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, and full extraction of the oil phase from both the salts brine and the desalinated HTL product (cases 1 and 3).

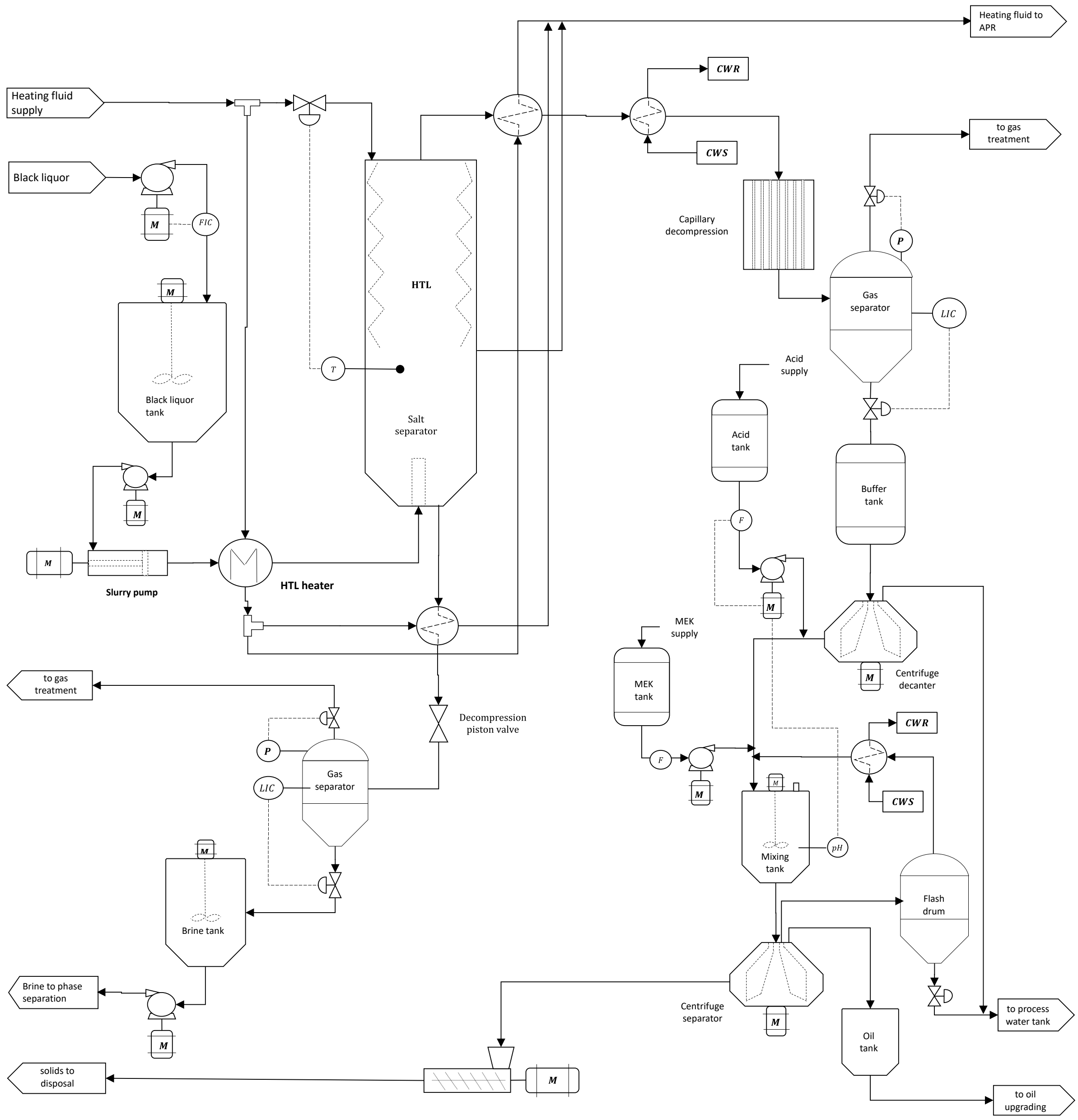

Figure 5: Process flow diagram representing the production of biocrude from black liquor based on integration of salt separation and hydrothermal liquefaction in the same reactor, extraction of the oil phase from desalinated HTL product only. The salts brine is cooled and depressurized before being fed into the recovery boiler at the pulp mill (case 2).

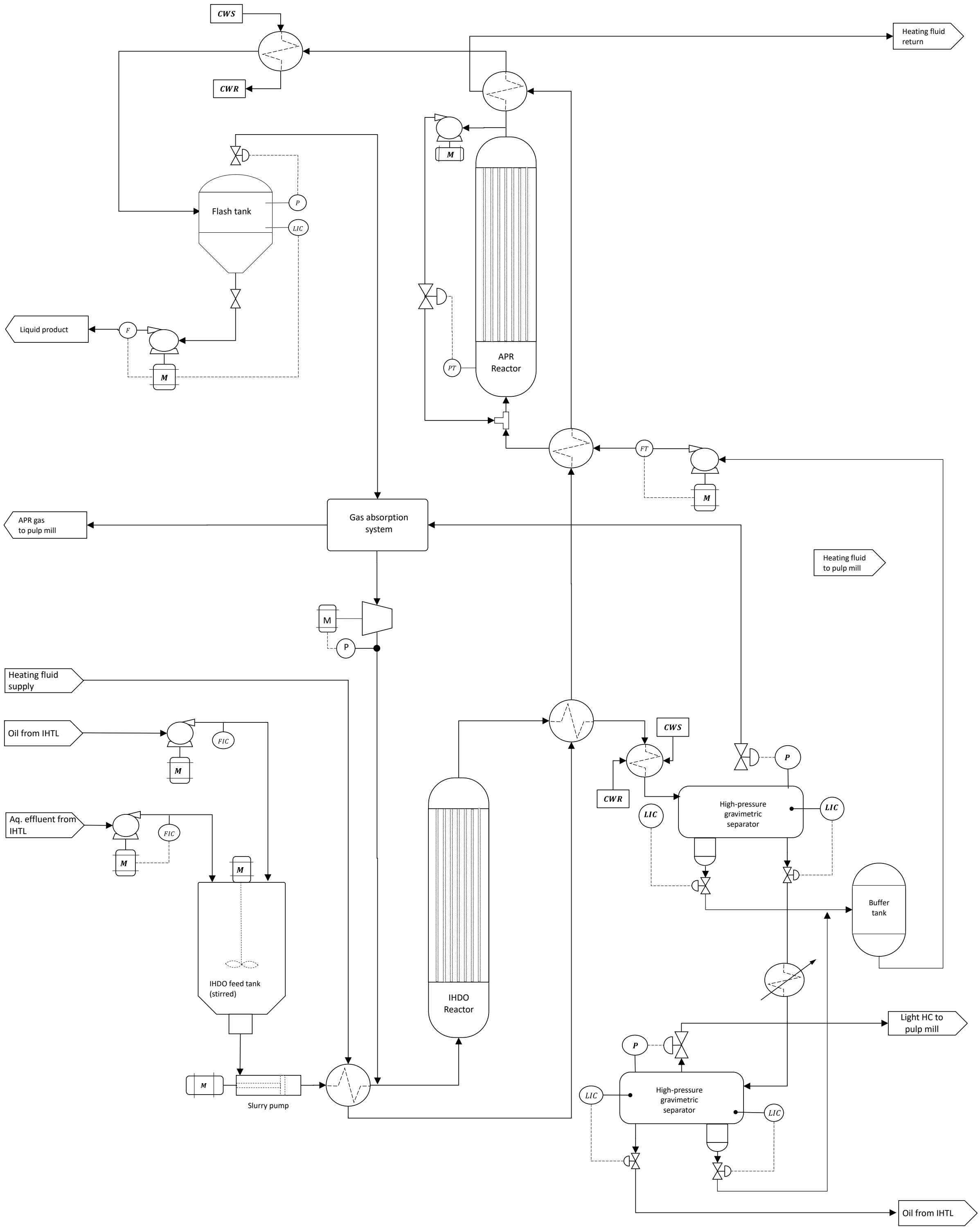

Figure 6: Process flow diagram representing the partial upgrading of HTL biocrude derived from black liquor hydrothermal deoxygenation with production of hydrogen by aqueous phase reforming of the process water (case 1 and 2).

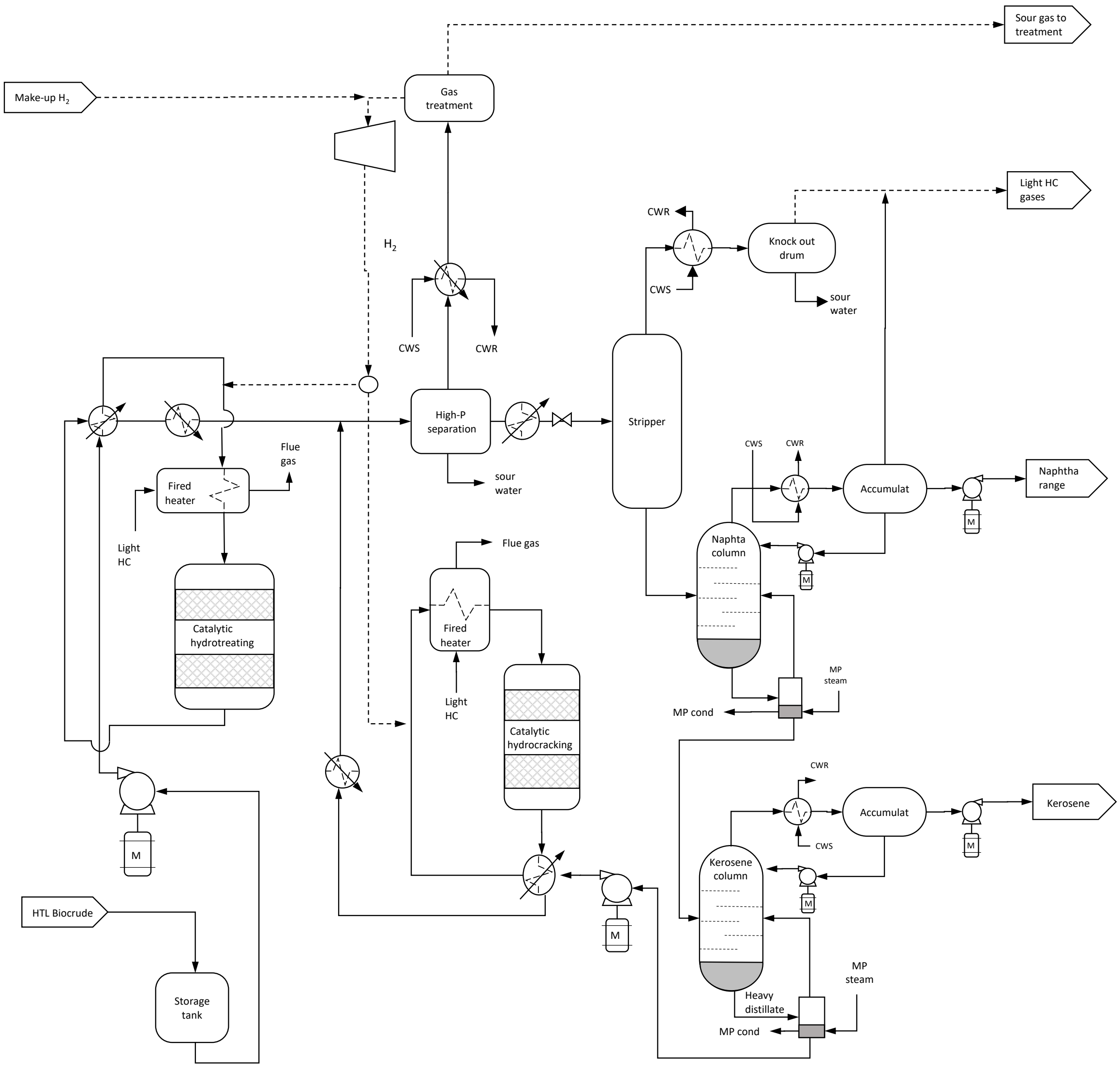

Figure 7: Process flow diagram representing the upgrading of biocrude derived from black liquor to kerosene and marine fuel based on conventional refinery processes.

