MODELLING OF COST OPTIMIZED HTL FUEL PRODUCTION BY PROCESS INTEGRATION

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Advanced biofuel production via hydrothermal liquefaction (HTL) can contribute to the mitigation of greenhouse gases from the transport sector. The HTL process is suitable for the conversion of a broad variety of organic feedstock including wet waste streams. The potential of transportation fuel production via sub-critical HTL of primary sewage sludge with integrated biocrude upgrading via hydrotreatment, and an energetic valorization of the aqueous phase via catalytic hydrothermal gasification is assessed. The thermal management of an integrated biofuel plant is optimized with respect to subsystem cost based on in-depth process modelling to quantify mass and energy flows and principles of pinch analysis to design a heat exchanger network. The cost optimized configuration has an installed heat exchanger surface of 1.32 m^2 related to 1 kg of processed sewage sludge and results in an overall process efficiency of 58.3 %.

Keywords: hydrothermal liquefaction, liquid biofuel, integration, modelling, sustainable aviation fuel

1 INTRODUCTION

HTL receives increasing attention in research and development as a highly versatile process that can convert a broad range of organic feedstock. The HTL process might represent a competitive alternative to other renewable fuel production processes, due to its promising ecological and economic performance [1].

In HTL, biomass is converted at temperatures of 300-420 °C and pressures of 150-350 bar into a highly viscous, dark oil, commonly referred to as biocrude [2,3]. This intermediate biocrude can be further upgraded to liquid hydrocarbon fuels via catalytic hydrotreatment [4]. Additionally the HTL process produces a solid, a gas phase containing mainly CO_2 and an aqueous phase consisting mainly of water and water-soluble components [5].

In comparison to other biofuel conversion technologies, HTL is particularly favorable for the conversion of biomasses that are readily available in aqueous slurries, e.g. sewage sludge, manure or microalgae, as their water content serves as reaction medium (solvent) and reactant.

The hydrothermal processing of waste materials represents a disposal process in addition to the fuel production path, which can contribute to a more cost-effective fuel production. Due to the fact, that the HTL process can also handle the disposal of waste materials, sewage sludge represents an example of an aqueous waste that is available at negative costs in many places [6] and thus is an attractive feedstock for HTL. However, the processing of sewage sludge involves some technical challenges that need to be solved. The composition of sewage sludge can vary significantly. When designing a HTL process, this must be taken into account, since the composition of the feedstock has a major impact on process performance parameters, e.g., yield and quality of biocrude as well as other product phases and energy efficiency.

When sewage sludge is used as HTL feedstock the biocrude usually contains a high share of mineral components and heteroatoms [7]. To comply with transportation fuel specifications an upgrading of the biocrude is mandatory.

Commercial implementation of a HTL process needs a preferably complete conversion of carbon-containing compounds into fuels. However, the aqueous phase contains up to 50 wt% of the carbon originally present in the HTL feedstock, with a total organic carbon value of 15 to 50 g/L [8]. Application of catalytic hydrothermal gasification (cHTG) represents a promising option to transform the organic content of the aqueous phase into a combustible gas. This can be used for on-site heat and power generation. In the case of sufficiently high product gas quantities, it can be considered to process the residual raw gas by means of gas scrubbing and sell methane on the market. Besides that, the combination of HTL and cHTG brings several additional benefits. cHTG simplifies subsequent aqueous phase treatment, as potentially toxic organic components are removed by conversion into useful products. Furthermore, methane from cHTG gas provides a cost-effective resource for producing H_2 via steam methane reforming. H_2 is required subsequently for biocrude upgrading. The herein considered HTL process chain is schematically depicted in Figure 1.

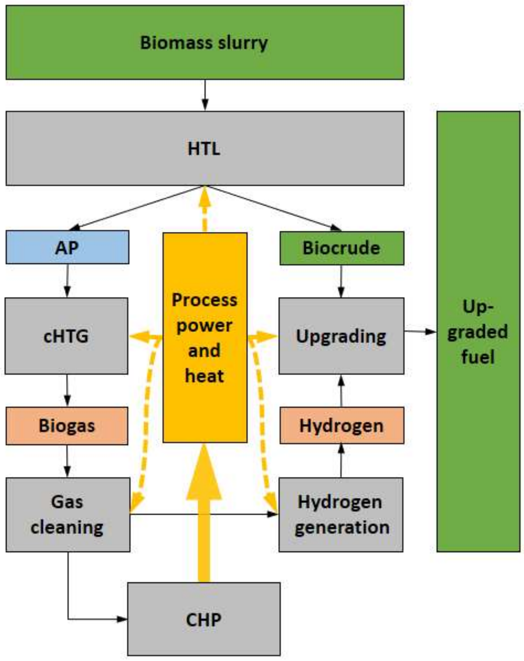

Figure 1: Process overview of the modelled and optimized HTL production chain (AP: aqueous phase; cHTG: catalytic hydrothermal liquefaction; CHP: combined power and heat plant).

