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Techno-economic assessment of the by-products contribution from non-catalytic hydrothermal liquefaction of lignocellulose residues

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ABSTRACT

Hydrothermal liquefaction of biomass continues to show promise in experimental and pilot scale tests for carbon partitioning toward desirable multi-phase organic products. Results from a techno-economic investigation are presented for a commercial scale stand-alone plant with primary production of renewable liquid fuels compatible with current transportation infrastructure. The plant feedstock was forest residues and the non-catalytic hydrothermal conditions were set to 330 °C and 210 bar. A sequential flowsheet was developed and simulated on Aspen Plus[®] that includes pre-treatment, hydrothermal liquefaction, fuel upgrading and residue recovery functional blocks. Different scenarios for the valorization of the liquefaction residue streams are examined to maximize organic recovery and eliminate process waste streams. The highest plant thermal efficiency on lower heating value basis was recorded for the polygeneration of renewable liquid fuels, Bio-char and hydrogen gas at 85.2%. The plant recorded a minimum selling price of 66 € per MWh of co-products. The break-even prices of the co-products under existing market conditions was found to be 1.03 € per kg of gasoline or 2.46 € per kg of hydrogen gas or 51.4 € per MWh of Bio-char.

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1. Introduction

Hydrothermal treatments (HTT) are considered promising thermochemical processes that have continued to establish advantageous nature for the conversion of biomass to a wide range of chemicals and fuels in lab and pilot scale processing [1–9]. The term hydrothermal, is often referred to aqueous processing of organics at elevated temperature profiles, beyond that of vapor pressure conditions [7]. Water solvency properties at such acute conditions vary significantly, and allow for diverse sets of degradation mechanisms for organics into solid, liquid and gaseous products. Temperature remains the governing parameter that defines product nature [2,3,7,8]; Hydrothermal Carbonization (HTC) between 200 and 270 °C yields a carbon rich solid char. Hydrothermal liquefaction (HTL) between 250 and 400 °C produces mostly long chain oxygenated hydrocarbons as part of a two-phase liquid product of organic compounds, soluble and insoluble in water. Finally, Supercritical Water Gasification (SCWG), referred to in some literature as Hydrothermal Gasification, at beyond 400 °C

favors complete degradation into synthetic gas [8]. Peterson et al. [2] in their review explained the processing opportunity of HTT that 'by moving from subcritical to supercritical temperatures at pressure above the critical, we can control both the rate of hydrolysis as well as phase partitioning and solubility of components so that more chemically and energetically favorable pathways to gaseous and liquid biofuels may be released.'

Through fine-tuned and novel bio-refinery process design, lignocellulose conversion under hydrothermal conditions could be devised to favor the production of liquid biofuels that could substitute for fossil-based counterparts [1,3,4,9]. Some earlier, and still ongoing, research has been dedicated to biomass indirect liquefaction pathways, the so called syngas to distillate synthesis [10]. However, significant efforts in the last two decades has dedicated resources to the investigation of the direct liquefaction (initial terminology dated back to the 1970s) or the HTL process. Process intensification is the main motivation behind that and is achieved by reducing processing steps through the utilization of the unique thermo-physical properties of the moisture substrate (or auxiliary water) in wet feedstock. The water content in the feed slurry acts as a multi-purposeful process medium, 'as a solvent, reactant and catalyst [7]'. Hydrothermal processes in general enable eliminating the processing penalty from the required drying loads in other

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thermochemical conversion pathways, such as pyrolysis or gasification. In the case of subcritical water processing conditions, both water's density and viscosity significantly drop, with the latter by more than 85% when compared to ambient conditions [6]. In addition, approaching the critical point of water, the specific heat capacity peaks at a theoretical infinity due to the absence of phase change. This significantly reduces mass and heat transfer limitations and allows for relatively more homogenous processing [8]. On the other hand, the ionic product of water increases by 2 orders of magnitude near to the critical point compared to ambient conditions [11,12]. Thus, chemical equilibrium, driven toward water dissociation/ionization, enables and catalyzes several reactions taking part in the organic hydrolysis process, including that of the highly stable lignocellulose structure [3,4,11,12].

In general, subcritical water processing aims to operate within temperature boundaries that allow the fragmentation and degradation of organic solids through both pyrolytic-like mechanisms and condensation pathways to a hydrophobic Biocrude product, while maintaining liquid phase [1]. The crystalline nature of cellulose monomer bonds, the largest component in wood-based feedstock, resists the traditional hydrolytic degradation route with acids and enzymes [13]. However, under hydrothermal conditions due to subcritical water's affinity to break linkage hydrogen bonding, a degraded and hydrolyzed set of product compounds, namely alcohols, furfurals, acids, cyclic ketones, methoxy-phenols and more condensed aromatic structures were reported experimentally [3]. The calorific value of the Biocrude product, characterized by lower heteroatom (O,N and S) concentration compared to the original biomass feedstock, could reach as high as 36 MJ/kg [1,5,14]. The HTL multi-phase product includes char and ash solid residue, water soluble organics, gas (over 90 wt% CO₂) and finally, Biocrude with reported yields ranging between 10 and 47% on mass basis [1,3–5]. The deoxygenated hydrolysate Biocrude, in comparison to pyrolysis oil, is a more viscous hydrophobic phase with less dissolved water, yet less dense energetically [1].