3. Scale-up and cost models

3.1 Equipment design models

3.1.1 IHTL reactor

The IHTL reactor has a tubular design with a heating jacket in the upper part to regulate the temperature of the hydrothermal liquefaction process. The size of the reactor is defined in terms of the total volume V_R, calculated from $V_R = (\dot{M}_{bl}/\rho_{bl})(t_P + t_{HTL})$ where $\dot{M}_{bl}$ and ρ_{bl} are the mass flow rate and density of the black liquor and t_P, t_{HTL} are characteristic residence times for the salt precipitation and hydrothermal liquefaction processes.

3.1.2 IHDO and APR reactors

The IHDO and APR reactors are designed as cylindrical vessels with tubular fixed bed catalyst support. The dimensions of the reactors are specified in terms of the reactor vessel height L_R, reactor vessel internal diameter D_R, number of tubes N_t, the tubes length L_t and internal tube diameter d_t. The main criterion considered for scaling-up the reactor designs is to preserve the similarity in the mass transfer along the catalyst bed, which is achieved by maintaining the Reynolds number inside the tubes $Re_t = 4\dot{M}_f/(N_t d_t \pi \mu_f)$ constant, where μ_f denotes the viscosity of the feed inside the tubes. The total internal volume of the tubes is specified as a function of the feed mass flow rate $\dot{M}_f$ and the weight hourly space velocity $WHSV$ of the catalyst from $V_t = L_t \pi\, d_t^2/4 = (\dot{M}_f/WHSV)\rho_{cat}^{-1}/(1-\phi_B)$, where ρ_{cat} is the density of the catalyst, ϕ_B is the porosity of the catalyst bed, and φ denote the ratio between the empty tubes to the catalyst bed volume. For both APR and IHDO reactors. It is assumed that the tubes are filled entirely with catalyst.

3.1.3 Hydrotreating and hydrocracking reactors

Both the hydrotreating and hydrocracking reactors arespecified by their volume V, calculated as a function of the feed mass flow rate $\dot{M}_f$ and the weight hourly space velocity $WHSV$ of the catalyst from $V = (\dot{M}_f^K/WHSV_{cat}^K)\rho_{cat}^{-1}\varphi/(1-\phi)$, where ρ_{cat} is the density of the catalyst and the parameters ϕ and φ denote, respectively, the porosity of the bed material and the empty volume to material filled volume ratio. For all catalytic reactors, the value of φ is constant and equal to 1.4.

3.1.4 Heat exchangers

Sizing of the heat exchangers has been defined in terms of the heat transfer area A_{th} calculated from the equation $A_{th} = \dot{Q}_{th}/(U_{th}\, LMTD)$, where $\dot{Q}_{th}$ is the thermal duty, $LMTD$ is the log mean temperature difference and U_{th} is the overall heat transfer coefficient. The thermal duty is evaluated from $\dot{Q}_{th} = \dot{M}_h c_{p,h}(T_h^{in} - T_h^{out})$ for heaters and $\dot{Q}_{th} = \dot{M}_c c_{p,c}(T_c^{in} - T_c^{out})$ for coolers, where the subscripts h and c denotes hold of cold sides of the heat exchanger, and $\dot{M}$, $c_p$denotes mass flow rate and specific heat capacity respectively. The scale-up of heat exchangers in this work assumed constant reference values of U_{th} for each type of heat exchanger.

3.1.5 Pumps

Dimensioning of pumps is defined by the total electric power calculated from $\dot{W}_{el} = (\dot{M}_f/\rho_f)\Delta P/\eta_m$ where η_m is the electric to mechanical power efficiency of the motor and $\Delta P = \Delta P_p + \Delta P_{losses} + \Delta P_H$ represents the total pressure increase, where ΔP_p is the increase of pressure between the pump feed and the one required by the main downstream process, ΔP_{lost} is the pressure drop in the piping and auxiliary equipment between the pump and the downstream process equipment, and $\Delta P_H = \rho_f g dz$ is the pressure difference due to change in elevation. This formulation assumes that the variation of the kinetic energy due to reduction or increase in flow velocities is negligible. Pressure losses have been calculated from $\Delta P_{\mathrm{losses}} = (K + f_{\mathrm{p}}\, L_{\mathrm{p}}/D_{\mathrm{p}})\rho v^2/2$, where v, ρ, L_p and D_p denote, respectively, the internal fluid velocity, the fluid density, the pipe length and the pipe diameter. The parameters f_p and K represents respectively an average friction coefficient for the piping and a lump pressure drop coefficient due to elbows, valves,fittings and heat exchangers.

3.1.6 Stirred tanks

Stirred tanks are specified by the total volume of the tank and the total electric power of the impeller. The tank volume is calculated from $V_T = (\dot{M}_f/\rho_f)t_R$ in terms of the residence time t_{R} and the feed mass flow rate $\dot{M}_f$ and density ρ_f. All stirred tank are assumed to be cylindrical with diameter and height calculated from $D = (4V_T/\pi k_{HD})^{1/2}$ and $L = k_{HD}(4V_T/\pi k_{HD})^{1/2}$, where k_{HD} is the height to diameter ratio which is assumed to be constant and equal to 2. The power consumption is calculated from $\dot{W}_{el}^{SR} = N_P \rho N_I^3 D_I^5/\eta_{el}^M$ where N_I is the impeller rotational speed (rpm), D_{I} is the impeller diameter, and $N_{\mathrm{P}} = 346.7/Re_I + 1.27$ is the impeller power number based on the Reynolds number $Re_I = (N_I/60)\, D_I^2 \rho_f/\mu_f$.

3.1.7 Storage tanks

The size of all tanks is defined by the total volume $V = (\dot{M}_f/\rho_f)t_R/\varphi$, calculated as a function of the feed mass flow rate $\dot{M}_f$ and density ρ_f, the residence time t_R, and the fraction of the total volume occupied by the material. For all storage equipment, the value of φ is constant and equal to 0.8.

3.1.8 HTL solids conveyor

Transport of the HTL solids is assumed to be carried out by one screw conveyor, dimensioned in terms of the total electric driving power which is calculated from $\dot{W}_{el} = (\dot{W}_N + \dot{W}_M + \dot{W}_H)/\eta_m$, where $\dot{W}_{\mathrm{N}} = DL/20$ is the empty power loss, $\dot{W}_M = f_{\mathrm{m}} g \dot{M}_f L$ is the load power due to friction losses caused by the weight of the material, and $\dot{W}_H = g\dot{M}_f H$ is the power required to overcome an elevation difference H. Here, the constant f_m is a progress resistance representing an artificial friction coefficient for the moving material, g is the gravitational constant, and η_m is the electric to mechanical power efficiency of the motor assumed to be constant and equal to 0.9.

3.1.9 Compressors and fans

Compressors are specified based on the total electric power calculated from $\dot{W}_{el} = (\dot{M}_g/\rho_g)P_i\left[(P_o/P_i)^{(1-1/k)} - 1\right][k/(k-1)]/\eta_m$, where $\dot{M}_g$ and ρ_g are the mass flow rate and density of the gas, P_i and P_o are the inlet and discharge pressure, η_{m} is the electric to mechanical power efficiency of the motor, and k is the polytropic coefficient assumed to be constant and equal to 0.8.

3.1.10 Centrifuge

The specification of the centrifuge is described in terms of the total electric power, calculated from $\dot{W}_{el} = k_w(\dot{M}_f/\rho_f)$, where k_w is the specific electricity consumption per unit volume of the input feed, assumed to be constant and equal to 1.4 kWh/m3.

3.2 Cost models

3.2.1 Equipment cost

The total equipment cost is defined as the sum of the purchase and installation costs for each equipment has been calculated from $C_{PI,k} = C_{P,k}^B(S_k/S_k^B)^{n_k}(I/I_B)f_{inst,k}$, where $C_{P,k}^B$ and S_k^B are the base-case equipment purchase cost and equipment size, S_k is the actual size of equipment, n_k is the equipment scale factor, $f_{inst,k}$ is the equipment installation factor, and I/I_B is the price index ratio between the actual year and the reference year where the base case purchase cost function is evaluated based on the Chemical Engineering Plant Cost Index (CEPCI). Table **3** lists the values for the equipment cost parameters used for calculating the installed cost of the equipment included in the biocrude production plant. All the plant costs are updated to 2022 with price indexes. Equipment cost for the upgrading plant is evaluated using Aspen Process Economic Analyzer [4].

3.2.2 Total permanent investment

The total capital costs are defined as the total permanent investment C_{TPI}, calculated from $C_{TPI} = (\sum_k C_{PI,k})[1 + f_{site} + f_{building} + f_{land}][1 + f_{cont} + f_{eng}][1 + f_{dev} + f_{com}]$, where $C_{PI,k}$ is the purchase and installation cost for a specific equipment k, and f_i represents additional costs factors including civil work associated with site preparation and process equipment building, offsite accessibility and services, contingency margin, contractors, land, royalties and patents. Representative values [3] for f_i are listed in Table 4.

3.2.3 Operating cost

The total operating costs are calculated on annual basis from $C_{OP} = C_{op,d} + C_{op,i} + C_{maint}$, where $C_{op,d}$ represents the variable direct operational cost dependent on the annual processing of feedstock, $C_{op,i}$ are the fixed indirect operational costs required for having the plant in activity, and C_{maint} are the maintenance costs. The direct operational costs are calculated from $C_{op,d} = C_{cons} + C_{em} + C_{res} + C_{ef} + C_{el}$, where C_{cons}, C_{em}, C_{res}, C_{ef}, C_{el} are annual costs of consumables, emissions to air, disposal of solid residues, disposal of effluents and electricity, respectively. These costs are calculated based on the individual rates of consumption or production obtained from the mass and energy flows reported in Section 6 with unit cost values shown in Table 15, and the annual operating time of the plant. The annual indirect operational costs have been calculated from $C_{op,i} = C_{labor} + C_{adm} + C_{insur} + C_{tax}$, where C_{labor}, C_{adm}, C_{insur} and C_{tax} are labor, administration, insurance and taxes, respectively. The total annual labour cost has been evaluated from $C_{labor} = \sum_j N_j[c_{r,j}(1 + f_{lb}) + f_{OH,j}c_{OH}]$, where the subscript j denotes the personnel categories, and N_j, $c_{r,j}$, f_{lb}, $f_{OH,j}$ and c_{OH} represent the annual man-hour of personnel required, the hourly rate, the labor burden factor, the overhead factor and the overhead cost factor, respectively. The personnel categories and the values for the labour cost used are shown in Table 4. The number of personnel required has been estimated based on individual main systems proportional to the purchase and installation costs, except for management which is assumed to be constant. It is also assumed that each area requires equal number of personnel per category, two regular operators, one skilled operator, one supervisor and two lab technicians. This work only considers overhead for regular and skilled operators associated to unplanned plant shutdowns due to equipment failures. The costs for administration and insurance are evaluated as a percentage of the total permanent investment according to Table 4.

3.2.4 Levelized cost of biocrude and minimum fuel selling price

The levelized cost of biocrude and the minimum fuel selling price (MFSP), denoted by c_{bc} and c_{bf}, are defined as the average prices for the biocrude and biofuels, respectively, per unit energy produced so that the overall net present value (NPV) for the total permanent investment over its lifetime becomes zero. Based on these definitions, c_{bc} and c_{bf} are calculated using the formulas

$$c_{bc} = \left(1/\dot{H}_F^{HTL}\right) \sum_{i=1}^{N} \left[(1+r)^{-i}\left(C_{TPI,i}^{PROD} + C_{OP,i}^{PROD} - C_{REV,i}^{PROD}\right)\right] / \sum_{i=1}^{N} \left[(1+r)^{-i} t_{p,i} \epsilon_{bc}\right] \quad (1)$$

and

$$c_{bf} = \left(1/\dot{H}_{bc}^{UPG}\right) \sum_{i=1}^{N} \left[(1+r)^{-i}\left(C_{TPI,i}^{UPG} + C_{OPI,i}^{UPG} + C_{bc,i}^{UPG} - C_{REV,i}^{UPG}\right)\right] / \sum_{i=1}^{N} \left[(1+r)^{-i} t_{p,i}(\epsilon_K + p_N \epsilon_N)\right]. \quad (2)$$

Here, r is the expected return of investment, $C_{TPI,i}^{K}$, $C_{OP,i}^{K}$ and $C_{REV,i}^{K}$ are the annual distributions of the annual total investment, operating costs, and revenues over the plant lifetime, with $K = PROD, UPG$ denoting the biocrude production plant and the biocrude upgrading plant respectively. In this notation, ϵ_{bc} is the annual energy efficiency of the black liquor to biocrude conversion, ϵ_D and ϵ_N is the annual energy efficiency of the biocrude to diesel and naphtha conversion, p_N is the market price of naphtha relative to diesel, and $t_{p,i}$ is the annual production time assumed to be 8000 hours. The financial assumptions used in Eq. (1) and (2) and the unit prices for calculation of revenues are shown in Table 4.