In order to optimally dimension the individual process steps, modelling of the entire process chain is required. This issue is addressed by an in-depth modelling of all individual process steps using Aspen Plus. Along this process chain, the most important reaction pathways are identified and mapped in order to provide accurate information about product yields and compositions.

This study was carried out as part of the EU project HyFlexFuel [9]. The model is based on thermodynamic and reaction kinetic data, and validated with experimental data, which allows comprehensive statements on mass and energy balances as well as statements regarding the composition of different products. In order to optimally arrange and dimension heat exchangers, a cost estimate of the HTL process is performed. An optimization of the overall system in terms of costs is carried out and the expected savings in heat demand are assessed. It is shown that an effective heat recovery is key for cost-competitive and environmentally friendly fuel production. Energy demand can be significantly reduced with the use of a heat management system covering the entire process. This study addresses the research gap of the process integration of individual subprocesses in an entire HTL process chain and the associated challenges in thermal management.

2 MATERIALS AND METHODS

2.1 Feedstocks

Primary sewage sludge representing a wet waste stream is considered in this study as feedstock (FS). Sewage sludge is typically provided as slurry ready to be used in HTL (with possibly prior adjustment of dry matter/water ratio).

The modelling was carried out with a dry matter (DM) content of 20 wt%, which is slightly higher than the current values in the experimental implementation of the HyFlexFuel project [9]. The high moisture content still allows pumping, facilitates high yields and energy efficiency and largely enables avoidance of coke formation [5].

The chemical composition of sewage sludge is represented by five basic biochemical groups, namely lipids, carbohydrates, proteins, lignin and ash. Table I shows the share of these components.

Table I: Biochemical composition of sewage sludge.

	Sewage sludge
Lipids	3.9 wt%, dry
Carbohydrates	45.0 wt%, dry
Protein	23.3 wt%, dry
Lignin	7.3 wt%, dry
Ash	20.5 wt%, dry

In the simulation, 21 model compounds are generated representing the biochemical groups (carbohydrates, lignin, lipids and proteins) by forming typical hydrolysis products. Databases containing thermodynamic and reaction kinetic data are available for these compounds. Table I quantifies the assumed feedstock composition and Figure 2 shows the hydrolysis of exemplary model compounds from the four biochemical groups. Ash is not involved in the simulation as a reaction partner.

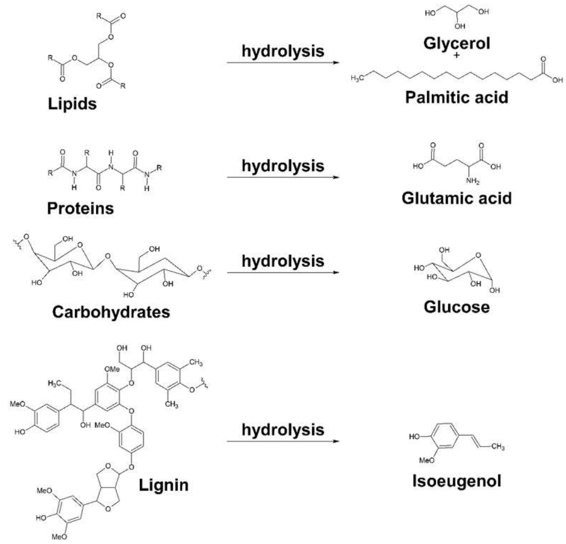

Figure 2: Hydrolysis of four biochemical groups for the modelling of the HTL of sewage sludge in Aspen Plus.

2.2 Process modelling

The modelling for the entire HTL production chain is performed using the simulation software Aspen Plus v10 [10]. The reactions of the HTL step, biocrude upgrading and cHTG processes are based on thermodynamic and kinetic data provided by Aspen Plus databases. Peng-Robinson equation of state with Boston-Matthias modification (PR-BM) was used as property method. Based on literature [11–14] , typical HTL reactions and biocrude compounds were identified and integrated in the Aspen model.