Akhtar et al. [3] and other recommended literature [4–6,9] reviewed the influence of the HTL reactor configuration on both Biocrude quality and yield. The consensus in most literature is that the reactive liquefaction temperature, residence times, rate of biomass heating, size of biomass particles and the recycle of aqueous residue streams have the largest influence, although with varying impact [1–3,5,7,9,15]. As of today several continuous and proof-of concept pilot systems are in place independently around the globe [1,16,17], and also, a commercial solution by Steeper Energy, ApS titled Hydrofaction™ Oil is reportedly offered [18]. It is worth to mention that the directly produced Biocrude is not considered an analogue of petroleum crude [19–21]. The different compositional structure (presence of fragmented and oxygenated hydrocarbons) and thermo-physical features (viscosity, stability etc.) deem the need for further upgrading to match current common fuel properties [22]. There have been intensive studies in literature on Biocrude upgrading and property enhancement, and various technologies have been either adapted from the petroleum/chemical refining industry or developed specifically for biomass-derived 'oils' for a wide range of applications [19,21,23,24].

HTL as a technology remains branded with "development potential" [25], as such efforts have been directed to the parameterization of the process [26]. However, the conclusion reached by Arturi et al. [26] after examining 34 peer reviewed experimental articles was that data in literature remains distorted and case specific studies would be needed for upscaling the HTL process. In recent years, the research scrutiny has been expanded to examine the technical and commercial opportunity for a complete plant layout from several feedstock; wood [27–29], microalgae [30–32], food waste [33], dairy effluent [34] and manure digestate [35]. In

the absence of technology standardization, different products or process configurations have also been taken into consideration; either a cut-off at the Biocrude production point [29,30,33,36,37] or further upgrading to match commercial fuel properties [27,28,31,32,34,35]. From a process design perspective, the main challenge for HTL remains energy recovery and carbon "losses" in residue streams. At desirable higher organic to water feeding conditions, more than 50% of carbon ends in the byproduct, either in the aqueous or solid phase [2,6,7,9]. Different solutions proposed in literature presented their own trade-offs and set of unique operational challenges. Catalytic processing reduces solid residues significantly, while, in some cases increases the aqueous and gas residues, as well as presenting challenges concerning catalyst deactivation and recovery [2,4]. In fact, reported catalytic activity could be offset by the fine tuning of the HTL reactor design through increasing the rate of heating followed by product flash quenching, which limits the organic substrates tendency to re-polymerize [17]. Complete or partial recycling of the aqueous solution is another proposed solution in literature to reduce auxiliary water and to allow miscible organics to act as a co-solvent [38]. However, the elimination of waste water treatment facility came at the expense of Biocrude quality, with reports of reactor clogging due to increased char formation and build-up of heavy organic compounds were reported in literature [39]. The objective of this article is to present and quantify the contribution of different HTL byproduct utilization scenarios through a techno-economic assessment (TEA) for a stand-alone non-catalytic plant. The primary biofuel products are renewable liquids, gasoline and diesel, compatible with existing transportation fuels infrastructure. The valorization of the solid residue to Bio-char and residue gas and aqueous streams to hydrogen gas co-products are investigated and compared to a stand-alone primary products pathway and reported catalytic HTL processing pathways in literature.

2. Materials and methods

The HTL case studies investigated are for renewable diesel and gasoline production in Finland from local lignocellulose residues. At first, based on reported experimental data for the HTL of a wide range of Northern hemisphere forest residues, a predictive model for the Biocrude chemical composition establishes the specific yields and closes the mass balance. The model findings offer an accurate representation of the thermo-physical properties and functional groups of the Biocrude, that later enable reliable modelling and simulation of a complete conceptual plant layout on Aspen Plus® commercial software [40]. To the author's knowledge, other similar system level modelling in literature have been mostly based on model compounds to represent the Biocrude [29,31,35–37] or simply rely on overall product phase yields [30,32–34] or a combination of both [28]. The different case study scenarios include a common Biocrude to renewable liquid fuels processing train, while differ concerning the byproduct streams. The solid residues, largely ignored in literature by the optimistic assumption of elimination by catalytic activity or as a non-value product, is sold as Bio-char for soil management and carbon sequestration purposes [41–43] or combusted onsite. The gas residue stream is utilized for onsite hydrogen generation to serve the Biocrude upgrading purpose, similar configurations are reported in other HTL plant designs [28,34,35]. The aqueous stream is treated in a wastewater treatment facility and recycled back to the plant entry point or introduced to a SCWG unit for hydrogen production as an additional plant biofuel yield; a similar configuration was adapted in Ref. [28]. All utilization scenarios are scrutinized with the pinch analysis method to establish the auxiliary utility demands and evaluate the overall thermal performance. Excess heat

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