Table 3: Parameters for calculating the purchase and installation cost for the equipment.

Equipment	Base specification		Material	Base purchase cost	L/Inst	Installation factor	Scale factor	Base Year
	S_k^B			$C_{P,k}^B$ (M€)		$f_{inst,k}$	n_k	
Screw conveyor	33.5	t/h	SS304	0.350	0.50	2.10	0.80	2002
Black liquor pump	45.0	kWe	SS316	0.175	0.50	2.47	0.70	2017
Black liquor tank	76.7	m^3	SS316	0.174	0.50	2.47	0.70	2010
Shell &tube heat exchanger	7.8	MW	SS316	0.080	0.50	1.24	0.60	2010
Stirred reactor tank	12.3	t/h	SS304	0.180	0.50	2.22	0.70	2009
Stirred reactor agitator	7.5	kW	SS316	0.027	0.50	1.85	0.50	2009
Solid storage silo	2821.0	kg/h	CS	0.055	0.50	2.10	0.70	2013
HTL pump	333.0	kW	SS347 / P91	0.470	0.50	2.84	0.80	2011
HTL preheater	17.5	MW	SS347 / P91	1.970	0.50	2.72	0.70	2012
HTL reactor	5.4	m^3	SS347 / P91	0.270	0.50	2.47	1.00	2013
HTL Product cooler	74.9	MW	SS347 / P91	5.540	0.50	2.72	0.70	2013
Gravimetric separation	12.0	l/s	CS	0.190	0.40	2.73	0.84	2007
Flash tank	20.0	m^3	CS	0.014	0.40	2.73	0.71	2007
Condenser	0.2	MW	SS304	0.067	0.50	1.67	0.53	2017
Centrifuge	500	kg/h	SS316	0.149	0.96	2.47	0.38	2007
Gas compressor	15.0	kW	CS	0.014	0.50	2.47	0.70	2017
Cooling water pump	3.7	kWe	CS	0.009	0.50	2.84	0.80	2013
Cylindrical atmospheric tank	1.5	m^3	CS	0.017	0.40	2.73	0.93	2007
Water cooler	0.4	MW	SS304	0.060	0.50	1.67	0.53	2017
Acid storage tank	1981	kg/h	SS316	0.100	0.40	1.73	0.71	2020
Acid pump	1981.0	kg/h	SS316	0.023	0.50	2.47	0.70	2010
HTL aqueous phase pump	73.5	kW	SS316	0.023	0.50	2.84	0.80	2009
APR reactor (tubular)	49.1	m^3/h	SS316	1.05	0.5	1.23	0.7	2018
IHDO reactor (tubular)	49.1	m^3/h	SS316	2.1	0.5	1.23	0.7	2018
PSA package	500	Kg-H_2 /h	SS304	1.6	0.5	1.23	0.7	2018
H2 compressor	107	kW	SS304	5.4	0.5	2.70	0.65	2005

Table 4: Unit prices considered for calculating operating costs

Parameter	Unit cost
Citric acid. €/ton	640
MEK. €/ton	1440
Catalyst hydrotreating, €/liter	31.4
Catalyst hydrocracking, €/liter	31.4
Process water disposal (upgrading) [€/m3]	83,0
Light hydrocarbons [€/kg]	1,0
H2 production cost (SMR) [€/kg]	3,8
Amine (MDEA) [€/kg]	2,9
Fresh water. €/ m^3	0.5
Electricity. €/kWh	1.0
labor average annual income. k€/year Managers O&M Manager Engineers Maintenance technician Shift supervisor Shift operators Administration Site and building maintenance	 162 88 96 59 66 59 37 37
Overhead factor (operators only). %	20
Labor overhead charge rate fraction	1.25
Annual operating time (h)	8000
Administration cost. % total permanent investment	2
Insurance cost. % total permanent investment	1
Loan interest rate. %	7
Return of investment. %	10
Equity to debt ratio	30/70
Plant lifetime. years	25
Construction time. years	2
Commissioning time. years	1

4. Analysis and results

This section the reports the scale-up and cost analysis biocrude production and upgrading at refinery based on the process design described in Section 2.

4.1 Biocrude production

4.1.1 Design parameters and assumptions

The overall black liquor to biocrude conversion process has been evaluated and described in terms of the overall mass and energy flows for HTL plant design scenarios corresponding to cases 1-3 as described in Section 2. The parameter representing the capacity of the biocrude production plant has been defined as the mass flow rate of black liquor diverted from the pulp mill to the HTL plant, denoted by $\dot{M}_{BL}^{HTL}$. To quantify a representative range of values for $\dot{M}_{BL}^{HTL}$, it has been considered a pulp mill with a reference annual bleached pulp capacity of 500 kt/year, which corresponds to an equivalent black liquor production of approximately 2000 dry-ton/day. As previously reported in deliverable D4.1, there is a limit for the maximum fraction of black liquor that can be diverted to a HTL plant integrated into the pulp mill which is associated to a reduction of the excess electricity production being zero. The black liquor composition and its calorific value are constant and specified in Table 5.

The operational parameters considered in the process analysis are listed in Table 6. In addition, the following assumptions have been used:

- The variation of all mass and enthalpy flow rates have a linear dependency on the plant capacity.
- Only the insoluble organic fraction, excluding salts, of the black liquor undertakes hydrothermal liquefaction. All dissolved salts in the aqueous phase prior liquefaction remains in the aqueous phase.
- There is no loss of solids, gas, aqueous or oil phases during the overall conversion.
- The pH control in the phase separation of both the desalinated HTL product and the salts brine is performed using citric acid.
- Methyl Ethyl Ketone (MEK) is used as the organic solvent used in the phase separation of both the desalinated HTL product and the salts brine. The is a recovery of MEK of 90% by flashing of the aqueous phase after three-phase centrifuge and condensation of the organic vapor.
- The aqueous phase that remains in the salts brine, oil and remaining solid residue after phase separation has the same chemical composition.
- All sulphur content in the gas phase after liquefaction or hydrodeoxygenation are in form of H_2S.
- The feed flow rate to the aqueous phase reforming system is assumed to be the same, corresponding to the liquid stream from the dewatering of the desalinated IHTL product. Due to absence of empirical data, the increase of process water and dissolved organics in the aqueous phase from IHDO is assumed to be negligible.

- It has been assumed that the hydrogen yields from the APR system matches the feed requirement by IHDO.

4.1.2 Baseline mass and energy flows

Tables 7 and 8 show the main mass and energy flows of the biocrude production process for the design cases 1-3. The calculations use the empirical models described previously [1]. Under the assumptions and operational parameters described above, the maximum mass and energy yields of oil that produced from IHTL relative to black liquor on dry basis are 23.9 % and 55.4 % respectively, assuming that the oil is extracted from both the salts brine and the desalinated HTL product. The mass yield of the salt brine before and after extraction of the oil has been estimated to be 32.6 and 24.3%, respectively, containing approximately 96% of the total salts in the black liquor. The yields of dry solids, gas, and dissolved material in the aqueous phases from the IHTL process are, respectively, 16.2, 6.3 and 44,4% on dry mass basis, which correspond to 4.3, 6.4 and 33,7 % on energy basis. Further upgrading of the total oil extracted from IHTL by hydrodeoxygenation (IHDO) decreases the mass and energy yields of oil to 21.1% and 50.4% while it increases the gas and dissolved material in the aqueous phase by 2.4 and 0.4 % on dry mass basis, equivalent to 10.6 and 12.6 % on energy basis.

4.1.3 Electric loads

Table 10-12 show the calculated electric loads (kW) of all the equipment included in the biocrude production plant as a function of the black liquor feed capacity for the design cases 1-3. The case 1 process design for the biocrude production plant exhibits the highest power consumption, approximately 538 kWh per dry ton, almost linear with the black liquor feed capacity indicating that the larger pressure drop due to larger plant layouts has a small effect in the overall power consumption. The electricity consumed by the separation of hydrogen from the sour gas after IHDO and the APR gas contributes largely and represents around 70% of the total power load. Discarding the extraction and further treatment of the oil contained in the salts brine (case 2) leads to a small reduction of the power consumption, about 4% when the oil content in the salts brine is assumed to be 10% of the total production in the IHTL process. For this case, since the power load of the gas treatment is very dominant, an increase in the oil fraction separated with the salts brine leads to a significant reduction of the total power consumption. Considering the production and full extraction of oil from the salts brine and the desalinated IHTL product only (case 3), the power load is around 108 kWh per dry ton of black liquor, where the main feed pump to the IHTL reactor and the centrifuges in the phase separation systems account to approximately 73% and 27% of the total power load.

4.1.4 Installed equipment cost

Tables 13-15 and Figure 8 show the calculated values of the installed cost for all equipment included in the process design of the biocrude production plant for cases 1-3 as a function of the black liquor feed capacity. The case 1 has the largest installed equipment cost of all cases, varying between 0.31 and 0.2 M€/dry-ton/day for plant capacities between 100 and 600 dry-ton/day. For this design scenario, the IHTL system accounts approximately for 50 % of the total equipment cost, with the main heater, IHTL reactor and IHTL product cooler contributing around 70% of the total cost for this system. Including the complete phase separation for extraction of oil from both the salts brine and the desalinated HTL product has a relatively small contribution to the total equipment cost, which represents approximately 10%. Partial upgrading of the IHTL oil by hydrodeoxygenation with local production of hydrogen via APR of the process water contributes approximately 40% to the total equipment cost. If the salts brine is taken back to the pulp mill without extraction of the oil, the total equipment cost is reduced by approximately 3%, assuming the oil contained in the salts brine is 10% of the total oil produced in the IHTL process. All cases have similar dependency of the equipment installed cost on the plant capacity, which corresponds to an overall scale factor of 0.7.

4.1.5 Operating cost

Calculations of the operating cost for the biocrude production plant for cases 1-3 as a function of the black liquor feed capacity are shown in Tables 16-18. These results indicate that including hydrodeoxygenation of the IHTL oil with local production of hydrogen by APR increases significantly the total operating cost due to the high electricity consumption by the gas treatment. Therefore, case 1 exhibits the highest operating cost, which ranges between 0.85 and 0.76 k€/dry-ton of black liquor for biocrude plant capacity between 100 and 600 dry-ton/day, with the cost of electricity accounting for 68% of the total operating cost. Another important contribution to the operating cost is the consumption of citric acid in the phase separation process, which represents approximately 7% of the total cost of consumables and utilities. Discarding the oil extraction from the salts brine (case 2), which reduces the IHTL oil feed to hydrodeoxygenation by 10% under the assumptions used in the analysis, leads to only a 5% decrease of the operating cost since the feed to the APR, and thus the gas produced from this process, is practically the same between cases 1 and 2. Considering only production and full extraction of the IHTL without further hydrodeoxygenation and APR leads to a significant reduction of the operating costs, which ranges between 0.32 and 0.27 k€/dry-ton of black liquor. Here, electricity still represents 42% of the total operating cost and the consumption of citric acid accounts for 23% of the total cost for consumables and utilities.

4.1.6 Levelized cost of biocrude

Results of the levelized cost of biocrude, together with a summary of total project costs, as a function of the biocrude production capacity for design cases 1-3 are shown numerically in Tables 16-18 and graphically in Figure 8. The cost of biocrude achieved only by production and full extraction of IHTL oil (case 3) is in the range of 2.1-1.7 €/Liter, under the assumptions and the black liquor feed capacities considered in the analysis. If hydrodeoxygenation of the total amount of IHTL oil and local production of hydrogen by APR of the process water are included in the biocrude production plant design (case 1), the cost of biocrude increases to 5.8-4.7 €/Liter mainly due to the large increase of operating costs. Reducing the oil extracted from IHTL by 10% (case 2) leads to a further increase of the cost of biocrude, which ranges between 6.1 and 5.1 €/Liter. This result shows that, when including partial upgrading of the IHTL oil in the biocrude production plant, lowering the oil production has a larger effect in increasing the cost of biocrude than the effect of reduction of capital and operating costs.