Since there is an equilibrium between gaseous CO_2 and dissolved CO_3^{2-}, which was not modelled, the assumption was made that 35% of CO_2 dissolves in the aqueous phase in the form of CO_3^{2-} [15]. The equilibrium of the reactions in the different RStoic reactors is adjusted according to analyses and experimental product yields. Table II gives an overview of the reactor models and the number of chemical compounds or reactions considered.

Table II: Considered model options for the different process steps using Aspen Plus (RStoic: Stoichiometric reactor model, RGibbs: Gibbs reactor model).

Process step	Reactor model	Number of defined reactions or compounds
HTL	RStoic	72 reactions
Upgrading	RStoic	92 reactions
cHTG	RStoic	128 reactions
CHP	RGibbs	21 compounds

Product yields are based on experimental results from the HyFlexFuel project. References are experimental measurements performed at the HTL pilot plant at Aarhus University [2], the gasification pilot plant at Paul Scherrer Institute [16] and the continuous biocrude upgrading unit at Aalborg University [4].

2.3 Process parameters

The integrated HTL plant has been designed as the size of a commercial one, which corresponds to a fuel production of 10 kt per year [17].
The feed slurry is heated in the HTL reactor to the reaction temperature of 350 °C at a pressure of 220 bar. This means that the HTL reactor is operated under sub-critical conditions. The gas resulting from the biomass conversion, consisting mainly of CO_2, is discharged, while the remaining product mixture is cooled to a temperature of 80 °C at ambient pressure and separated into biocrude, aqueous phase and a solid residue applying a gravimetric phase separation. The main product biocrude is subsequently upgraded and refined into marketable fuels and chemicals at 400 °C and 70 bar. This is done with a hydrogen surplus of 25 mol per kg biocrude [18]. Remaining hydrogen in the refinery gas is recycled and made usable again for upgrading.

The HTL aqueous phase is treated by cHTG under super-critical conditions at 450 °C and 280 bar. The biogas produced in the cHTG process mainly consists of CH_4 and CO_2, and can contain trace amounts of particulate matter or sulfidic and chloric compounds (e.g. H_2S, HCl). Therefore, a gas treatment process is included in the model. The purified biogas is used in a steam reforming process to produce the H_2 needed for the upgrading at temperatures of 500 °C and a pressure of 20 bar. An energetic use of the methane for the in-process heat demand is modelled. The exhaust gas (~800 °C) of the combustion is used for heat recovery.The energy demand of the pumps was modelled to be 12 W/L.

2.4 Modelling of heat exchangers

The required heat and cold demands are evaluated for the individual process steps using Aspen Plus. In order to achieve the best energetic solution for the arrangement of heat exchangers, process integration was performed using principles of pinch analysis in Aspen Energy Analyzer v10. For ΔT_{min} a value of 5 °C was assumed in the modelling.

Counter current shortcut recuperators are used as heat exchangers. The heat exchanger surface A is derived from the following correlation

$$\mathrm{Q} = \mathrm{UA}\Delta\mathrm{T}_{\mathrm{LM}},$$

whereat Q corresponds to the heat flow in the heat exchanger. The overall heat transfer coefficient U is assumed to be 15 W/(m^2·K) for all heat exchangers. The logarithmic mean temperature difference ΔT_{LM} represents the driving force for the heat exchange between process streams and is given by

$$\Delta\mathrm{T}_{\mathrm{LM}} = \frac{\Delta\mathrm{T}_1 - \Delta\mathrm{T}_2}{\ln\left(\frac{\Delta\mathrm{T}_1}{\Delta\mathrm{T}_2}\right)},$$

where ΔT1 corresponds to the temperature difference of the hot stream, $\Delta T2$ to the temperature difference of the cold stream. Two references for process optimization are taken into account. For comparison, "No HR" describes a hypothetical HTL process chain where no heat recovery (HR) is applied. For "sep HR" it is assumed that there is internal heat recovery in the individual sub-processes considered with a recovery rate of 75 %. "HEN" considers a fully integrated heat recovery.

2.5 Cost modelling

An optimization with respect to the lowest overall costs of an integrated HTL process is aimed for. Based on the computed mass and energy balances, the operating costs and the achievable profits of a HTL production are estimated. The assumed costs for feedstock and energy supply as well as the expected revenues from sales of the HTL products are shown in Table III.

Table III: Considered operating costs and revenues for fuel production by HTL.