4.2 Biocrude upgrading.

4.2.1 Design parameters and assumptions

The scale parameter is defined as the mass flow rate of the biocrude feed entering the upgrading process, denoted by $\dot{M}_{bc}^{UPG}$. A representative range for $\dot{M}_{bc}^{UPG}$ used in the analysis has been estimated to be 100-2500 ton/day. This range is equivalent to the maximum biocrude production rate from 5-25 pulp mills all with an annual bleached pulp capacity of 500 kt/year.

Analysis of the upgrading process has been performed using the process design parameters listed in Table **19.** In addition, the following assumptions have been used:

- The biocrude composition and calorific value is constant, independent on the feed capacity, and corresponding to Table 10.
- Hydrotreating and hydrocracking processes have been assumed to be adiabatic.
- Reduced S from the feed in hydrotreating and hydrocracking processes are transferred to the gas-phase as H2S, respectively.
- Reduced O from the feed in the guard reactor, hydrotreating and hydrocracking processes forms CO2 by decarboxylation and water.
- Reduced heteroatoms other than S and O remain in the catalyst bed (adsorbed) in solid phase.
- The S and O heteroatoms in the oil phase after hydrotreating remain in the distillate residue from fractionation.
- The main liquid organic products from fractionation are naphtha, kerosene, and a distillate residue. The boiling temperature range and high heating values for those fractions are specified in Table 20.

4.2.2 Baseline mass and energy flows

Tables 21-22 show the main mass and energy flows for the overall biocrude production process. Assuming that the whole distillate residue from the kerosene column is further treated by hydrocracking, the total yields of naphtha and kerosene products, relative to the biocrude feed, are estimated to be 24.2% and 54.4% on dry mass basis and 29.9 and 66% on energy basis. Under this assumption, the overall upgrading process requires approximately 13 kg of make-up hydrogen per ton of biocrude, 80% of which is consumed by the hydrotreating process and the remaining by hydrocracking. The total heat demand is around 2.1 MJ per kg of biocrude feed where the heating of the biocrude before hydrotreating and the reboiler of the naphtha column bottoms are the main contributions.

4.2.3 Electric loads

Calculations of the equipment electric loads for the biocrude upgrading at refinery as a function of the biocrude feed capacity are shown in Table 23. The total electric load for the overall upgrading process is approximately 10.6 kWh per ton of biocrude feed and exhibits a linear dependency with the feed capacity. The main contribution to the total electric load corresponds to the hydrotreating system which represents around 50% of the total, where the power of the main biocrude pump consumes about 65% of the power load of this system. The other major contributors to the power load are the hydrocracking system (25%) mainly from the pressurization of the distillate residue, and the gas treatment and compression (22%) from the amine system and compression of the hydrogen feed to the catalytic processes.

4.2.4 Installed equipment cost

Table 24 shows the calculated values of the installed cost for all the equipment considered in the biocrude upgrading process design. Figure 10 shows graphically the variation with the feed capacity of the specific equipment installed cost per unit biocrude mass flow rate for the total upgrading process (left) and the distribution among systems (right). For the range of biocrude feed capacity considered in the analysis, total cost of equipment required for the upgrading varies between 0.16 and 0.054 M€/ton/day. The hydrocracking system accounts for 30% of the total equipment cost, being the main contribution above the hydrotreating system. This is due to both the high fraction of distillate residue and higher installed cost of the hydrocracking reactor compared to hydrotreating. The fractionation system has also a significant contribution, above hydrotreating at smaller upgrading plant capacities, approximately 26.5% at biocrude feed capacity of 100 dry-ton/day. However, as the plant capacity increases, the contribution of the hydrotreating unit to the total equipment cost increases over fractionation due to higher scale factor of the former. The contribution of the cleaning and recycling the hydrogen-rich sour gas is also significant in the lower capacity range, approximately 20%, but decreases rapidly with the size of the upgrading plant, being approximately 8% at a biocrude feed capacity of 2500 dry-ton/day.

4.2.5 Operating cost

Table 25 shows the calculated values of the variation of the operating costs of the overall upgrading process with the biocrude feed capacity. The total operating cost behaves almost linear with the capacity of the upgrading plant, varying between 5.9 and 1.6 k€ per dry-ton for biocrude cost varying between 1 and 6€/Liter and for biocrude feed capacity in the range of 100 to 2500 dry-ton per day. Consumables other than the biocrude and utilities account for 17.5 % of the operating cost, where the electricity and the make-up hydrogen are the main contributions. Assuming an operational lifetime of two years for the catalysts, their annual contribution to the consumables and utilities costs is about 8%. Fixed operating costs have a small contribution to the operating costs, about 2.5 % of the total, manly from labour and maintenance.

4.2.6 Minimum fuel selling price

Calculated values of the minimum fuel selling prices (MFSP) for naphtha and kerosene as a function of the upgrading plant capacity are shown in Tables 25-26, together with a summary of the total investment, operating costs and revenues. Considering production of naphtha and kerosene only and assuming the kerosene has a market value 1.5 higher than naphtha on energy basis, the minimum fuel selling prices (MFSP) are in the range 10.9-2.1 €/Liter for kerosene and 7.1 and 1.3 €/Liter with biocrude feed capacities considered in the analysis, based on a specific biocrude cost varying between 1 and 6 €/Liter.

Table 5: Black liquor composition used in the analysis

Dry matter, DM (%wt.)	15.8
Salts (%wt. dry)	*24.7*
HHV (MJ/kg) (dry basis)	13.3
Elemental analysis	
Carbon, C (%wt dry)	33.89
Hydrogen, H (%wt dry)	4.13
Oxygen, O (%wt dry)	31.07
Nitrogen, N (%wt dry)	0.00
Sulfur, S (%wt dry)	6.13
Phosphorous, P (g/kg dry)	0.05
Calcium, Ca (g/kg dry)	
Aluminium, Al (g/kg dry)	0.01
Iron, Fe (g/kg dry)	
Magnesium, Mg (g/kg dry)	0.30
Potassium, K (g/kg dry)	37.29
Chlorine, Cl (g/kg dry)	0.48
Sodium, Na (g/kg dry)	209.57
Silicon, Si (g/kg dry)	0.02
Manganese, Mn (mg/kg dry)	91
Composition of salts	
NaOH (%wt dry)	5.91
Na_2S (%wt dry)	0
NaHS (%wt dry)	9.44
Na_2SO_4 (%wt dry)	2.75
Na_2SO_3 (%wt dry)	0
$Na_2S_2O_3$ (%wt dry)	0.29
Na_2CO_3 (%wt dry)	3.36
K_2CO_3 (%wt dry)	2.92

Table 6: Process design parameters for the HTL plant

Process design parameter	Value
Black liquor feed temperature (°C) [a]	105
Black liquor feed pressure (bar-a)	5
Temperature at salt precipitation process (°C)	350
Pressure at salt precipitation process (bar-g)	320
Temperature at HTL process (°C)	350
Pressure at HTL process (bar-g)	320
Salts brine temperature after cooling (°C)	150
Salts brine pressure after depressurization (bar-g)	30,0
Temperature gas separation of desalinated HTL product (°C)	150
Pressure gas separation of desalinated HTL product (bar-g)	30
Temperature first centrifugation of desalinated HTL product (°C)	150
Flashing temperature aqueous phase from desalinated HTL product (°C)	105
Flashing pressure aqueous phase from desalinated HTL product (bar-g)	30
IHDO temperature (°C)	350
IHDO pressure (bar-g)	320
Temperature at HP phase separation after IHDO (°C)	50
Pressure at HP phase separation after IHDO (bar-g)	320
Temperature at LP phase separation after IHDO (°C)	150
Pressure at LP phase separation after IHDO (bar-g)	30
Temperature at aqueous phase reforming (°C)	275
Pressure at aqueous phase reforming (bar-a)	30
Thermal fluid	Pressurized water
Thermal fluid supply temperature (°C)	400
Thermal fluid supply pressure (bar-g)	15

Table 7: Main mass flows for the biocrude production per dry ton of black liquor

Biocrude production process design scenario		Case 1	Case 2	Case 3
Oil product	Dry kg/s	0,211	0,190	0,239
Salts brine	Dry kg/s	0,243	0,326	0,243
Total solids from desalinated HTL product	Dry kg/s	0,162	0,162	0,162
Gas separated from desalinated HTL product	Dry kg/s	0,063	0,063	0,063
Make-up hydrogen feed to IHDO	Dry g/s	11,9	10,7	-
Sour gas from IHDO product	Dry g/s	9,66	8,70	-
Light hydrocarbons from IHDO product	Dry g/s	1,40	1,26	-
Gas from APR	Dry g/s	17,28	14,08	-
Dissolved material in liquid from APR	Dry kg/s	0,156	0,127	-
Dissolved material in return liquid effluent	Dry kg/s	0,375	0,300	0,364
Total acid to phase separation	Dry kg/s	0,066	0,050	0,066
Total organic solvent to phase separation	Dry kg/s	0,0048	0,0043	0,0048

Table 8: Main energy flows for the biocrude production per MJ chemical energy of black liquor

Biocrude production process design scenario		Case 1	Case 2	Case 3
Oil product	MJ/s	0.504	0.453	0,554
Salts brine	MJ/s	0.011	0,134	0.011
Total solids from desalinated HTL product	MJ/s	0.043	0.043	0.043
Gas separated from desalinated HTL product	MJ/s	0.054	0.054	0.054
Make-up hydrogen feed to IHDO	MJ/s	0.128	0.115	-
Sour gas from IHDO product	MJ/s	0.103	0.093	-
Light hydrocarbons from IHDO product	kJ/s	6.3	5.7	-
Gas from APR	MJ/s	0.022	0.018	-
Dissolved material in return liquid effluent	MJ/s	0,385	0,314	0,338

Table 9: Oil-phase composition and calorific value

	Unit	Biocrude from HTL	Organic liquid from IHDO
High heating value, HHV	MJ/kg	30.85	31.72
Carbon, C	% wt. dry	83.53	83.06
Hydrogen, H	% wt. dry	7.025	9.255
Oxygen, O	% wt. dry	7.285	5.300
Nitrogen, N	% wt. dry	0.00	0.00
Sulfur, S	mg/kg dry	3.00	0.014
Potassium, K	mg/kg dry	1.740	1.927
Sodium, Na	mg/kg dry	19.809	21.93

Table 10**:** Electric loads (kW) as a function of the plant capacity for the biocrude production process based on the design case 1

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Total Electric load (kW)	2239,4	4249,1	6397,3	8558	10730,1	12912,7
Slurry preparation and HTL	**348,8**	**716,1**	**1097,6**	**1491,6**	**1896,9**	**2312,7**
Black liquor supply pump	1,6	2,8	3,8	4,8	5,7	6,6
Black liquor tank, agitator	16,1	51,0	100,2	161,8	234,7	318,0
Black liquor pre-charger pump	0,1	0,2	0,3	0,4	0,5	0,6
Black liquor HP pump	329,5	659,0	988,5	1318,0	1647,5	1976,9
Salt brine tank, agitator	1,5	3,1	4,8	6,6	8,5	10,5
Phase separation from desalinated HTL product	**186,8**	**373,8**	**560,7**	**747,8**	**934,9**	**1122,0**
Centrifuge solids decanter	105,1	210,2	315,3	420,4	525,5	630,6
Mixing vessel before second centrifuge	14,8	29,6	44,4	59,2	74,0	88,8
Centrifuge oil-water separation	65,8	131,6	197,3	263,1	328,9	394,6
solids conveyor	1,2	2,4	3,7	5,1	6,6	8,0
Phase separation from salts brine	**23,4**	**46,9**	**70,3**	**93,8**	**117,2**	**140,6**
Mixing vessel before second centrifuge	4,3	8,6	12,9	17,2	21,5	25,8
Centrifuge oil-water separation	19,1	38,3	57,4	76,5	95,7	114,8
Hydrodeoxygenation	**100,7**	**114,5**	**128,4**	**142,2**	**156,0**	**169,9**
Mixing tank	13,8	27,7	41,5	55,4	69,2	83,0
Feed pump	86,8	86,8	86,8	86,8	86,8	86,8
Aqueous Phase Reforming	**0,1**	**0,2**	**0,4**	**0,5**	**0,6**	**0,7**
Process water pump	0,1	0,2	0,4	0,5	0,6	0,7
Gas treatment	**1579,5**	**3159,1**	**4738,7**	**6318,2**	**7897,8**	**9477,3**
PSA PACKAGE	1552,3	3104,6	4657,0	6209,3	7761,6	9313,9
H2-rich recycle compressor	27,2	54,5	81,7	108,9	136,2	163,4