	Cost/Revenue	Ref.
Sewage sludge	-160 €/t	[6]
Energy supply	0.077 €/kWh	[19]
Waste water treatment	1.77 €/m^3	[20]
Gas treatment	0.10 €/m^3	[21]
Naphtha	-797 €/m^3	[22]
Kerosene	-569 €/m^3	[22]
Diesel	-756 €/m^3	[22]

Negative values indicate that the position is associated with negative costs or revenues. Since HTL also represents a disposal process for sewage sludge, it is assumed that sewage sludge is available at negative costs. The disposal cost of 160 €/t serves as reference [6]. The costs of the heat exchangers (C_{HEX}) are estimated with 3000 €/m^2. To relate the investment costs of the heat exchangers to the fuel costs, the present value PV_{HEX} over n years is evaluated

$$\mathrm{PV}_{\mathrm{HEX}} = \mathrm{C}_{\mathrm{HEX}}(1+i)^n.$$

Variable i describes inflation, which is assumed 2 % per year. The lifetime n of the heat exchangers is assumed to be a period of 10 years. Economy of Scale is considered for the heat exchangers by using the following equation in relation to the quotient of new (Q_n) and reference capacity (Q_r)

$$\mathrm{PV}_{\mathrm{HEX,Scale}} = \mathrm{PV}_{\mathrm{HEX}}\left(\frac{\mathrm{Q_n}}{\mathrm{Q_r}}\right)^{\mathrm{m}}.$$

The correlation exponent m is derived from the cost and capacity of large and small heat exchangers.

$$\mathrm{m} = \frac{\log \mathrm{C_l} - \log \mathrm{C_s}}{\log \mathrm{Q_l} - \log \mathrm{Q_s}}$$

The costs for the different heat exchanger sizes are estimated according to Keshavarzian et al. [23].

It is assumed that further investment and operating costs are constant and are not considered in the cost optimization.

2.6 Assessment of process performance

Since biocrude is the primary target product in the HTL process, biocrude yield is often used as central metric to improve the plant economics. The biocrude yield is expressed as the ratio of the obtained biocrude to the feedstock input

$$\gamma_{\mathrm{bc}} = \frac{\mathrm{m_{biocrude,dry}}}{\mathrm{m_{FS,dry}}}.$$

However, the metric yield of biocrude neglects the quality of the yielded products. The product quality is reflected, e.g., in their specific energy, typically measured as higher heating value (HHV). The HHVs of the feedstocks and biocrude are calculated according to the equation proposed by Milne [24] based on elemental composition and ash content (elemental and ash fraction in %; HHV in MJ/kg).

$$\mathrm{HHV_{Milne}} = 0.341\,\mathrm{C} + 1.322\,\mathrm{H} - 0.12\,\mathrm{O} - 0.12\,\mathrm{N} + 0.0686\,\mathrm{S} - 0.0153\,\mathrm{ash}$$

Since the entire HTL process chain is considered, the fuel yield is determined using the following equation

$$\gamma_{\mathrm{fuel}} = \frac{m_{\mathrm{fuel,dry}}}{m_{\mathrm{FS,dry}}}.$$

The total efficiency of a HTL process is defined as the ratio of the energy output of the upgraded fuel and the sum of the energy content of the feedstock, the required electrical power P_{el} and the process heat H.

$$\eta_{\mathrm{total}} = \frac{\mathrm{HHV_{ubc}} \cdot m_{\mathrm{ubc}}}{\mathrm{HHV_{FS}} \cdot m_{\mathrm{FS}} + P_{\mathrm{el}} + H}$$

3 RESULTS

3.1 Elemental compositions

The elemental analysis of the product streams are used to validate the model of the HTL process chain. The comparison of the calculated elemental composition of feedstock, biocrude and upgraded biocrude with the respective elemental analyses from literature values is shown in Figure 3.

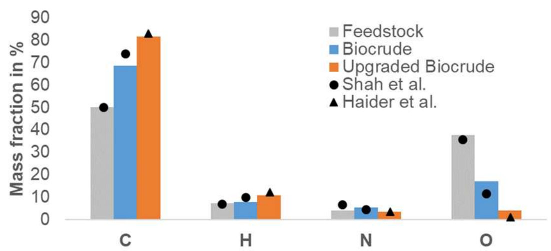

Figure 3: Composition of feedstock (grey), biocrude (blue) and upgraded biocrude (orange) using sewage sludge HTL as feedstock [4,12].

Keeping in mind that literature values can differ quite significantly, the calculated model values of the elemental analysis are in reasonable agreement. According to the experimental results, the modelling clearly shows that an energetic upgrading of sewage sludge takes place. The relative carbon content can be increased by 47.5 % during biocrude production, while an additional upgrading process leads to an increase of 71.2 %.

The portion of heteroatoms could be reduced significantly during HTL processing and the subsequent upgrading process. The modeled upgraded biocrude shows nitrogen contents of 3.5 wt% and oxygen contents of 4.0 wt%. The sulfur content in the upgraded biocrude was modeled at 0.094 wt%.

In order to estimate which amounts of final products can be expected from a distillation of the upgraded biocrude, the individual components of the upgraded biocrude were grouped according to their boiling points. Plotting the cumulated masses of the individual components of the modeled upgraded biocrude with respect to their boiling points leads to a simulated distillation curve, which is shown in Figure 4. The calculated boiling point distribution is compared with experimental results obtained from literature [4].

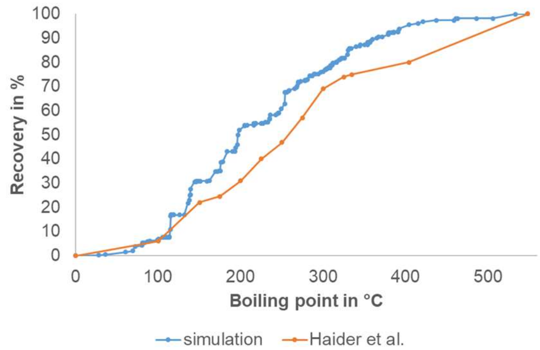

Figure 4: Comparison of simulated distillation curves from upgraded biocrude of sewage sludge (blue: model, orange: simulated distillation of upgraded biocrude experimentally produced from sewage sludge) [4].

As can be seen in Figure 4, there are two model components that do occur in striking amounts. The first component is ethylbenzene, which has a boiling point of 136.0 °C and is present in the upgraded biocrude with an amount of 13.7 wt%. The second component is 3-ethylindole with a boiling point of 270.0 °C and a weight fraction of 11.4 wt%. Ethylbenzene is derived from the amino acid phenylalanine by decarboxylation and deamination, while 3-ethylindole is the product of decarboxylation and deamination of the amino acid tryptophan. Due to the limited number of amino acids considered in the feedstock and the narrow range of reactions for amino acids, these high values are observed. Although the upgraded biocrude is modeled with 266 model components, this finding shows, that in order to achieve an even smoother distributed model, more initial feedstock components and a greater variety of reaction pathways have to be considered.

Nevertheless, the amounts of obtained product fractions are in an acceptable range compared to the experimental results, since simulated distillation curves can vary significantly when applying different conditions.

The boiling point ranges of the product groups naphtha (32-150 °C), kerosene (150-250 °C) and diesel (250-350 °C) serve to determine the fuel product quantities that can be processed from upgraded biocrude. Thus, 36.2 wt% naphtha, 18.2 wt% kerosene and 33.2 wt% diesel can be produced from the upgraded biocrude.

3.2 Mass balances

Based on the modeling, a mass balance is calculated covering the entire HTL process chain for the application of sewage sludge. The outputs of the respective process steps are listed in Table IV, compared to the basic plant layout in Figure 1.

Table IV: Mass balance for products and intermediate products of an integrated HTL process chain based on an HTL input of 1 kg sewage sludge feedstock (dry) and 4 kg water.

Product	Mass fraction
HTL	
Biocrude	0.342 kg/kg_{FS}
Solids	0.144 kg/kg_{FS},
Waste gas	0.109 kg/kg_{FS}
Aqueous phase	4.407 kg/kg_{FS}
Hydrotreating	
HT gas phase	0.072 kg/kg_{FS}
HT wastewater	0.047 kg/kg_{FS}
Upgraded biocrude	0.240 kg/kg_{FS}
cHTG	
cHTG wastewater	4.170 kg/kg_{FS}
cHTG gas	0.237 kg/kg_{FS}
Gas cleaning	
Methane	0.099 kg/kg_{FS}
Waste gas	0.138 kg/kg_{FS}
Steam reforming	
Hyrdogen	0.012 kg/kg_{FS}
Reforming gas	0.091 kg/kg_{FS}
CHP	
Waste gas	0.693 kg/kg_{FS}