Table 11**:** Electric loads (kW) as a function of the plant capacity for the biocrude production process based on the design case 2

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Total Electric load (kW)	2153,5	4249,1	6397,3	8558	10730,1	12912,7
Slurry preparation and HTL	**348,8**	**716,1**	**1097,6**	**1491,6**	**1896,9**	**2312,7**
Black liquor supply pump	1,6	2,8	3,8	4,8	5,7	6,6
Black liquor tank, agitator	16,1	51,0	100,2	161,8	234,7	318,0
Black liquor pre-charger pump	0,1	0,2	0,3	0,4	0,5	0,6
Black liquor HP pump	329,5	659,0	988,5	1318,0	1647,5	1976,9
Salt brine tank, agitator	1,5	3,1	4,8	6,6	8,5	10,5
Phase separation from desalinated HTL product	**186,8**	**373,8**	**560,7**	**747,8**	**934,9**	**1122,0**
Centrifuge solids decanter	105,1	210,2	315,3	420,4	525,5	630,6
Mixing vessel before second centrifuge	14,8	29,6	44,4	59,2	74,0	88,8
Centrifuge oil-water separation	65,8	131,6	197,3	263,1	328,9	394,6
solids conveyor	1,2	2,4	3,7	5,1	6,6	8,0
Salts brine extraction	**4,3**	**8,6**	**12,9**	**17,2**	**21,5**	**25,8**
Buffer tank	4,3	8,6	12,9	17,2	21,5	25,8
Hydrodeoxygenation	**90,6**	**103,1**	**115,5**	**128,0**	**140,4**	**152,9**
Mixing tank	12,5	24,9	37,4	49,8	62,3	74,7
Feed pump	78,1	78,1	78,1	78,1	78,1	78,1
Aqueous Phase Reforming	**0,1**	**0,2**	**0,4**	**0,5**	**0,6**	**0,7**
Process water pump	0,1	0,2	0,4	0,5	0,6	0,7
Gas treatment	**1522,8**	**3045,7**	**4568,7**	**6091,6**	**7614,5**	**9137,4**
PSA PACKAGE	1496,6	2993,2	4490,0	5986,6	7483,2	8979,8
H2-rich recycle compressor	26,2	52,5	78,8	105,0	131,3	157,5

Table 12**:** Electric loads (kW) as a function of the plant capacity for the biocrude production process based on the design case 3

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Total Electric load (kW)	2239,4	4249,1	6397,3	8558	10730,1	12912,7
Slurry preparation and HTL	**348,8**	**716,1**	**1097,6**	**1491,6**	**1896,9**	**2312,7**
Black liquor supply pump	1,6	2,8	3,8	4,8	5,7	6,6
Black liquor tank, agitator	16,1	51,0	100,2	161,8	234,7	318,0
Black liquor pre-charger pump	0,1	0,2	0,3	0,4	0,5	0,6
Black liquor HP pump	329,5	659,0	988,5	1318,0	1647,5	1976,9
Salt brine tank, agitator	1,5	3,1	4,8	6,6	8,5	10,5
Phase separation from desalinated HTL product	**186,8**	**373,8**	**560,7**	**747,8**	**934,9**	**1122,0**
Centrifuge solids decanter	105,1	210,2	315,3	420,4	525,5	630,6
Mixing vessel before second centrifuge	14,8	29,6	44,4	59,2	74,0	88,8
Centrifuge oil-water separation	65,8	131,6	197,3	263,1	328,9	394,6
solids conveyor	1,2	2,4	3,7	5,1	6,6	8,0
Phase separation from salts brine	**23,4**	**46,9**	**70,3**	**93,8**	**117,2**	**140,6**
Mixing vessel before second centrifuge	4,3	8,6	12,9	17,2	21,5	25,8
Centrifuge oil-water separation	19,1	38,3	57,4	76,5	95,7	114,8

Table 13: Installed equipment cost for the HTL plant as a function of black liquor capacity for the biocrude production design case 1.

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Total plant equipment installed cost (M$)	31,23	52,09	69,95	87,91	104,30	119,28
Salt separation and HTL	**15,12**	**26,78**	**37,58**	**47,88**	**57,86**	**67,58**
Black liquor supply pump	0,07	0,113	0,151	0,184	0,215	0,245
Black liquor tank, vessel	0,856	1,391	1,848	2,26	2,642	3,001
Black liquor tank, agitator	0,011	0,015	0,019	0,022	0,024	0,027
Black liquor pre-charger pump	0,007	0,012	0,016	0,019	0,022	0,025
Black liquor HP pump	1,675	2,916	4,034	5,077	6,07	7,023
Black liquor pre-heater	0,683	1,11	1,474	1,803	2,108	2,395
Main HTL heater	3,131	5,087	6,756	8,263	9,66	10,975
IHTL reactor	5,387	10,773	16,16	21,546	26,933	32,32
IHTL product cooler	3,30	5,36	7,12	8,71	10,18	11,57
Phase separation from desalinated HTL product	**3,09**	**4,71**	**6,06**	**7,27**	**8,38**	**9,42**
Capillary decompression	0,782	1,271	1,688	2,064	2,413	2,742
Gas separation	0,216	0,386	0,542	0,691	0,833	0,971
Buffer tank emulssion	0,286	0,464	0,616	0,754	0,881	1,001
Centrifuge solids decanter	0,552	0,718	0,837	0,934	1,017	1,090
Mixing vessel before second centrifuge	0,232	0,377	0,501	0,613	0,716	0,814
Centrifuge oil-water separation	0,462	0,601	0,701	0,782	0,851	0,912
flash tank aq. effl from centrifuges	0,023	0,037	0,050	0,061	0,071	0,081
Vapor cooler	0,094	0,136	0,169	0,196	0,221	0,243
Oil tank	0,415	0,674	0,895	1,094	1,279	1,453
solids conveyor	0,028	0,048	0,066	0,084	0,100	0,116
Phase separation from salts brine	**1,19**	**1,83**	**2,37**	**2,84**	**3,28**	**3,69**
Salts brine cooler	0,746	1,213	1,611	1,97	2,303	2,616
Salt brine stirred tank	0,10	0,16	0,21	0,26	0,30	0,34
Centrifuge oil-water separation	0,29	0,38	0,44	0,49	0,53	0,57
flash tank after second centrifuge	0,010	0,016	0,021	0,026	0,030	0,034
Vapor cooler	0,049	0,071	0,088	0,102	0,115	0,126
Hydrodeoxygenation	**7,13**	**11,25**	**14,02**	**17,82**	**20,65**	**22,55**
Mixing tank	0,18	0,29	0,38	0,46	0,54	0,62
IHDO feed pump	0,68	1,18	1,63	2,06	2,46	2,84
IHDO feed heater	2,67	3,86	3,27	5,57	3,49	4,72
Heating fluid pump	0,021	0,041	0,041	0,062	0,062	0,062
IHDO reactor	1,71	2,77	3,68	4,5	5,26	5,98
IHDO product cooler	1,28	2,08	3,6	3,38	6,7	5,85
High pressure separator	0,5	0,87	1,2	1,51	1,81	2,09
Low pressure separator	0,09	0,16	0,22	0,28	0,33	0,39
Aqueous Phase Reforming	**3,58**	**5,52**	**7,13**	**8,56**	**9,87**	**11,08**
Process water buffer tank	0,16	0,26	0,35	0,43	0,5	0,56
Process water pump	0,04	0,06	0,07	0,09	0,1	0,11
Heater	1,86	2,68	3,33	3,87	4,36	4,8
APR reactor	0,77	1,26	1,67	2,04	2,39	2,71
Treated water cooler	0,057	0,087	0,111	0,131	0,15	0,168
Flash tank	0,47	0,82	1,13	1,42	1,7	1,97
Treated water storage tank	0,218	0,354	0,47	0,574	0,671	0,763
Gas treatment	**1,13**	**1,99**	**2,78**	**3,53**	**4,25**	**4,95**
Gas cooler	0,056	0,082	0,101	0,118	0,132	0,146
PSA PACKAGE	0,383	0,622	0,826	1,01	1,181	1,342
H2-rich recycle compressor	0,69	1,288	1,855	2,403	2,938	3,462

Table 14: Installed equipment cost for the HTL plant as a function of black liquor capacity for the biocrude production design case 2.

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Total plant equipment installed cost (M$)	30,38	50,81	68,39	86,01	102,13	116,91
Slurry preparation and HTL	**15,12**	**26,78**	**37,58**	**47,88**	**57,86**	**67,58**
Black liquor supply pump	0,07	0,113	0,151	0,184	0,215	0,245
Black liquor tank, vessel	0,856	1,391	1,848	2,26	2,642	3,001
Black liquor tank, agitator	0,011	0,015	0,019	0,022	0,024	0,027
Black liquor pre-charger pump	0,007	0,012	0,016	0,019	0,022	0,025
Black liquor HP pump	1,675	2,916	4,034	5,077	6,07	7,023
Black liquor pre-heater	0,683	1,11	1,474	1,803	2,108	2,395
Main HTL heater	3,131	5,087	6,756	8,263	9,66	10,975
IHTL reactor	5,387	10,773	16,16	21,546	26,933	32,32
IHTL product cooler	3,30	5,36	7,12	8,71	10,18	11,57
Phase separation from desalinated HTL product	**3,09**	**4,71**	**6,07**	**7,27**	**8,38**	**9,42**
Capillary decompression	0,782	1,271	1,688	2,064	2,413	2,742
Gas separation	0,216	0,386	0,542	0,691	0,833	0,971
Buffer tank emulssion	0,286	0,464	0,616	0,754	0,881	1,001
Centrifuge solids decanter	0,552	0,718	0,837	0,934	1,017	1,09
Mixing vessel before second centrifuge	0,232	0,377	0,501	0,613	0,716	0,814
Centrifuge oil-water separation	0,462	0,601	0,701	0,782	0,851	0,912
flash tank aq. effl from centrifuges	0,023	0,037	0,05	0,061	0,071	0,081
Vapor cooler	0,094	0,136	0,169	0,196	0,221	0,243
Oil tank	0,415	0,674	0,895	1,094	1,279	1,453
solids conveyor	0,028	0,048	0,066	0,084	0,1	0,116
Salts brine extraction	**0,84**	**1,37**	**1,82**	**2,23**	**2,60**	**2,96**
Salts brine cooler	0,746	1,213	1,611	1,97	2,303	2,616
Salt brine tank stirred tank	0,10	0,16	0,21	0,26	0,30	0,34
Hydrodeoxygenation	**6,62**	**10,44**	**13,01**	**16,53**	**19,16**	**20,92**
Mixing tank	0,17	0,27	0,35	0,43	0,50	0,58
IHDO feed pump	0,63	1,08	1,50	1,89	2,26	2,61
IHDO feed heater	2,48	3,59	3,04	5,17	3,24	4,38
Heating fluid pump	0,02	0,04	0,04	0,06	0,06	0,06
IHDO reactor	1,59	2,57	3,42	4,18	4,89	5,55
IHDO product cooler	1,19	1,93	3,34	3,14	6,22	5,43
High pressure separator	0,46	0,81	1,11	1,40	1,68	1,94
Low pressure separator	0,08	0,15	0,20	0,26	0,31	0,36
Aqueous Phase Reforming	**3,58**	**5,52**	**7,13**	**8,56**	**9,87**	**11,08**
Process water buffer tank	0,16	0,26	0,35	0,43	0,5	0,56
Process water pump	0,04	0,06	0,07	0,09	0,1	0,11
Heater	1,86	2,68	3,33	3,87	4,36	4,8
APR reactor	0,77	1,26	1,67	2,04	2,39	2,71
Treated water cooler	0,057	0,087	0,111	0,131	0,15	0,168
Flash tank	0,47	0,82	1,13	1,42	1,7	1,97
Treated water storage tank	0,218	0,354	0,47	0,574	0,671	0,763
Gas treatment	**1,13**	**1,99**	**2,78**	**3,53**	**4,25**	**4,95**
Gas cooler	0,056	0,082	0,101	0,118	0,132	0,146
PSA PACKAGE	0,383	0,622	0,826	1,01	1,181	1,342
H2-rich recycle compressor	0,69	1,288	1,855	2,403	2,938	3,462

Table 15: Installed equipment cost for the HTL plant as a function of black liquor capacity for the biocrude production design case 3.