As modeling input for the HTL, sewage sludge with a DM content of 20 wt% was considered. Regarding the HTL mass balance it becomes clear that large amounts of aqueous phase are produced in addition to the desired biocrude. The model shows that 33.8 wt% of the introduced carbon ends up in the aqueous phase. This underlines the high relevance of the treatment of the aqueous phase in a HTL process chain. cHTG has proved to be a suitable process for the processing of the aqueous HTL phase in the modeling. 68.7 wt% of the carbon can be made usable again after upconcentration by a membrane and removal of the sulphur-containing components by a sulphur trap. In this way, treatment of the aqueous phase is also facilitated, since a large part of the salts is concentrated and the organic content in the wastewater is considerably reduced. The product of the cHTG is a methane-containing lean gas with a calorific value of 29.1 MJ/kg. The cHTG gas is similar in composition to biogas and is suitable in combination with gas purification for H_2 production as well as for energetic use.

The estimations suggest that it is sufficient to use 33 % of the purified biogas to cover the hydrogen demand in upgrading. The remaining biogas is used in a CHP unit to cover the electricity and heat demand for the HTL process chain.

3.3 Energy balances

The energy balance is derived from the mass flows of the overall HTL process chain, enthalpy flows associated with the heating and cooling demand as well as specific energy requirements of pumps, compressors and turbines. Heat recovery options are not taken into account when establishing the energy balance. Rather, the energy balance is compiled in order to show where integrated heat management is appropriate. The results of the energy balance are shown in Figure 5 in form of a Sankey diagram.

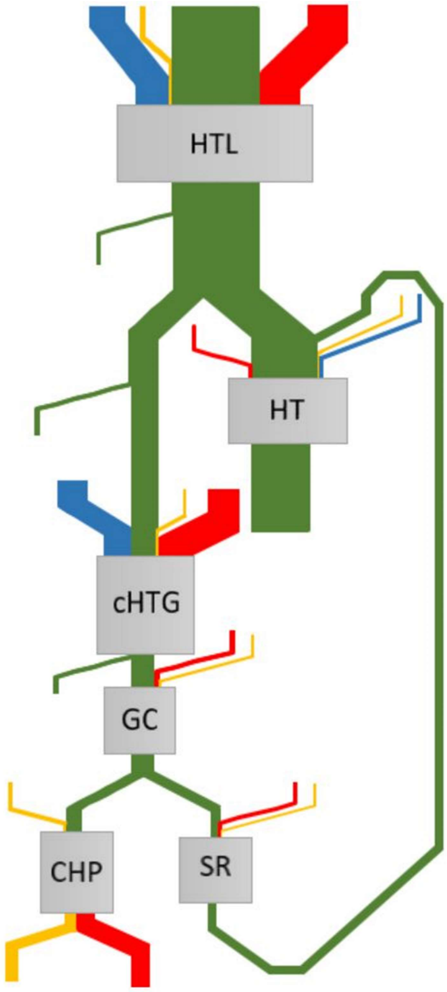

Figure 5: Energy balance for an HTL process chain using sewage sludge as HTL feed. Green: Energy in streams, blue: cooling demand, red: heating demand, yellow: electrical power (HTL: hydrothermal liquefaction; HT: hydrotreating; cHTG: catalytic hydrothermal liquefaction; GC: gas cleaning; CHP: combined power and heat plant; SR: steam reforming).

The figure illustrates the energy level of the streams and the type of energy (heating, cooling, electrical) required for the various sub-processes. Sewage sludge represents the HTL input biomass. For the HTL process significant quantities of heat (shown in red) are required. For the subsequent phase separation, which takes place at low temperatures, high amounts of cooling energy are needed. The demand for electrical energy is mainly due to the pumping. A minor part of the carbon is lost in the solid phase, which is not used energetically.

Considerable quantities of heating and cooling energy are also required for the cHTG as large quantities of aqueous phase have to be heated and gas as well as remaining water has to be separated. The electrical energy demand arises mainly from gas cleaning and concentration.

In a CHP process, electrical energy is needed to run pumps, but mainly to compress air for an effective combustion process. The electrical energy produced by CHP is far greater.

The hydrogen for biocrude upgrading is produced by a steam reforming process. The upgrading process requires more cooling than heating energy, as the deoxygenation during hydrotreatment is a exothermic process [25]. The hydrogenation reaction thus releases heat and less additional heat needs to be provided to maintain the process conditions.

The electrical energy generated by gas combustion is sufficient to cover the demand of an HTL process chain. Therefore, no additional electric energy source is needed in an integrated HTL process chain. The computed heat and cold flows in the energy balance form the basis for the implementation of an integrated heat management, which covers the entire HTL process chain and offers potential to increase process efficiency.