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Total plant equipment installed cost (M$)	19,40	33,32	46,01	58,00	69,52	80,69
Salt separation and HTL	**15,12**	**26,78**	**37,58**	**47,88**	**57,86**	**67,58**
Black liquor supply pump	0,07	0,113	0,151	0,184	0,215	0,245
Black liquor tank, vessel	0,856	1,391	1,848	2,26	2,642	3,001
Black liquor tank, agitator	0,011	0,015	0,019	0,022	0,024	0,027
Black liquor pre-charger pump	0,007	0,012	0,016	0,019	0,022	0,025
Black liquor HP pump	1,675	2,916	4,034	5,077	6,07	7,023
Black liquor pre-heater	0,683	1,11	1,474	1,803	2,108	2,395
Main HTL heater	3,131	5,087	6,756	8,263	9,66	10,975
IHTL reactor	5,387	10,773	16,16	21,546	26,933	32,32
IHTL product cooler	3,30	5,36	7,12	8,71	10,18	11,57
Phase separation from desalinated HTL product	**3,09**	**4,71**	**6,06**	**7,27**	**8,38**	**9,42**
Capillary decompression	0,782	1,271	1,688	2,064	2,413	2,742
Gas separation	0,216	0,386	0,542	0,691	0,833	0,971
Buffer tank emulssion	0,286	0,464	0,616	0,754	0,881	1,001
Centrifuge solids decanter	0,552	0,718	0,837	0,934	1,017	1,090
Mixing vessel before second centrifuge	0,232	0,377	0,501	0,613	0,716	0,814
Centrifuge oil-water separation	0,462	0,601	0,701	0,782	0,851	0,912
flash tank aq. effl from centrifuges	0,023	0,037	0,050	0,061	0,071	0,081
Vapor cooler	0,094	0,136	0,169	0,196	0,221	0,243
Oil tank	0,415	0,674	0,895	1,094	1,279	1,453
solids conveyor	0,028	0,048	0,066	0,084	0,100	0,116
Phase separation from salts brine	**1,19**	**1,83**	**2,37**	**2,84**	**3,28**	**3,69**
Salts brine cooler	0,746	1,213	1,611	1,97	2,303	2,616
Salt brine stirred tank	0,10	0,16	0,21	0,26	0,30	0,34
Centrifuge oil-water separation	0,29	0,38	0,44	0,49	0,53	0,57
flash tank after second centrifuge	0,010	0,016	0,021	0,026	0,030	0,034
Vapor cooler	0,049	0,071	0,088	0,102	0,115	0,126

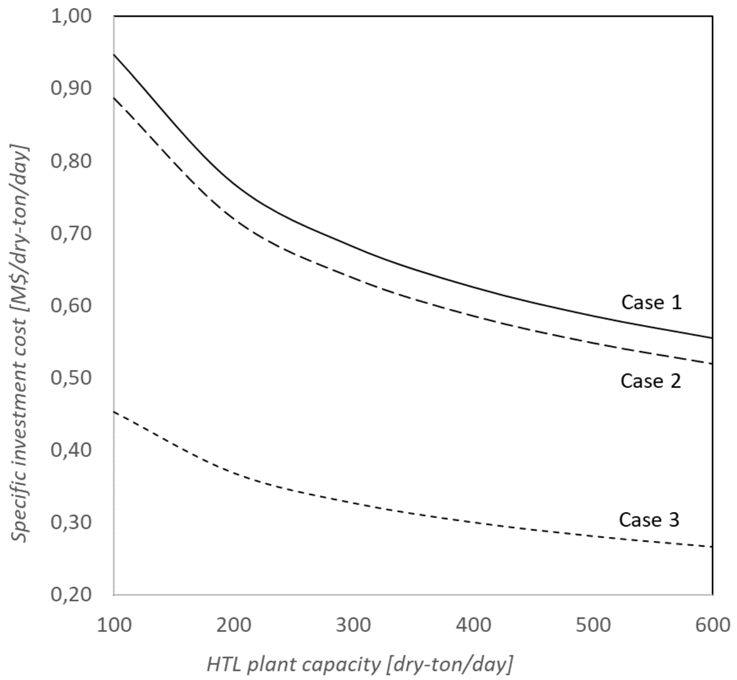

Figure 8: Variation of the specific installed equipment cost per unit mass flow rate of dry black liquor entering the HTL plant as a function of the black liquor feed capacity for the case 1 (solid line), case 2 (long dashed line) and case 3 (short-dashed line) scenarios for the HTL plant design.

Table 16: Overall project cost as a function of black liquor capacity for the biocrude production plant design case 1.

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Annual energy oil production (GJ)	223386	446772	670158	893545	1116931	1340317
Annual volume oil production (liter)	6758	13516	20274	27032	33790	40548
Annual mass oil production (kg)	7042	14085	21127	28170	35212	42255
Annual operating cost (M€/year)	29,53	54,97	79,70	104,04	128,16	152,11
Consumables and utilities	20,1	39,7	59,4	79,2	99,1	119,1
Electricity	17,9	35,3	52,8	70,4	88,0	105,8
Acid to phase separation	1,43	2,85	4,28	5,71	7,14	8,56
MEK (phase separation)	0,23	0,46	0,69	0,92	1,15	1,38
APR Catalyst	0,30	0,61	0,91	1,21	1,52	1,82
IHDO Catalyst	0,25	0,50	0,76	1,01	1,26	1,51
Fresh water	0	0	0	0	0	0
Labor	1,12	1,82	2,41	2,95	3,46	3,93
Maintenance	7,20	11,69	15,55	19,03	22,27	25,32
Insurance and taxes	0,36	0,58	0,78	0,95	1,11	1,27
Administration and Services	0,72	1,17	1,55	1,90	2,23	2,53
Annual Income (M$/year)	0,00	0,00	0,00	0,00	0,00	0,00
Total permanent investment (M$/year)	82,44	143,62	198,95	254,58	307,16	357,01
Equipment insllated cost	31,23	52,09	69,95	87,91	104,30	119,28
Chemicals (Initial batch)	20,13	39,71	59,40	79,20	99,09	119,06
Piping	2,03	3,39	4,55	5,71	6,78	7,75
Electrical system	1,56	2,60	3,50	4,40	5,21	5,96
Instrumentation & control system	1,41	2,34	3,15	3,96	4,69	5,37
Project Costs	26,08	43,49	58,41	73,40	87,09	99,60
Land	3,12	5,21	6,99	8,79	10,43	11,93
Site preparation	1,72	2,86	3,85	4,84	5,74	6,56
Foundation and buildings	6,25	10,42	13,99	17,58	20,86	23,86
Plant engineering	4,69	7,81	10,49	13,19	15,64	17,89
Contingency	6,25	10,42	13,99	17,58	20,86	23,86
Project development and licenses	0,94	1,56	2,10	2,64	3,13	3,58
Commissioning	3,12	5,21	6,99	8,79	10,43	11,93
Cost of biocrude (€/GJ)	**189,63**	**173,50**	**165,80**	**161,54**	**158,40**	**155,90**
Cost of biocrude (€/liter)	**5,77**	**5,28**	**5,05**	**4,92**	**4,82**	**4,75**

Table 17: Overall project cost as a function of black liquor capacity for the biocrude production plant design case 2.

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Annual energy oil production (GJ)	201047	402095	603142	804189	1005236	1206284
Annual volume oil production (liter)	6082	12164	18246	24328	30411	36493
Annual mass oil production (kg)	6338	12676	19015	25353	31691	38029
Annual operating cost (M€/year)	28,22	52,65	76,40	99,81	123,01	146,06
Consumables and utilities	19,4	38,4	57,4	76,6	95,8	115,1
Electricity	17,2	34,0	50,8	67,8	84,9	102,0
Acid to phase separation	1,43	2,85	4,28	5,71	7,14	8,56
MEK (phase separation)	0,23	0,46	0,69	0,92	1,15	1,38
APR Catalyst	0,30	0,61	0,91	1,21	1,52	1,82
IHDO Catalyst	0,23	0,45	0,68	0,91	1,13	1,36
Fresh water	0	0	0	0	0	0
Labor	1,05	1,70	2,26	2,77	3,24	3,68
Maintenance	6,74	10,95	14,55	17,81	20,84	23,69
Insurance and taxes	0,34	0,55	0,73	0,89	1,04	1,18
Administration and Services	0,67	1,09	1,46	1,78	2,08	2,37
Annual Income (M$/year)	**0,00**	**0,00**	**0,00**	**0,00**	**0,00**	**0,00**
Total permanent investment (M$/year)	**80,02**	**139,73**	**193,84**	**248,14**	**299,54**	**348,38**
Equipment insllated cost	30,38	50,81	68,39	86,01	102,13	116,91
Chemicals (Initial batch)	19,41	38,35	57,41	76,56	95,80	115,13
Piping	1,97	3,30	4,45	5,59	6,64	7,60
Electrical system	1,52	2,54	3,42	4,30	5,11	5,85
Instrumentation & control system	1,37	2,29	3,08	3,87	4,60	5,26
Project Costs	25,36	42,43	57,10	71,81	85,27	97,62
Land	3,04	5,08	6,84	8,60	10,21	11,69
Site preparation	1,67	2,79	3,76	4,73	5,62	6,43
Foundation and buildings	6,08	10,16	13,68	17,20	20,43	23,38
Plant engineering	4,56	7,62	10,26	12,90	15,32	17,54
Contingency	6,08	10,16	13,68	17,20	20,43	23,38
Project development and licenses	0,91	1,52	2,05	2,58	3,06	3,51
Commissioning	3,04	5,08	6,84	8,60	10,21	11,69
Cost of biocrude (€/GJ)	**202,18**	**185,35**	**177,31**	**172,86**	**169,57**	**166,97**
Cost of biocrude (€/liter)	**6,15**	**5,64**	**5,40**	**5,26**	**5,16**	**5,08**

Table 18: Overall project cost as a function of black liquor capacity for the biocrude production plant design case 3.

	Plant capacity [dry ton/day]					
	100	200	300	400	500	600
Annual energy oil production (GJ)	245639	491277	736916	982555	1228193	1473832
Annual volume oil production (liter)	7641	15281	22922	30563	38203	45844
Annual mass oil production (kg)	7962	15925	23887	31849	39812	47774
Annual operating cost (M€/year)	10,62	19,73	28,54	37,23	45,85	54,44
Consumables and utilities	6,1	12,4	18,8	25,3	31,9	38,5
Electricity	4,5	9,1	13,8	18,7	23,6	28,6
Acid to phase separation	1,43	2,85	4,28	5,71	7,14	8,56
MEK (phase separation)	0,23	0,46	0,69	0,92	1,15	1,38
APR Catalyst	-	-	-	-	-	-
IHDO Catalyst	-	-	-	-	-	-
Fresh water	-	-	-	-	-	-
Labor	0,53	0,87	1,16	1,42	1,66	1,89
Maintenance	3,44	5,61	7,47	9,14	10,71	12,18
Insurance and taxes	0,17	0,28	0,37	0,46	0,54	0,61
Administration and Services	0,34	0,56	0,75	0,91	1,07	1,22
Annual Income (M$/year)	**0,00**	**0,00**	**0,00**	**0,00**	**0,00**	**0,00**
Total permanent investment (M$/year)	**44,83**	**78,89**	**110,60**	**141,00**	**170,57**	**199,52**
Equipment insllated cost	19,40	33,32	46,01	58,00	69,52	80,69
Chemicals (Initial batch)	6,13	12,41	18,80	25,29	31,87	38,54
Piping	1,26	2,17	2,99	3,77	4,52	5,25
Electrical system	0,97	1,67	2,30	2,90	3,48	4,03
Instrumentation & control system	0,87	1,50	2,07	2,61	3,13	3,63
Project Costs	16,20	27,83	38,42	48,43	58,05	67,38
Land	1,94	3,33	4,60	5,80	6,95	8,07
Site preparation	1,07	1,83	2,53	3,19	3,82	4,44
Foundation and buildings	3,88	6,66	9,20	11,60	13,90	16,14
Plant engineering	2,91	5,00	6,90	8,70	10,43	12,10
Contingency	3,88	6,66	9,20	11,60	13,90	16,14
Project development and licenses	0,58	1,00	1,38	1,74	2,09	2,42
Commissioning	1,94	3,33	4,60	5,80	6,95	8,07
Cost of biocrude (€/GJ)	**70,48**	**64,22**	**61,29**	**59,50**	**58,28**	**57,39**
Cost of biocrude (€/liter)	**2,09**	**1,90**	**1,81**	**1,76**	**1,73**	**1,70**

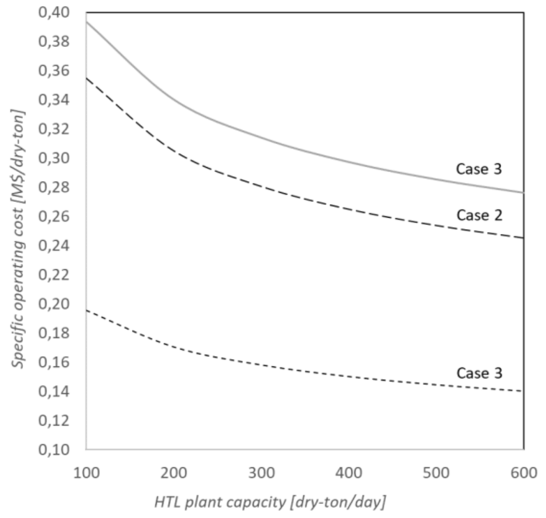

Figure 9: Variation of the specific installed equipment cost per unit mass flow rate of dry black liquor entering the HTL plant as a function of the black liquor feed capacity for the case 1 (solid line), case 2 (long dashed line) and case 3 (short-dashed line) scenarios for the HTL plant design.

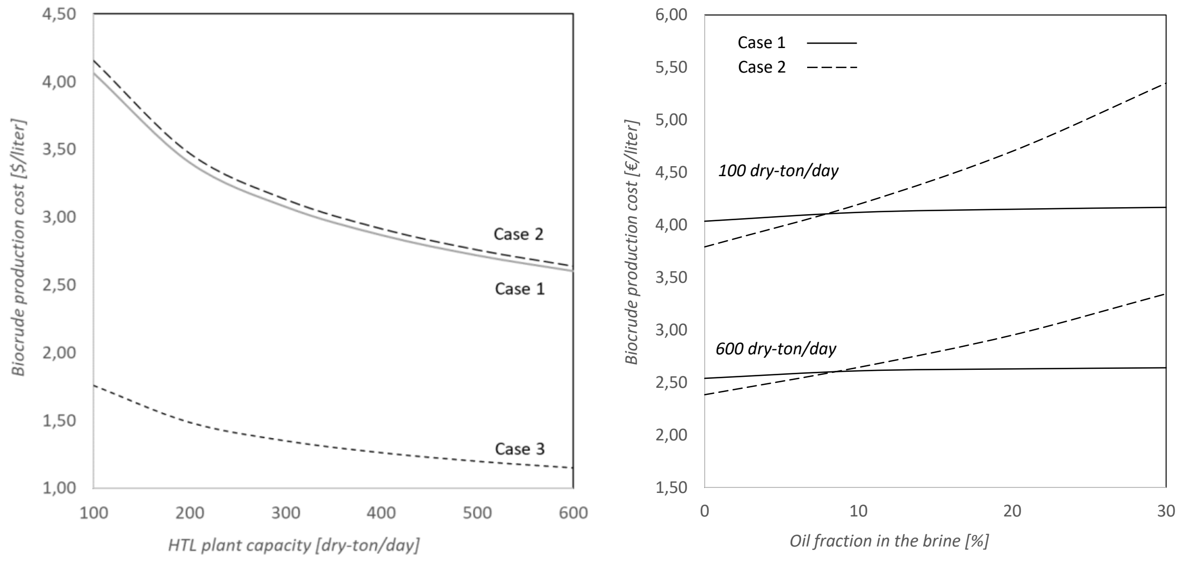

Figure 10: Variation of the specific installed equipment cost per unit mass flow rate of dry black liquor entering the HTL plant as a function of the black liquor feed capacity for the case 1 (solid line), case 2 (long dashed line) and case 3 (short-dashed line) scenarios for the HTL plant design.

Table 19: Process parameters for the overall biocrude upgrading process considered in the analysis.

Process parameter	Unit	Value
Storage capacity biocrude	h	148
Preheated biocrude temperature before pumping	deg. C	40
Hydrotreating temperature	deg. C	360
Hydrotreating pressure	bar	100
Hydrotreating hydrogen consumption	kg/kg feed	0,11
Hydrotreating hydrogen reacted	%wt feed	0,2
Hydrotreating catalyst WHSV	kg/kg/h	0,4
Hydrotreating catalyst lifetime	h	16000
Hydrotreater volume	m3	28
Hydrocracking temperature	deg. C	390
Hydrocracking pressure	bar	100
Hydrocracking hydrogen consumption	kg/kg feed	0,09
Hydrocracking hydrogen reacted	%wt feed	1,2
Hydrocracking catalyst WHSV	kg/kg/h	0,4
Hydrocracking catalyst lifetime	h	16000
Hydrocracker volume	m3	7
Temperature at three-phase high-pressure separator	deg. C	40
Pressure at three-phase high-pressure separator	bar	100
Temperature at three-phase low-pressure separator	deg. C	113
Pressure at three-phase low-pressure separator	bar	2
Inlet temperature at distillation column	deg. C	113
Pressure distillation column	bar	2
Inlet temperature at distillation column	deg. C	256
Pressure distillation column	bar	1,1
Naphtha cut-off temperature at distillation	deg. C	180
Kerosene cut-off temperature at distillation	deg. C	220

Table 20: Range of boiling point and reference high heating values of the distillation fractions considered in the analysis

Distillation fraction	Light hydrocarbons	Naphtha	Kerosene	Distillate residue
TBP (deg. C)	< 80	80-180	180-300	> 300
Carbon number	< C_5	$C_6 - C_{10}$	$C_{10} - C_{17}$	> C_{17}
Specific gravity	0.66	0.78	0.82	0.9
Carbon (% wt.)	82.43	84. 5	86.21	87.1
Hydrogen (% wt.)	16.1	14.2	13.5	12.9
Molecular weight (g/mol)	102	130	200	425
HHV (MJ/kg)	48.5	46.7	45.8	44.3

Table 21: Main mass flows calculated per unit mass (ton) of biocrude feed.

Light hyrocarbons	ton	0,071
Naphtha product	ton	0,242
Kerosene product	ton	0,544
Distillate residue from fractionation	ton	0,471
Process water	Liter	67,778
Sour gas from separation	kg	8,056
Total make-up H_2	kg	13,194
H_2 consumption in hydrotreating	kg	10,167
H_2 consumption in hydrocracking	kg	3,028
Light gas consumption	kg	46,056
Light gas consumption fired heater hydrotreating	kg	36,028
Light gas consumption fired heater hydrocracking	kg	10,028
Excess light gas	kg	24,5
Make-up amine consumption	kg	0,281

Table 22: Main energy flows per unit energy (MJ) biocrude feed.

Chemical energy light gases	MJ	0,091
Chemical energy naphtha	MJ	0,299
Chemical energy kerosene	MJ	0,244
Chemical energy diesel	MJ	0,416
Chemical energy H2 consumed	MJ	0,049
Heating biocrude before hydrotreating (FH1)	kJ	21
Heating reboiler distillation column 1	kJ	23
Heating reboiler distillation column 2	kJ	4
Heating organic liquid before hydrocracking (FH2)	kJ	6

Table 23**:** Electric loads (kW) as a function of the plant capacity for the biocrude upgrading process for feed capacity range of 100-2500 dry ton / day

	Plant capacity [dry ton/day]					
	100	500	1000	1500	2000	2500
Total Electric load (kW)	44,00	220,00	440,00	660,00	880,00	1100,01
Hydrotreating	**22,45**	**112,27**	**224,53**	**336,80**	**449,07**	**561,34**
Biocrude pump	15,92	79,58	159,17	238,75	318,33	397,92
Fuel gas compressor	0,49	2,43	4,86	7,29	9,72	12,15
Combustion air fan	2,38	11,92	23,84	35,76	47,68	59,61
Exhaust fan	3,67	18,33	36,66	55,00	73,33	91,66
Phase separation and fractionation	**0,67**	**3,35**	**6,70**	**10,05**	**13,40**	**16,75**
condensed naphtha reflux pump	0,34	1,68	3,35	5,03	6,70	8,38
condensed kerosene reflux pump	0,33	1,67	3,35	5,02	6,70	8,37
Hydrocracking	**11,18**	**55,91**	**111,82**	**167,73**	**223,64**	**279,56**
Heavy distillate pump	6,51	32,56	65,12	97,69	130,25	162,81
Fuel gas compressor	0,23	1,13	2,26	3,39	4,51	5,64
Combustion air fan	1,81	9,07	18,13	27,20	36,27	45,33
Exhaust compressor/fan	2,63	13,15	26,31	39,46	52,62	65,77
Gas treatment and hydrogen recycle	9,69	48,47	96,94	145,42	193,89	242,36
AMINE PACKAGE	5,53	27,64	55,27	82,91	110,55	138,19
H2-rich recycle compressor	2,51	12,53	25,05	37,58	50,11	62,63
Makeup H2 compressor	1,66	8,31	16,62	24,93	33,23	41,54

Table 24: Installed equipment costs (M€) as a function of the biocrude feed capacity

Biocrude capacity [dry ton/day]	100	500	1000	1500	2000	2500
Total equipment installed cost	15,896	36,834	61,848	86,812	110,718	135,547
Hydrotreating	3,481	10,952	20,158	29,481	38,047	46,902
Hydrotreater	2,064	8,794	17,127	25,557	33,451	41,420
Pump 1	0,718	0,792	0,872	0,960	1,084	1,112
Heat exchanger 1	0,211	0,346	0,582	0,910	0,948	1,381
Fired heater 1	0,489	1,020	1,577	2,054	2,564	2,990
Gas treatment	3,319	4,903	6,296	7,789	9,228	10,635
Heat exchanger 3	0,219	0,536	0,914	1,300	1,936	2,247
Compressor	1,834	2,178	2,391	2,807	3,051	3,261
Heater 2	0,105	0,208	0,320	0,419	0,482	0,605
Heater 1	0,093	0,157	0,210	0,239	0,257	0,304
Pump 2	0,184	0,351	0,464	0,599	0,676	0,814
Regenerator column tower	0,312	0,489	0,658	0,799	0,928	1,165
Reboiler	0,119	0,215	0,302	0,376	0,502	0,585
Condenser pump	0,033	0,043	0,057	0,059	0,060	0,068
Condenser drum	0,114	0,173	0,234	0,263	0,286	0,320
Condenser HE	0,098	0,206	0,253	0,314	0,348	0,420
Absorber column tower	0,210	0,348	0,495	0,616	0,701	0,847
Hydrocracking	4,879	13,173	24,313	35,624	45,729	56,955
Hydrocracker	3,404	11,285	22,181	33,255	43,037	53,720
Pump 3	0,951	0,962	0,972	1,022	1,041	1,066
Heat exchanger 3	0,232	0,372	0,412	0,436	0,652	1,013
Fired heater 2	0,292	0,554	0,749	0,912	0,999	1,156
Fractionation	4,217	7,807	11,081	13,917	17,713	21,056
Diesel column reboiler	0,121	0,183	0,229	0,255	0,306	0,342
Diesel column tower	0,618	0,651	0,847	0,952	1,079	1,306
Diesel column condenser	0,065	0,122	0,123	0,125	0,125	0,150
Diesel column drum	0,253	0,402	0,484	0,561	0,618	0,724
Diesel column pump	0,035	0,045	0,059	0,061	0,065	0,071
Naphtha column Reboiler	0,190	0,357	0,511	0,739	0,897	1,132
Naphtha column tower	0,899	1,729	2,353	3,138	3,881	4,874
Naphtha column condenser	0,079	0,103	0,126	0,157	0,164	0,171
Naphtha column pump	0,050	0,082	0,094	0,104	0,107	0,138
Naphtha column drum	0,191	0,307	0,421	0,485	0,527	0,610
HP separator	0,440	1,136	1,958	2,564	3,421	4,003
LP separator	0,248	0,430	0,499	0,579	0,686	0,730
Heat exchanger 2	0,094	0,107	0,136	0,145	0,151	0,190
Cooler 1	0,229	0,624	0,929	1,019	1,519	1,619
Cooler 2	0,115	0,123	0,150	0,153	0,153	0,156
Knockout drum	0,235	0,406	0,469	0,543	0,644	0,682
Oil tank	0,356	0,999	1,694	2,339	3,371	4,159