3.4 Heat exchangers

To increase process efficiency, power and heat demands are minimized. The energy balance has shown that the entire electric energy demand of an HTL process chain can be covered by the energetic use of the cHTG gas. In order to minimize the heat demand, a HEN is developed comprising the entire HTL process chain. A list and a ranking regarding the heating or cooling duty of the cold and hot process streams serves as a basis for an optimal configuration and design of heat exchangers according to the pinch principles. In order to keep the process as simple as possible and to optimize costs with regard to the required heat exchanger surface area, only those streams were considered that have an enthalpy greater than 1000 kJ/kg_{FS}. This results in the list of hot and cold flows considered in Table V.

Table V: Input for the pinch analysis.

Name	Inlet T in °C	Outlet T in °C	Enthalpy in $kJ/kg_{FS, dry}$
HTL slurry in	25	350	7044
HTL slurry out	350	80	5852
cHTG in	80	450	5896
cHTG out	450	80	5896
CHP out	810	25	4999

By identifying the pinch temperature and starting to design the heat transfer network around this point, the energy targets can be met by transferring heat between hot and cold flows. The arrangement of the heat exchangers and the required heat exchange surface is shown in Table VI.

Table VI: Configuration of the modeled heat exchangers.

Stream in, hot	Stream in, cold	ΔT_{LM} in °C	Area in m^2/kg_{FS}
cHTG out	HTL slurry in	318	0.34
HTL slurry out	cHTG in	316	0.34
CHP out	HTL slurry in	151	0.20

The two heat exchangers comprising the cHTG and the HTL are of equal size with 0.34 m^2/kg_{FS}. The heat exchanger used to preheat the HTL slurry is slightly smaller with 0.20 m^2/kg_{FS}. As cumulated heat exchange surface, 0.88 m^2 per kg feedstock (dry) is computed.

3.5 Cost optimization

The calculated heat exchange surface on the one hand and the costs and sales revenues of the modeled inputs and outputs on the other hand serve as a basis for cost optimization and an adjustment of the heat exchanger surface. The costs and sales revenues are referred to as OPEX. With an increase of the heat exchanger surface the costs for the heat demand decrease. CH_4, which is not energetically converted for in-process use, can be sold and provides an additional revenue in the case of fuel production via HTL. On the other hand, an increase in the heat exchange surface results in an increase in the investment costs (CAPEX) of a HTL plant. In an optimization approach the most cost effective heat exchange surface is calculated. Since the economy of scale is considered in the calculation of the heat exchange surfaces, a HTL process chain on an industrial scale that allows to produce 10 kt biofuel per year is assumed. The cost functions of heat exchange surfaces (CAPEX HEX) as well as the relative prices of the raw materials used and the relative revenues for the products (OPEX) are shown in Figure 6.

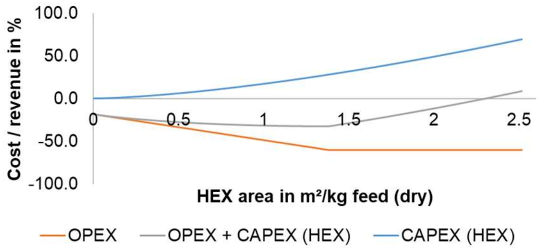

Figure 6: Cost optimization for processing sewage sludge via HTL related to a feedstock input of 1 kg sewage sludge (dry).

If the costs for feedstock, products and investment costs are summarized, an optimum heat exchange surface of 1.32 m^2 per kg sewage sludge results.

3.6 Process efficiency

The optimized heat exchanger surface in the model is used to evaluate the process efficiency. Figure 7 summarizes the computed efficiencies for different heat recovery options and HTL process configurations.

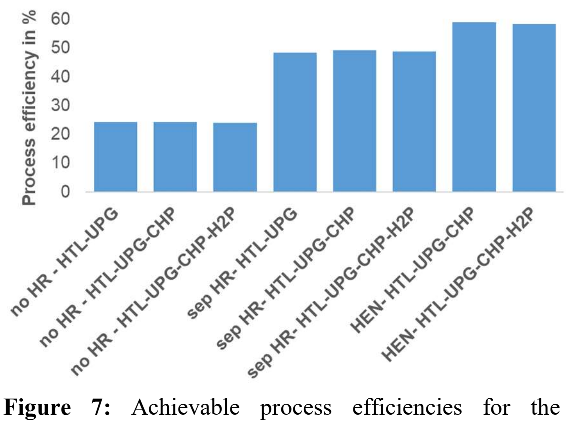

Figure 7: Achievable process efficiencies for the configurations considered: no heat recovery (no HR), individual heat recovery in each process step (sep HR), heat exchanger network (HEN).