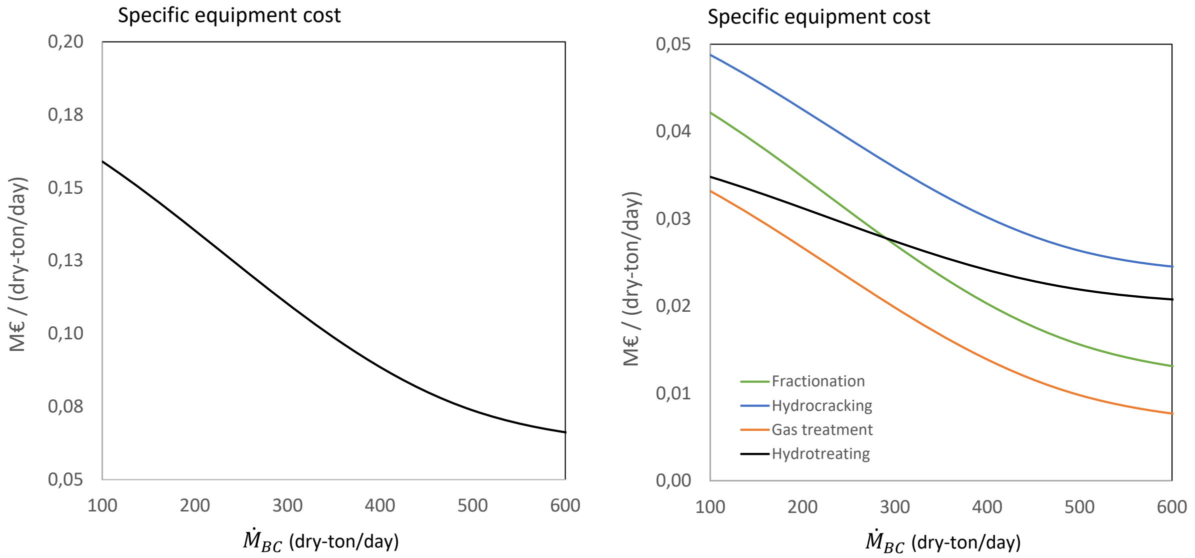

Figure 11: Variation of the specific installed equipment cost per unit mass flow rate of biocrude feed as a function of the biocrude upgrading capacity: (left) total; (right) distribution among main systems, i.e. hydrotreating, separation and fractionation, hydrocracking, and sour gas treatment with H2 recirculation

Table 25: Distribution of the annual operating cost (M€) for the upgrading of biocrude to naphtha and kerosene as a function of the biocrude feed capacity, for a specific biocrude cost of 1 €/Liter

Biocrude capacity [dry ton/day]	100	500	1000	1500	2000	2500
Naphtha capacity (MW)	21,79	108,97	217,94	326,91	435,88	544,85
Kerosene capacity (MW)	6,88	34,38	68,75	103,13	137,50	171,88
Total annual operating costs (M$/year)	**42,99**	**211,67**	**420,88**	**630,95**	**840,99**	**1051,07**
Biocrude	34,364	171,82	343,64	515,46	687,29	859,11
Consumables and utilities	7,526	37,628	75,267	112,887	150,489	188,111
Catalyst	0,599	2,997	5,995	8,992	11,989	14,987
Make-up hydrogen	1,045	5,224	10,448	15,673	20,897	26,121
Amine	0,028	0,138	0,277	0,415	0,554	0,692
Fresh water	0,135	0,673	1,347	2,02	2,693	3,366
Process water disposal	0,22	1,1	2,2	3,3	4,4	5,5
Electricity	4,977	24,885	49,77	74,655	99,54	124,425
Steam	0,522	2,611	5,23	7,832	10,416	13,02
Labour	0,31	0,38	0,47	0,56	0,64	0,73
Maintenance	0,32	0,74	1,24	1,74	2,21	2,71
Insurance and taxes	0,32	0,74	0,18	0,21	0,24	0,27
Administration and services	0,16	0,37	0,09	0,10	0,12	0,14
Total permanent investment (M$)	**69,67**	**238,53**	**446,45**	**653,28**	**858,80**	**1065,33**
Installed equipment	10,838	28,17	50,336	73,49	94,861	117,104
Piping	2,474	4,904	6,83	8,189	10,106	12,205
Electrical system	2,058	2,571	3,052	3,427	3,786	4,098
Instrumentation & control system	4,973	6,013	6,942	7,458	8,013	8,465
Insulation and painting	0,53	0,926	1,229	1,407	1,613	1,828
Land and site preparation	0,487	1,056	1,618	2,119	2,607	3,086
Foundation and buildings	2,269	3,232	3,99	4,547	5,283	5,962
Design, Engineering Procurement	4,385	6,058	7,57	8,696	9,762	10,79
Contingencies	5,041	9,729	15,121	20,36	25,415	30,621
Project development and licenses	1,627	2,936	4,415	5,836	7,214	8,619
Commissioning	34,99	172,94	345,34	517,75	690,14	862,55
Total Income (M$/year)	**1,375**	**6,873**	**13,746**	**20,619**	**27,491**	**34,364**
Minimum fuel selling price [€/GJ], naphtha	40,87	37,96	37,41	37,27	37,20	37,15
Minimum fuel selling price [€/GJ], kerosene	61,30	56,93	56,12	55,91	55,79	55,73
Minimum fuel selling price [€/liter], naphtha	1,49	1,38	1,36	1,36	1,35	1,35
Minimum fuel selling price [€/liter], kerosene	2,30	2,14	2,11	2,10	2,10	2,09

Table 26: Distribution of the annual operating cost (M€) for the upgrading of biocrude to naphtha and kerosene as a function of the biocrude feed capacity, for a specific biocrude cost of 6 €/Liter

Biocrude capacity [dry ton/day]	100	500	1000	1500	2000	2500
Naphtha capacity (MW)	21,79	108,97	217,94	326,91	435,88	544,85
Kerosene capacity (MW)	6,88	34,38	68,75	103,13	137,50	171,88
Distillate residue capacity (MW)	12,58	62,91	125,81	188,72	251,62	314,53
Total annual operating costs (M$/year)	**214,81**	**1070,78**	**2139,09**	**3208,28**	**4277,42**	**5346,60**
Biocrude	206,2	1030,9	2061,9	3092,8	4123,7	5154,6
Consumables and utilities	7,526	37,628	75,267	112,887	150,489	188,111
Catalyst	0,599	2,997	5,995	8,992	11,989	14,987
Make-up hydrogen	1,045	5,224	10,448	15,673	20,897	26,121
Amine	0,028	0,138	0,277	0,415	0,554	0,692
Fresh water	0,135	0,673	1,347	2,02	2,693	3,366
Process water disposal	0,22	1,1	2,2	3,3	4,4	5,5
Electricity	4,977	24,885	49,77	74,655	99,54	124,425
Steam	0,522	2,611	5,23	7,832	10,416	13,02
Labour	0,31	0,38	0,47	0,56	0,64	0,73
Maintenance	0,32	0,74	1,24	1,74	2,21	2,71
Insurance and taxes	0,32	0,74	0,18	0,21	0,24	0,27
Administration and services	0,16	0,37	0,09	0,10	0,12	0,14
Total permanent investment (M$)	**69,67**	**238,53**	**446,45**	**653,28**	**858,80**	**1065,33**
Installed equipment	10,838	28,17	50,336	73,49	94,861	117,104
Piping	2,474	4,904	6,83	8,189	10,106	12,205
Electrical system	2,058	2,571	3,052	3,427	3,786	4,098
Instrumentation & control system	4,973	6,013	6,942	7,458	8,013	8,465
Insulation and painting	0,53	0,926	1,229	1,407	1,613	1,828
Land and site preparation	0,487	1,056	1,618	2,119	2,607	3,086
Foundation and buildings	2,269	3,232	3,99	4,547	5,283	5,962
Design, Engineering Procurement	4,385	6,058	7,57	8,696	9,762	10,79
Contingencies	5,041	9,729	15,121	20,36	25,415	30,621
Project development and licenses	1,627	2,936	4,415	5,836	7,214	8,619
Commissioning	34,99	172,94	345,34	517,75	690,14	862,55
Total Income (M$/year)	**1,375**	**6,873**	**13,746**	**20,619**	**27,491**	**34,364**
Minimum fuel selling price [€/GJ], naphtha	193,70	190,79	190,24	190,10	190,03	189,98
Minimum fuel selling price [€/GJ], kerosene	290,55	286,18	285,36	285,15	285,04	284,98
Minimum fuel selling price [€/liter], naphtha	7,06	6,95	6,93	6,92	6,92	6,92
Minimum fuel selling price [€/liter], kerosene	10,91	10,75	10,72	10,71	10,70	10,70

5. Conclusions

This deliverable addresses the scale-up and costing of the complete technological pathway proposed in the BL2F project for production of liquid biofuels from black liquor diverted from Kraft pulp mills. The overall conversion from black liquor to biofuels includes a decentralized HTL plant integrated into the pulp mill, where the black liquor is converted by hydrothermal liquefaction to an oil-phase product or biocrude, and further upgrading of the HTL biocrude in a refinery.

The production of biocrude from black liquor includes several innovative processes: 1) integration in one reactor of the precipitation of salts followed by hydrothermal liquefaction (IHTL); 2) partial hydrodeoxygenation of the desalinated HTL oil phase in aqueous phase also under near critical-water conditions (IHDO); 3) aqueous phase reforming (APR) of dissolved organic components present in the HTL process water to produce hydrogen required by the IHDO process. These processes exhibit several risks identified during the experimental work in the BL2F project. In the IHTL process, the precipitation of the salts and the hydrothermal liquefaction of the organic fraction overlap within the temperature range investigated, and the salts brine and desalinated IHTL product form a quite stable solutions containing oil, solid and aqueous phases. Main challenges for the IHDO and the APR processes are catalyst deactivation and the clogging of the catalyst reactor bed, the later imposing stringent requirements for the feed to be free of solids. For the APR process, another challenge is the low hydrogen production due to either poor reforming rate or low hydrogen selectivity, to supply the local demand of hydrogen by the IHDO process. In view of the risks identified, the process design for the conversion of black liquor to biocrude has been updated and contemplates three different cases: 1) full extraction of oil from the salts brine and the desalinated IHTL product, further hydrodeoxygenation of the oil and local production of hydrogen by APR of process water from IHDO; 2) modification of case 1 only extracting the oil contained in the desalinated IHTL product; 3) discard from case 1 the hydrodeoxygenation of the oil and APR of the process water, the biocrude product being the oil extracted from IHTL. This case exhibits the lowest cost of biocrude, in the range of 2.1-1.7 €/Liter for black liquor feed capacities between 100 and 600 dry ton per day. Including hydrodeoxygenation of the total amount of IHTL oil with local production of hydrogen with full treatment of the process water in APR leads to a significant increase of the cost of biocrude, to 5.8-4.7 €/Liter, mainly due to the large increase in operating costs. Discarding the oil extracted from the salts brine leads to a further increase of the cost of biocrude, which ranges between 6.1 and 5.1 €/Liter when the oil content in the salts brine is 10% of the total.

The overall upgrading process considered naphtha and kerosene as the main products, the process design including hydrotreating of the biocrude for reduction of S and O heteroatoms, catalytic hydrocracking of the distillate residue separated during fractionation of the hydrotreating and hydrocracking organic liquid products to naphtha and kerosene, and treatment of the hydrogen-rich sour gas for separation of CO_2 and H_2S and recycling of the remaining hydrogen back to the catalytic processes. Assuming that the whole distillate residue from the kerosene column is further treated by hydrocracking, the total yields of naphtha and kerosene products, relative to the biocrude feed, are estimated to be 24.2% and 54.4% on dry mass basis and 29.9 and 66% on energy basis. Scale-up and cost analysis of the upgrading of biocrude considers a capacity range at refinery of 100-2500 ton of biocrude per day, equivalent to the maximum biocrude production rate from 5-25 reference pulp mills all with an annual bleached pulp capacity of 500 kt/year. Considering production of naphtha and kerosene only and assuming the kerosene has a market value 1.5 higher than naphtha on energy basis, the minimum fuel selling prices (MFSP) are in the range 10.9-2.1 €/Liter for kerosene and 7.1 and 1.3 €/Liter with biocrude feed capacities considered in the analysis, based on a specific biocrude cost varying between 1 and 6 €/Liter.

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