The first considered heat exchange configuration is the no heat recovery option (NoHR). This means that the process streams are spatially connected between the individual process steps, but there is no heat exchange between hot and cold streams.

The second heat recovery configuration describes heat exchangers that only comprise an individual process step. For example the heat exchanger of the HTL pilot plant at

Aarhus university performs with a heat recovery of 80 % [2]. For this consideration, it is assumed that in each high-temperature process step, individual heat recovery is performed with a heat recovery efficiency of 75 %.

The heat exchange network, which includes an energetic optimization of the heat exchanger assembly and a cost optimization of the heat exchanger surface, forms the third system configuration considered.

In addition, the process efficiency was determined for the following three process configurations: Firstly, for a HTL process coupled with an upgrading, secondly this process with additionally an energetic use of the aqueous phase by cHTG and CHP and finally a process in which a part of the methane produced is used for the provision of internally required hydrogen.

From the data it is clear that the efficiency with separate heat recovery for all process configurations is significantly higher than with no heat recovery. The efficiency can be further improved by using an integrated heat exchanger network. For the first process configuration the heat exchanger network is not applicable, because in the HTL process no additional waste heat can be used for upgrading.

The overall best process efficiency with a value of 58.8 % can be achieved with a process configuration in which an energetic use of the aqueous phase is carried out and hydrogen is added externally.

For the case of an in-process hydrogen supply and an application of a heat exchanger network, a process efficiency of 58.3 % can be calculated.

Since this configuration can be favourable from an economic point of view and the efficiency is only slightly lower, this a promising option for an HTL process chain.

4 DISCUSSION

The field of HTL process models can be differentiated by several characteristics, two of them being the use of a reaction network and the consideration of a process chain connected to the HTL process. Previous studies of the HTL process including a reaction network are mainly based on HTL batch experiments. Most of these studies use a small number of model compounds and do not consider a process chain connected to the HTL process [26]. The most prominent system analyses of continuous HTL processes and process chains were performed by the Pacific Northwest National Laboratory [27]. However, no reaction network is considered and product yields and compositions are specified according to experimental results.

The herein described HTL process model combines the two characteristics of modeling a reaction network as well as a HTL process chain.

Intensive heat recovery can significantly increase the efficiency of a fully integrated HTL process. However, some aspects have to be taken into account when implementing integrated heat recovery into HTL that are not examined in this study.

Since high temperatures (exceeding 800 °C in turbine flue gas) occur in the process, care must be taken to select suitable materials when implementing a system. However, no negative material effects are to be expected when using suitable heat resistant steel on affected system parts.

A further factor that has to be considered in the technical implementation of a HEN is the possibility of precipitation of mineral components when the product streams are cooled. At low temperatures salts in the pipes or in the heat exchangers can precipitate, leading to an increased corrosion risk as well as a reduced heat exchange efficiency. In addition, when dimensioning a HEN, limitations due to the specific HTL plant need be taken into account. Heat exchangers may not be installed at every point in the system in the desired dimension. However, this can only be assessed when designing and dimensioning a plant in detail.

5 CONCLUSIONS

In this study a HTL process including a valorization of the HTL products to marketable fuels is modeled. A numerical system model for a HTL production chain including biocrude upgrading and energetic valorization of the aqueous phase via cHTG is introduced. Based on the modeled energy and mass flows a heat exchanger network is designed to thermally integrate the individual process steps and to optimize the utilization of combustible gases that evolve from the process chain for hydrogen, heat and power generation. The process efficiency was determined for a cost-optimized heat recovery system and two reference scenarios.

The results indicate that the energetic valorization of the organic content in the aqueous phase is important for HTL plants, and point to the benefits of close thermal integration in particular for the HTL and cHTG subsystems. Hydrogen generation from cHTG gas for biocrude upgrading via hydrotreatment is another potential benefit from plant integration. However, the potential for thermal integration of the upgrading step seems to be limited. Further investigations are required to weigh the benefits of on-site integration of upgrading against the advantages of centralized biocrude upgrading facilities with improved economy of scale or co-processing of HTL biocrudes in refineries.

The modelling lays the basis of upcoming techno-economic and environmental system analyses of integrated HTL plants for transportation fuel production from sewage sludge and various other organic feedstock.

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8 LOGO SPACE